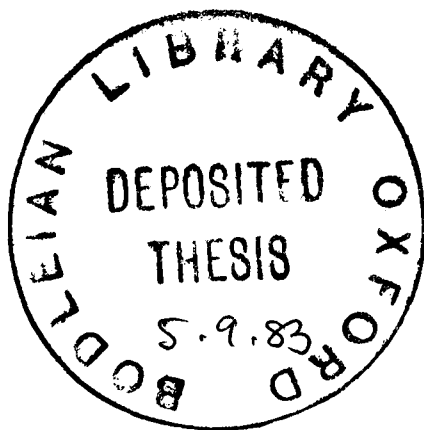


Studies
of
Rare Gas Halide Lasers

by
Daniel Christopher Hogan
Linacre College
Oxford

A thesis submitted for the degree of
Doctor of Philosophy
at the University of Oxford



The Clarendon Laboratory,
Oxford,
Hilary Term, 1983.

To my Mother and Father

and

To my wife, Myszka

who had the good sense to go to Paris while I was writing this thesis.

Studies of rare-gas halide lasers

by

D.C.Hogan

Linacre College, Oxford.

A thesis submitted for the degree of Doctor of Philosophy
at the University of Oxford, Hilary Term, 1983.

Abstract

This thesis presents the results of a study of the mechanisms responsible for limiting the laser pulse duration obtainable in xenon chloride lasers which are excited by UV-preionized, self-sustained gas discharges.

The xenon chloride laser system, the principal emission band of which is centred around 308 nm, belongs to the class of high pressure gas lasers known as 'rare-gas halides' (RGH). RGH lasers are now well known for their high peak power output at a number of wavelengths from 193 nm to 353 nm in the ultraviolet region of the spectrum. To date, however, they have only been operated in the pulsed mode with laser pulse durations of ~1000 ns for devices employing electron beam excitation and ~30 ns for devices employing transverse discharge excitation. There is no a priori kinetic limitation which prevents RGH lasers from operating in the CW mode, and an attempt to extend the duration of the laser pulse would enable the quality of laser output to be improved.

The laser pulse duration of a discharge excited XeCl^* laser was extended by about one order of magnitude - to 270 ns FWHM - by the use of a distributed resistance electrode to stabilize the discharge. The typical gas mixture used in the laser was ~2 atm of Ne (buffer gas), ~25 mbar of Xe, and 2.5 mbar of HCl. However, the laser pulse duration obtained was considerably shorter than the 500 ns duration, 2000 A peak current, discharge excitation pulse.

The cause of this difference between the duration of the laser output pulse and the discharge current pulse was found by carrying out a comprehensive parametric study of the laser, combined with a detailed spectroscopic analysis and the results of a semi-empirical computer model. Two interrelated factors were identified as being responsible for the short duration of the laser output: namely, a temporal collapse of the discharge volume and a spatially non-uniform depletion of the HCl within this volume. The experimental results presented here contradict an earlier theory which ascribed the onset of discharge instabilities in RGH lasers to step-wise ionization of the minority rare gas atoms, and which attributed stability enhancement properties to the electronegative halogen gases used in RGH lasers.

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Glossary of Abbreviations, Symbols, Acronyms, and Terms

NON SI UNITS

The SI system of units is used throughout, except for the following:

atm	-	atmosphere = $1.0125 \times 10^5 \text{ Nm}^{-2}$
mbar	-	1010.25 mbar = 1 atmosphere
psi	-	Pound per Square Inch = $6.895 \times 10^3 \text{ Nm}^{-2}$
psig	-	Pounds per Square Inch Gauge above atmospheric pressure
torr	-	760 torr = 1 atmosphere
Td	-	Townsend, the unit of $E/N = 10^{-17} \text{ V-cm}^2$
$\text{\AA}$	-	Angstrom = 10^{-10} m
cm^{-1}	-	wavenumbers
cm^3	-	10^{-6} m^3
eV	-	electron volt = $1.602 \times 10^{-19} \text{ J}$

SYMBOLS

ϵ	-	mean electron energy
ν_i	-	an ionization rate
ν	-	frequency
σ	-	a cross section
τ	-	a lifetime
λ	-	a wavelength
$\Delta\tau$	-	a time interval
Φ	-	a stimulated photon
γ	-	transient loss
Φ_{output}	-	output photon flux
Φ_{ic}	-	intra-cavity photon flux
σ_ν	-	absorption cross section
$\delta\nu$	-	linewidth
Ψ	-	gas relaxation time
τ_D	-	lifetime against destruction
A_a	-	Avagadro's number
A_{21}	-	a transition probability
c	-	the speed of light
c_v	-	molar specific heat
e	-	the electronic charge
E/p	-	average electric field divided by pressure in torr
E/N	-	average electric field divided by gas number density
f_{21}	-	an oscillator strength
h	-	Planck's constant
I	-	a current
k_ν	-	absorption coefficient
ℓ_g	-	gain length
ℓ_c	-	cavity length
m_e	-	the electron rest mass
n_e	-	electron density
ΔQ	-	specific input energy
R_b	-	ballast resistance
t_{LP}	-	laser pulse duration (FWHM)
t_{DT}	-	double transit time of a cable PFN
Δt	-	a time interval
ΔT	-	change in temperature
T_e	-	electron temperature
$\frac{v}{V}\text{drift}$	-	electron drift velocity
V	-	a voltage
XeI	-	a neutral xenon atom
XeII^*	-	a xenon ion
X^*	-	an excited state of a species 'X'
$[X]$	-	the population or number density of a species 'X'
$\frac{d}{dt}(X)_{\text{cr}}$	-	the rate of creation of a species 'X'

ACRONYMS

FWHM - Full Width at Half Maximum
 CW - Continuous Wave
 H.V. - High Voltage
 IR - InfraRed
 LPD - Long Pulse Duration laser - the device described in chapter 4
 OMA - Optical Multichannel Analyser
 PFN - Pulse Forming Network
 PCB - Printed Circuit Board
 RGH - Rare Gas Halide
 SPC - Static Parametric Contraction
 SRS - Stimatewd Raman Scattering
 TVC - Temporal Volume Constriction
 UV - UltraViolet
 XUV - eXtreme UltraViolet

TERMINOLOGY

Active volume -	the region in which the discharge exists at a point in time
Ballast Resistance -	the equivalent series resistance presented to the discharge circuit by a distributed resistance electrode
e-beam -	electron beam
Ion channel -	three body ion-ion recombination of Xe^+ with Cl^- to form XeCl^*
Neutral or excited channel -	XeCl^* formation via reactions involving HCl and a xenon neutral excited state atom
Pseudo-resistance -	since a discharge is a non-ohmic device the term 'resistance' is not applicable. A pseudo-resistance is the instantaneous ratio of voltage to current.
Shot -	a discharge event

C H A P T E R O N E

Introduction

1.1 THE CONTEXT OF THE PRESENT WORK:

This thesis presents the results of a study of the physical mechanisms responsible for limiting the laser pulse duration obtainable from transverse discharge excited xenon chloride (XeCl^*) lasers.

The XeCl^* laser system belongs to a class of gas lasers known as 'rare-gas halides' (RGH) which are commonly grouped under the more general title of 'excimer' lasers. The word excimer is a contraction of EXCited diMER. The heteronuclear RGH molecule is actually an excited complex; the feature they share with the true excimer lasers (eg. Xe_2) is that the laser operates on a transition from an electronically excited molecular state to a dissociative or very weakly bound ground state. This similarity has encouraged the use of the term 'excimer laser' to describe both true excimers and the RGH 'exciplexes'.

The first report of measurements of the fluorescence emitted when excited rare-gas halide molecules undergo spontaneous transitions to their dissociative ground state was made by Golde and Thrush[1974]. Very shortly thereafter, Velazco and Setzer[1975] suggested that the rare gas halide excimers might well be capable of producing laser action in the ultraviolet. Later that same year laser action on xenon bromide at 282 nm was reported by Brau and Ewing[1975]. From that time a considerable effort has been mounted on a worldwide scale to develop these lasers to their full potential. The reason for this immediate and widespread interest in RGH lasers was their promised (and now proven) potential for efficient generation of coherent radiation at very high peak powers at a number of wavelengths from 193nm to 353nm. This broad spectral region in the ultraviolet had previously been almost devoid of high power laser transitions.

Only the nitrogen (C-state 337nm) laser system and frequency doubled visible laser radiation provided output there. A comprehensive review of early developments in the field can be found in the monograph edited by Rhodes[1979] or the shorter review by Shaw[1979].

As is usual after the first discovery of a new laser system, it was soon apparent that there were going to be some difficulties in achieving the high performance capabilities which were initially forecast for RGH lasers. Specifically these lasers had (and still have) only been operated as pulsed devices - a feature which is responsible for their generally poor performance in terms of beam divergence and linewidth (see section 1.3 below). Laser pulse durations were (and still are) limited to values less than $\sim 1 \mu\text{s}$ for devices employing electron beam (e-beam) excitation and $\sim 20 \text{ ns}$ for the more generally useful transverse discharge-excited devices.

There is no obvious a priori kinetic limitation on RGH laser pulse duration as is the case in, for example, the neutral N_2 laser (C-B transition) at 337.1 nm; here, the lower laser level is metastable and cannot be cleared sufficiently rapidly to allow laser oscillation for more than $\sim 30 \text{ ns}$ - CW operation is completely out of the question (see e.g. Willett[1974], p.342).

Two considerations motivated the studies described here. Firstly, there were clearly considerable technological advantages to be gained by increasing the laser pulse duration of the RGH system. Secondly, there was the more academic question of whether the limited pulse durations reported for the various RGH lasers were merely a result of the difficulty of maintaining stable excitation for a long period of time or due to some, then unaccounted for, kinetic mechanism. Although it then seemed that CW operation of RGH lasers was theoretically possible, the large value of the minimum specific input power ($\sim 10\text{--}100 \text{ kW/cm}^3$; see below) required to maintain laser action was enough to stifle any hope of achieving fully CW operation. Even if a sufficiently large power supply could be found, the problem of disposing of the energy converted to heat ($\sim 99\%$ of total energy input) would be far from easy with state-of-the-art technology.

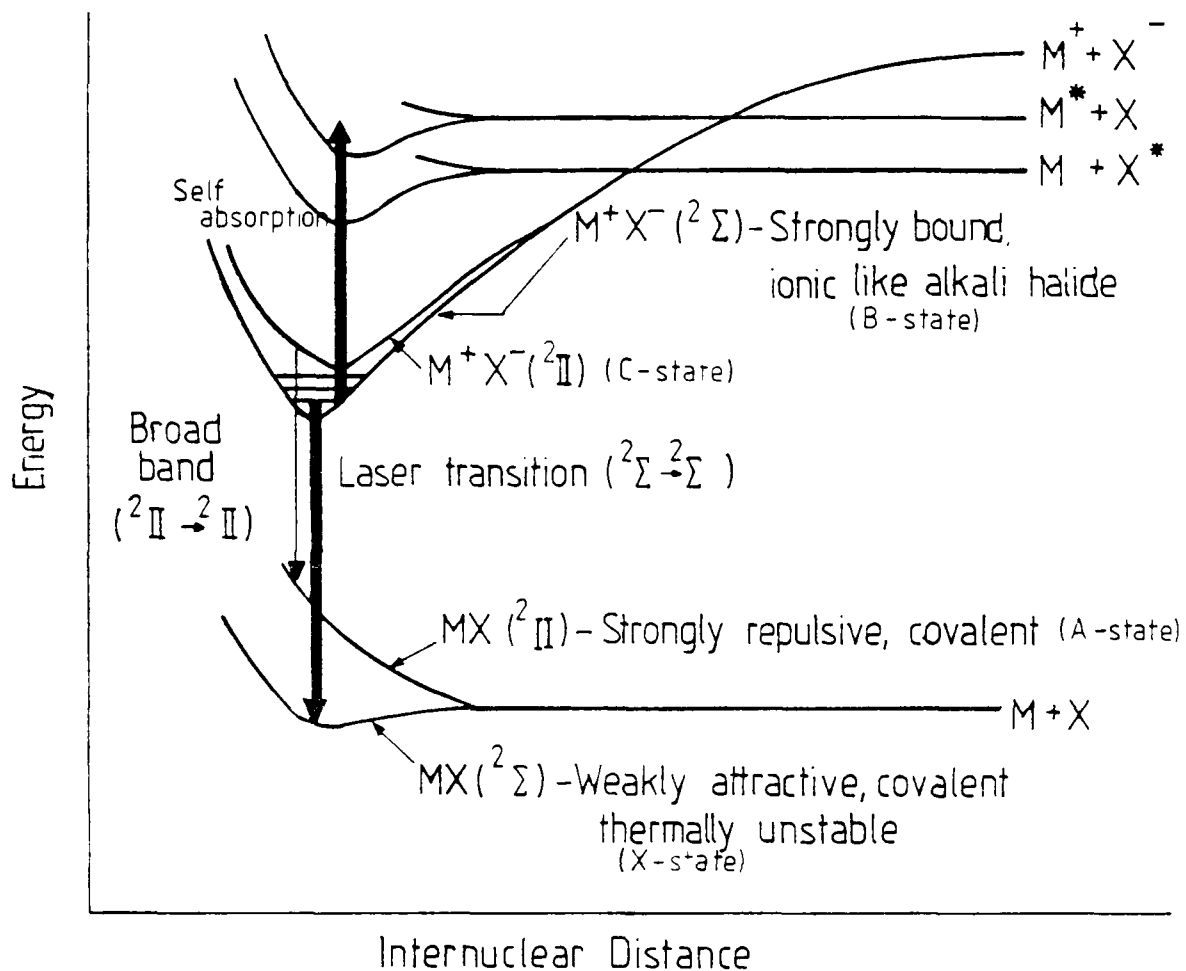


FIGURE 1.1: Generalised rare gas (M)–halide (X) energy level diagram (after Rhodes[1979]).

1.2 RGH LASERS - A BRIEF REVIEW:

1.2.1 RGH kinetics:

There have already been many reviews published in the literature about RGH lasers - including 'detailed' descriptions of the kinetic processes. Consequently, only a very brief description of the RGH system will be given here and that will be with particular reference to XeCl^{*}. The reader is referred either to Rhodes[1979] or Kearsley[1980] for a more thorough description of both 'true' excimer systems (e.g. Xe₂) and the RGH system.

(a) RGH molecular spectroscopy:

Figure 1.1 presents a generalised energy level diagram for RGH molecules. In the spectrum of all the RGH molecules yet observed in fluorescence or as lasers (see table 1.1), emission resulting from a transition from the 'B' state to the 'X' state produces the most intense spectral feature. There have been reports, however, of relatively weak laser action on the 'C' - 'A' transition of XeF at ~490 nm (see e.g.Fisher et al.[1979a] or Bischel et al.[1979]).

In all known RGH molecules, the ground state is covalently bonded and correlates to ground state ¹S rare gas and ²P halogen atoms at infinite inter-nuclear separation. The ground state manifold has two states of which the ²Σ has the lowest energy. This state is called the 'X' state and is generally

Table 1.1
Observed emission from rare gas halides

<u>Rare gas</u> <u>Halogen</u>	He	Ne	Ar	Kr	Xe
F	-	(F) 108	(L) 193	(L) 249	(L) 353
Cl	-	-	(F [*]) 175	(L) 222	(L) 308
Br	-	-	-	(F) 206	(L) 282
I	-	-	-	-	(F) 253

Wavelength region of B - X emission band indicated in nanometres
(L)= lasing reported; (F)= fluorescence reported;

* lasing was reported by Waynant[1977] but no further reports have since appeared.

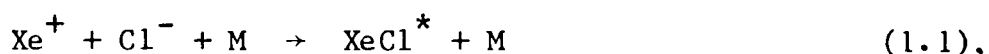
repulsive or very weakly bound. In the case of XeCl^* the binding energy is of the order of 250 cm^{-1} and so it is thermally unstable at room temperatures. On the other hand, the XeF^* ground state is actually quite strongly bound ($\sim 1065 \text{ cm}^{-1}$) and improvements in laser performance have been observed by Hsia et al.[1979] when the temperature of the bulk gas is raised. These authors attribute the improvement mainly to increased vibrational relaxation of the upper level but believe that increased thermal dissociation of the lower level also plays some part. The $^2\Pi$ state of the ground state manifold is always repulsive as indicated. Since it forms the first state above the ground state it is referred to as the 'A' state.

The upper laser level is an ionically bound, charge-transfer state correlating to the ^2P rare gas positive ion and ^1S halogen negative ion at infinite internuclear separation. The structure of the ionic upper level manifold is similar to that of the ground state manifold with the $^2\Sigma$ state lying lowest. By convention this is called the 'B' state. The $^2\Pi$ state gives rise to the 'C' (lowest energy) and 'D' states. As is discussed by Brau[1979], the rare gas halides (in the B state) bear a marked resemblance to the ground state of the halide of the alkali metal occupying the neighbouring position in the periodic table (e.g. KrF correlates to RbF) in terms of bonding and of mode of formation. Useful predictions about reaction kinetics of the rare gas halides can be made by arguing from these analogies.

The emission spectrum of each of the electronic transitions of the rare gas halides is either a quasi-continuum or comprises several broad vibrational bands. The gain profile is consequently very broad and limited tunability ($\sim 1 \text{ nm}$) has been reported for most RGH lasers (see e.g. Caro et al.[1982]).

(b) RGH molecule formation reactions:

Two reaction channels leading to the formation of RGH excited molecules have received the most attention. The first is the three-body ion-ion recombination reaction which in the case of XeCl^* (a star will be used to denote an excited state species throughout this thesis) is:



where M is a third body (in the gas mixtures of interest here M = He or Ne). The reaction described by eq.1.1 will be referred to throughout this thesis as the 'ion channel'. It now seems likely that three-body ion-ion recombination dominates the production of the upper laser level molecules in all the RGH lasers. This would account for the fact that all RGH lasers produce their optimum output in high pressure (>2 atm) gas mixtures.[†] In discharge excited systems the gases usually employed to provide high pressure buffering of the rare gas halogen mixture are neon and helium. The theory of three-body ion-ion recombination stems from the original work by Langevin (for pressures above ~1 atm) and that by Thompson (for pressures below ~1 atm: see e.g.von Engel[1965] for a discussion). A more general theory recently published by Flannery[1978a,b,c] is valid in the high as well as the low pressure regime. Rate coefficients for the ion channel have not been determined experimentally for any of the RGH systems. Most computer models of RGH lasers rely on predictions from Flannery's formula. Fortunately however (see below and in chapter 6), this is not a serious limitation since it would seem that the ion channel production rate is virtually independent of the rate coefficient for the reaction as the coefficient is varied over ~3 orders of magnitude.

The other major formation channel for XeCl^* production,



in which an excited state of neutral xenon (Xe^*) reacts with the halogen donor species (usually HCl) to form the B state molecule, does not require the presence of a third body since any excess energy is carried off by the hydrogen atom which is a by-product. The reaction described by eq.1.2, which will be referred to here as the 'neutral channel', was shown by Kearsley[1980] in his experiments with a short pulse duration discharge excited XeCl^* to contribute (at most) 15% to the total XeCl^* (B state) production. Results of experiments on a long pulse duration system which are presented later in this thesis support Kearsley's findings. It is worth noting that the formation of XeCl^* via this

[†]But not for the obvious reason. See discussion in chapters 6 and 7.

channel when the xenon excited state in question is one of those of the lowest configuration requires the presence of vibrationally excited HCl: the reaction of one of these xenon excited states with ground state HCl is slightly endothermic for XeCl^* production and results merely in the dissociation of HCl into H and Cl.

That HCl rather than Cl_2 is the preferred and most widely used halogen donor for XeCl is a result of the general experience that the XeCl^* system shows better performance when HCl is the halogen donor. One factor responsible for the superiority of HCl is that, unlike Cl_2 , it does not have significant absorption at the laser wavelength of 308 nm. Some workers have used CCl_4 as a halogen donor but have found that it also does not perform as well as HCl. This is probably due to the fact that the rate of attachment of electrons to CCl_4 - to form the halogen precursor species, Cl^- , for the ion channel reaction - is much slower than in HCl. It was also concluded by Kearsley[1980] that the rate of formation of Cl^- by dissociative attachment of electrons to HCl,



is the rate limiting step in the production of the XeCl B state molecule. That conclusion is confirmed in this thesis.

(c) Early thoughts on pulse length limitations:

One mechanism which could be foreseen as a barrier to obtaining long pulse durations (and which was evident at an early stage(Brau[1979])) was depletion of the halogen donor species. This mechanism is expected to occur on time scales of order 10 μs . The rate of formation of negative halogen ions from the neutral atom is very slow in comparison to their rate of formation by dissociative electron attachment to halogen bearing molecular species. Reformation of the molecular species from the by-products after dissociation of the RGH molecules is so slow (see comments in chapter 6) that it can be totally disregarded on the time scales normally associated with RGH excitation. Only a very limited ($\sim 0.1\%$ in the gas mixture) concentration of halogen donor can be tolerated in state-of-the-art lasers because of a combination of increased passive optical

absorption of the laser radiation inside the active medium (rare gas fluorides), quenching of the upper laser level (Fisher et al.[1979b]), and increased discharge instability (Hogan et al.[1983]). Significant 'burn-up' of the halogen fuel - with consequential laser pulse termination - is therefore a very real possibility.

Further discussion of XeCl kinetics will be deferred until chapter 6. The reader's attention is drawn, however, to the comprehensive compilation of data relevant to RGH kinetics by McDaniel et al.[1977, 1980, 1982]. If kinetic rates are quoted in the body of this thesis without direct reference, the original source will be found in McDaniel's compilation.

1.2.2 RGH laser excitation methods:

The most important consideration in the excitation of RGH lasers is the requirement for a very large specific power input to the gas volume. This need, which is common to all broadband lasers, is a consequence of the comparatively low values of their gain cross sections ($2.5 \times 10^{-16} \text{cm}^2$ for XeCl^* : see e.g. Levin[1981]). RGH lasers therefore require very high populations in order to produce a reasonable optical gain coefficient. Since the upper level lifetime, τ , of all RGHs is very short ($\sim 10 \text{ ns}$ for XeCl^*), a large continuous energy input is implied if lasing is to be achieved.

Take, for example, an XeCl^* laser with a gain length, ℓ_g , of 50 cm and an optical cavity which transmits a fraction, 'f', of 4% of the radiation out of the cavity in one double transit. The minimum excitation rate, χ , required to overcome this loss due to output is

$$\chi = f / (2\ell_g \times \sigma \times \tau) = 0.04 \times (2 \times 50 \times 2.5 \times 10^{-16} \times 10^{-8})^{-1} = 1.6 \times 10^{20} \text{cm}^{-3} \text{s}^{-1}.$$

where σ is the gain cross-section. Assume now that the overall efficiency, Ξ , is $\sim 1\%$, and E_{in} the minimum energy investment to create an XeCl^* B state molecule is $\sim 12 \text{ eV}$ ($1.9 \times 10^{-18} \text{ J}$ - to ionize Xe). The calculated threshold input power is

$$P_{\text{threshold}} = \frac{1}{\Xi} \chi E_{\text{in}} = 100 \times 1.6 \times 10^{20} \times 1.9 \times 10^{-18} = 30 \text{ kWcm}^{-3}.$$

This is an extremely generous underestimate because it ignores losses due to transient absorptions and processes leading to quenching of the upper laser

level. To date, there have been no reports in the literature of lasing being achieved in the XeCl^* system for specific power input levels below $\sim 100 \text{ kW cm}^{-3}$.

The two pumping schemes which have been most commonly employed for RGH lasers are electron beam excitation and transverse discharge excitation of the high pressure gas mixture. Both of these methods rely on electron impact to excite or to ionize the gas mixture - although the method of production of electrons differs. Recently, radio-frequency (RF) and microwave excitation techniques (Young[1982], Wisoff[1982], Waynant[1983]) have been developed. The output power of these devices is disappointing low (500W peak), and it does not appear that scaling to larger volumes is feasible.

(a) Electron-beam excitation:

The first reports of laser action from RGH systems were those by Brau and Ewing[1975], Ault et al.[1975], and Tisone et al.[1975]. In all three cases a beam of high energy electrons was used to excite the laser gas mixture. Very briefly, in this pumping scheme a beam of very high energy electrons (100-1000 keV) is injected into the high pressure gas mixture which consists mainly of a buffer gas of high atomic number (e.g. argon). This beam of electrons causes many ionization events along its path and secondary electrons go on to excite or ionize further atoms. The high density buffer gas absorbs most of the energy. The energy is then channelled through a number of highly efficient reactions (charge exchange etc.) to form the laser species precursors.

The major advantage of e-beam excitation is the fact that power input is independent of the reaction kinetics within the gas volume; the problems of maintaining the volume of gas contributing to upper laser level formation (the 'active volume') which are paramount in discharge excited systems are non-existent in pure e-beam pumped devices. The longest laser pulse durations for all the RGH systems have been achieved with e-beam devices. For example, a 1 μs FWHM laser pulse duration from the XeCl^* system has been reported by Moody et al.[1981].

At the time of writing, the largest peak power reported in the open literature for a rare-gas halide is 4 GW (160 J in a 40 ns pulse) from KrF^* by

Edwards et al.[1983].[†] Their device is a large volume e-beam excited laser. Development is underway at the Lawrence Livermore National Laboratory in the USA to produce an e-beam device capable of producing a 1-2 kJ laser pulse with a duration of the order of tens of nanoseconds.

Although very large single pulse energies can be obtained from e-beam excited devices, they do have shortcomings. Apart from the basic considerations of their greater cost and size which make them unattractive for general laboratory use, they are capable of only comparatively low pulse repetition frequencies and pose a significant radiation hazard.[‡] Furthermore, as is discussed by Kearsley[1980], the quantum efficiency for XeCl^* production is lower in e-beam pumped devices than those employing discharge excitation - the overall efficiency of the discharge excited laser can therefore be expected to be higher with consequent savings in operating costs.

(b) Discharge excitation:

From the point of view of producing a generally useful laboratory tool, discharge excitation is a far more attractive method of pumping high pressure gas lasers. The fact that energy is supplied by discharge within the gas volume - rather than being supplied from some external source and therefore decoupled from the active volume - introduces major difficulties however. The problems associated with maintaining uniform excitation over a large active volume for a sufficiently long period to achieve lasing are by no means trivial. Although the large volume discharge technology for discharge excited CO_2 lasers was well advanced by the time the RGH system was discovered, it was a year before the first report of the successful operation of a discharge pumped RGH laser (Burnham[1976]). The method used then (and which continues to be employed in commercially produced discharge excited RGH lasers) was to run very short duration (~20 ns), high power deposition discharges in the gas volume in the hope that the maximum laser pulse energy would be extracted before the onset of discharge instabilities.

[†] Not counting amplification by a RGH of picosecond pulses from other lasers.

[‡] Because of intense X-ray production when the e-beam strikes the gas vessel.

The design criteria for discharge excited RGH lasers are far more exacting than for CO₂ systems for two reasons: the presence of extremely 'agressive' electron-attaching halogen donor gases in the laser gas mixture; the higher specific input powers required to operate the RGH laser in comparison to the CO₂ laser. The propensity for discharges containing electronegative gases to become unstable in very short times has been known for many years and has been the subject of intense study in the low-pressure (below 500 mbar) regime (see e.g. Emeleus and Wolsey[1970]). A discussion of discharge stability and design criteria for discharge-excited RGH lasers will be presented in chapter 2.

1.3 THE NEED FOR LONG PULSE DURATION RGH LASERS:

The output radiation from short pulse duration discharge excited RGH lasers, of the type normally produced commercially, typically has a divergence some two orders of magnitude greater than the ultimate due to diffraction from the laser aperture. The linewidth, $\delta\nu$, of the radiation is normally some 500 times greater than that which might be predicted by the Fourier transform of the laser pulse duration, t_{LP} ,

$$\delta\nu \approx \frac{1}{2\pi t_{LP}} \quad (1.4),$$

The poor quality of this output - which could be said to be not much better than a high power flashlamp - is a result of the high gains, large volumes, and short pulse durations typical of the discharge excited laser. In most cases the ~20ns pulse duration is only sufficient to allow the laser radiation to circulate two or three times within any optical cavity placed around the discharge. This is far too small a number of round trips for a well defined spatial mode structure to develop in the stable optical resonators (see e.g. Yariv[1971] p.64) normally employed with lasers.

Although some improvement in beam divergence (for a stable resonator configuration) might be expected if the pulse duration were extended, it was concluded by Caro[1981](p.13) that the combination of high gain and large volumes in RGH systems makes unstable resonators a better choice even for relatively long laser pulse duration systems. Diffraction limited output from

short pulse duration RGH lasers has been achieved by both Caro[1981] and Smith[1982] using unstable resonator cavities.

Output of narrower linewidth than that which can presently be achieved with tuned short pulse RGH lasers (for applications such as high resolution spectroscopy) would result from systems with longer pulse durations. Tuning elements within a laser cavity introduce high loss for wavelengths which are to be suppressed. It is clear that an increase in laser pulse duration with the consequent increase in the number of cavity round trips will enhance oscillation at the desired wavelength. The possibility of encouraging well developed longitudinal cavity modes by increasing the laser pulse duration brings into prospect passive mode-locking of RGH lasers. The ultrashort, high-power UV pulses thus produced would find application in many fields - not least of which would be experiments into laser induced fusion. Corkum and Taylor[1982] have recently demonstrated the use of an XeCl^* laser to amplify a frequency-doubled 2ps pulse from a CW mode-locked dye laser to a peak power of ~ 20 GW.

One of the major uses for commercial RGH lasers has been as sources of high power radiation for pumping dye lasers. Corney et al.[1979] have used a short pulse duration XeCl^* laser to obtain a linewidth close to the Fourier limit from a dye laser. Longer pulse durations from the 'pump' laser would enable a further reduction in linewidth. There are applications for good beam quality, narrow linewidth RGH lasers in fields such as long range communications, photochemistry (plutonium isotope enrichment for example), photolithography, and non-linear generation of XUV radiation. The reader is referred to the work of Caro[1981] for a description of the application of RGH lasers to non-linear optics and that of Smith[1982] for a detailed discussion of the efficacy of using RGH lasers to generate other wavelength radiations by Stimulated Raman Scattering (SRS).

1.4 THE OBJECTIVES OF THIS STUDY:

Given the need for long pulse duration RGH lasers, the object of this study was to extend - as far as was possible - the laser pulse duration from a

device employing transverse discharge excitation.

For a number of reasons the XeCl^* (308 nm) system was the only one which was investigated. In particular, it was realised at a very early stage that the device would have to be pumped at comparatively low power levels to maximise the laser pulse duration. The reduced gain of the system would necessitate the use of an optical cavity with mirror reflectivities far greater than those then in use (short pulse duration, high gain, systems operate successfully with output coupler reflectivities as low as 10%). At the time the only system for which high reflectivity optics were readily available was XeCl^* . Since the technology of manufacturing mirrors in the wavelength region of interest for RGH lasers is relatively new, a summary of the methods used to manufacture the mirrors used here is included as appendix 1. Another reason that only XeCl^* was studied was that it was known that the useful lifetime of a single gas fill was longer in XeCl^* than in the rare-gas fluorides. This was an attractive property if experimental measurements of any significant duration were to be carried out on the laser. Also, it was feared that changing the gas mixture between the fluorides and chlorides would have undesirable gas contamination effects as a result of the halogen donor gas reacting with the walls of the gas containment vessel.

1.5 OUTLINE OF THE WORK TO BE PRESENTED IN THIS THESIS:

Since the first part of the investigation was to design and construct a long pulse duration RGH laser, it is relevant to outline the state of knowledge about discharge excitation and discharge stability in these systems at the time the study was commenced. Consequently, the next chapter reviews the general problem of discharge stability in RGH lasers and the various stability enhancement methods which have been used in high pressure gas discharges.

The third chapter describes the design and performance of a prototype long pulse duration XeCl^* laser. This laser was used to prove the efficacy of resistive stabilization of the discharge in extending laser pulse durations.

In chapter 4, the design and performance of a resistively stabilised device

which was the focus of the rest of the investigation is discussed in detail. The chapter includes discussion of the effect that changing parameters such as the gas mixture composition, the resistance of the ballasted electrode, and the level of power deposition into the discharge, had on the laser output. Results of an investigation of the spatial uniformity of the discharge as a function of time and as a function of the concentration of the halogen donor gas, HCl, are also presented at this stage.

Although the laser described in chapter 4 produced the longest laser pulse durations (270ns FWHM) yet reported for any self-sustained discharge excited RGH laser, the laser output pulse was observed to terminate well before the end of the electrical excitation pulse. The investigation and eventual identification of the cause of this premature termination forms the basis of chapters 5 and 6.

In chapter 5 results of an extensive spectroscopic investigation of the laser discharge are presented. This investigation included:

1. Time-resolved measurements of transient absorptions at various UV wavelengths.
2. Time-resolved emission spectra of XeI and XeII transitions under various discharge conditions.
3. The time-resolved measurement of the XeI resonance and metastable populations to verify Kearsley's conclusion about the dominance of the ion channel (eq.1.1) in the production of XeCl^* B state molecules.

Chapter 6 approaches the problem of identification of the laser termination mechanism from a more theoretical standpoint. It presents a 'semi-empirical' numerical model of the kinetics of the long pulse laser. This model was developed to provide a more rigorous insight into the gas chemistry than could be obtained by performing 'back of the envelope' calculations.

The final chapter contains a summary of the achievements of the study and a discussion of the pulse termination mechanism. It also includes proposals for further work, and a discussion of the prospects for further extensions of the laser pulse duration obtainable from discharge excited RGH lasers.

C H A P T E R T W O

Discharge stability and other problems in the discharge-excitation of RGH lasers

2.1 INTRODUCTION:

The discovery and subsequent development of electrically excited gas lasers have proven to be major stimuli to gas discharge research in recent years. However, nearly all of the fundamental research into gas discharges has been carried out in the low pressure regime (less than say 100 mbar) and most of this on single species gas mixtures. The reason for this lies in the rapid increase in the complexity of systems which operate at, say, slightly higher pressures or with one additional species. Consequently, although the onset of discharge instabilities in high power gas discharge lasers is probably the single most important problem to be solved to obtain improvements in laser performance characteristics, and in spite of the fact that these instabilities have been studied by countless investigators for many years, with few exceptions, the theoretical studies of the phenomena have addressed mechanisms which are difficult to pin down experimentally and have invoked explanations which are frequently **contradictory**. The present author therefore makes no apology for the fact that the discussion which follows will be qualitative and based to some degree on 'conventional wisdom'.

The discovery of the CO_2 laser and the realisation of the fact that its performance could be enhanced by operating it at comparatively high pressures (~1 atmosphere), led to the development of transverse discharge excitation by Beaulieu[1970] and others (the reader is referred to Bekefi[1976] for a review by DeMaria of CO_2 laser excitation). Longitudinal discharge excitation (electric field parallel to optic axis) is useful only for low pressure

discharge applications. This is because of the high voltages required to achieve breakdown and the propensity for the discharge, once initiated, not to fill the volume but to contract down to a 'snaking' arc. Uniform field breakdown in most gases follows Paschen's Law, in which the voltage, V_b , required to achieve breakdown is expressed as a function of the product ' $N \times d$ ' where ' N ' is the gas number density and ' d ' the length of the discharge gap.[†] For example, the static field breakdown voltage of argon (which is sometimes used as a buffer gas in discharge RGH lasers) is ~6kV for a 1 cm gap at atmospheric pressure (see eg. Blair[1978]). Although the breakdown behaviour of argon does deviate from similarity rules at high breakdown voltages, it is clear that for a 1 m discharge length (typical of longitudinal discharge devices) the voltage which must be applied to achieve breakdown is ~600 kV.

The use of a transverse discharge ('transverse' because the electric field is transverse to the optic axis) alleviates the problems associated with requiring extremely high voltages to achieve breakdown. In this case the discharge gap is usually 1-3cm which means that voltages some two orders of magnitude less than longitudinal devices are required. The optical gain length for lasing and the volume of the discharge are increased by encouraging the discharge (by various means) to operate along the entire length (10-100cm say) of the transverse electrodes.

2.2 DISCHARGE STABILITY:

2.2.1 General:

Many of the problems with the onset of discharge instabilities (spatial non-uniformities) are closely linked with those of initiating a uniform discharge between the transverse electrodes in the first place: if a uniform

[†]Given that gas breakdown characteristics are dependent on three variables - the applied voltage, the gas number density, and the discharge gap - the quantity E/N , which is electric field at some point divided by the gas number density and which thus combines all three, is quite useful. Once breakdown has been achieved, the value of E/N , and therefore the mean electron energy, required to sustain a discharge at a particular current density is determined by the nature of the electron production and loss processes.

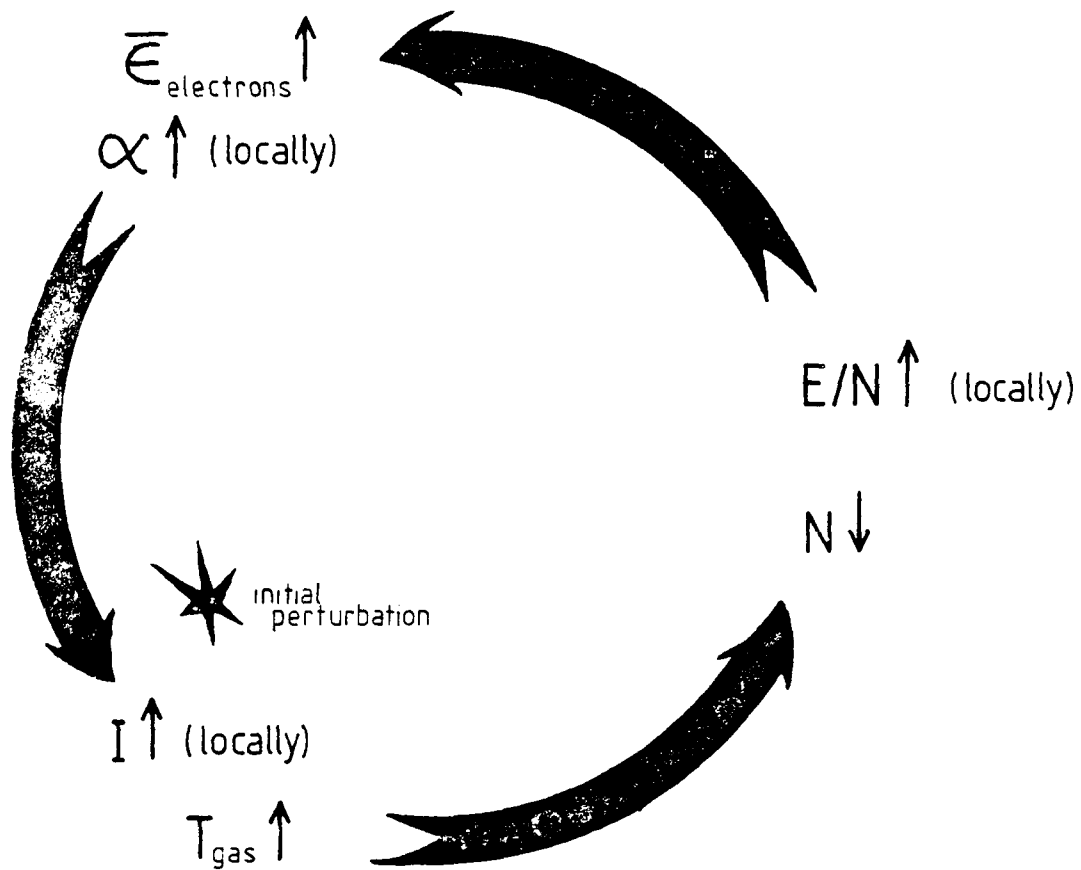


FIGURE 2.1: Qualitative description of the growth of a thermal discharge instability. $\bar{E}_{\text{electrons}}$ is the mean electron energy, α is the primary ionization coefficient, and 'I' is the current.

discharge is not initiated, it will invariably collapse into a number of bright, high current arcs in a very short time. Even when a spatially uniform discharge is initiated it is often difficult to maintain that uniformity. The time taken for the formation of gross arcs is variable, from nanoseconds to milliseconds and beyond, and is very much dependent on the composition of the gas mixture.

Figure 2.1 illustrates, very qualitatively, how a thermal instability can lead to a transition from the type of large volume glow discharge ideally suited to laser excitation to a small volume, high current arc. Initially, a uniform glow discharge is assumed to be established. At some point in time, a local increase in electron density may occur (by whatever means). The local gas temperature consequently increases above that of the surrounding region and the local density decreases. As a result of this decrease in density, E/N rises locally. The rate of production of electrons, being dependent on the mean electron energy and therefore E/N , also rises. The result of this is an even larger difference between the electron density in that localised region and that in the rest of the discharge volume. The greater current flow there results in further local heating of the gas, further local increases in the E/N value relative to that prevailing in the surrounding gas volume, and a positive feedback loop is established. All the while, more and more of the total (power supply impedance limited) current will begin to flow in this localised region. Since the gas density is lower here, the average electric field required to sustain this total current flow will be falling. That is, the voltage at all points on the electrode will decrease and, as a result, the rate of creation of electrons will fall below their rate of loss (loss due to processes such as recombination) in the volume outside the perturbed region and the discharge will cease to exist there. It is clear from this discussion that, given an initially uniform discharge, spatial non-uniformities do take time to develop. It must be emphasised that this model is oversimplified. It does, however, introduce the ideas of positive feedback and current runaway in a discharge - both of which are applicable to other instability mechanisms.

One solution to the problem, which was implemented very early in the

development of CO_2 lasers, is to apply only a very short electrical excitation pulse to the gas volume and to extract the laser energy before the instabilities have had time to grow. This is the philosophy most commonly applied in the discharge excitation of RGH lasers. It is the general experience, however, that it is far more difficult to initiate and sustain a spatially uniform RGH discharge than a CO_2 discharge. Consequently, the term 'short excitation pulse' in RGH lasers usually means a pulse lasting not more than, say, 50 ns foot-to-foot, whereas for a CO_2 laser it would be, say, 1-2 μs .

2.2.2 Theories of discharge instability in RGH lasers:

The most striking difference between a RGH laser discharge and a CO_2 laser discharge is the extraordinary measures which must be taken to prevent the formation of hard arcs in the RGH laser compared with the relative ease with which this can be done in CO_2 systems.

A simple visual observation of a short pulse RGH laser discharge reveals a dramatic increase in discharge non-uniformity as the partial pressure of the halogen donor species is increased - whether it is HCl , F_2 or one of the other species commonly employed for this purpose. A discharge containing just the mixture of rare gases required for RGH laser operation (i.e. no halogen present), which is initiated under identical conditions to a full laser mixture, invariably appears to be a totally uniform glow.

Many studies have been made of the stability of low pressure discharges containing electronegative gases reported in the literature (see for example Emeleus and Wolsey[1967], Edgley and von Engel[1980], Nobato and Kando[1979]). All these studies comment on the far stronger constriction which is observed in electron attaching gas discharges than is seen for the pure rare gases or of mixtures of the rare gases. Douglas-Hamilton and Mani[1973] have reported the occurrence of an electron attachment plasma instability in high pressure CO_2 laser discharges.

It is somewhat surprising, therefore, that the only theory dealing specifically with the onset of discharge instabilities in RGH discharges yet published (Daugherty, Mangano and Jacob[1976]), states categorically that the

only reason such discharges are stable is because the electron-attaching halogen species is present. The implication is that the discharge of a gas mixture containing a mixture of the rare gases and no halogen is totally unstable.[†]

The experimental results which will be presented in this thesis indicate clearly that Daugherty et al.'s stability criterion has no basis in reality. Webb[1976a] has shown that Daugherty et al.'s theory has obvious theoretical flaws. Since Webb's comments were not published in the open literature it is worthwhile to mention them briefly here.

The final conclusion of the theory of Daugherty et al., which identifies an instability due to two-step ionization of the rare gas, is that stability in the RGH (specifically KrF^{*}) discharge is achieved when the following condition is met:

$$\beta > 2\nu_{i0} \quad (2.1),$$

where β is the rate of loss of electrons due attachment to the halogen donor species, and ν_{i0} is the equilibrium ionization rate of Kr metastable atoms by electron impact, per electron per second.

The most fundamental objection to the theory of Daugherty et al. is that it deals only with a homogeneous (nonspatially dependent) runaway of electron density - it does not predict break-up of the discharge into filamentary arcs. The second objection is that a term describing the depletion of metastable Kr by ionization (ionization of Kr metastables is the primary source of electron production in the theory) is ignored in the equation governing the ambient Kr metastable density. Neglect of this term is not justified (for reasons discussed below). If included its effect would be to relax considerably the stability requirement on the value of β .

Daugherty et al. do state that:

".....Further, spatial non-uniformities probably play an important role in the onset of the instability. In the analysis we have assumed that the discharge is spatially uniform."

[†]Mangano[1977] specifically confirmed that this was the belief of himself and his co-authors in response to a direct question by C.E.Webb on stability in the absence of halogen bearing species.

Although it could be argued that this statement effectively prevents criticism via the first objection raised in the preceeding paragraph, inclusion of such a caveat is very much a case of 'having your cake and eating it too'. The need to emphasize the point arises because other workers (eg. Brown and Nighan[1978,1979]) have been encouraged to use Daugherty et al.'s criterion to explain stability in their RGH devices - thus reinforcing its credibility. The solution to preventing the formation of the type of global instability discussed by Daugherty et al. is simple. One need merely introduce a current limiting resistor into the circuit in series with the discharge. If the basic assumptions of this theory were correct, one could use it to explain current runaway in, say, a standard Hg discharge lamp. The inclusion of a current limiting resistor in the power circuit of these devices is essential to their proper operation.†

In arriving at their stability criterion, Daugherty et al. make the following assumptions regarding electron production and loss:

(1) the ionization rate ν_i is dominated by the ionization of metastable Kr atoms (Kr^*) with a rate constant $\langle\sigma v\rangle_i$, so that

$$\nu_i = \langle\sigma v\rangle_i [\text{Kr}^*] \quad (2.2)^\ddagger$$

(2) Kr^* atoms are themselves excited by electron impact on ground state Kr atoms with a rate constant $\langle\sigma v\rangle_*$.

(3) the rate of destruction of Kr^* is dominated by electron density independent processes, such as diffusive loss and destruction by the fluorine donor species in collisions which lead to KrF^* formation. This rate can be represented by a characteristic destruction lifetime, τ_d .

(4) the ionization rate of Kr^* by electrons is assumed in this model not to be a significant term for the loss of Kr^* .

With these assumptions, the rate of change of Kr^* can be written

$$\frac{d}{dt} [\text{Kr}^*] = \langle\sigma v\rangle_* [\text{Kr}] n_e - \frac{[\text{Kr}^*]}{\tau_d} \quad (2.3),$$

†This author wishes to thank CEW for describing in vivid detail, and on numerous occasions, the dramatic consequences of failing to provide such a lamp with a current limiting resistor.

‡Square brackets denote a population or number density.

where n_e is the electron density. In order to see how the stability criterion described by eq.2.1 is obtained, the reader is referred to appendix 7 for details of the argument from this point.

It is immediately obvious how a strong positive feedback loop, leading to current runaway, is initiated in this model. A global increase in n_e leads to an increased density of Kr^* . Since the ionization $[Kr^*]$ is the dominant source of electron production, the rate of production of electrons increases as a result. This further increase in n_e results in the production of an even greater $[Kr^*]$ ad infinitum. The role of the electron attaching gas in this scenario is to stop this type of runaway by introducing a large source of loss for electrons. As F_2 dissociation proceeds during the discharge pulse, however, the ability of the gas to stop this 'runaway' is diminished. Brown and Nighan[1978] confirm that they interpret the stability criterion in this way by stating:

"Consequently, the electron density rises and, additionally, there results an increase in the concentration of rare gas metastables, which are produced primarily by electron impact and lost primarily by reaction with F_2 ."

An increase in the density of Kr metastable atoms as the electron density increases is crucial to the speed with which the instability grows in Daugherty et al.'s theory. At the electron densities typical for the optimum excitation of RGH systems ($\sim 10^{15} \text{ cm}^{-3}$), conventional analyses of discharge excitation and loss processes would suggest that the number density of a metastable species is totally independent of (i.e. 'saturated with respect to') the electron density.

A more realistic version of the rate process balance giving the ratio of the density of atoms in an upper state, N_j , to that in a lower state, N_i , assuming domination of excitation by electron impact, would be:

$$\frac{N_j}{N_i} = \frac{n_e S_{ji}}{n_e d_{ji} + A'_{ji}} \quad (2.4),$$

where S_{ij} , the rate coefficient for excitation, and d_{ji} , the total electron collision destruction rate of level 'j' (either by ionization or quenching back

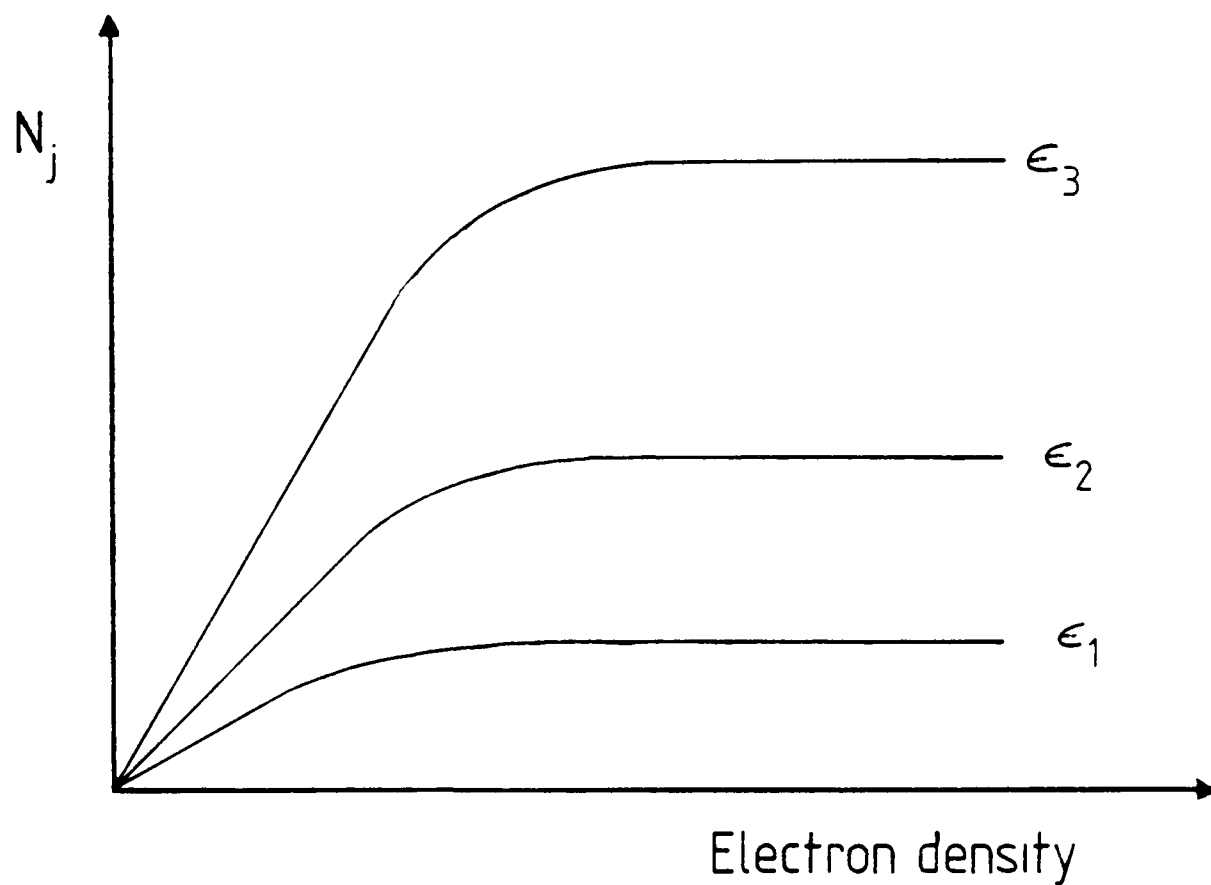


FIGURE 2.2: Schematic of the dependence of the population, N_j , of an excited state, j , of a neutral atom on electron density for a number of mean electron energies, $\epsilon_1 < \epsilon_2 < \epsilon_3$. 'Saturation' of the population at high electron densities is evident.

to level 'i' - the term neglected by Daugherty et al.), are both functions of the mean electron energy, and $A'_{ji}^{\dagger}$ is a coefficient representing losses by processes not involving electrons.

In the presence of high electron densities, $n_e d_{ji} \gg A'_{ji}$, and the n_e in the numerator and denominator of eq.2.4 cancel, leaving an n_e independent population in the upper level. S_{ji} and d_{ji} are, however, still dependent on the mean electron energy. The density of electrons required to achieve saturation therefore depends on the value of this energy (and therefore E/N). This effect is shown schematically in figure 2.2. The reader is referred to texts on the subject of excitation for a more detailed discussion (see e.g. Webb[1976b]).

There is no reason to believe that any fundamental difference exists between the kinetic processes at work within the KrF^* system and those in the XeCl^* system. The results presented in chapter 5 of this thesis show conclusively that saturation of the rare gas metastable population with respect to the electron density does indeed occur. Furthermore, in contradiction to the postulations of Brown and Nighan quoted above, but nevertheless in agreement with basic theory, the effect of a depleted halogen donor concentration is shown to result in a lower rare gas metastable density. This occurs because, with a lower halogen concentration, the rate of loss of electrons from the discharge is lower. The E/N required to sustain current flow in the face of electron loss is lower, with the consequence that the mean electron energy (and therefore the metastable production rate) is reduced.

[†] Usually, the coefficient represents loss due to radiative decay; in cases where a transition directly back to the ground state is not allowed (metastable), or radiatively trapped (resonance level), it is still possible to define a decay probability. In the low pressure longitudinal discharge this will represent the reciprocal of the diffusion time of ions to the wall; in this case, it would represent losses due to volume recombination or any other loss process not involving electrons.

2.3 DISCHARGE STABILIZATION METHODS:

Discharge stabilization methods can be classified broadly as being either active or passive techniques. Passive stabilisation techniques would include:

- (1) Careful profiling of the transverse electrodes. This is done in order to obtain as uniform an electric field as is practicable within the discharge gap.
- (2) Replacement of the gas between the electrodes after each discharge event in high repetition rate devices. This is done in an effort to remove regions of gas which may be hotter and therefore have a lower density than other regions.
- (3) Preionization of the discharge region by irradiating it with X-rays, high-intensity UV radiation, or a short pulse electron beam, before switching the discharge on. This is done in order to provide a uniform seed of free electrons throughout the volume to encourage the electron avalanche to grow equally fast in all regions.
- (4) Provision of a very fast rising, short duration discharge excitation pulse. Basically this method is an attempt to avoid the problem by switching the discharge off before instabilities have time to grow. However, the fast rise of the excitation pulse also aids the formation of a uniform discharge.

All of these methods provide 'preventive medicine' for the stability problem but do not actively inhibit the formation of spatial inhomogeneities after initiation of the discharge. Active stabilisation methods would include:

- (5) The provision of an external source of electrons which actually dominates electron production. This is often termed an e-beam sustained discharge. The degree of inhibition of formation of spatial inhomogeneities depends on the degree to which the external source dominates electron production. The most efficient CO_2 lasers are produced using this technique because the optimum E/N for excitation of the laser levels is far below the minimum E/N required for self-sustained discharge operation (see eg. DeMaria[1976]).

(6) The provision of a spatially distributed resistance within the electrode area. This provides current limitation on a local basis. If a local increase in electron density should occur, E/N drops locally due to an increased voltage drop across the ballast resistance in that region. Positive feedback is thus inhibited. The degree of inhibition depends on the local value of the resistance. Recently, Sze et al.[1983], have developed this idea a stage further by replacing the localised resistance with a localised inductance.

Methods (3),(4) and (6) above are of special relevance to this study and will now be discussed in more detail.

2.3.1 Preionization:

The provision of a source of preionizing radiation - to create a large ($\sim 10^8 \text{ cm}^{-3}$) density of free electrons in the gas volume before (and during) breakdown of the discharge gap - aids considerably the formation of a spatially uniform glow discharge.

The breakdown mechanism is characterized by a large number of successive electron avalanches that originate from the cascade growth of electron density. The limited diffusive spreading ability of the secondary electrons and ions associated with each primary electron (i.e. the free electrons existing - or additionally provided by preionization - within the gas before application of the electric field) means that, in the case of an un-preionized discharge region, the avalanching tends to occur non-uniformly within the volume. The running discharge then consists of a few randomly situated filamentary arc channels. The explanation used by Palmer[1974], for example, to account for the beneficial effects of preionization was that, given a high enough density of spatially-uniformly distributed primary electrons, there would be significant spatial overlap of the primary electron avalanches. The result of this overlap would be avalanching at all places within the discharge volume (assuming that the electric field between the electrodes has some degree of spatial uniformity) and, consequently, the development of a spatially uniform glow discharge. It is pointed out by Levatter and Lin[1980] that there are some deficiencies in the

description given by Palmer and the reader is referred to their paper for a more complete discussion.

Three sources of preionization are in general use for discharge excited RGH lasers: UV light, X-rays, and e-beams. The degree of preionization which can be obtained from e-beams is variable - from a seeding of electrons equivalent to that obtained with UV light, up to the extent that the electron density within the volume is sufficiently high that electron avalanching is not required on application of the electric field. Devices utilizing e-beam preionization are not noticeably less complicated than pure e-beam devices.

The two most useful sources of preionization are X-rays and UV light, of which UV sources are the simplest to manufacture. X-rays do have one large advantage over UV sources, however: they have a greater penetration into the gas than the short wavelength UV light and metals are effectively transparent to them. As a result, much larger volume self-sustained discharges have been achieved with X-ray preionized devices. Furthermore, relatively simple X-ray sources for preionization applications are becoming readily available commercially. The reader is referred to the literature for further information (see eg. Shields and Alcock[1982], Lin et al.[1981], Levatter and Li[1981], Lin and Levatter[1979], and Tallman and Bigio[1983]). At the time of writing, it would seem that X-ray preionization offers the most potential for further development of discharge-excited RGH lasers.

UV preionization was (and still is) the technique most commonly employed for initiating stable RGH discharges. It has generally been found that the radiation from a high current arc provides suitable radiation for preionization purposes. There have been no reports in the literature to date, however, of the optimum spectral characteristics of UV preionization sources for RGH laser discharges. McKen et al.[1976] looked at questions of importance in CO₂ laser systems and a parallel study for RGH lasers clearly needs to be undertaken.

One arc source is not sufficient to provide the necessary uniformity of free electrons throughout the volume. The provision of an array of arcs situated at regular spacings (1-3 cm, say) along the entire length of the

discharge volume, and situated as close as is practicable to the discharge region, is usually found to be adequate. It is possible to obtain large volume discharges with UV sources (Watanabe et al.[1981], Watanabe and Endoh[1982]). Such devices usually require the preionization to be situated behind a semi-transparent mesh electrode to minimize the distance travelled by the UV radiation before reaching the actual discharge region. Kearsley[1980] and Smith[1982] both discuss requirements on the timing of the preionizing radiation relative to application of the main discharge pulse in a 'standard' short pulse duration RGH laser.

2.3.2 Distributed resistance electrodes (resistive ballasting):

The idea of providing of an electrode consisting of discrete resistive elements distributed along its length to assist the production and maintenance of spatially uniform transverse discharges was used in the first transverse discharge excited CO_2 laser (Beaulieu[1970]). There is a crucial difference between this type of resistive ballasting and the inclusion of a single current limiting resistor in series with the discharge (as described above). The difference lies in the fact that the distributed resistance electrode inhibits current growth on a local basis, whereas the current limiting resistor restricts the total current which is allowed to flow in the discharge as a whole. Inhibition of the growth of current on a local basis aids the maintenance of the spatial uniformity of a discharge. Limitation of the total current merely determines the maximum current which could flow in a single arc column.

Various techniques of providing this spatially distributed resistive ballasting have been developed. They include, for example, the use of:

- (1) a row of carbon resistors (Beaulieu[1970])
- (2) a row of pencil leads (Fujii et al.[1981] for high pulse repetition frequency operation of an XeCl^* laser)
- (3) continuous electrodes manufactured from amorphous Ge or Si (Gibson et al. [1977a,b,c] for CO_2 and HF lasers)
- (4) continuous electrodes manufactured from conducting plastics (Solimeno et al.[1974] for HF lasers)

(5) discrete metal pin electrodes with one end facing the discharge gap and the other in contact with an electrolytic solution (Pummer and Kompa[1972] for HF lasers, Gower[1973] for CO_2 lasers, and Hogan et al.[1980] for an XeCl^* laser).

The continuous electrodes are obviously very attractive from the point of view of obtaining a uniform electrode profile. Field enhancement at the edges of the elements of discrete electrodes is unavoidable. An initial study of the use of Ge electrodes for RGH lasers by Dr.A.J.Andrews of the Clarendon Laboratory indicated that this material was unsuitable for two reasons. Firstly, the propensity for RGH lasers to arc meant that breakage of the Ge electrode - should an arc occur - would be a continuing problem. Secondly, the material was observed to be etched badly by the highly reactive halogen donors present in RGH gas mixtures. Any advantage in uniformity over discrete element electrodes would therefore be lost as soon as the Ge became pitted.

Carbon resistors have also been observed to suffer catastrophic failure in the presence of arcing. Furthermore, and in common with graphite or pencil lead elements, the resistance of the elements cannot be easily altered.

Ballasting of a metal pin electrode with an electrolytic solution (such as NaCl or CuSO_4) also has problems but these are mainly to do with their initial construction (obtaining vacuum seals so that the electrolyte does not seep into the gas containment vessel). They do have a number of advantages over the other types of resistive electrode. In the event of a hard arc causing dielectric failure of the resistive ballasting, for example, liquid electrolyte electrodes are 'self-healing'. For the purposes of an investigation of the optimum ballast resistance, changing the concentration of electrolyte in the solution provides a simple method of altering the resistance without needing to open the gas volume to the air.

2.3.3 Fast discharge excitation:

(a) General:

Fast pulse excitation of CO_2 lasers was a widely recognised and well developed method of obtaining large peak power laser pulses before the discovery

of the RGH laser systems. The development of discharge excited RGH lasers has consequently been based to a very great extent on knowledge gained from CO_2 laser excitation.

The basic idea of fast pulse excitation is to initiate a uniform discharge very quickly and to terminate excitation on time scales which match the formation time of instabilities. Furthermore, by initiating the discharge quickly, a population inversion is obtained more rapidly and lasing occurs sooner.

Fast initiation, itself, aids the formation of a uniform glow discharge; in order to achieve it, a large 'overvoltage' - a voltage far in excess of the uniform field breakdown voltage, V_b (section 2.1), of the gas mixture - must be applied to the discharge gap. If the voltage does not rise sufficiently rapidly, breakdown will occur before the desired overvoltage is achieved. The time available to achieve the desired overvoltage will depend on a number of factors, including the composition of the gas mixture, the density of free electrons within the discharge region, and the value of the desired overvoltage itself (see eg. Morgan[1978]). A large overvoltage helps the formation of a fast, uniform discharge in two ways. Firstly, by maximising E/N , the overall rate of ionization of the gas is increased and the discharge is initiated more rapidly (in practice the speed with which the total current flow can rise is quite often limited by the electrical driver circuit). Secondly, small differences in E/N , resulting from (say) non-uniformities in the average electric field in the discharge region, will have a smaller effect on the avalanching process at high voltages. The rate coefficients for excitation and ionization of Kr by electron impact, for example, exhibit an approximately exponential dependence on E/N at the values of E/N typical of an established discharge, at higher values of E/N this dependence is not so strong (Greene and Brau[1978]).

(b) A 'standard' short pulse duration RGH laser:

The preceeding discussion indicates why it is desirable to establish a

Capacitive Transfer Circuit

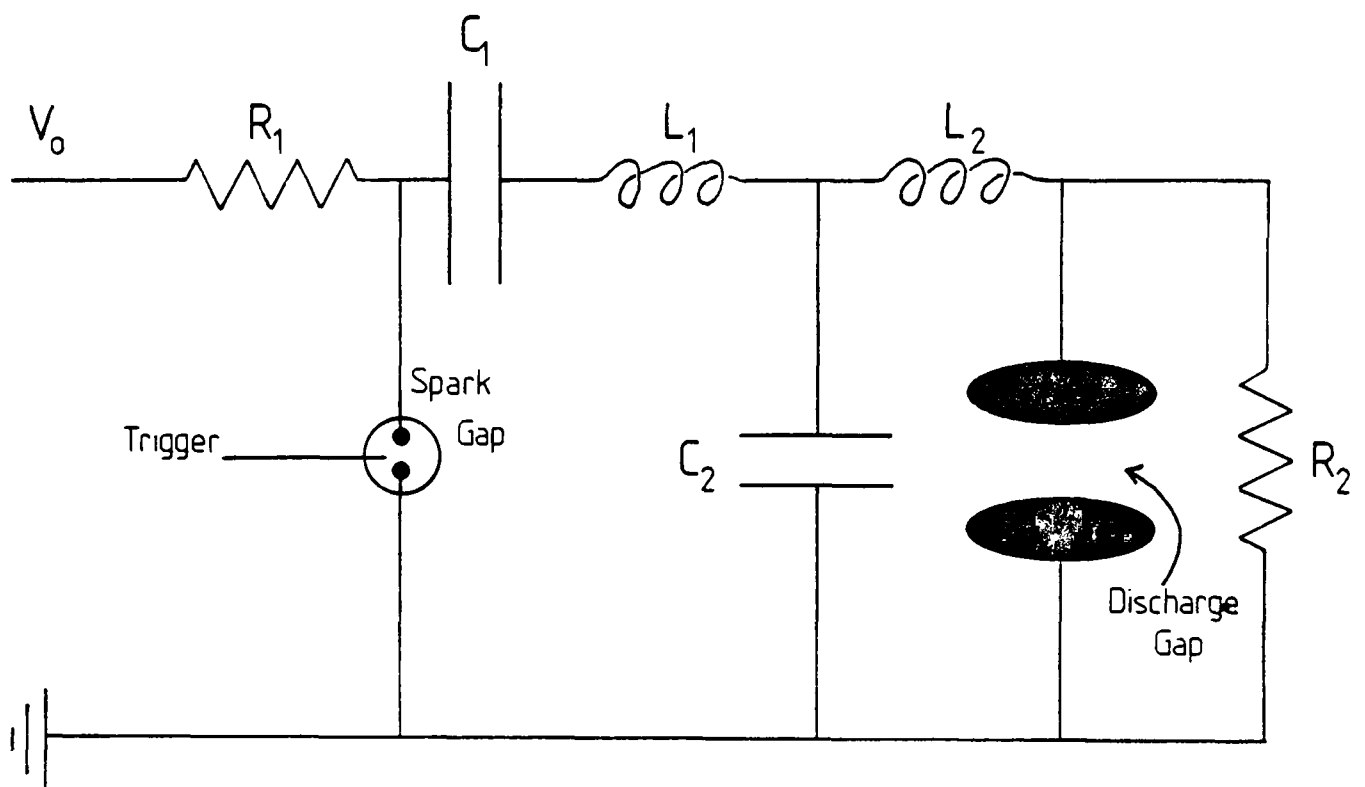


FIGURE 2.3

discharge quickly. In this paragraph the means by which this can be achieved in practice will be outlined. The most convenient approach is simply to describe the operation of a 'standard' short pulse duration laser system of the type developed by Kearsley[1980].

The simplest method of providing an electrical excitation pulse of short duration is to discharge a capacitor, which has previously been charged to some high voltage, through the laser discharge. This is the type of arrangement which was used by Beaulieu[1970] for his CO₂ laser. It is matter of general knowledge, however, that the current risetime in such a circuit is far too slow for the initiation of uniform RGH laser discharges at the high specific power densities ($\sim 100 \text{ kW cm}^{-3}$) required to achieve lasing. As is discussed in detail by Kearsley[1980](p.178ff), the major reason for this is the 'large' inherent inductance of such a circuit. It is clear from the expression,

$$V_o = L \frac{dI}{dt} \quad (2.5),$$

where V_o is the voltage at breakdown ($\sim 30 \text{ kV}$), I is the total discharge current, and L is the inherent inductance, that to achieve total currents of $\sim 1 \text{ kA}$ in times of $\sim 10 \text{ ns}$, i.e. $\frac{dI}{dt} = 10^{13} \text{ As}^{-1}$, that inductances of the order of a few nH are extremely important.

The speed of the excitation may be increased considerably by using a 'capacitive transfer' circuit (see figure 2.3). In this scheme C_2 is a bank of (say 10 off 2 nF) small 'peaking' capacitors which is placed as close as possible to the transverse electrodes. The main energy storage capacitor, C_1 , pulse charges C_2 when the spark gap is fired. At some time before the initial voltage, V_o , on the storage capacitor develops across the resistor R_1 , the discharge gap breaks down. At this point the bank of capacitors C_2 begins to discharge. The current risetime in this case is determined mainly by the much smaller enclosed inductance loop, L_2 , rather than by the previously large ($\sim 200 \text{ nH}$) inductance, L_1 , of the external circuit (see Bushnell et al.[1976]).

Two of the bank of internal peaking capacitors can be seen in the cross sectional diagram of a 'standard' short pulse laser presented in figure 2.4. Unlike figure 2.3, this diagram illustrates the small, but ingenious,

A "STANDARD" RGH LASER HEAD (Discharge-excited)

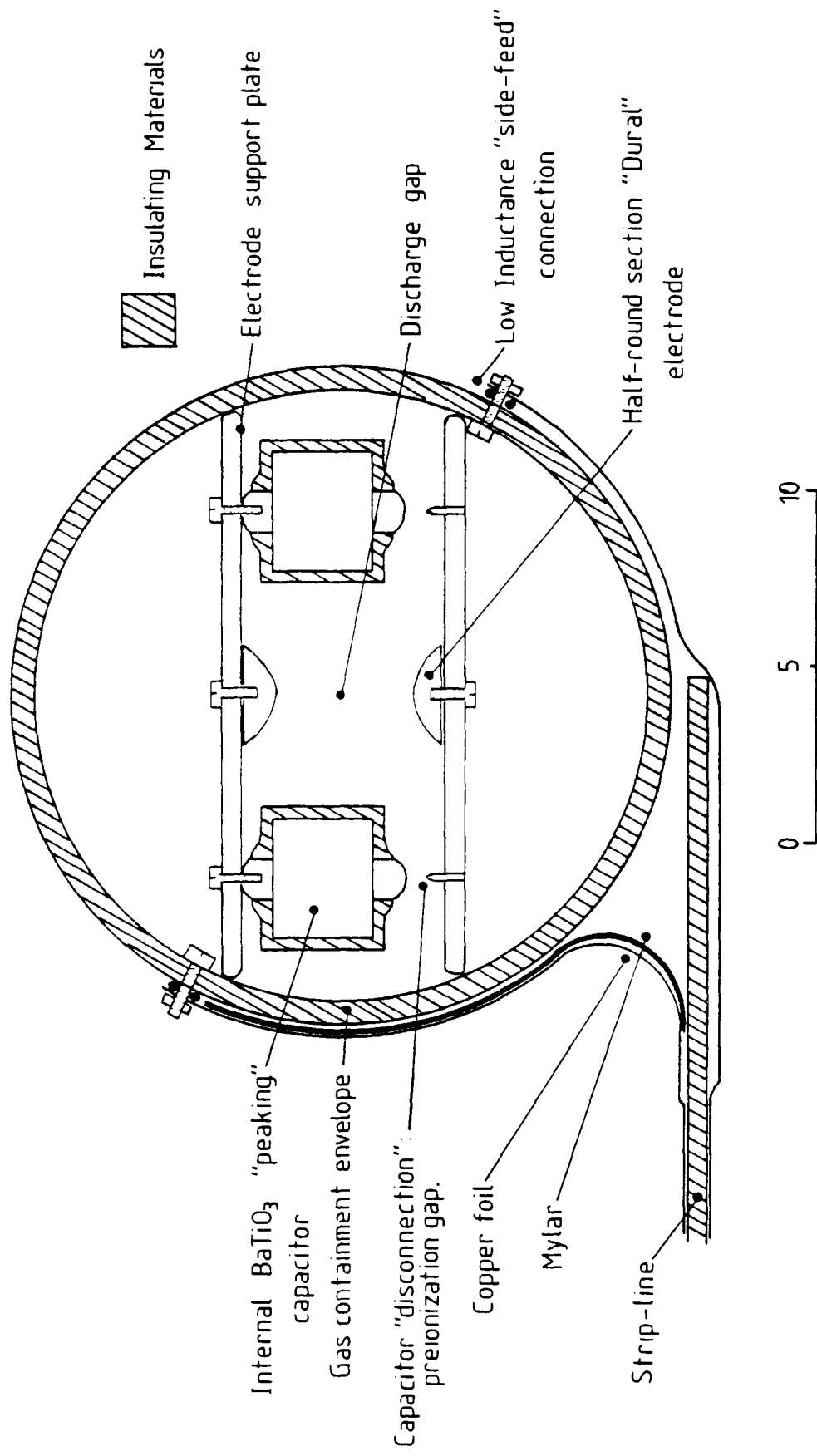


FIGURE 2.4

modification to the circuit which was developed by Kearsley[1979] to enable 'automatic' preionization of the discharge volume prior to breakdown of the discharge gap: small gaps are left between one stub of each of the peaking capacitors and one of the electrode support plates. During the initial charging of the peaking capacitors by the external energy storage capacitor, arcs form at each of these gaps (otherwise known as 'disconnections') flooding the discharge region with high intensity UV radiation. Prior to this development, UV-preionized, short pulse discharge, RGH lasers had required an extra circuit to provide preionization from a separate linear array of spark gaps.

For reasons that will be discussed later, however, the use of internal capacitors was not desirable for obtaining fast risetime, long pulse duration discharge excitation. The development of long pulse duration discharge excited RGH lasers by other workers in the field will be commented upon at appropriate stages in the remainder of this thesis.

2.4 SUMMARY:

The problems of maintaining a spatially uniform transverse discharge is the principal hurdle to be overcome in the extension of the laser pulse duration of discharge excited RGH lasers. The development of pulsed excitation techniques for CO₂ lasers provides a solid basis for the development of RGH discharge devices.

Three practical methods of obtaining and maintaining stable RGH discharges for time scales longer than the 10-20 ns typical of short pulse RGH lasers were identified:

- (1) UV preionization of the discharge region prior to breakdown;
- (2) application of a fast rising electrical excitation pulse to the discharge region;
- (3) the use of distributed resistance electrodes to aid the maintenance of the uniform discharge procured via (1) and (2) above.

The remainder of this thesis describes the development and subsequent investigation of a device which incorporated all three of these discharge stabilization techniques.

C H A P T E R T H R E E

A prototype, resistively-ballasted electrode, XeCl^* , Laser: design, construction, and performance.

3.1 GENERAL REMARKS:

The various methods which have been reported in the literature for the promotion of discharge uniformity in high pressure discharges were outlined in the previous chapter. Consideration of the relative merits of these methods indicated that resistively ballasted devices employing electrolytic solutions as the ballasting medium had considerable promise. Consequently the design and construction of such a device was undertaken; it was hoped that the performance of the device would prove quickly whether or not distributed resistive ballasting per se was a suitable method for the stabilization of the arc-prone RGH discharge.

The simplest way to tackle the problem was to replace one of the 'half-round' electrodes in an existing 'standard' laser assembly (see section 2.3.3) with a resistive pin-array electrode. In its normal configuration, the 'standard' laser delivered laser pulses with durations of ~15 ns FWHM. This was in spite of the fact that the power delivered to the discharge by the driver circuit lasted for over 200 ns foot-to-foot. The reason for this premature termination of the laser was believed to be due to the onset of discharge instabilities which drastically curtailed the (electrically) pumped volume. Two factors engendered this belief:

1. The laser pulse appeared in the initial stages of the discharge-
before a collapse of the discharge voltage.
2. Observation of the uniformity of the discharge by eye revealed the
presence of some isolated arc channels. This observation must be

qualified with two caveats: firstly, observation by eye gives a time-integrated view of the discharge uniformity so conclusions cannot be drawn about how early or late within the pulse such features occur; secondly, the construction of the laser envelope made this kind of observation difficult - the discharge could be viewed only at a small angle along the optic axis thus only major non-uniformities could be seen.

If discharge instabilities were the problem then it could be hoped that the insertion of a resistively ballasted electrode would ameliorate the effect. If this were successful the project could then proceed to the development of a RGH laser device designed specifically for the generation of long-duration output pulses.

3.2 DESIGN AND CONSTRUCTION OF THE PROTOTYPE ELECTRODE:

3.2.1 Design constraints:

The design constraints of a pin-array electrode ballasted with an electrolytic solution require the provision of:

1. an array (in this case a single longitudinal row) of metal pins embedded in a block of insulating material to form the main body of the electrolyte 'reservoir' or 'bath';
2. a conducting strip on the side of the bath opposite to that of the pin array to provide the electrical path to the driver circuit (referred to hereinafter as 'the backing plate');
3. a sufficiently small size and spacing for the pins of the array such that the assembly behaves in a manner which is not significantly different from a 'continuous' electrode. This requirement must be weighed against the need to have the pins spaced as far apart as possible to avoid significant conduction between adjacent pins (not only within the electrolyte but also to avoid surface 'tracking' on the side presented to the gas volume);
4. a thickness of electrolyte sandwiched between the pin ends and the

PROTOTYPE BALLASTED ELECTRODE (cut-away relief)

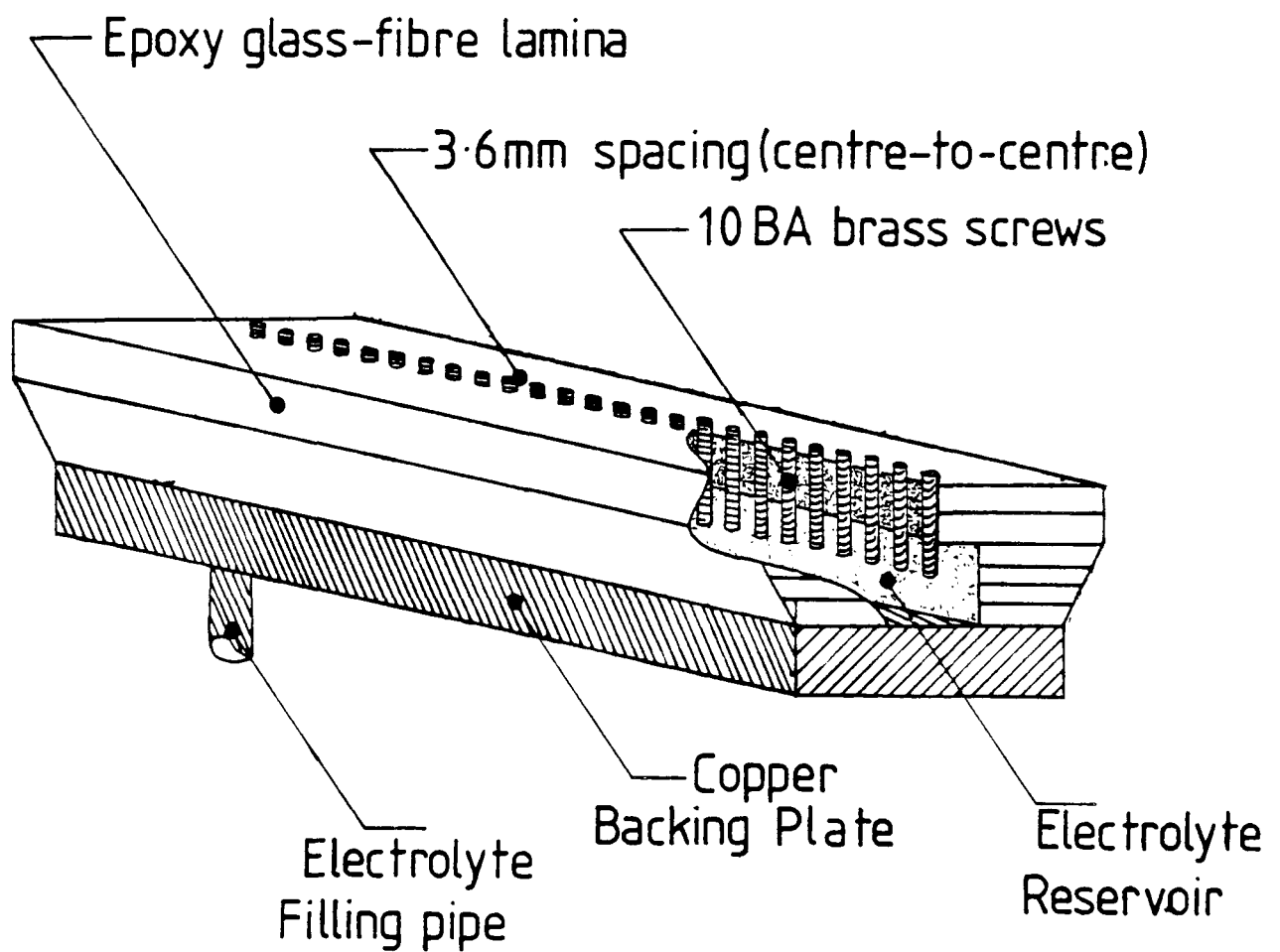


Figure 3.1

backing plate which must be sufficient to avoid recurrent dielectric failure of the electrolytic solution.

The first two requirements are relatively straightforward except for the problem of obtaining vacuum integrity at the insulator-conductor interfaces. The pin size was determined by constraints in obtaining vacuum integrity, the optimum pin spacing was similarly found by 'trial and error'. The separation between the backing plate and the pin array was estimated using data contained in notes written by J.C.Martin and others [1970] which deal with pulsed breakdown of deionized water: for uniform electric fields, the breakdown field is given approximately by

$$Ft^{1/3}A^{1/10} = k \quad (3.1),$$

where F is the field in MV/cm, t is in microseconds, and A is the electrode area in cm^2 . The constant ' k ' depends on the properties of the dielectric medium and for water, $k=0.3$. In the case of a ballasted electrode, however, the 'electrode' combination of ballasted pins and backing plate does not result in a uniform field so a field enhancement factor must be included.

3.2.2 Construction Details:

The greatest problem to be overcome in the construction of both the prototype and the long pulse duration laser (chapter 4) was to obtain a vacuum-tight seal between the conductors and insulators of the electrode. In the prototype over 230 metal pins penetrated the insulating support member and a seal over 2 m in length had to be made between the support member and a metallic backing plate.

Initial tests using a PVC support member, a copper backing plate, and round-headed tailors' pins, bonded together with 'Araldite' epoxy adhesive were unsuccessful: Araldite can be bonded to copper which has been specially treated, but it cannot be made to bond well to PVC. It was found that the pins were pushed out of their support plate under the pressure of the electrolyte behind them when the laser chamber was evacuated. The electrode did operate for a sufficiently long time, however, to observe surface tracking between the pin heads. It was clear that the spacing of $\sim 0.75\text{mm}$ between the closest points of

PROTOTYPE RESISTIVELY BALLASTED ELECTRODE

XeCl* LASER

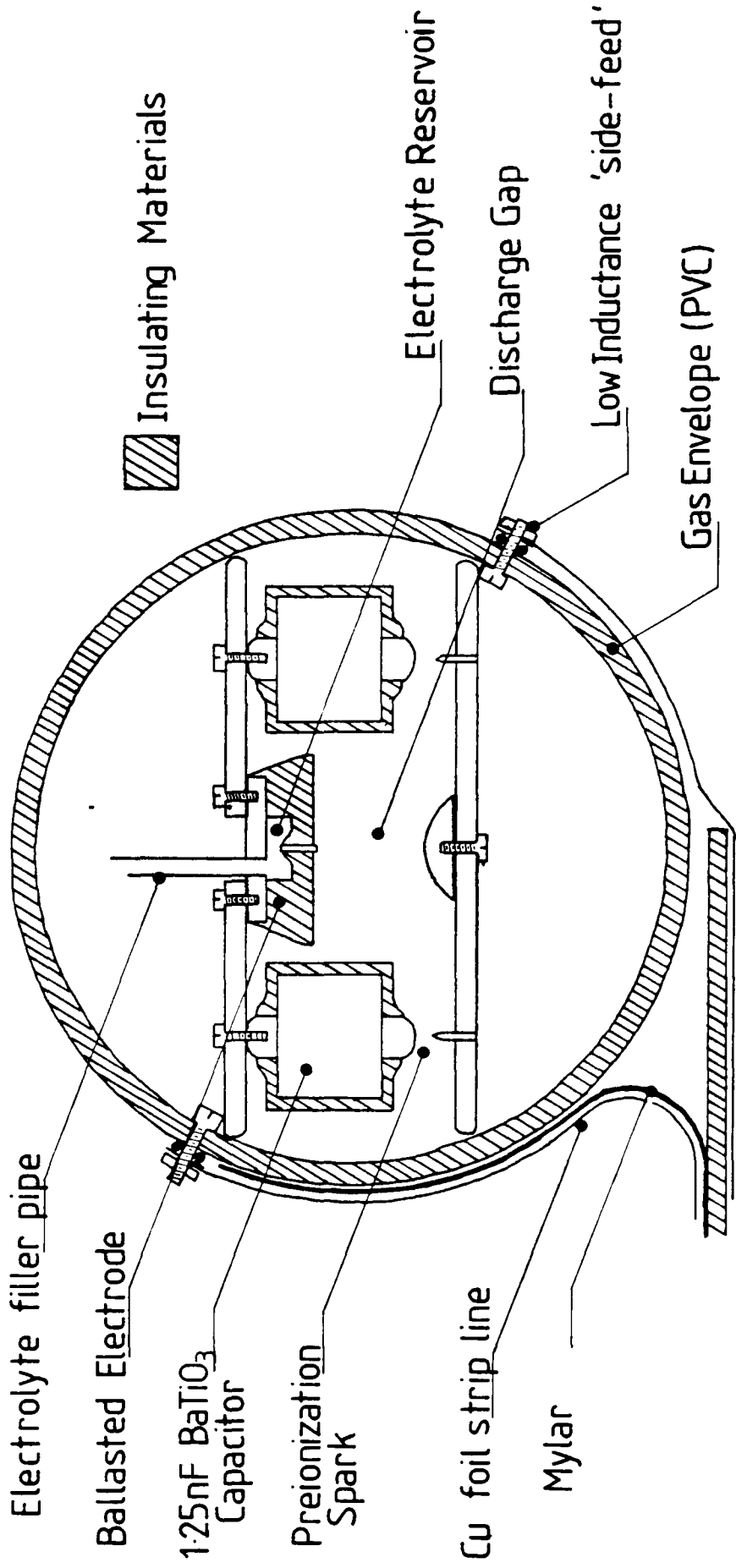


Figure 3.2

adjacent pins was not large enough.

A satisfactory solution to the second of these difficulties was to use small (10BA) brass screws - instead of pins - with a one-screw-diameter space left between adjacent screws. The more serious vacuum seal problem was overcome by using a laminate of commercially available printed-circuit board (PCB) as the insulator. PCB consists of 1.5 mm thick epoxy glass-fibre sheet coated on each side with a thin layer of copper. The electrode finally used for this preliminary evaluation is shown in cutaway relief in figure 3.1 and in cross-section, in position within the laser head, in figure 3.2 .

The glass-fibre laminate consisted of six 30 mm x 1 m strips of PCB. All but one of these strips had all of their copper cladding removed by immersion in nitric acid. Only one side of the cladding was removed from the sixth strip. A 5 mm slot was milled down the length of this and three other strips of the PCB for the eventual formation of the electrolyte containment bath. An effective seal between the copper backing plate and the PCB (on which the cladding had been retained) was attained by using soft solder. The other three slotted strips were then bonded to this assembly with araldite. The two remaining strips were also fixed together to form a reasonably thick pin-array support member. Approximately 230 holes were drilled and tapped along the length of this member at spacings of ~ 3.6 mm: 10BA brass screws were inserted into these holes and then glued into position. The screws were made uniform in length by careful milling and finally each screw tip was polished to reduce field enhancement from sharp edges. When the pin-array was affixed to the backing plate assembly a gap of ~ 3.5 mm remained between the (internal) tips of the screws and the exposed copper of the backing plate. This gap was bridged by the electrolytic solution (aqueous copper sulphate solution), which could be introduced via a system of PTFE pipework connected to outside the laser gas envelope: in this way the electrolyte could be changed without dismantling the laser. The ballasted electrode and the remaining 'conventional' electrode of 'half-round' section aluminium defined the limits of the transverse discharge which had a gap of ~ 23 mm and a length of ~ 820 mm.

PROTOTYPE LASER: DRIVER CIRCUIT SCHEMATIC

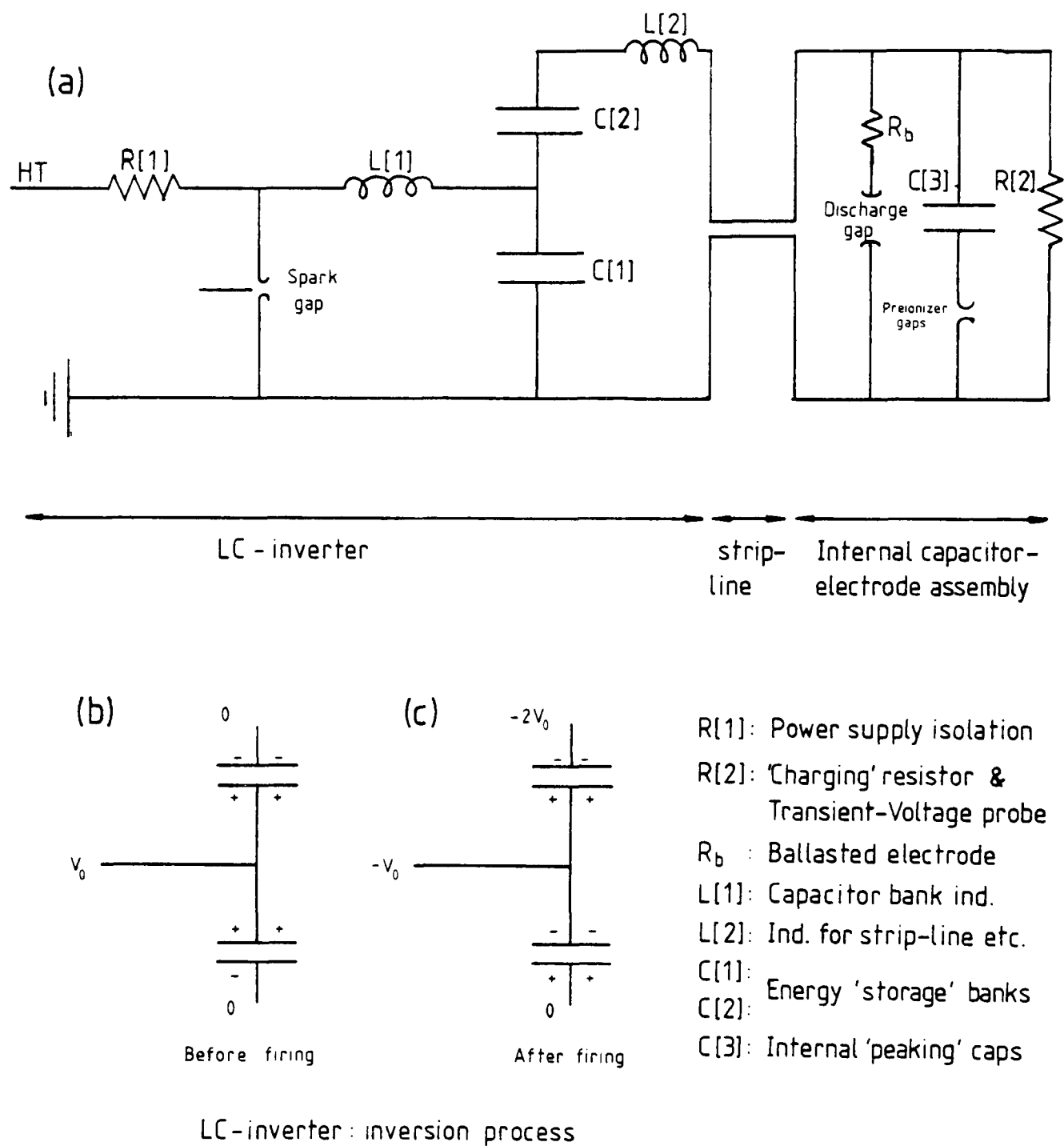


Figure 3-3

3.3 THE DRIVER CIRCUIT:

The discharge driver circuit comprised two parts: an LC-inversion network to provide the main energy storage, and an internal bank of 'peaking' capacitors which provided UV-preionization by the automatic preionization scheme (section 2.3.3(b)).

The internal bank (a part of which is shown in figure 3.2) consisted of eighteen 1.25 nF barium titanate capacitors spaced evenly along both sides of the discharge gap. A circuit diagram illustrating the principles of operation of an LC-inversion network is given in figure 3.3 . The following description comes from Kearsley[1980]:

'A pair of capacitors C[1] and C[2] are connected in series. The common terminal is charged to a high voltage V[0] (figure 3.3(b)). The spark gap is fired and charge flows through the inductance L[1] reducing the voltage across C[1] so that eventually it inverts in polarity to -V[0]. The voltage across the laser channel thus changes from zero to -2V[0] (figure 3.3(c)). The laser discharge gap will, in practice, undergo breakdown before the voltage has fully inverted and charge will begin to flow through L[2]. If the capacitors are low inductance capacitor banks (so that the individual inductances of the capacitors appear in parallel) then the inductances L[1] and L[2] may be small and a fast discharge results.'

In the present case, the speed of the peaking capacitor bank was the major factor in determining the speed of the discharge. Each of the capacitor banks, C[1] and C[2], contained thirty-five closely spaced 2.5 nF Barium Titanate capacitors. In order to keep the inductance L[2] small it was necessary to use low inductance 'strip-lines' to connect the network to the laser head: part of the strip-line used is also shown in figure 3.2 . The stray inductance in the circuit can be estimated by the approximation:

$$L \approx \mu_0 A / \ell \quad \text{Henry} \quad (3.2),$$

where 'A' is the cross-sectional area of the enclosed loop in m² and 'ℓ' is its length in metres. The inductance of the current loop associated with each row of capacitors and the electrode is twice the total inductance associated with the entire internal-capacitor-electrode assembly. The cross-sectional area was ~5cm x 5cm and the length was ~1 m. Therefore, the total inductance of the assembly,

$$L_{\text{total}} = 0.5 \times (4\pi \times 10^{-7} \times 25 \times 10^{-4}) = 1.5 \text{ nH}.$$

PROTOTYPE LASER

Typical Discharge Voltage Pulse

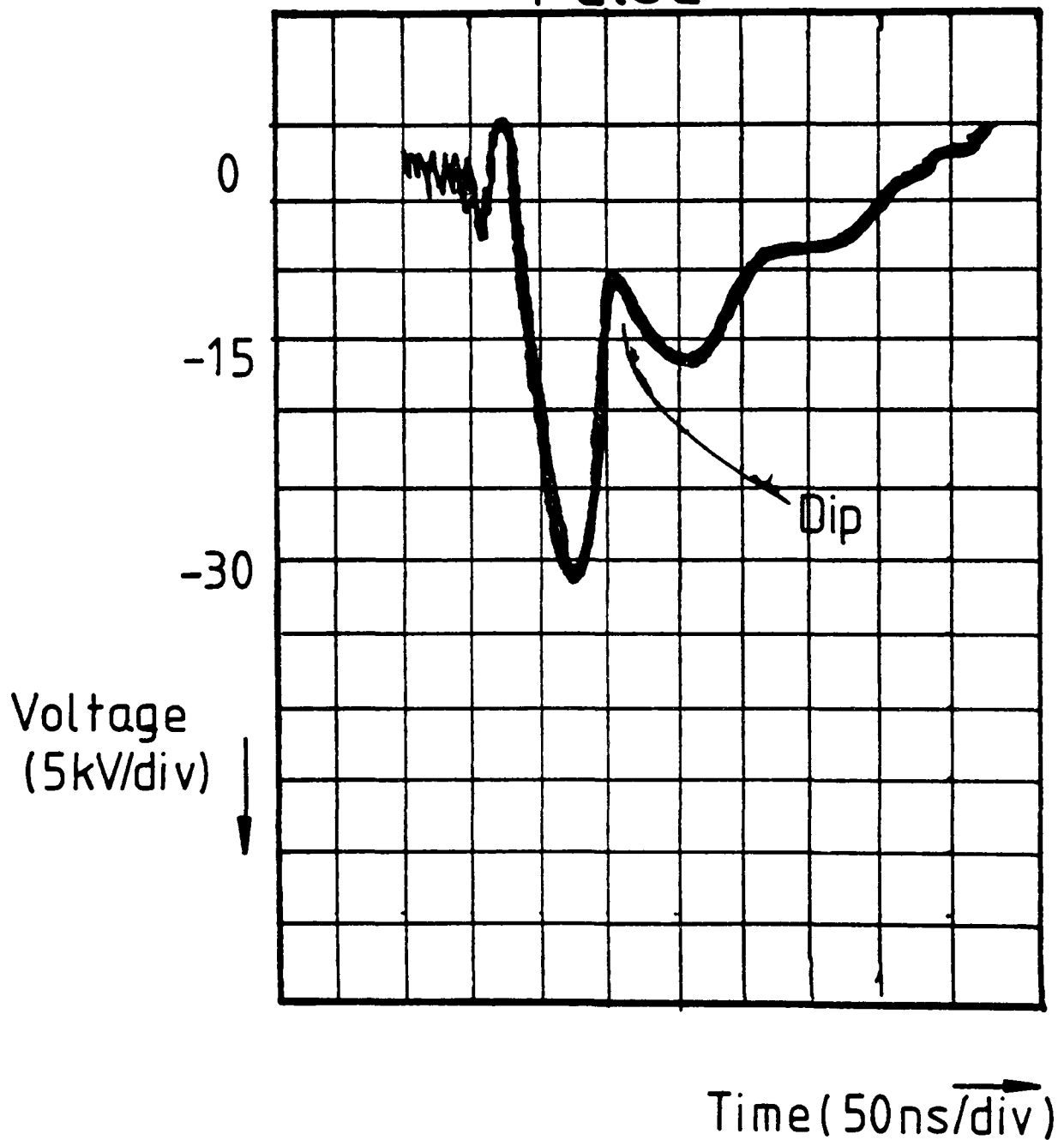


Figure 3.4

Inductances can be similarly estimated for the strip lines (perspex spacer ~ 5 mm thick; width ~ 30 cm) and spark gaps (~ 20 nH). The inductance, $L[2]$ is unlikely to be less than 100 nH when the internal inductance of the capacitors and stray inductance are taken into account. The two capacitor banks, $C[1]$ and $C[2]$, were usually charged to ~ 30 kV before firing the spark gap.[†] The sequence of events following the firing of the spark gap began as described above; at sometime during the voltage inversion process the internal capacitor bank would start to charge from the LC-inverter. During this charging phase, arcs formed at each of the gaps between an internal capacitor and its opposing pin and flooded the volume with high intensity UV radiation. This radiation provided a relatively uniform seeding of free electrons throughout the volume before the breakdown of the main discharge which occurred shortly afterwards.

The transient voltage appearing across the internal electrode-capacitor assembly was measured using a low-inductance voltage divider connected across the gap. A typical voltage trace is shown in figure 3.4.[‡] This figure shows the initial build up of voltage as the internal capacitor bank charges, the subsequent breakdown of the discharge gap, and, later, a rise in voltage as the external 'energy-storage' circuit starts to deliver power to the discharge volume directly. In its 'standard' (i.e. unballasted) configuration the discharge time of the internal capacitor bank was shorter than the risetime of the LC-inverter network. This arrangement allows the power input into the discharge to fall to zero momentarily. The fact that the discharge driver circuit did not provide a steady deposition of power over a well defined period of time considerably complicated any interpretation of the observed behaviour of the laser output: on some shots a second and weaker laser pulse was observed at the time of the second voltage peak with the laser in its 'standard' (unballasted) configuration.

Direct measurement of the discharge current in this laser was not possible. Any measuring device placed within the capacitor-electrode assembly

[†] A charging voltage of 30 kV corresponds to a stored energy of ~ 40 J.
[‡] Obtained with the ballasted electrode in place and recorded on Tektronix 466 fast storage oscilloscope.

would have not only perturbed the operation of the device by the introduction of extra inductance but it would have almost certainly caused undesirable 'flashover' problems. A very rough estimate of the total current in the discharge can be made using the relation:

$$I_d = \frac{d}{dt} Q_c = C \frac{d}{dt} V_c \quad (3.3),$$

where Q_c is the charge, and V_c the voltage, on the capacitor bank. Ideally, eq.3.3 should be used over a region where $d^2V/dt^2=0$ so that inductive effects can be ignored. The capacitance, C , of the internal capacitor bank, is 22.5 nF and dV/dt can be obtained from the measured discharge voltage. For this estimate the voltage pulse measured with a 125 ohm-cm electrolyte in the ballasted electrode (3.75 ohm equivalent series resistance) was used. The measured value of $dV/dt \approx 0.6\text{kV/ns}$ yields:

$$I \approx 13\text{kA}.$$

However, this can be considered an order of magnitude estimate only in view of the uncertainty in the measured value of dV/dt and the assumption that $d^2V/dt^2=0$.

3.4 THE PERFORMANCE OF THE PROTOTYPE LASER:

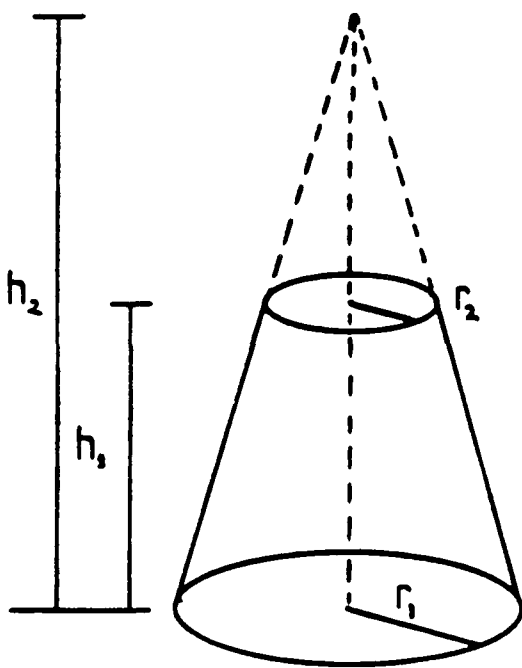
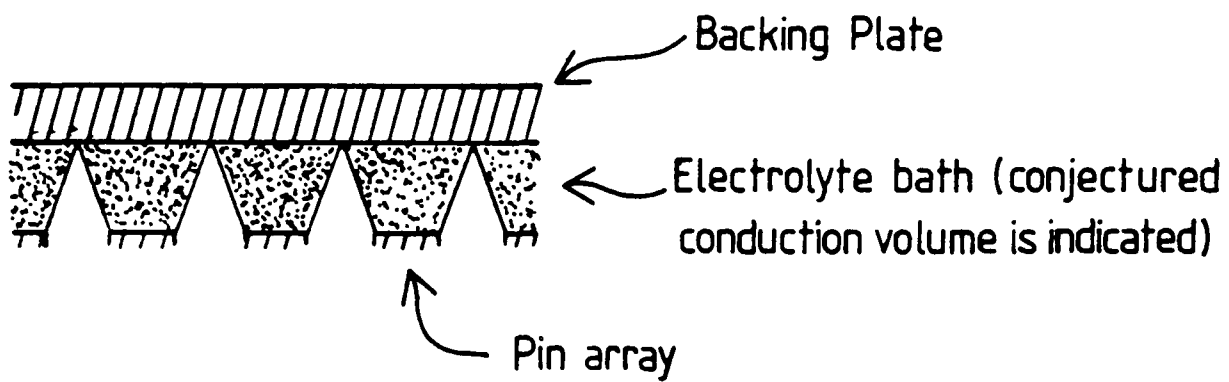
An investigation was carried out to determine the sensitivity of the XeCl^* laser pulse duration and energy to the variation of the composition of the gas mixture, the charging voltage applied to the LC-inverter, and the concentration (and therefore the resistance) of the copper sulphate ballasting solution.

3.4.1 Experimental Details:

Measurements of the temporal evolution of the laser pulse were carried out using a photodiode with a sub-nanosecond risetime (ITL type no. FD50M20UVE48) in conjunction with a Tektronix 519 oscilloscope. Pulse energies were determined using a calibrated energy meter (GenTec type ED500).

The conductivity of electrolyte solutions was measured with a Wayne-Kerr bridge and a conductivity cell. The problems associated with the accurate measurement of high conductivities are well known (Braunstein[1971]): for this reason an uncertainty of $\pm 30\%$ was estimated for these measurements. An

Geometry used in the estimation of individual pin resistance of an electrolyte array



The conduction volume is a truncated right cone.

The resistance, $R = \frac{\rho l}{A}$, where ρ is the resistivity in $\Omega\text{-cm}$, l is the distance in cm, and A is the cross-sectional area.

$$\begin{aligned} \text{In this case, } \int dR &= \int \frac{\rho l}{A(l)} dl \\ &= \frac{\rho h_1}{\pi r_1 r_2} \end{aligned}$$

Figure 3.5

estimate of the equivalent series resistance that the ballasted electrode would introduce into the circuit was made at the design stage. The approximations used in arriving at the estimate are indicated in figure 3.5. It was assumed that each individual screw would draw current through a volume of electrolyte as large as was possible without overlapping the conduction volume of its nearest neighbour. Also, it seemed likely that the current paths between the backing plate and the end of each screw would be reasonably direct.

The resistance of the conduction volume associated with each screw is not only dependent on the separation between the backing plate and the end of each screw (h_1), but also on the h_1 dependent cross-sectional area through which the charge must flow. The appropriate integration yields the result that the individual screw resistance,

$$R_s = \rho h_1 / \pi r_1 r_2 \quad (3.4),$$

where ' ρ ' is the resistivity of the electrolyte in ohm-cm, r_1 is half the pin-to-pin spacing in cm, and r_2 is the radius of the end of each ballasted screw in cm. For the case of the prototype electrode: $r_1=0.18$ cm, $r_2=0.09$ cm, and $h_1=0.35$ cm. This results in an estimate for the individual screw resistance, R_s , of $\sim 7\rho$ ohm, and an equivalent series resistance for the whole 230 pin electrode, R_b , of $\sim \rho/33$ ohm.

A flow of electrolyte solution through the electrode during operation of the laser was maintained by means of a small peristaltic pump connected to a feeder tank of ~ 100 cm³ volume. An estimate of the rise in temperature experienced by the electrolyte during each discharge shot was made because of concern that local heating of the electrolyte may have been excessive: after repetitive operation small bubbles would appear in the return flow line and the solution was noticeably warm. However, a rough estimate of the rise in temperature for a single shot in one small conduction volume (fig 3.5) arrived at the value of 0.2°C using previously estimated values of current and resistance. It was found later that small leaks existed between the copper backing plate and the PCB laminate of the ballasted electrode. It was concluded that gas from the volume was being forced into the electrolyte reservoir to

PROTOTYPE XeCl Laser

Laser Output vs. Resistivity of Ballast Solⁿ.

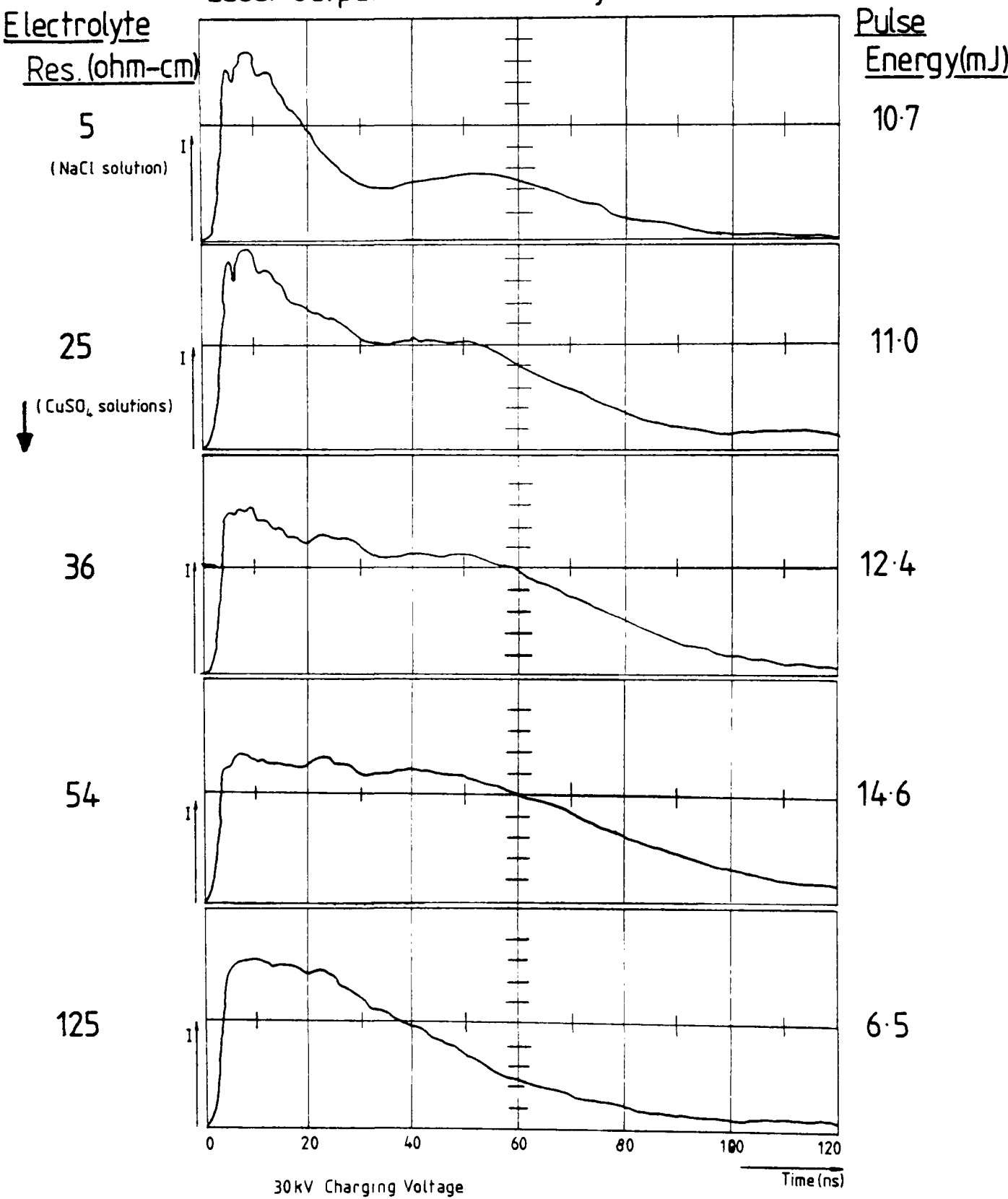


Figure 3.6

appear later as the bubbles in the return flow line.

The optical cavity consisted of two silica blanks (Spectrosil B - nominal quarter-wave flatness) with 'soft' dielectric coatings which were mounted directly onto the end flanges of the gas envelope. One disc was coated to provide a nominal reflectivity of 99.6%, the other had a nominal 90% reflectivity. The uncoated face of each disc was presented to the gas mixture since these coatings were easily etched by HCl (see appendix 1).

The optimum gas mixture was found to be ~3.5 mbar HCl, ~16 mbar Xe, and 2.33 atm He in the parametric study of gas composition described below. Unless otherwise stated this mixture was used to obtain all the results presented in this chapter. Details of the purity of gases used in all these experiments are given in appendix 3.

3.4.2 The dependence of the laser pulse duration on the ballast resistance:

The investigation of the dependence of the laser pulse duration on the strength of the ballast resistance was the most important facet of this initial feasibility study. It was mainly because very encouraging early results were obtained that it was decided to continue with the 'resistive stabilization' approach to the generation of long pulse duration RGH lasers. Representative output waveforms are shown in figure 3.6. The maximum pulse duration (80 ns FWHM) obtained was, at that time, only 20 ns shorter than that obtained in a purpose-built X-ray preionized device reported by Lin and Levatter[1979].

As is evident from figure 3.6, both the laser pulse duration (measured at FWHM) and the pulse energy^{initially} exhibit a continuous increase as the ballast resistance is increased. The optimum value of ballast resistivity was found to be ~50 ohm-cm. This corresponds to an individual pin resistance of ~350 ohm (from eq.3.2). At very high resistivities (≥ 125 ohm-cm) the pulse duration was curtailed and the energy was correspondingly diminished. This was by no means unexpected since the ballasting solution and the discharge were 'competing' for the power delivered by the driver circuit:

$$P_{\text{discharge}} = P_{\text{total}} - (I^2 R_{\text{ballast}}) \quad (3.5).$$

A balance had to be sought between the beneficial effects of discharge

PROTOTYPE XeCl LASER

Laser output vs. LC-inverter Charging Voltage

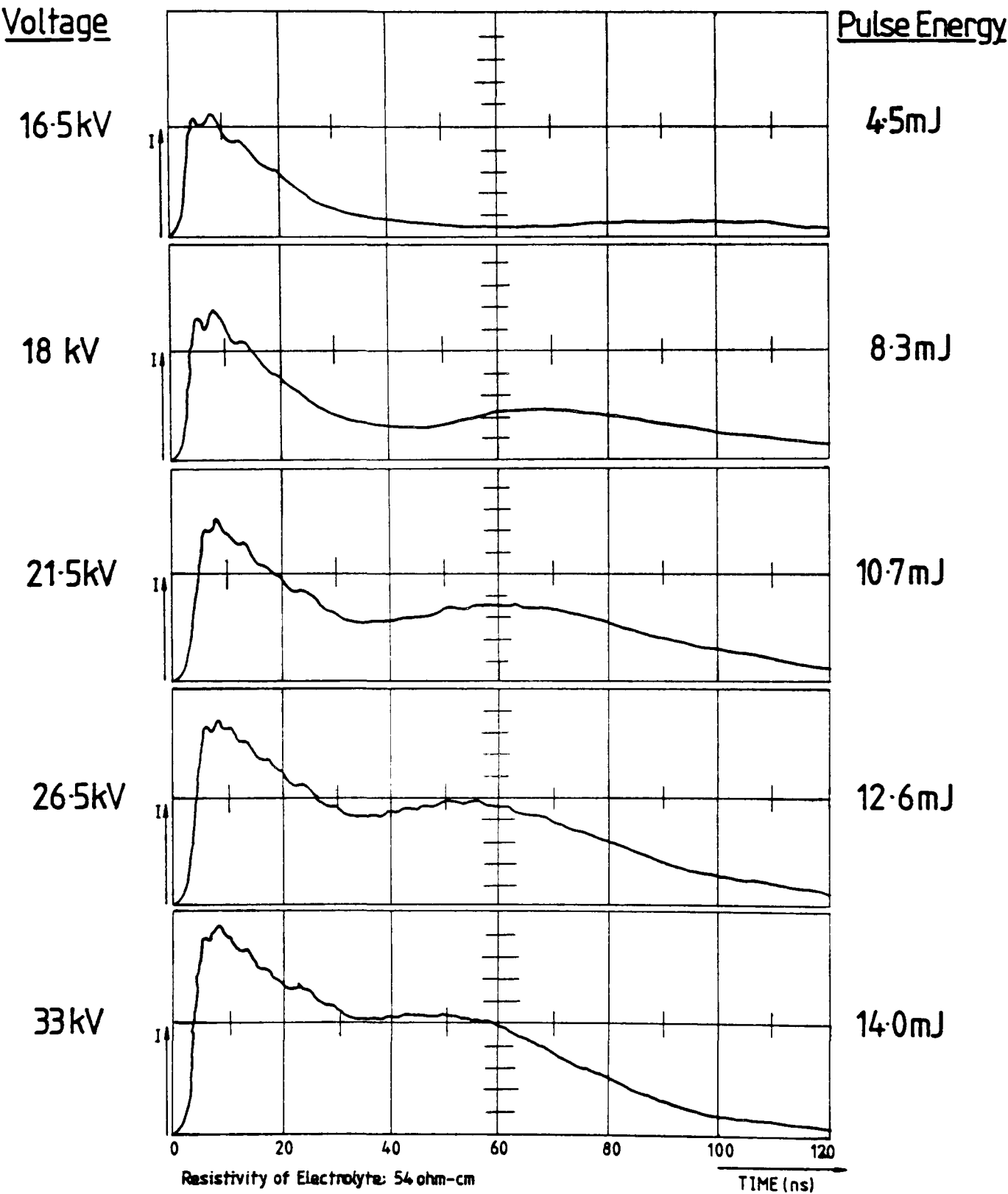


Figure 3.7

stabilization and the disadvantage of lower power deposition in the active volume. Pulse energies obtained from the device, when fitted with two conventional electrodes, were often as much as an order of magnitude greater than the energies obtained with the ballasted electrode in place. Overall, the discharge uniformity, as observed by eye, was considerably better than had been the case without resistive stabilization: high current bright arc channels between the electrodes seemed to be entirely absent. For very high resistivity solutions - especially above 125 ohm-cm - arcing occurred frequently. However, these arc channels connected points between the electrode support plates to which the ballasted electrode and its 'conventional' partner were attached. Clearly, the preferred path for current flow was not between the electrodes even though the extra discharge path was ~ 1.5 cm. This problem had to be taken into account in the design of the laser described in chapter 4.

Interpretation of the dependence of laser pulse duration on ballast resistance cannot be made in terms of 'discharge stabilization' alone; the ballast resistance also affects the characteristic discharge time of the internal capacitor-electrode assembly. As the ballast resistance was raised the discharge time of this assembly became longer. The consequence of this was that power was delivered to the discharge at a more constant rate because the internal circuit was still delivering power in the period before the slower LC-inverter circuit began to power the discharge (the dip in figure 3.4 is shallower).

3.4.3 The laser pulse duration as a function of the charging voltage:

The results presented in figure 3.7 illustrate the effect that varying the value of the voltage initially applied (the charging voltage) to the LC-inverter capacitor banks had on the laser pulse duration and energy. The general trend is one of improvement in laser output with increasing voltage. As with the variation of the ballast resistance, the interpretation of this trend is not straightforward since the magnitude of the charging voltage affects not only the amount of energy eventually deposited in the active volume but also the 'risetime' of the voltage across the internal electrode-capacitor assembly and

consequently, as mentioned in chapter 2, the discharge stability.

In order to discriminate between the relative importance of energy deposition and discharge stability when the charging voltage is altered, two small experiments were carried out whilst holding the charging voltage constant. Firstly, an extra 250 nF capacitance was inserted in each of the two capacitor banks of the LC-inverter with care taken to minimize any increase in circuit inductance. Although the main purpose was to detect differences due solely to increased energy deposition, in practice, no definite separation of responsibility for the very poor laser output observed could be obtained. The LC-inverter circuit was very sensitive to small changes in inductance and so, even with care, the addition of extra capacitance inevitably produced a decrease in the voltage risetime as well. The results of the second experiment admitted a more 'clear-cut' interpretation. With the LC-inverter in its original state, the series inductance between the LC-inverter and the internal electrode-capacitor assembly (L[2] in figure 3.3) was increased. This would have a substantial effect on the voltage risetime but a negligible effect on the energy deposition. The inductance increase was achieved by separating the conductors of the normally low inductance strip-line connecting the two circuit elements (figure 3.2) so as to produce a large enclosed loop. The deleterious effect on laser output was not as pronounced as in the first experiment but the degradation in both pulse duration and energy was significant.

A preliminary hypothesis could now be put forward to explain the results of figure 3.7: the major effect of an increase in the charging voltage^t is the production of a more stable discharge through a decrease in voltage risetime. A decrease in voltage risetime would be expected to increase the intensity of UV preionizing radiation and minimize the effects of non-uniformities in, for example, the electrode profiles. No definitive statement could be made at this stage regarding the consequent increase of energy deposition in the active volume with charging voltage.

3.4.4 Gas mixture studies:

Laser pulse duration and energy were found to be sensitive to the

concentrations of all three constituent gases: xenon, hydrogen chloride, and either helium or neon as the buffer gas.

Laser output was particularly strongly affected by small changes in the concentration of the halogen donor gas, HCl. The optimum partial pressure of HCl was found to lie between 2.5 and 3.5 mbar. An order of magnitude decrease in pulse energy was observed for minor (~ 2 mbar) excursions outside this range. Problems with gas lifetime, which were mentioned briefly in chapter 1, seem to have been initially associated with loss of HCl within the volume. Decanting small amounts of HCl into an 'stale' gas mixture would revive the laser to its initial performance. However this was not a permanent solution: eventually, addition of more HCl had no effect on laser output. The build-up of UV absorbing molecules within the volume which reduce the efficiency of the preionizing process, as reported by McKee et al.[1980], provides a plausible explanation of this phenomenon.

The ideal partial pressure of xenon in the gas mixture was found to be anything above 12 mbar. Below this value a gradual decrease in pulse energy was observed, above it (up to 30 mbar) very little difference was discerned. The high cost of xenon (£12/litre-atm) encouraged the use of only as much of the gas as was necessary.

Helium and neon were both tested as buffer gases: no evidence was gained which indicated that one was better than the other. Again, cost considerations meant that helium was chosen for most tests. Argon was also tried as a buffer gas but heavy arcing within the volume meant that it was not used in any further investigations. A steady increase in both pulse duration and energy (5.1 to 13.5 mJ) was observed over the range of pressure (atmospheric to 30 psig) investigated with helium and neon. It was originally thought that this increase in output with buffer gas pressure would not be unexpected if, as concluded by Kearsley[1980], three body ion-ion recombination (eq.1.1) is the major XeCl^* formation channel. An increase in the number density of the buffer gas would, at first sight, be expected to enhance the rate of formation of XeCl^* . However, as is discussed later in this thesis, this formation rate is virtually

independent of everything except the rate of formation of Cl^- by electron attachment. The reader will have to await the discussion of later chapters for an explanation of the effect of increasing the buffer gas pressure.

3.5 SUMMARY:

This initial feasibility study was quite successful considering the objectives set. The efficacy of resistive stabilization in the production of long pulse duration XeCl^* lasers was demonstrated by the attainment of pulse durations of ~ 80 ns FWHM. The results of this project have been published (Hogan et al.[1980]).

Technological difficulties with the manufacture of the ballasted electrode were overcome and areas of concern for the design of a 'dedicated' long-pulse device were identified. An investigation of laser pulse duration as a function of the ballast resistance of the electrode indicated that this parameter has a strong influence on discharge uniformity. However, the strong influence that the ballast resistance had on the characteristic rise and decay times of the driver circuit meant that some doubt existed as to the relative contributions of enhanced stability and temporally more uniform power input towards increased laser pulse duration.

Further evidence supporting the usefulness of resistive stabilization in RGH lasers was obtained toward the end of this feasibility study by Fujii et al.[1981] at the Clarendon Laboratory. In those experiments the ballasted electrode comprised an array of resistive pins made of pencil leads. This device was employed to demonstrate high pulse repetition frequency (~ 200 Hz) operation of an XeCl^* laser. Fujii et al. found that resistive stabilization inhibited arc formation from gas density and temperature fluctuations within the discharge volume. If a hot column of gas from a preceding discharge event remained in place between an unballasted pair of electrodes, it would form a favoured path for arc formation. The provision of resistive ballasting inhibited the degree to which such regions could form such paths.

C H A P T E R F O U R

A long pulse duration (LPD) XeCl* Laser: design, construction, and performance.

4.1 DESIGN CONSTRAINTS:

The overall objective of the work described in this thesis was to find if there was a fundamental limitation to the maximum pulse length obtainable from a RGH laser - apart from those associated with providing sufficient input power and dissipating the heat consequently generated. To achieve this goal it was not only necessary to design and construct a device capable of sustaining a stable discharge of long duration, but also to provide the device with a sufficient degree of 'flexibility' that extensive parametric characterization was possible.

The ease with which the resistivity of electrolyte ballasted electrodes could be altered to suit the demands of discharge stability and those of the driver circuit was the major reason for continuing with that method. The fact that liquid resistors are 'self-healing' in the event of failure of the resistive material was also considered to be a bonus - reliable operation was essential from the point of view of saving time and from that of reproducibility of results.

The prototype electrode, though successful in the limited objectives set for it, suffered from limitations both in its performance as a device and in the ease with which measurements could be made since:

1. although the discharge voltage could be measured satisfactorily, no simple method of measuring the discharge current was available because of the presence of the internal 'peaking' capacitor bank;

LONG DURATION XeCl DISCHARGE DEVICE

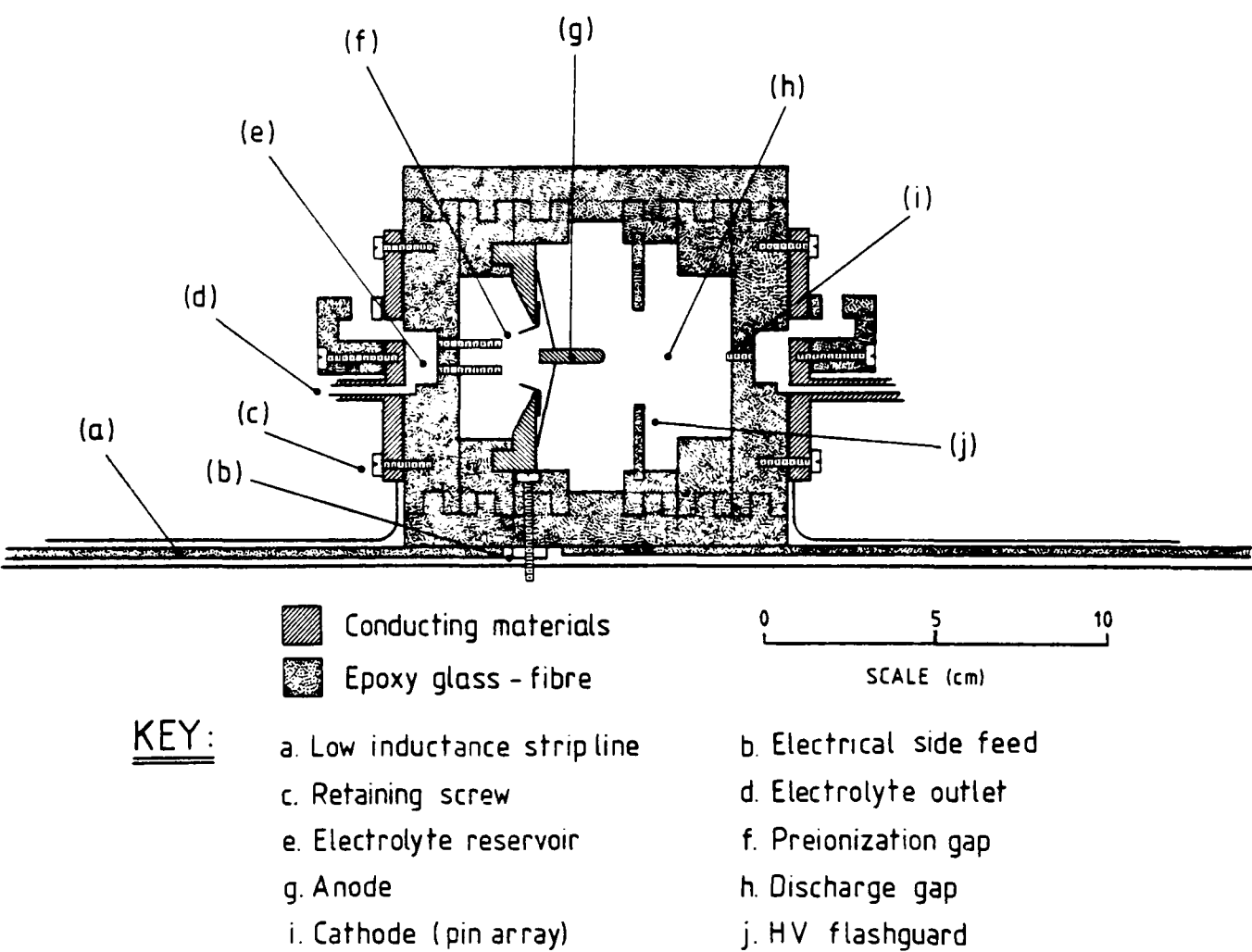


Figure 4.1

2. visual observation of the discharge to assess its spatial uniformity was difficult: the discharge could be viewed only at a grazing angle to the optic axis;
3. measurements of emission through a thin section of discharge - to determine the importance (or otherwise) of transient absorption - could not be made;
4. at high values of ballast resistance, breakdown tended occur between the ballasted electrode support plate and the 'earth-return' electrode or its support plate. Arcs also 'tracked' across the surface of insulators connecting pulsed HT and earthed elements inside the laser envelope;
5. some electrolyte leakage was evident between the copper backing plate and the glass-fibre laminate;
6. interpretation of the variation of laser pulse duration with, for example, the ballast resistance in the prototype laser was not straightforward because the driver circuit did not supply power to the discharge in a well defined manner. A fast pulse forming network (PFN) with the facility for alteration of both the duration and magnitude of the power delivered to the circuit - without altering the shape of the electrical pulse - would overcome this limitation. Matching a 'lumped' circuit element such as the 'peaking' capacitor bank to any external circuit in order to provide smooth continuous power input would be difficult;
7. if 'peaking' capacitors were to be excluded to satisfy (1) and (6) above then a new preionization scheme would have to be instituted. Also, the cross-section of the laser head would have to be small to minimize enclosed inductance loops (important from the point of view of minimizing current risetime).

In view of these shortcomings of the prototype device, the design and construction of a new laser (the long pulse device) which would avoid most, if not all, of these shortcomings was undertaken.

DEVICE FOR LONG DURATION XeCl LASER PULSE STUDIES

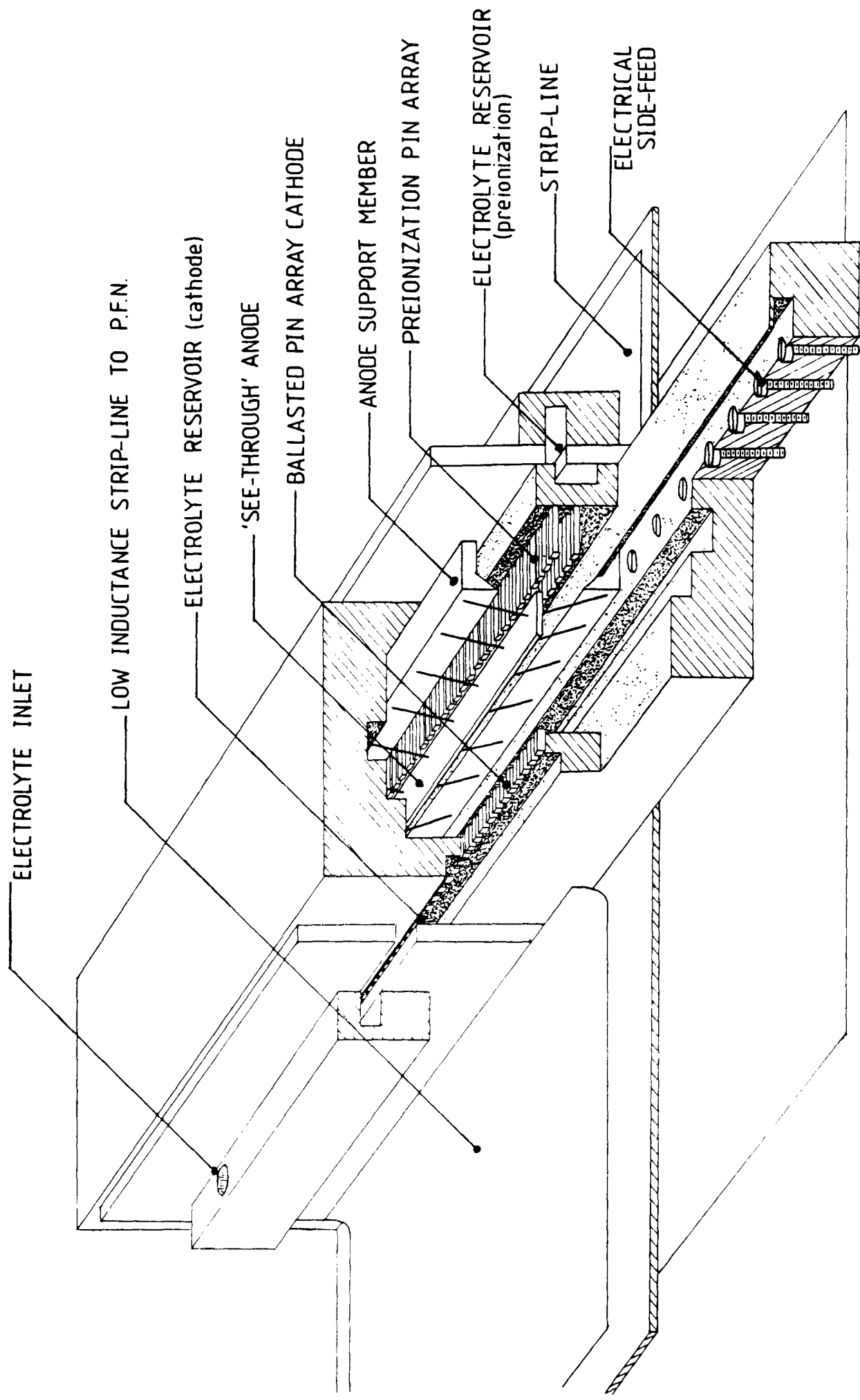


Figure 4.2

4.2 CONSTRUCTION:

The design of the long pulse duration (LPD) laser is shown schematically in cross-section in figure 4.1. Two more figures, 4.2 and 4.3, help to provide a clear view of the overall structure: figure 4.2 shows the device in cutaway relief and figure 4.3 illustrates the detail of the (removable) unballasted electrode. In this section the degree of compliance of the laser head design with the desiderata specified above will be discussed.

The first point to note is the absence of internal 'peaking' capacitors. The current loop runs from the ballasted electrode, fig.4.1(i), to the unballasted electrode(g), through the side feed screws(b) and back along the low-inductance strip-line to the ballasted electrode. This current loop encloses a similar cross-sectional area to that enclosed by the current loop in the prototype laser, shown in figure 3.2, which runs from one electrode to the other then back through the internal capacitor bank. However, because the length of the device is approximately half that of the prototype and, unlike the prototype, current is fed from one side only, the inductance of the head (eq.3.4) is ~ 8 nH. Two objectives were therefore achieved:

1. discharge current could be measured, for example, by placing a small resistor in series with the discharge circuit and external to the gas envelope;
2. if it should be found to be necessary, 'peaking' capacitors could be included in the circuit without having a significantly larger inherent circuit inductance.

The ballasted electrode was embedded in one wall of the laser envelope so that the electrolyte reservoir, together with its backing plate, could be positioned external to the volume. The problem of flashover breakdown causing conduction between objects other than the ballasted electrode was eliminated - no other conductors were present. Electrolyte leakage was also no longer a problem since only the tips of the individual ballasted screws penetrated the gas volume. Electrical flashguards (figure 4.1(j)) increased the distance a surface tracking arc had to follow - its likelihood of occurrence was thus

"See-through" Anode: Study Device (relief)

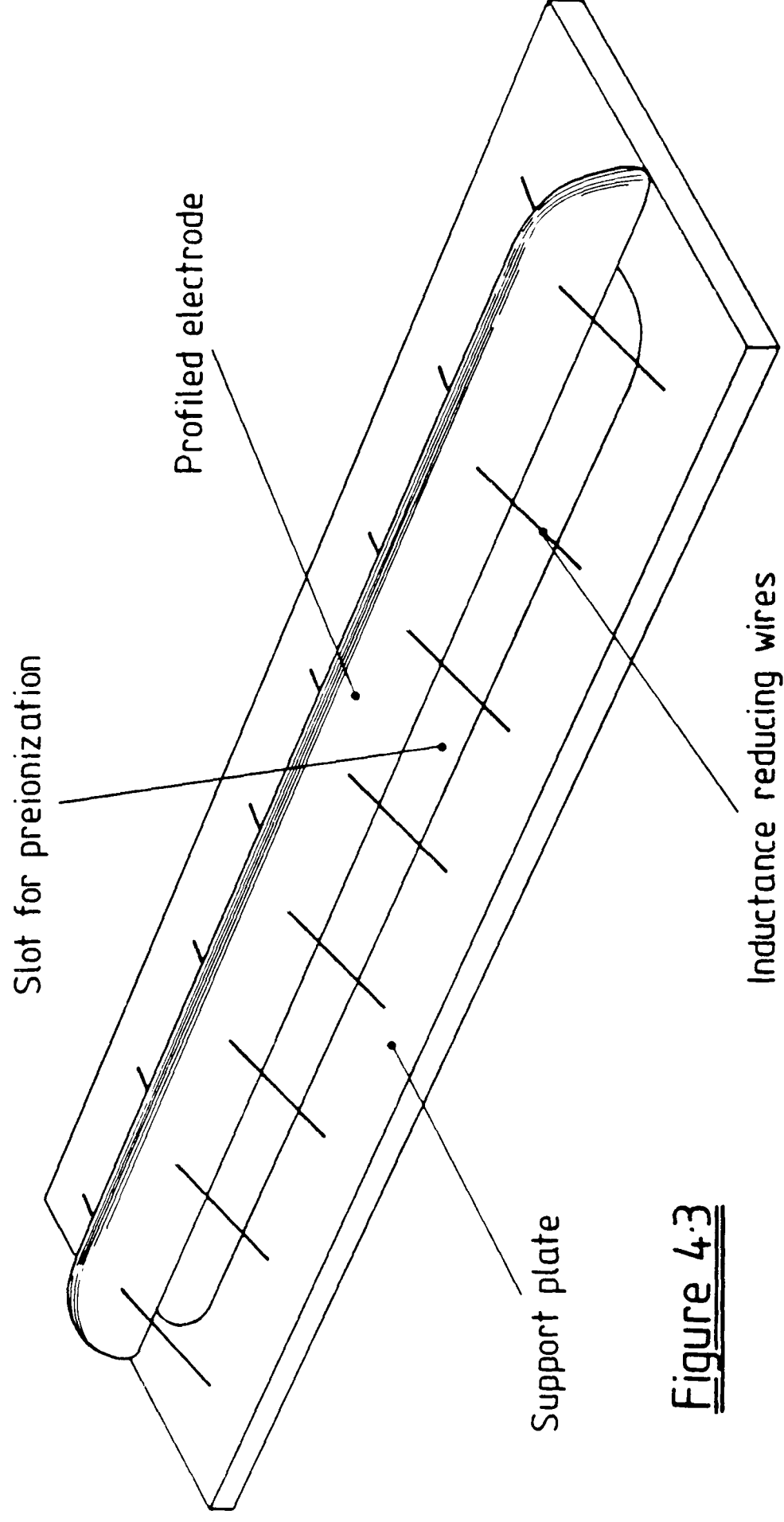


Figure 4:3

Table 4.1
Details of LPD laser head

Gas envelope:

Material:	1.3 cm thick epoxy glassfibre sheets bonded together with 'Araldite' epoxy adhesive
Enclosed volume:	~1 litre
Depth of Electrolyte reservoirs:	~7 mm

Cathode: (ballasted pin array - single row)

Material:	Brass screws (10BA) ~1.8 mm diameter
Length:	~38 cm
Inter-screw spacing:	~1.8 mm
Total number of screws:	~105

Anode: ('see-through')

Material:	Copper bar, profiled and polished
Length:	~39 cm
Width:	~3.2 mm

Preionization: (three rows of ballasted pins)

Material:	Brass screws (10BA)
Length of Array:	~39 cm
Total number of screws:	~330 (only two rows of pins contributed to preionization.)
'Spark' gap:	~3 mm

Discharge:

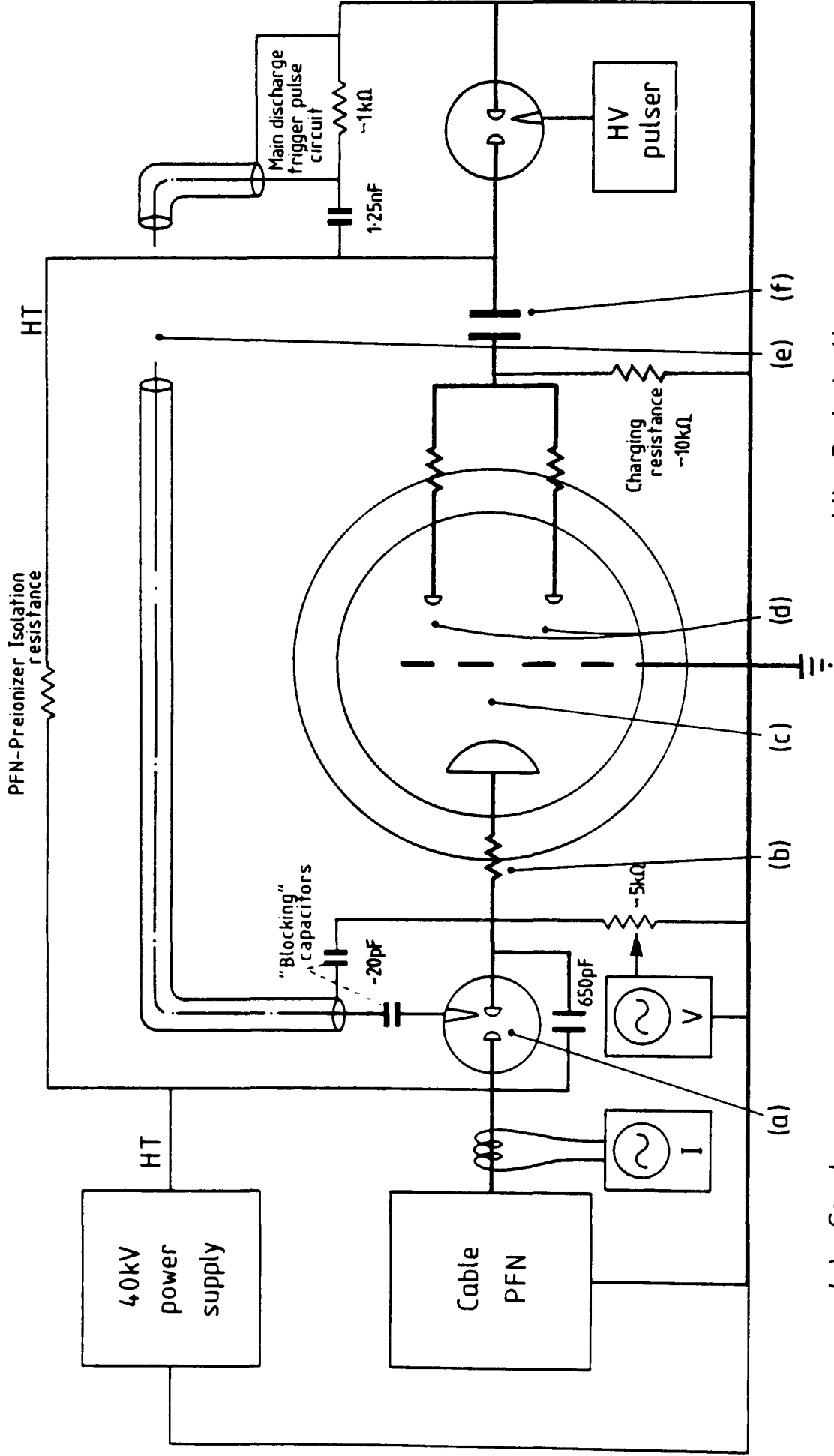
Length:	~38 cm
Width:	~1.8 mm (diameter of each ballasted screw)
Gap:	~2.2 cm
Discharge volume (LxWxG)	~15 cm ³

reduced.

Preionization was provided by arcs which bridged two rows of ~100 screws (figure 4.1(f)) and thin, sharp-edged, copper sheets attached to a 'see-through' unballasted electrode. The preionization pins were also ballasted by an electrolyte bath to ensure that an arc was formed at each of the 200 pins. Extremely uniform preionization, powered by a circuit separate from that for the discharge, was thus provided along the entire discharge length.

A 'sidelight' window (not shown in the figures) was provided in the top wall of the laser head (that wall nearest the letter '(g)' in figure 4.1). This permitted the observation of the discharge in a direction perpendicular to both the optic axis and the direction of the transverse electric discharge gap.

L.P.D LASER : DRIVER CIRCUIT SCHEMATIC



- (a) Spark gap
- (b) Resistive ballasting
- (c) Main discharge gap
- (d) Preionization arcs
- (e) Preionizer - main discharge delay line
- (f) 20 nF capacitor PREIONIZER circuit

Figure 4.4

Consequently, visual observation of discharge uniformity and quantitative measurements of emission were able to be made through a thin (~ 2 mm) section of discharge.

The specific details of construction of the laser head are not crucial to the discussion which follows and, consequently, they have been included as an appendix (appendix 2). Since certain comments made therein will explain more clearly how this particular design was arrived at, the reader is recommended to read it at a later stage. Table 4.1 provides a summary of specific details of dimensions of the LPD laser head.

4.3 THE ELECTRICAL CIRCUITS:

The absence of a 'peaking' capacitor bank meant that the 'automatic preionization' method could not be used; therefore, two circuits would have to be provided for the LPD laser: one circuit to drive the main discharge and one to drive the preionization array. Experiments performed by Smith[1982] with a flashboard (sliding-spark) preionization scheme indicated that there was a non-zero optimum value for the delay time between the firing of the preionization circuit and that of the main discharge in his laser: a value which was of the order of tens of nanoseconds. Therefore, a low-jitter method had to be found to fire the two circuits of the LPD laser. Fortunately this problem was one which had already been encountered by Kearsley[1980] and the solution was straightforward. Two spark gaps, each of which combined the features of 'triggertron' and 'field enhancement' gaps, were used with excellent results. One was triggered by a 'normal' low voltage circuit, and it in turn provided a sharply rising, high-voltage (~ 30 kV) initiation pulse for the other at an optimized and well defined (< 5 ns jitter) delay: the duration of the delay was determined by the length of a coaxial cable delay line. Figure 4.4 shows schematically the entire electrical circuit for the LPD laser. The discharge driver and preionization driver circuits will be described separately.

4.3.1 The discharge driver circuit

It was realised that the requirements for the discharge driver circuit for

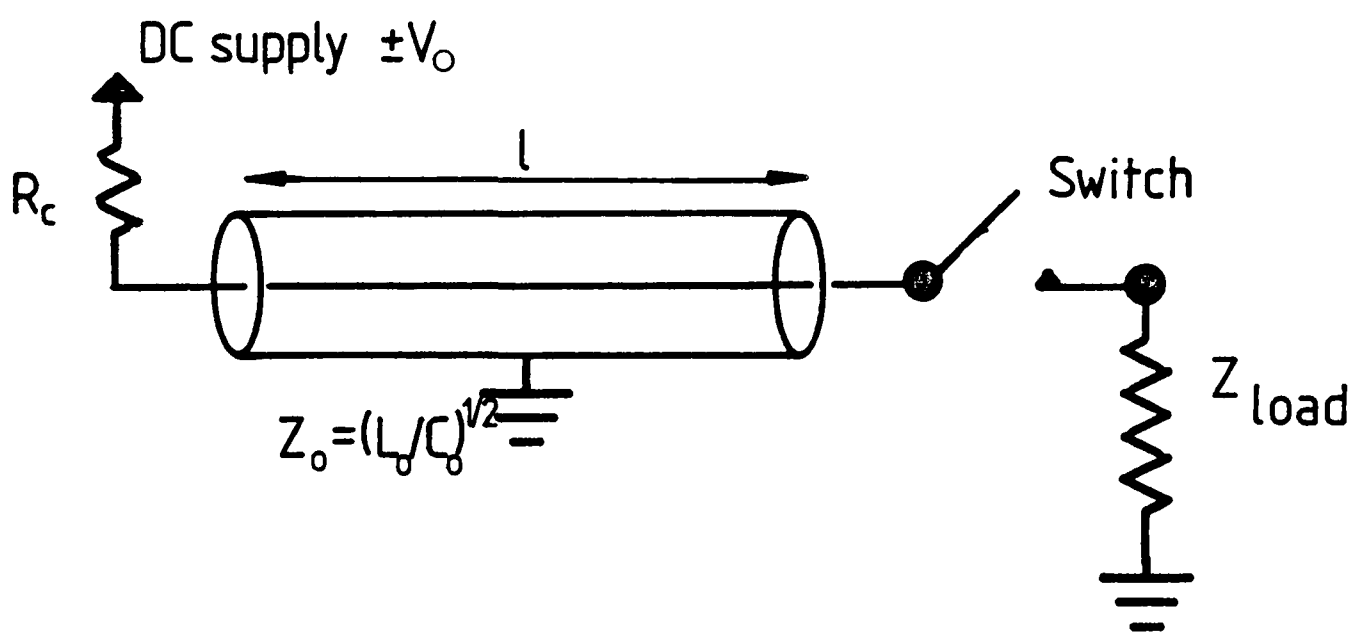


Figure 4-5: Schematic of a cable PFN

this LPD laser would best be served by a coaxial cable pulse forming network (cable PFN). Such a device would be capable not only of delivering temporally 'square' power input pulses to the discharge but it would also be a simple matter to alter both the magnitude and the duration of the pulses.

(a) Cable Pulse Forming Networks:

The principles of operation of cable PFNs (otherwise known as discharge line generators) are described by Lewis and Wells[1954] and by Craggs and Meek[1954]. However it will be useful to recall some of their properties here.

A loss-free, open-ended transmission line (figure 4.5) is initially charged to a DC potential V_0 through a resistance R_c (which is sufficiently large to isolate the supply from the line on the time scales required to discharge the line. When the switch is closed a voltage,

$$V_L = Z_L V_0 / (Z_L + Z_0) \quad (4.1),$$

is developed across the load, where Z_L is the impedance of the load and Z_0 is the characteristic impedance of the line. A current,

$$I = V_0 / (Z_L + Z_0) \quad (4.2),$$

flows through Z_L . The pulse current is sustained by a voltage wave, $V_0 - Z_L I$, which travels in the cable with a velocity

$$v_{\text{propagation}} = \frac{1}{\sqrt{L_0 C_0}} \quad (4.3),$$

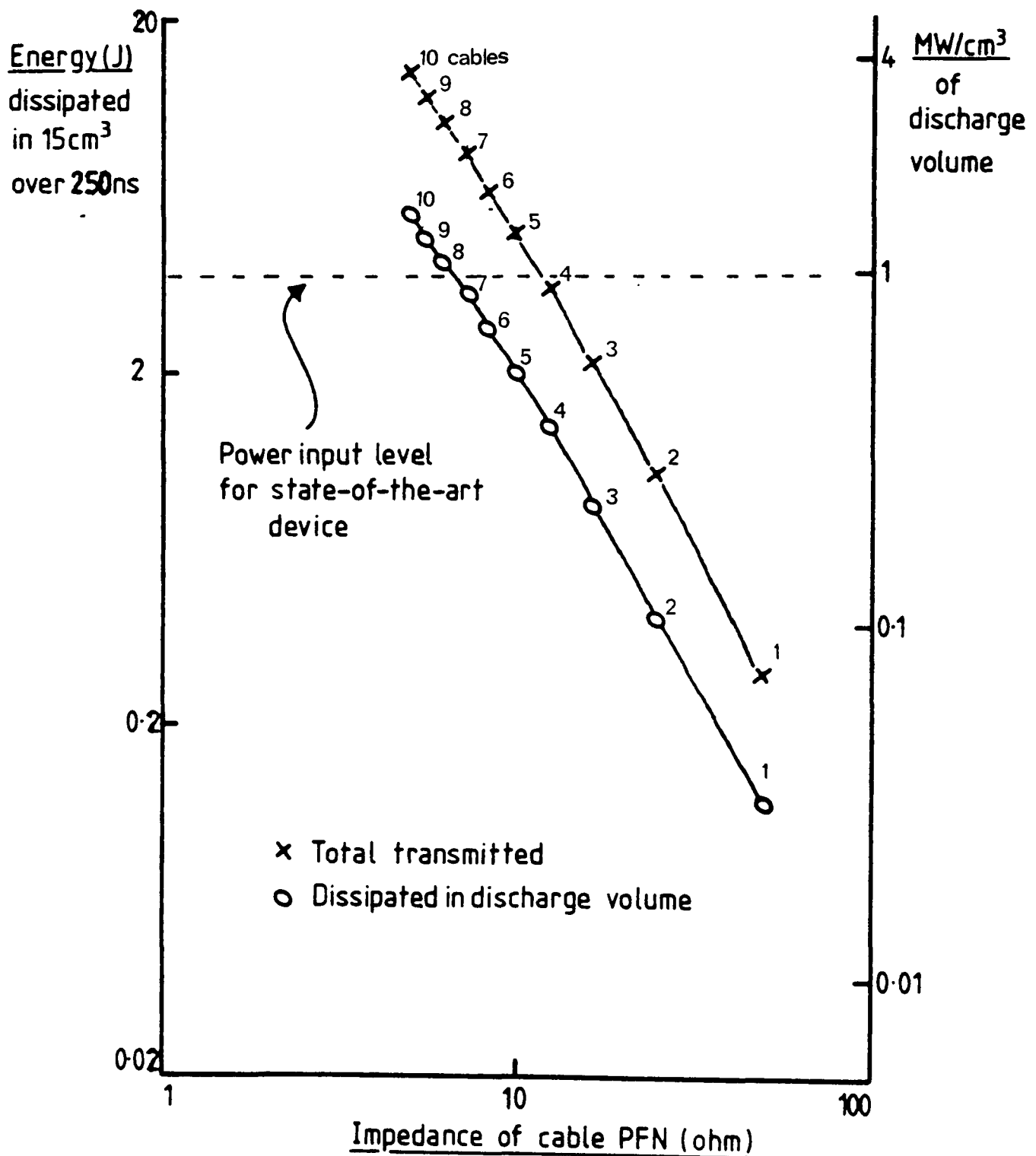
where L_0 is the inductance/unit length and C_0 the capacitance/unit length of the transmission line. This wave is reflected at the far, open-circuited, end of the cable and returns after a time,

$$t_{DT} = 2\ell \sqrt{L_0 C_0} \quad (4.4),$$

in such a sense as to reduce the current in the load, where ' ℓ ' is the length of the cable. This time is known as the 'double transit time' of the line. In the case of an impedance 'matched' line and load ($Z_L = Z_0$), all of the energy stored in the line is deposited in the load and the pulse is terminated after a time equal to t_{DT} has elapsed.

If the load impedance differs from the characteristic impedance of the line, reflection of voltage and current will occur at the termination. A voltage reflection coefficient can be defined:

Estimation of power delivered to a 'mismatched' load by cable-PFNs of different impedances



Charge Voltage=35kV; Z_0 (1 cable)=50ohm; 12.5m cables
 Z_{load} =2.5 ohm :- 1 ohm discharge; 1.5 ohm ballast.

Figure 4.6

$$\rho \text{ (voltage)} = (Z_L - Z_0)/(Z_L + Z_0) \quad (4.5).$$

In the case of a purely resistive load, R_L , a power reflection coefficient can be obtained from eq.4.5:

$$\rho \text{ (power reflected)} = (R_L - Z_0)^2/(R_L + Z_0)^2 \quad (4.6).$$

The fraction of power transmitted to the load,

$$\rho \text{ (power transmitted)} = 1 - (R_L - Z_0)^2/(R_L + Z_0)^2 \quad (4.7),$$

is, therefore, a strong function of the impedance match of the load to the line.

Equation 4.7 was of prime importance in the determination of the suitability of cable-PFNs for powering the LPD laser. The values of current and voltage estimated by Kearsley[1980] for his 'standard' XeCl^* laser indicated that the instantaneous ratio of voltage to current (or 'pseudo' resistance) of that discharge was ~ 1 ohm. If commercially-available high voltage coaxial cables (usually $Z_0 = 50$ ohm) were to be used, the question arose of whether sufficient power could be provided to the discharge for lasing to occur by a PFN consisting of less than (say) twenty such cables. Kearsley[1980] suggested that a minimum input power of $\sim 1 \text{ MW/cm}^3$ would be necessary for efficient operation of an XeCl^* laser.

The energy, E_{line} , stored in a transmission line, of length ' ℓ ' and charged to a voltage V_0 , is given by the relation

$$E_{\text{line}} = \frac{1}{2} \ell C_0 V_0^2 \quad (4.8).$$

The energy, E_n , stored in a PFN consisting of ' n ' such cables connected in parallel is simply

$$E_n = \frac{1}{2} n \ell C_0 V_0^2 \quad (4.9),$$

and the impedance, Z_{PFN} , of the pulse forming network is

$$Z_{\text{PFN}} = Z_0/n \quad (4.10),$$

since the characteristic inductances and capacitances of the cables are in parallel.

Equations 4.7, 4.9, and 4.10 can be combined to provide an estimate of the power transmitted to a load of constant (purely resistive) impedance as a function of an ' n '-cable-PFN charged to some initial voltage V_0 . Figure 4.6 shows the results of such an estimate for cable-PFNs consisting of from one to

ten 50 ohm coaxial cables connected in parallel and charged to 35 kV. The cables used throughout were 'Uniradio 67' H.V. coaxial cables with $C_0=100\text{pF/m}$ and $L_0=250\text{nH/m}$. A load resistance of 2.5 ohm was assumed: made up of a 1.5 ohm equivalent series resistance of the ballasted electrode and a 1 ohm pseudo-resistance for the discharge. A double transit time of 250 ns was assumed for each cable (25 m lengths). Two curves are shown in figure 4.6: one shows the total power delivered to the load, the other shows the power delivered to the discharge alone as a function of the PFN impedance. The abscissae give the power dissipated in MW/cm^3 and the total energy deposited in the 15cm^3 active volume of the LPD laser over 250 ns. A dashed line is drawn at the 1MW/cm^3 power input level - indicating that a PFN consisting of at least eight of these cables should be capable of delivering the minimum input power for efficient operation estimated by Kearsley[1980].

(b) Specific Details:

Initially it was decided to use a 'pulse-charged' rather than a 'DC-charged' cable PFN.[†] This was done by connecting one terminal of a 70 nF capacitor to the inner conductors of the cables (at what would normally have been their far, open-circuited end); the other terminal was connected (via a suitable charging resistor) to a H.V., D.C. supply. A spark gap connecting the high voltage side of the capacitor to 'earth' was fired and produced voltage inversion on the capacitor. This pulsed voltage source continuously charged the whole PFN until at some critical voltage (quite close to that initially applied to the capacitor, but of opposite polarity) the discharge gap, acting initially as the 'switch' but later as the 'load', broke down. The 'overvoltage' achieved before breakdown was sufficient to obtain uniform discharge initiation.

This 'pulse-charged' PFN was not suitable for two reasons:

1. The temporally square power input pulses, desired for unambiguous interpretation of the laser performance, were not achieved - this was

[†] In order to take advantage of the fact that the discharge gap could be used as both the 'switch' and the 'load' of figure 4.5. It was because of concern over the speed of the spark gaps available that DC charging was not used at first. In the event this concern did not prove to be warranted.

Coaxial cable - Strip line: low inductance connection

('cut-away' relief)

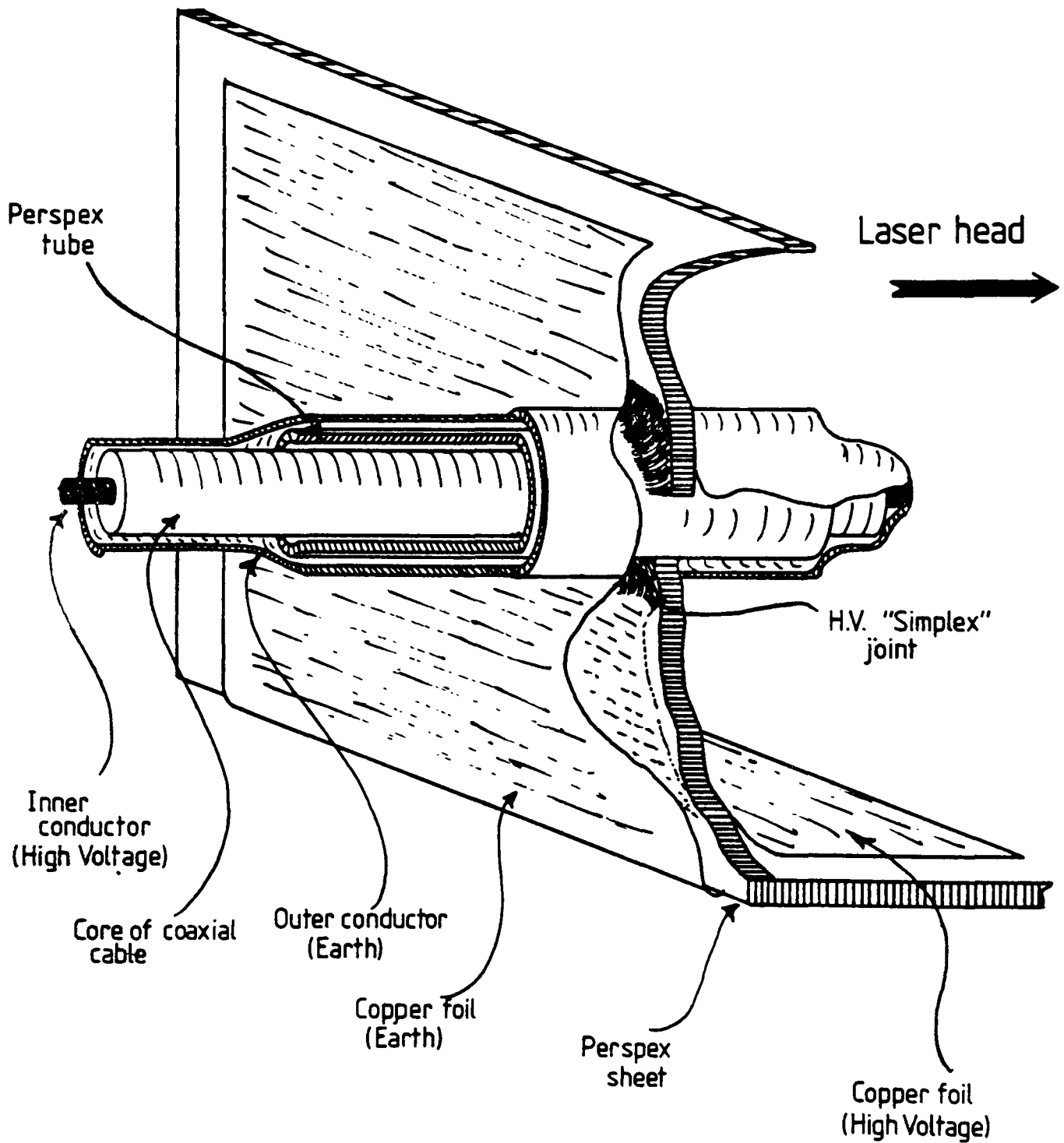


Figure 4.7

mainly due to a very slow current rise-time at discharge initiation.

2. The energy stored in the capacitor (and therefore eventually deposited in the load) was sufficient to cause a shock wave which invariably ejected the electrolytic solution from its containment reservoir and sprayed it all over the high voltage circuitry. The subsequent D.C. flashover problems were, to say the least, time wasting.

The first problem was not a result of using a pulse charged configuration as such but rather one due to poor connections between the cables of the PFN and the strip-line (fig.4.1) connecting them to the discharge gap. Straightforward connection of the inner conductor of each cable to one side of the strip-line and the outer conductor to the other resulted in the presence of a large stray inductance. This inductive component of the load, although contributing little to the total load impedance before breakdown, was sufficiently large in the breakdown phase to slow the current rise-time significantly.[†] The solution to this problem was to provide a low inductance connection between the cables and the strip-line: such a connection is shown schematically in figure 4.7. Perspex tubes were bonded into flat perspex sheet using a fast setting methyl-methacrylate bonding agent ('Simplex'); the tubes and the sheet were clad in copper foil and the inner conductor of each cable was passed down one of the tubes. The inner and outer conductors of the cables were then soldered to their respective copper foils.

In order to overcome the second problem it was decided to try to operate the PFN in the DC-charged mode so that only that energy stored in the cables would be transferred to the load. The discharge cannot be used as its own switch in this regime. The maximum charging rate of the line when coupled to a typical high voltage D.C. supply is so slow that breakdown would almost certainly occur as soon as the minimum breakdown potential is reached. The low voltage achieved will not only result in the initiation of a discharge with poor uniformity because of the low breakdown voltage (section 2.3.3(a)) but also the energy stored in the PFN - and subsequently delivered to the discharge will be

[†] The impedance of the gap before breakdown is $\sim 10^{10}$ ohm.

low. An 'in series' switch must therefore be provided. A small (500pF) capacitor placed in parallel with a spark gap of the 'standard' type described by Kearsley[1980] encouraged very fast breakdown of the gap and also reduced timing jitter with the preionization circuit. Care was also taken in the 'termination' of the 'open-circuited' ends of the cables: sleeves made of perspex tube, sealed at one end, enclosed the inner conductor of each cable. These sleeves were clad in copper foil which then attached to the outer conductor of each cable.

Typical voltage and current waveforms from a 250ns pulse-charged, 5 ohm PFN and from a 500ns DC-charged, 12.5 ohm PFN are provided for comparison in plate 4.1. The magnitudes of voltage and current (in the first double transit) are comparable in the two cases - $\sim 2\text{kA}$ and $\sim 10\text{kV}$. As can be seen, the current rise-time in the pulse-charged line was comparatively long. On the other hand the short current rise-time and the signal-to-noise quality of the current and voltage waveforms obtained from the DC-charged PFN exceeded all expectations: the clarity of the recorded waveforms and 'smoothness' of power dissipation achieved was of enormous benefit in the understanding of the performance of the LPD laser.

All subsequent experiments were carried out with a DC-charged PFN consisting, unless otherwise stated, of four 50m (500ns double transit time), 50 ohm coaxial cables. The power input level achieved with this PFN was about 0.7 MW/cm^3 - significantly higher than that predicted for a 12.5 ohm PFN in figure 4.6. This discrepancy can be accounted for by the fact that the effective 'resistance' of the discharge was much higher ($\sim 2.5\text{ ohm}$ - neon buffer; $\sim 4.7\text{ ohm}$ - helium buffer) in the LPD laser than was estimated by Kearsley[1980] for his laser ($\sim 1\text{ ohm}$): consequently the 'line-load mismatch' was not as great. The difference between the actual and estimated discharge resistance can be accounted for by the much larger cross-sectional area available for current flow and higher electron density in Kearsley's laser. The $\sim 0.7\text{ MW/cm}^3$ power input level from the 4-cable PFN is slightly lower than the 1 MW/cm^3 target set initially but, as is shown later, increasing power deposition beyond this level

resulted in laser pulses of shorter duration.

4.3.2 The Preionization Circuit:

Two hypotheses, both based upon experimental observations by Dr.A.J.Kearsley and others at the Clarendon Laboratory, formed the basis for the design of the preionizer circuit. The first was that discharge uniformity was sensitive to both the spatial uniformity of the preionizing radiation and to its timing with respect to the onset of the main discharge. The second was that long duration (~1 microsecond) preionization pulses exhibited no advantage over short duration (~100ns) pulses.

It was hoped that the 220 ballasted preionization pins along the length of the discharge would provide preionization of good spatial uniformity when powered by a simple capacitive discharge circuit of short pulse duration. The capacitor bank initially tested consisted of eight 2.5 nF BaTiO₃ capacitors connected in parallel and incorporated into a strip line to increase the speed of the circuit. However, problems with electrolyte 'ejection' - as with the pulse-charged PFN - forced a reduction to five 2.5 nF capacitors. A strip line connected the capacitors to the preionization pins on one side and to a spark gap on the other; DC high voltage was applied between the spark gap and the power supply (the same power supply which charged the cable-PFN).[†] When the preionizer circuit spark gap was fired the voltage on the capacitor bank was inverted and a high voltage developed across the gap between the preionization pins and the 'see-through' anode inside the gas envelope. The visible (and near UV) light pulse emitted from the preionization spark array was observed to have an overall duration (foot-to-foot) of ~120 ns.

4.4 THE OPTICAL CAVITY:

Various optical cavities were employed. Initially, 50 mm diameter silica flats (soft-dielectric coated with nominal reflectivities of 99.6% and 96%) were used. These mirrors were mounted directly onto the end flanges of the gas

[†] A large resistance (~10 k-ohm) was placed between the PFN and the preionizer circuit to isolate one from the other.

envelope. Although a limited amount of adjustment (barely sufficient to enable alignment) was possible without compromising the integrity of the gas volume, this arrangement was inconvenient from the point of view of changing mirror reflectivities. It was also undesirable to open the 'passivated' volume to the atmosphere.

At various times either a set of windows with a 3° off-axis tilt or a set tilted at the full Brewster angle were used in conjunction with externally mounted mirrors. For the majority of experiments in which lasing was to be observed 3 m radius of curvature concave mirrors (96%:99.6% reflectivities) and Brewster angle windows were standard: not surprisingly, it was found that this confocal cavity was far easier to align than a plane-plane cavity. Furthermore, slightly longer laser pulse durations were obtained due to a small, but easily discernible, decrease in the cavity 'build-up' time.

Lasing could be obtained with output coupling mirrors having as little as ~25% reflectivity but, at this low reflectivity, both pulse energy and duration were curtailed - again due to increased cavity 'build-up' times. The use of a 50% output coupling mirror resulted in pulse energies approximately twice as large as those obtained with the standard - 96% reflectivity - output coupler (with only a 20% reduction in FWHM laser pulse duration).

Windows with a 3° off-axis tilt were used in all absorption experiments which required the passage of a 'reference' beam through an unexcited region of the gas volume.

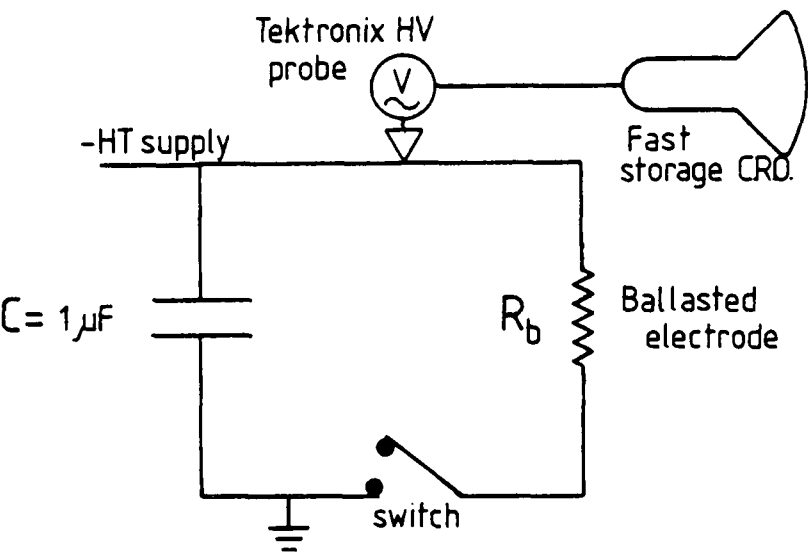
4.5 EXPERIMENTAL DETAILS:

4.5.1 Ballast resistance calibration:

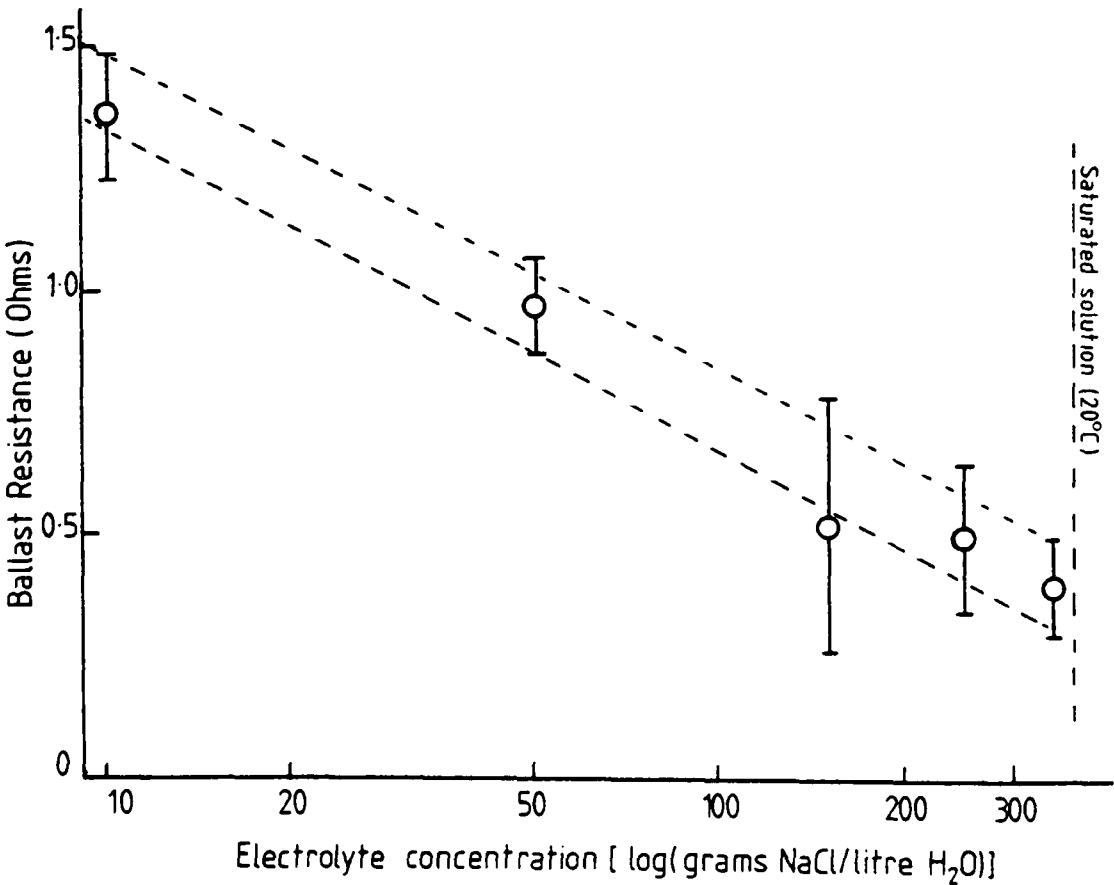
The equivalent series resistance of the ballasted electrode was measured in the LPD laser by measuring the decay of voltage in an RC circuit in which the ballasted electrode was the resistive element. The PFN was disconnected from the laser head and a $1\ \mu\text{F}$ capacitor was connected in series with the ballasted electrode on the 'backing plate' side. A lead was connected to an assembly which 'shorted out' all the pins of the ballasted array on the side which faced

Determination of the variation of the series equivalent resistance of the ballasted electrode with electrolyte concentration.

(a) circuit schematic



(b) results



(a) Figure 4.8 (b) Figure 4.9

the anode (care was taken to minimize contact resistance). This lead was touched to earth to discharge the RC circuit formed by the capacitor and the electrode (figure 4.8). A 'Tektronix' HV probe was used to provide a voltage decay waveform on a 466 storage oscilloscope.

In any RC series circuit initially charged to a voltage V_0 , the voltage, $V(t)$, measured across R at a time t is given by:

$$V(t) = V_0 e^{-(t/RC)} \quad (4.11).$$

If $V(t + \Delta t)/V(t) = e^{-1}$ then

$$R = \Delta t / C \quad (4.12).$$

The ballast resistance was determined using eq.4.12 for different concentration aqueous solutions of NaCl placed in the ballasted electrode. The time taken for voltage to decay to 1/e of its original value was measured five times for each solution and the standard deviation of the resultant resistances was taken as a measure of the error. The results thus obtained are presented in figure 4.9: the rather large error bars are indicative of the uncertainty in the measurement the '1/e points' from the oscilloscope traces.

Predicted resistances for the ballasted bath, obtained by using the approximation derived in chapter 3 (eq.3.3) and tabulated values of conductivity vs. concentration (Hodgman[1962]) at 18°C (no measurement was made of the conductivity of each solution) were in good agreement with the experimentally determined resistance. Higher resistances were determined by extrapolation of the plot of measured resistance in figure 4.9.

4.5.2 Voltage and current measurements:

The discharge current was measured using a Rogowski coil (Pearson type no 411) which was mounted coaxially with the inner conductor of one of the cables of the PFN near its connection to the strip-line. The calibration factor, quoted by the manufacturer for conversion of the measured voltage to current, was 10 A/V. The total current is related to the measured current by:

$$I_{\text{total}} = I_{\text{measured}} \times n_{\text{cables}} \quad (4.13).$$

The voltage was measured using a low inductance voltage divider of the type described by Kearsley[1980] which was connected across the discharge and

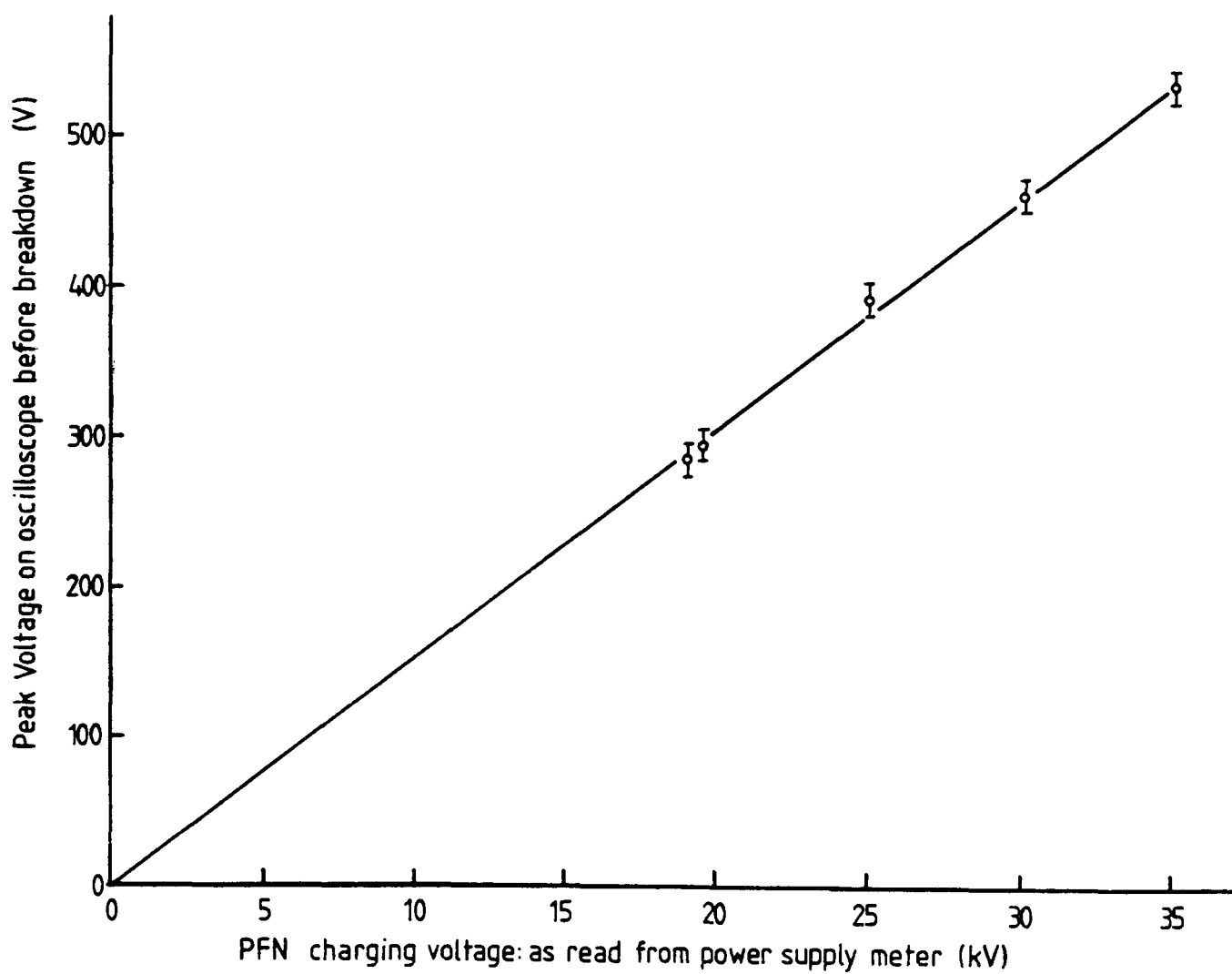
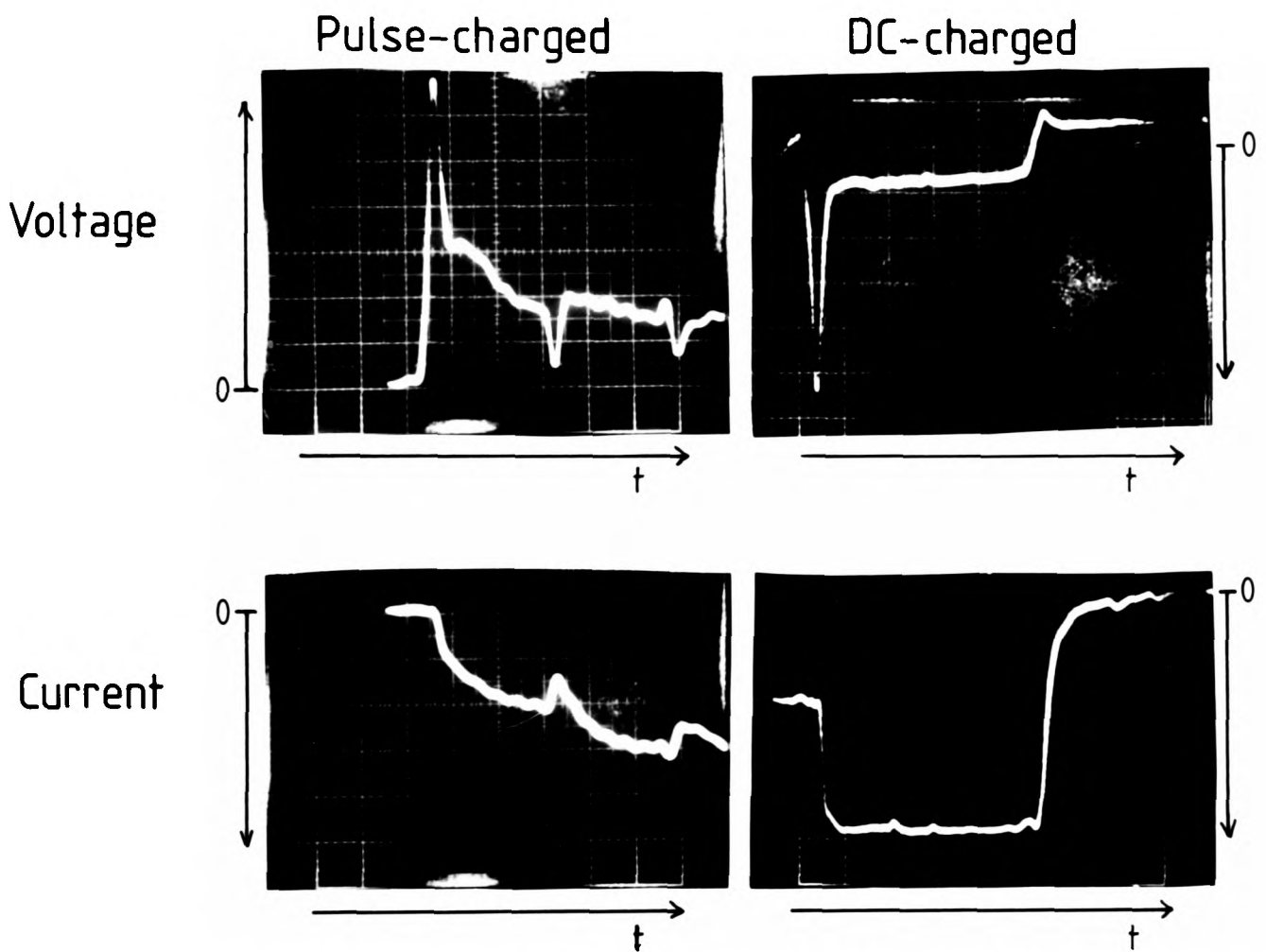


FIGURE 4.10: Voltage divider calibration curve

Comparison of Voltage & Current pulse shapes from pulse-charged & D.C.-charged cable-P.F.N.s



Time scale: 100ns/division. Pulse magnitudes are NOT relative.

Pulse-charged PFN: 10×25metre, 50ohm cables.

DC-charged PFN: 4×50metre, 50ohm cables.

Trace of pulse-charged PFN's voltage is inverse polarity of real voltage.

Plate 4.1

the ballasted electrode. The actual voltage appearing across the discharge gap was thus obtained indirectly by subtracting that part of the measured voltage which was dropped across the ballasted electrode:

$$V_{\text{discharge}} = V_{\text{measured}} - (R_{\text{ballast}} \times I_{\text{total}}) \quad (4.14).$$

Calibration of the divider was carried out in situ. The laser head was filled with argon to a pressure of ~ 1 atmosphere. The higher breakdown voltage of argon, as compared to that of helium or neon, allowed a delay of ~ 200 ns between the breakdown of the spark gap and that of the discharge (plate 4.2). The peak voltage as measured at the oscilloscope was assumed to be directly related to that initially applied to the PFN (measured by the voltage meter on the power supply). Figure 4.10 shows the voltage on the divider as a function of the initial charging voltage of the PFN. All current and voltage waveforms were recorded on a 'Tektronix' 466 storage oscilloscope except in the later stages of this work: by that time a fast transient digitizer (Biomation 8100) had been borrowed from the Rutherford-Appleton Laboratory. It was found to be convenient to record voltage and current in this way for manipulation by computer to obtain 'true' discharge voltage and power input waveforms. This equipment is described in greater detail in section 5.3. The results presented in figures 4.11-4.14 and 4.17 were obtained using this apparatus.

No problems were experienced with RF noise from the discharge and its associated circuitry. The entire laser assembly was contained within a 'Faraday cage' laboratory (its walls, floor, doors, and ceiling were clad in copper). The signals from the voltage and current probes were passed out of this 'Faraday cage' via well earthed coaxial 'feed-throughs'. Light was passed through waveguide portholes in the wall into an adjoining laboratory for detection.

Voltage and current traces are presented where it is considered necessary. The impedance mismatch between the 'load' and 'PFN' resulted in several distinct voltage and current after-pulses ('rings') through the discharge. Only the time history up to one double transit time of the cables will normally be considered since no emission at 308 nm was observed in the later 'rings' of each pulse. Voltage and current waveforms obtained from a

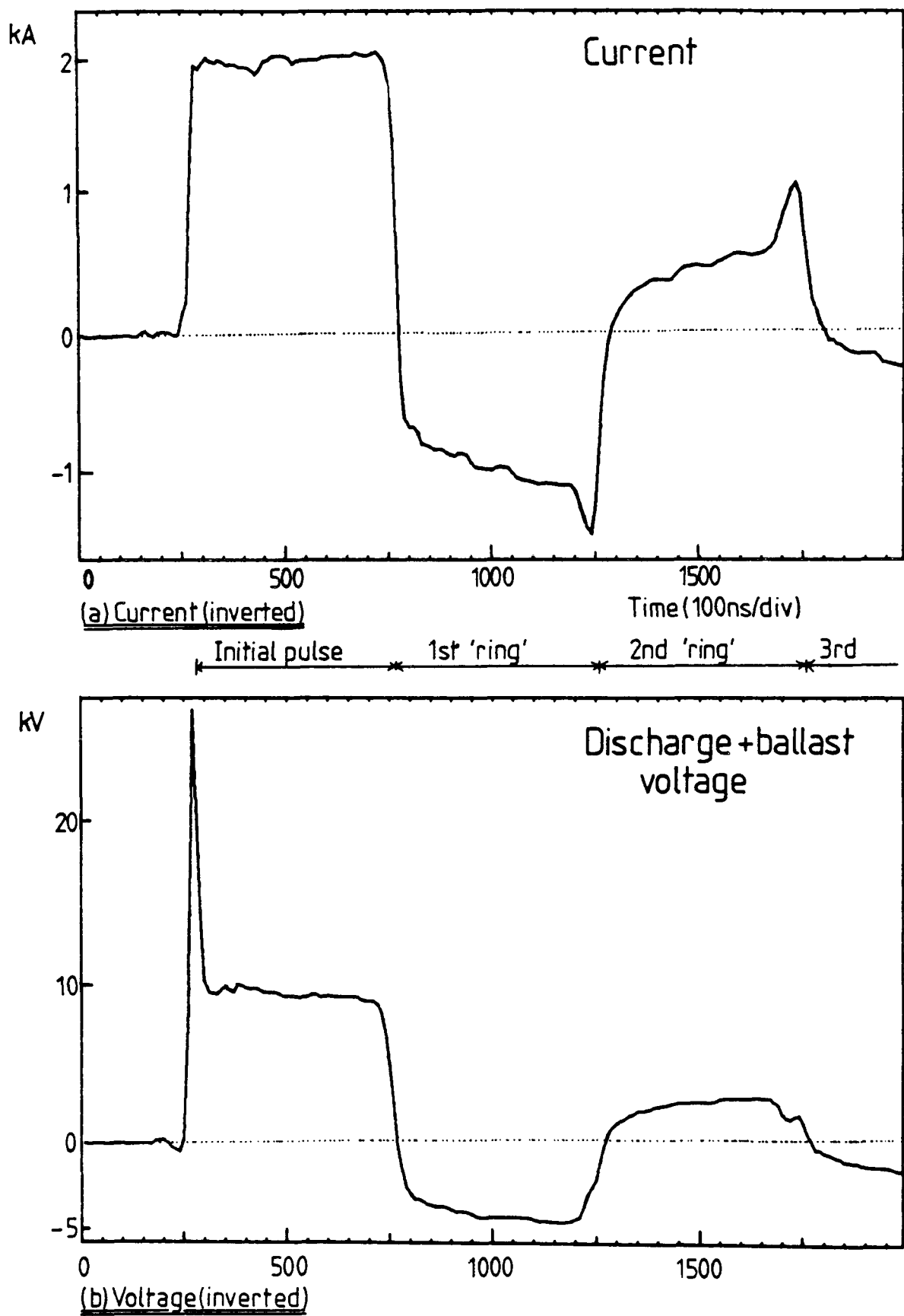


FIGURE 4.11: Temporal evolution of current & voltage in a Xe (25mbar) plus Ne (2.33atm) discharge driven by a 4-cable PFN. Figure shows 'ringing' of V & I at double transit periods.

Breakdown delay in pure Argon

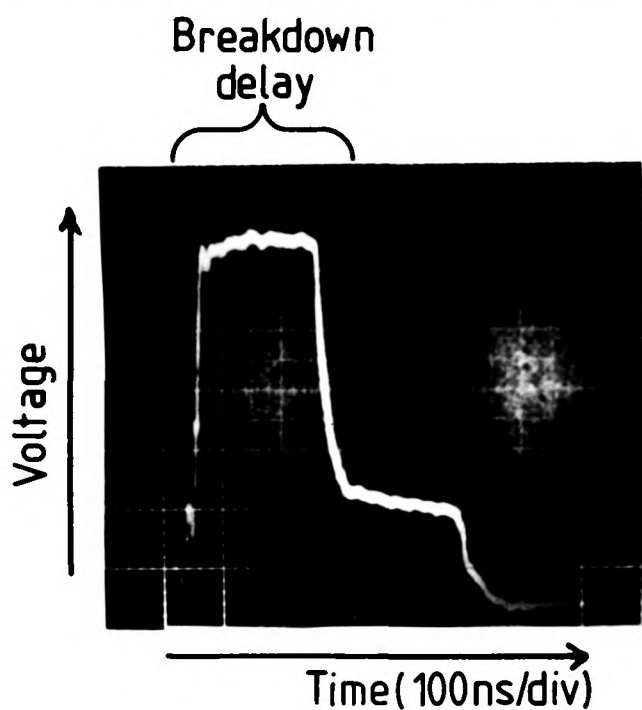


PLATE 4.2: Temporal evolution of voltage in pure argon (2.3 atm) showing delay in breakdown which was useful in calibrating voltage divider.

discharge containing 25 mbar of xenon and 2.3 atmosphere of neon are shown in figure 4.11. These traces exhibit the expected voltage and current inversion at each double transit period of ~ 500 ns which is typical of an 'under-terminated' transmission line. Voltage and current waveforms from gas mixtures containing HCl exhibited the same general appearance in the 'ringing' phase.

4.5.3 Light detection:

Generally the temporal evolution of XeCl^* emission at 308 nm was measured with a fast (sub-nanosecond) risetime UV photodiode of the same type used for the prototype laser. The intensity of 308 nm emission, both spontaneous and stimulated, so strongly dominated the output of the discharge that the intensity of emission at other wavelengths was negligible by comparison. Consequently, many optical measurements could be made without the need for wavelength discrimination.

Care had to be taken to avoid saturation of the photodiode in the detection of lasing. Two methods, both of which gave similar results, were used to reduce the signal level:

1. The light was reflected off glass microscope slides (up to three in sequence) before being allowed to fall on the photodiode;
2. UV fluorescent paper was placed over the aperture of the photodiode and the visible fluorescence due to the 308 nm emission was detected.

The fact that both these methods produced identical results meant that the second, and somewhat simpler, method was generally used without concern about the decay time of the UV fluorescer with which the paper was impregnated.

As will be discussed in section 4.6.1, it was quickly realised that termination of the laser pulse well before the end of the electrical excitation pulse was a persistent feature and that its cause was an overall decrease in formation of the XeCl B-state molecules. In view of this fact, direct monitoring and optimization of this population via its spontaneous emission was clearly a more straightforward and reliable indicator of the real quantity of interest: concern about mirror alignment and degradation of mirror reflectivity, which also affect laser performance, could thereby be eliminated. Consequently,

some of the results are couched in terms of laser pulse duration and/or energy whilst others are presented in terms of the relative intensity of spontaneous 308 nm emission. The spontaneous emission was generally measured as 'endlight' (along the optic axis) and was subject to single pass gain (estimated to be a net 200-300% over the 38cm discharge path). All spontaneous emission traces presented in plates were obtained from 'sidelight' emission and therefore were not subject to this single pass gain. There is, however, no doubt that the three quantities provide essentially the same information.

4.5.4 Comments on gas mixture composition:

The small gas volume enclosed by the LPD laser head (~1 litre) resulted in problems with the mixing of the component gases and the useful 'lifetime' of a single gas-fill. When the laser was filled with the gas mixture, two different gauges were used: a low-pressure gauge (Leybold-Heraeus) which gave readings in the 0-1000 mbar region on a logarithmic scale and a high-pressure gauge (Budenberg) for pressures in the range from atmospheric to 40 psig (3.5 atm). If the amounts of HCl and xenon in the gas mixture were to be known accurately then those two gases had to be admitted to the (evacuated) gas volume before the (high-pressure) buffer gas. The consequence of this was that the HCl and xenon were forced out of the laser head and into the (closed-off) vacuum lines (which were of comparable volume) when the buffer was added: a gas circulating compressor had to be added to the system to ensure reliable and reproducible operation. Later, a large (~40 litre) gas reservoir was also added to extend the useful lifetime of a single gas-fill.

The prohibitively high cost of xenon required that HCl partial pressure optimization had to be carried out using a small decanting volume which would allow the addition of HCl to the laser chamber after the (high-pressure) buffer gas had been added. Unfortunately, this meant a considerable reduction in the accuracy with which the HCl partial pressure could be measured. Some accurate tests were performed in the 1-4 mbar HCl partial pressure region; generally, however, values quoted in the text for HCl partial pressures are subject to an inaccuracy of ~1 mbar. When the value of a particular parameter is not stated

Table 4.2
Optimum parameter values for the LPD laser

<u>Driver Circuit:</u>	1. 4x50m, 50 ohm HV coaxial cables (Uniradio 67) in parallel ($Z_{PFN}=12.5$ ohm) 2. 35kV PFN charging voltage. 3. Double transit time ~ 500 ns (50 m length)
<u>Gas Mixture:</u>	1. 2.5 mbar HCl 2. 25 mbar xenon 3. 2.33 atm neon
<u>Ballasted Electrode:</u>	1. 10g/l aqueous solution of NaCl corresponding to: 2. equivalent series resistance of ~ 1.5 ohm 3. individual pin resistance of ~ 165 ohm
<u>Preionization:</u>	1. Fast (~ 100 ns) capacitive discharge into ~ 220 individual arcs spaced evenly along the length of the discharge. 2. total capacitance of circuit = 12.5 nF 3. charging voltage was the same as PFN at all times 4. ~ 40 ns delay between peak of light emission from preionization array and the onset of the discharge current pulse.
<u>Discharge:</u>	1. normal running voltage ~ 8 kV (less ~ 3 kV for voltage drop across ballast). 2. normal running current ~ 2 kA 3. ~ 0.65 MW/cm³ input power (initial stages) This was ~ 0.9 MW/cm³ for a helium buffered gas mixture with all other parameters unchanged.
<u>Optical Cavity:</u>	1. Brewster angle windows 2. 96% and 99.6% nominal reflectivity mirrors: 3 m radius of curvature; concave; Spectrosil 'B' grade silica; 25 mm diameter; 'hard' dielectric coatings 3. Separation of mirrors ~ 40 cm
<u>Lasing:</u>	1. Maximum XeCl* laser pulse duration = 270 ns FWHM with a pulse energy of ~ 5 mJ 2. ~ 12 mJ pulse energy, 200 ns FWHM with 50% reflectivity output coupler

explicitly it can be assumed that it has the value given in Table 4.2.

4.6 PARAMETRIC INVESTIGATIONS:

4.6.1 Typical results:

At this point it is worthwhile to anticipate some later results and examine the performance of the LPD laser at a stage of its development when parameters such as gas mixture, ballast resistance, and power input had been

optimized for the longest laser pulse duration.

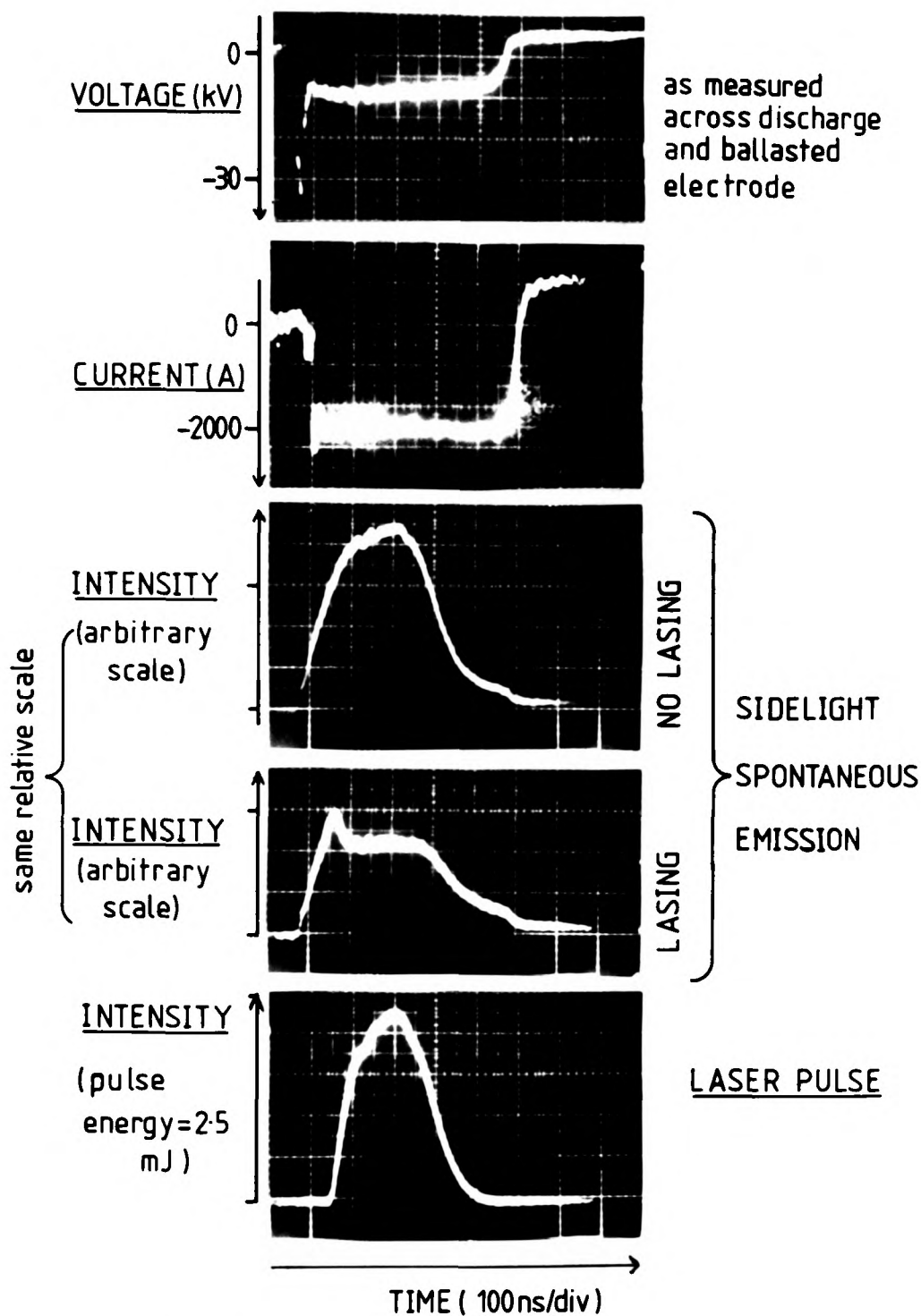
The longest pulse duration obtained from the LPD laser was 270 ns FWHM. Other self-sustained discharge devices, reported in the literature, which have come close (200 ns FWHM) to this pulse duration are a large-volume, high pulse energy, X-ray preionized device reported by Levatter et al.[1981], and a very low pulse energy microwave excited laser reported by Young et al.[1982]. This work will be discussed later but an important point to note now is that there is a distinct similarity in the behaviour of both these devices and the LPD laser: all show a premature termination of laser emission before the completion of the discharge excitation pulse.

The results presented in plate 4.3, though not optimum from the point of view of the longest pulse duration obtained, serve well to illustrate the nature of the problem which forms the focus of all further work discussed in this thesis. The temporal evolution of five quantities appears in this plate: the voltage developed across the discharge and the ballasted electrode; the current flowing through the discharge circuit; the 'sidelight' spontaneous emission at 308nm - observed in one case in the presence of lasing and in the other with the optical cavity removed; the laser emission. All of these traces are referenced in time with respect to one another with an accuracy of ± 10 ns. The voltage exhibits a fast rising leading edge before the breakdown of the discharge gap and then a constant 'running' voltage of ~ 8 kV is achieved. The current behaves in a similar manner: the current risetime is very fast (sub-nanosecond) and reaches a quasi 'steady-state' value at which it remains for the full duration of the PFN double transit time. As far as can be ascertained, then, energy is being deposited in the discharge at a constant rate for a full 500 ns. However, it is clear from plate 4.3 that both the spontaneous and stimulated 308 nm emissions terminate abruptly long before the end of the power input pulse.

This then is the paradox that the remainder of this thesis sets out to resolve: why do the spontaneous emission and laser power show this sudden decrease?

The laser pulse duration is somewhat shorter, at 200 ns FWHM, than that of

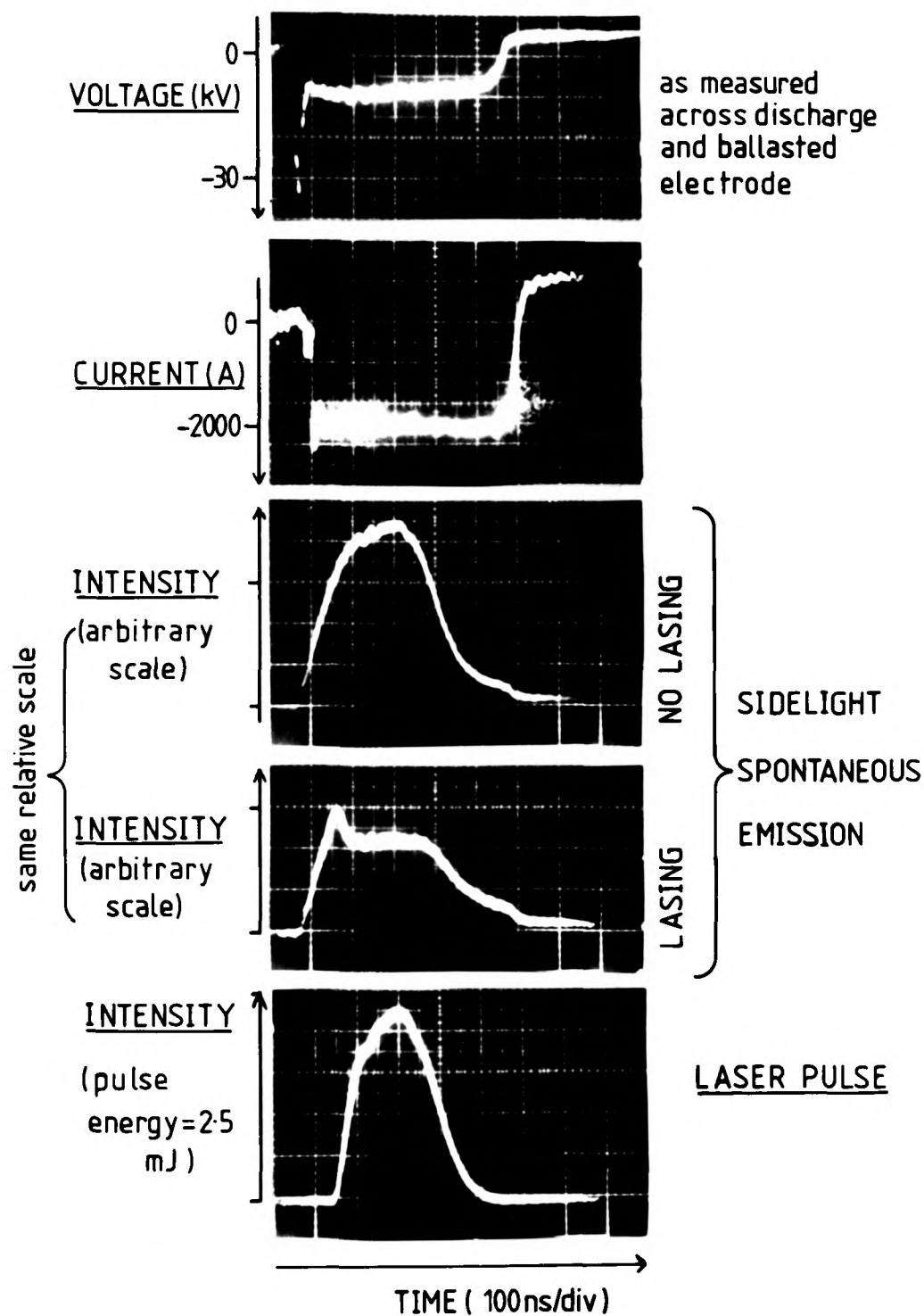
Time referenced Voltage, Current, & XeCl emission
from LPD laser (typical but not optimum)



GAS MIXTURE: 2mbar HCl; 25mbar Xe; 2.3atm. Neon
OPTICAL CAVITY: brewster windows; 96 % / 99.6% mirrors
BALLAST RESISTANCE: ~1.4ohm

Plate 4.3

Time referenced Voltage, Current, & XeCl emission
from LPD laser (typical but not optimum)



GAS MIXTURE: 2 mbar HCl; 25 mbar Xe; 2.3 atm. Neon
OPTICAL CAVITY: brewster windows; 96 % / 99.6 % mirrors
BALLAST RESISTANCE: ~1.4 ohm

Plate 4.3

the spontaneous emission at ~ 300 ns FWHM. This difference can be accounted for almost entirely by the cavity build-up time of ~ 100 ns which is clearly evident in plate 4.3 (the precise value of this delay was observed to be sensitive to changes in the cavity length and mirror reflectivity). The evolution of spontaneous emission in the presence of lasing shows the expected 'burn-down' in intensity due to the influence of stimulated emission. Since the spontaneous emission was observed in sidelight, through a very thin (~ 2 mm) section of discharge, it can be interpreted as a monitor of the upper laser level population: the plateau of the 'burn-down' could be considered as being indicative of the 'threshold' upper level population required for lasing. The laser pulse, during the termination phase, follows the reduction in spontaneous emission precisely and it does not terminate until the threshold level is reached.

These facts conclusively rule out the possibility of an increase of the population of transient absorbing species within the active volume as being the sole agency responsible for the extinction of the laser pulse.[†]

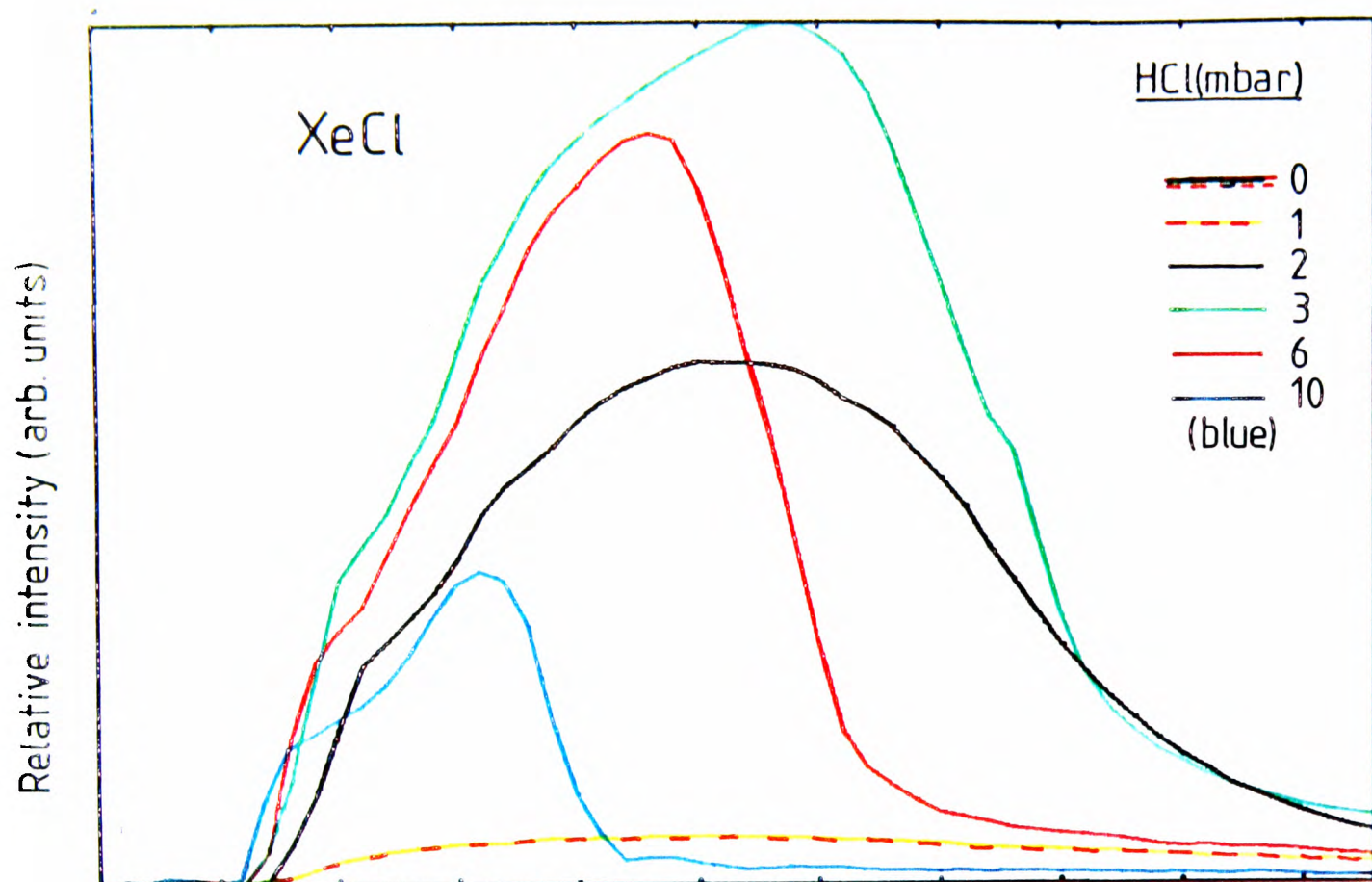
The spontaneous emission in the axial direction, which arises therefore throughout the 38 cm depth of plasma, was also monitored via the end windows of the discharge tube. It showed a very similar temporal evolution to that observed in 'sidelight'. If an increase in the density of a transient absorbing species was responsible for the laser pulse termination then the evolution of the normalised intensities of these emissions would be expected to differ according to Beer's law with appropriate integration:

$$I_{\nu}(\ell) = I_{\nu}(0)e^{-k_{\nu}\ell} \quad (4.15),$$

where $I_{\nu}(\ell)$ is the transmitted intensity at frequency ν , $I_{\nu}(0)$ is the incident intensity at frequency ν , ' ℓ ' is the length of the absorbing medium traversed, and k_{ν} is the absorption coefficient at frequency ν .

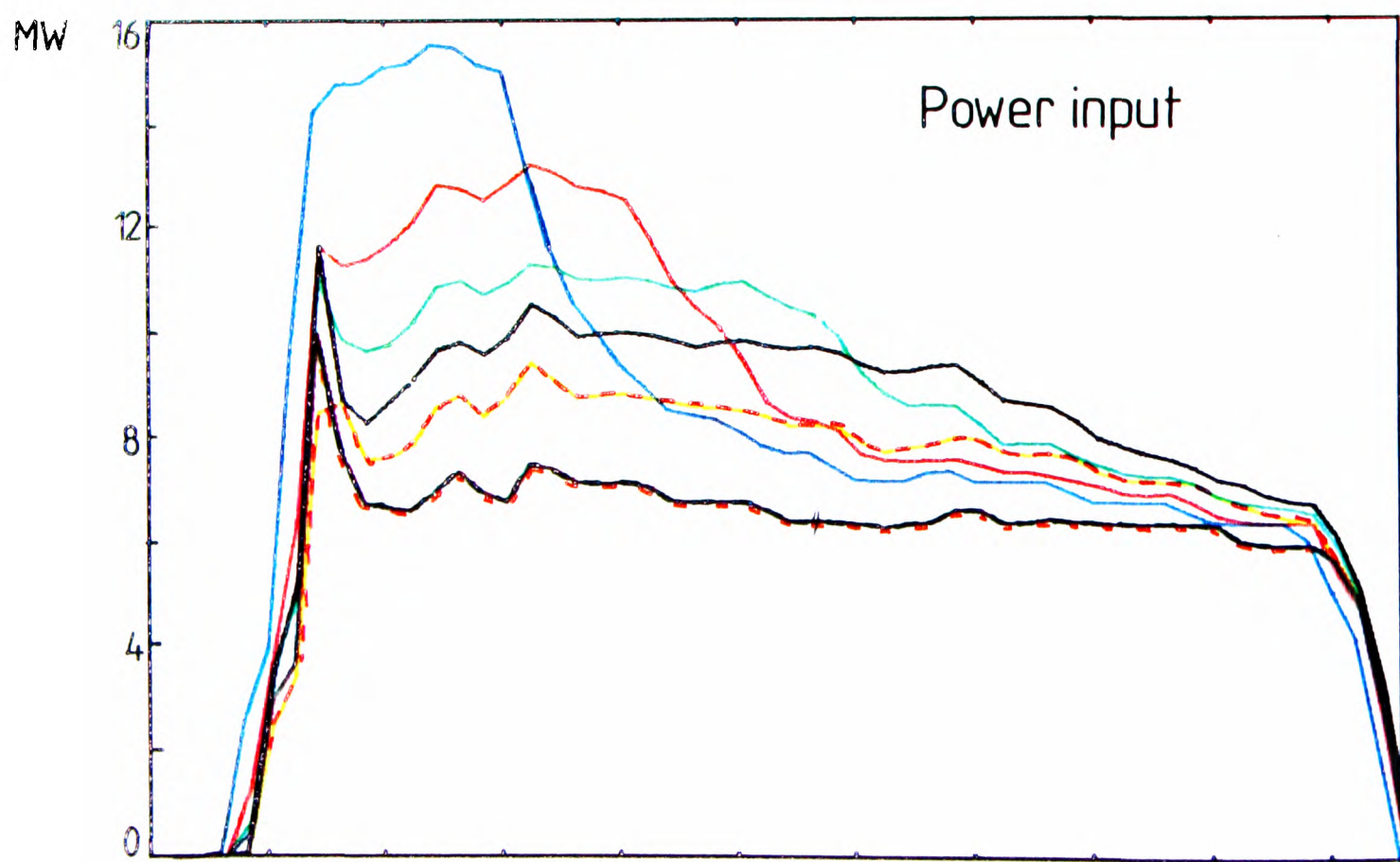
Judicious application of Occam's razor suggests that the obvious candidate on which to lay the blame for this premature termination is some mechanism which

[†]It is nevertheless true that if transient absorbers were not present the laser pulse would be expected to follow the termination of the spontaneous emission more closely.



(a) XeCl(308nm) spontaneous emission

Time (50ns/div)



(b) Total power input into discharge

Time (50ns/div)

FIGURE 4-12: Spontaneous XeCl emission vs. HCl partial pressure. 4-cable PFN used throughout. Neon buffer.

prevents the formation of the XeCl^* B-state. This will be discussed more fully in section 4.8.

4.6.2 Studies of the effect of varying the composition of the laser gas mixture on the XeCl^* emission - results:

(a) Results of the variation of the xenon partial pressure:

The partial pressure of xenon in the gas mixture was found to have relatively little effect on laser pulse duration and energy. Laser output deteriorated more sharply when decreasing the partial below ~12 mbar than in the range above 12mbar. No improvement or deterioration in output was detected for partial pressures above 25 mbar and below 100 mbar. Consequently, in view of the high cost of xenon, a 25 mbar partial pressure was used in all further experiments.

(b) Results of the variation of the HCl partial pressure:

The dependence of laser output on the HCl partial pressure was not as marked in the LPD laser as it was in the prototype: nevertheless the dependence was strong. Figures 4.12 and 4.13 illustrate this dependence on HCl partial pressure. Figure 4.12 shows the temporal evolution of XeCl^* spontaneous emission (a) and the power delivered to the discharge (b) for a number of HCl partial pressures. Also shown in figure 4.12(b) is the temporal evolution of the power deposited in a discharge which contains no HCl: the only parameter which was changed in obtaining all these power waveforms was the HCl partial pressure. Figure 4.13 contains traces of the current (a) and voltage (b) waveforms from which the power curves in figure 4.12(b) were obtained by point by point multiplication:

$$\text{Power}(t) = \text{Voltage}(t) \times \text{Current}(t) \quad (4.16).$$

The voltage traces in figure 4.13(a) represent the voltage drop across the discharge alone. The component due to the ballast resistance, $I \times R_b$, was removed. The interesting features of figure 4.12(a) are:

- (1) In the initial stages of the discharge ($t < 100$ ns) the 308nm emission does not increase with increasing HCl partial pressure, above a 3 mbar partial pressure, as might be expected.

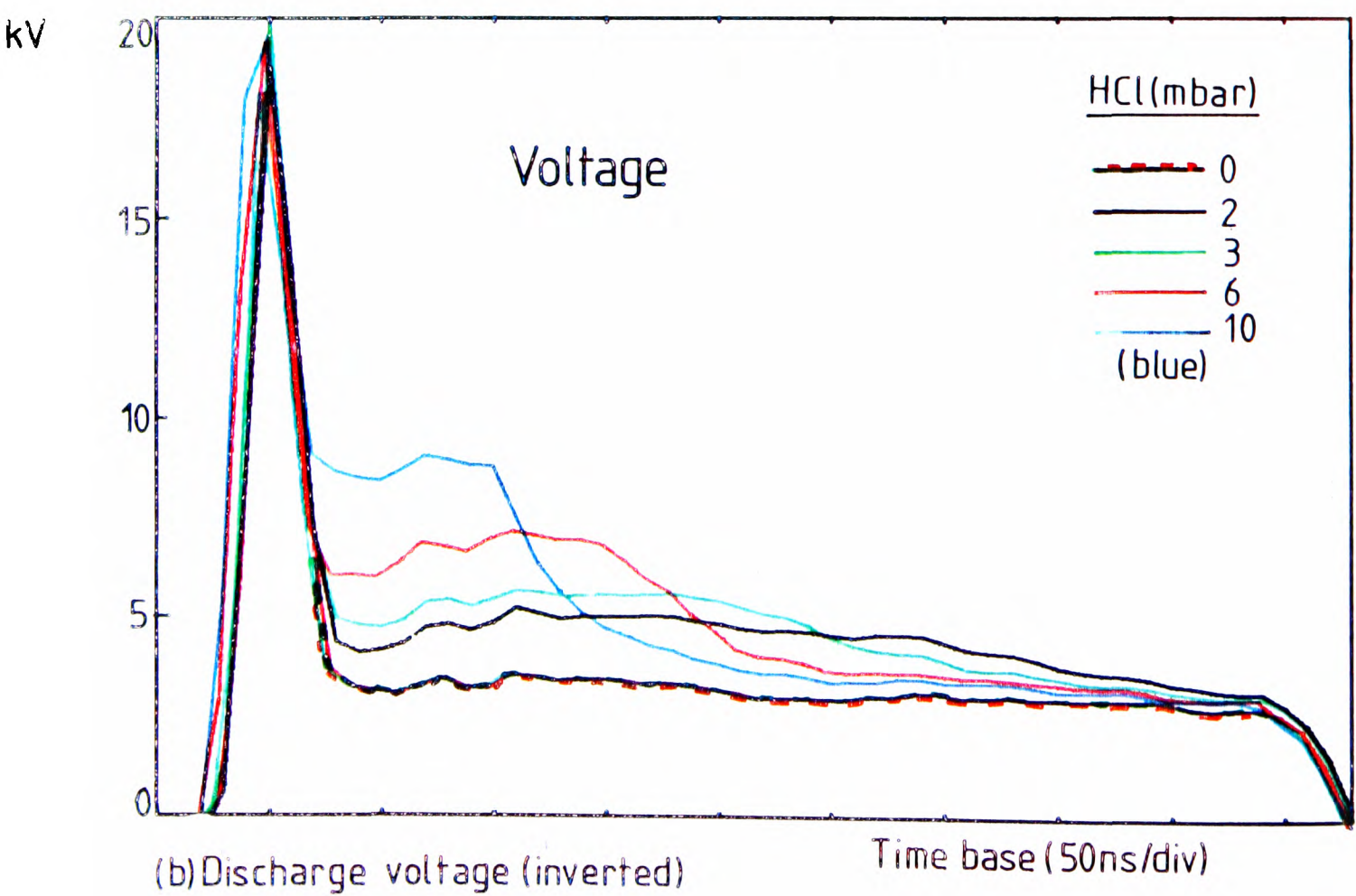
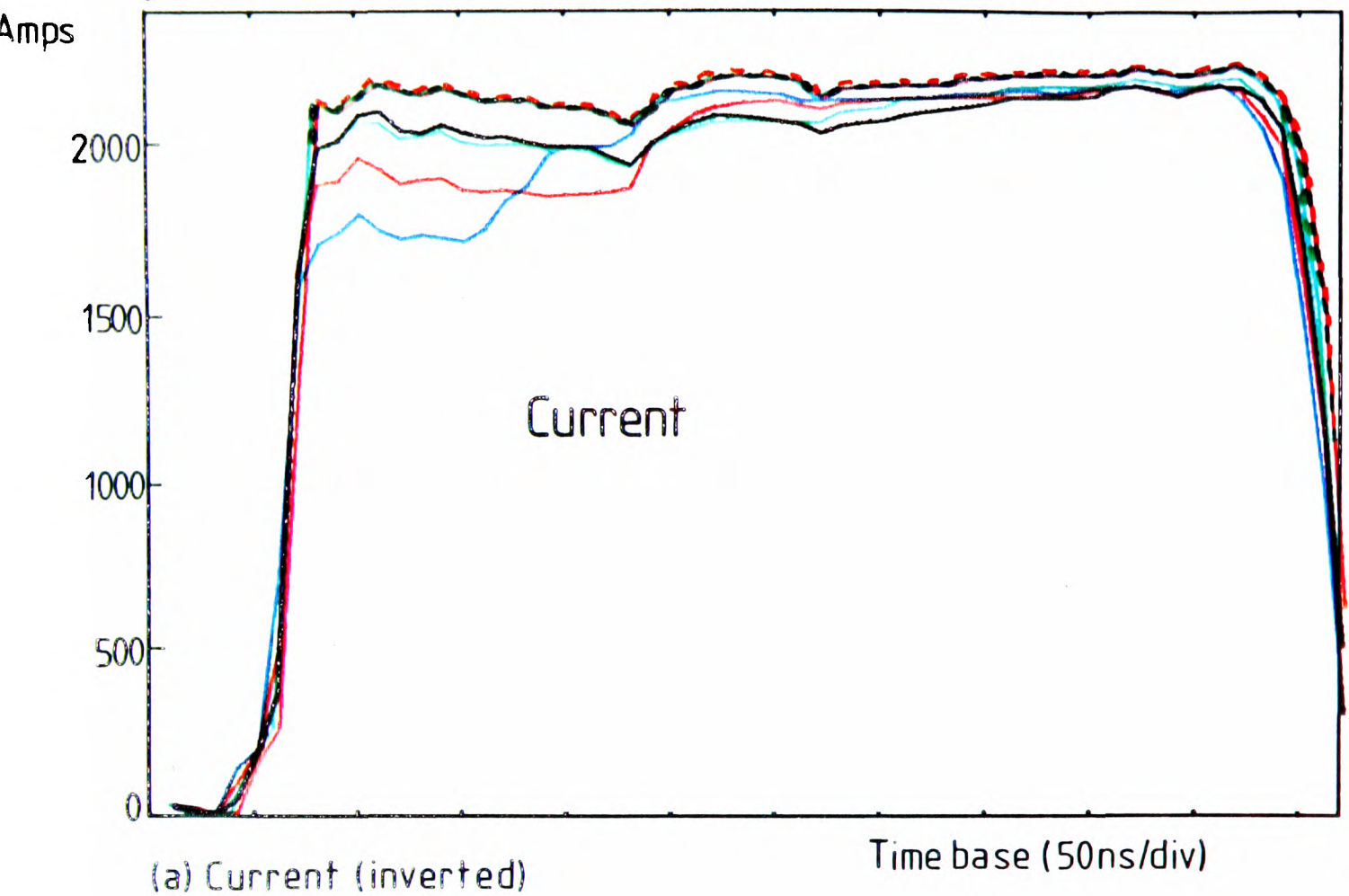


FIGURE 4-13: Discharge voltage and current vs. HCl partial pressure.
(cf. fig.4-12)

- (2) The spontaneous 308 nm emission exhibits progressively earlier premature termination with increasing HCl partial pressure.

The points to note from figure 4.13 are:

- (1) In the initial stages of the discharge, the voltage becomes progressively higher, and the current corresponding lower as the partial pressure of HCl is increased.
- (2) For a 10 mbar HCl partial pressure, a very sharp collapse in the discharge voltage and a corresponding rise in the discharge current takes place at the same time that the spontaneous emission intensity begins to fall.
- (3) The change in voltage and current becomes progressively less dramatic for lower HCl partial pressures: the common feature is that all the voltage and current traces tend towards the values of voltage and current of a discharge which contains no HCl.

(c) Results of the variation of the total buffer gas pressure and differences between He and Ne:

The performance of the LPD laser was tested as a function of pressure when using either helium or neon as the buffer gas. Although no difference in the relative performance of the two gases had been detected in the prototype laser, reports in the literature of improved lasing performance in XeCl^* for neon rather than helium buffered mixtures (e.g. Sze[1979]) encouraged a further search for some difference between the two gases.[†]

Consistently longer laser pulse durations and higher pulse energies were obtained from neon buffered gas mixtures than those obtained from helium buffered gas mixtures in the LPD laser. The results presented in figure 4.14 show that a continual (though non-linear) increase in the laser pulse duration and energy was exhibited by neon buffered mixtures in the pressure range up to 3.7 atmospheres.[‡] At pressures above 2.3 atmospheres the increase in laser pulse

[†] Argon was tried again, but with disappointing results, the main problem seemed to be that a stable, uniform discharge could not be obtained with this gas.

[‡] It was considered prudent, for reasons of safety, to restrict the pressure in the LPD laser head to this value.

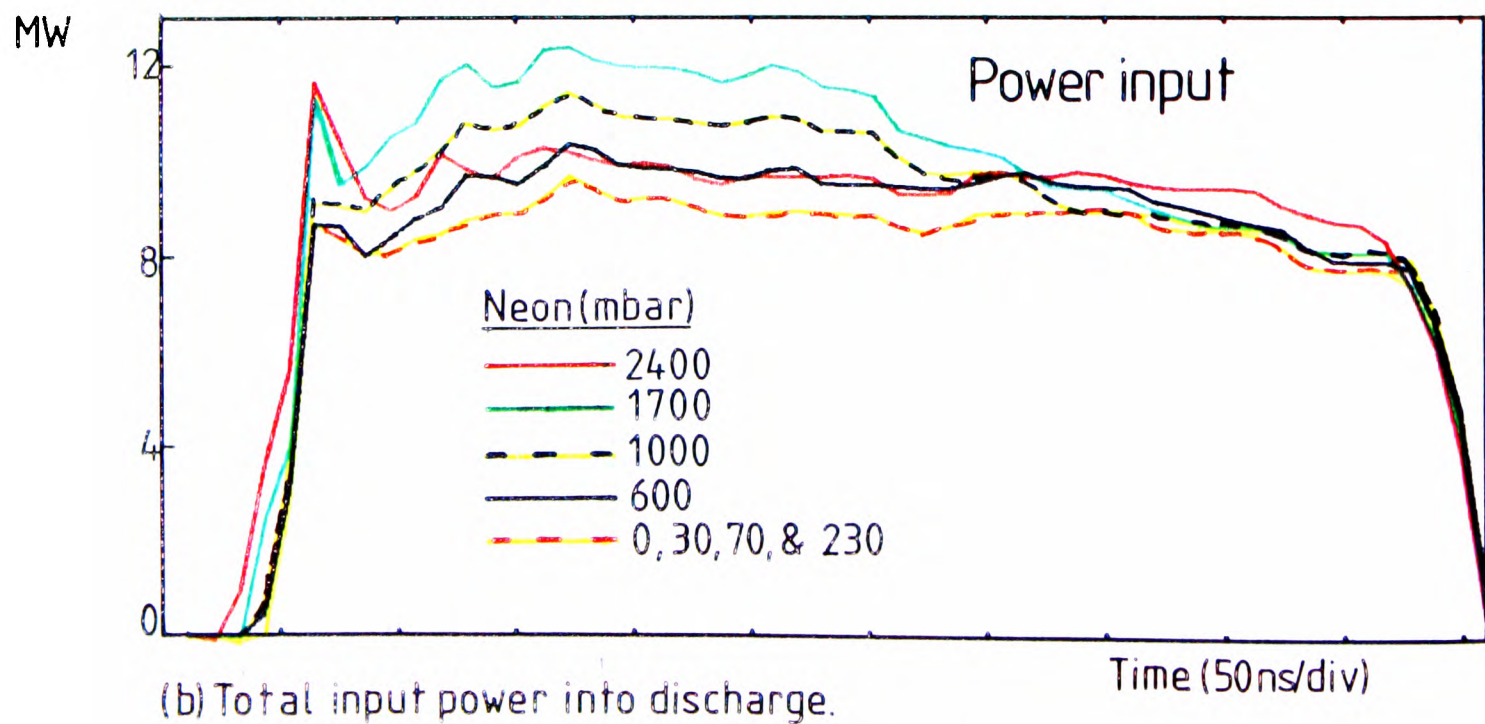
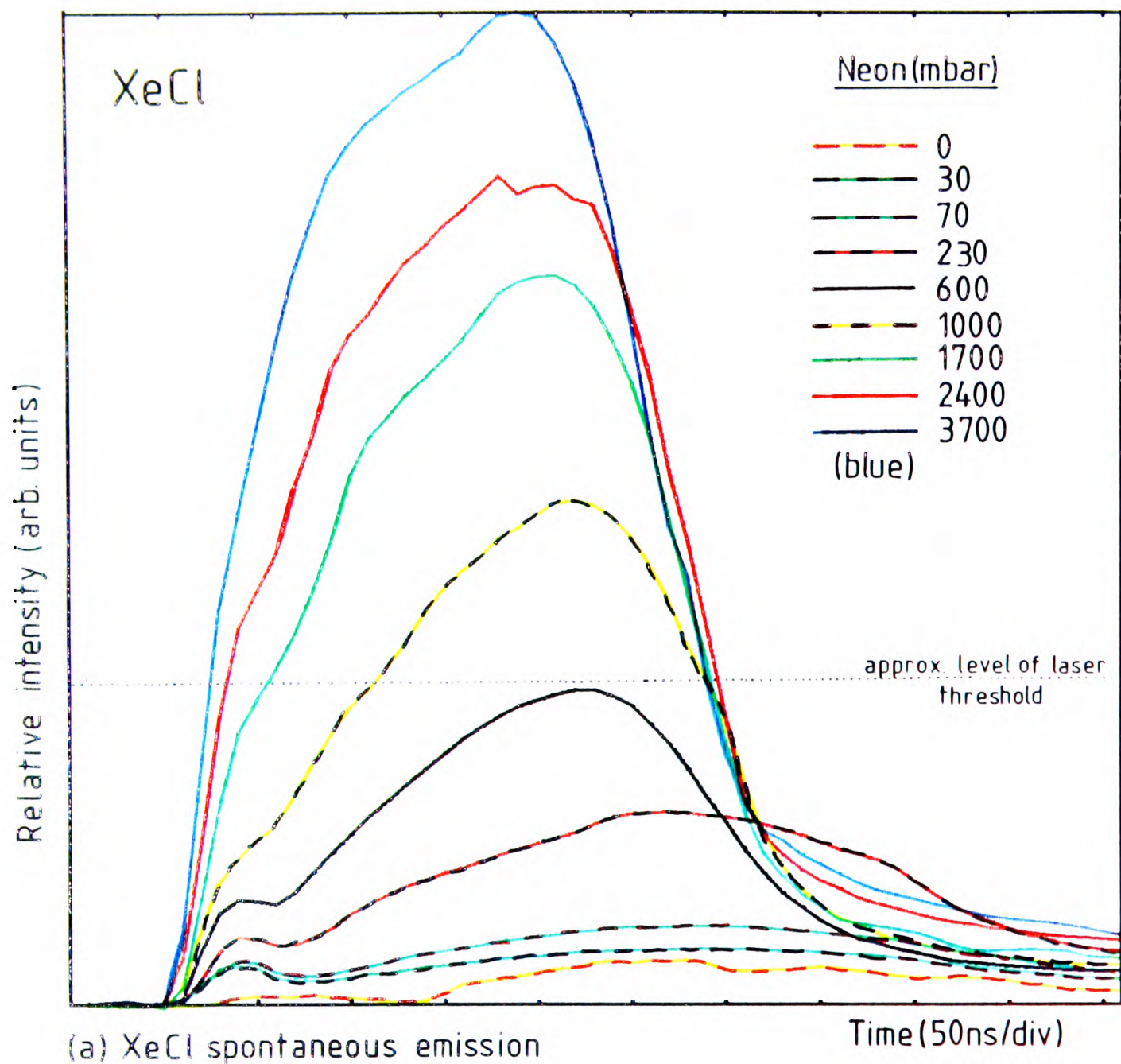


FIGURE 4-14 : XeCl spontaneous emission vs. Neon buffer gas partial pressure. Other gas pressures and parameters are outlined in Table 4-2.
(Resolution in time of ~30ns)

duration was due entirely to a decrease in the cavity build-up time: the onset of premature pulse termination was virtually the same except at very low pressures (<1 atm). This effect is illustrated clearly in figure 4.14. where a very approximate level of laser threshold[†] for the LPD laser is shown. The intensity of XeCl^* emission reaches threshold in a shorter time for higher buffer gas pressures.

Figure 4.14(b) shows the variation in total input power for the discharge as a function of the neon buffer gas pressure. The input powers for neon pressures in the range 0-230 mbar are too close to be resolved on this scale, however, as with the traces up to 1700 mbar, the general trend is an increase in input power with increasing pressure. The voltage and current traces from which the power traces were derived showed a slight continual decrease in current with increasing pressure (~5% over the entire range) but a proportionately greater increase in voltage with pressure: except in the case of a 2400 mbar (2.33 atmospheres) neon pressure where a sharp drop in voltage was observed (with a consequent lower total input power).

The maximum laser pulse duration obtained from a helium buffered gas mixture was ~200 ns FWHM which was about 70 ns less than that obtained from a neon buffered mixture; the helium buffered mixture pulse energy was ~2/3 the pulse energy of a neon buffered mixture. Furthermore, the laser output was observed to deteriorate at helium pressures above 2.33 atmospheres. The observed 308 nm spontaneous emission and V-I waveforms for a helium buffered mixture at a pressure of 2.33 atmospheres can be compared with those from a neon buffered mixture in plate 4.4.

As has been noted before, a change in the power input level is observed to coincide with the onset of termination of the light pulse: the helium buffered mixture exhibits an earlier onset of pulse termination and a corresponding earlier transition to a lower power input level than the neon buffered mixture. The higher power input level in helium in the early stages of the discharge is

[†] Estimated on the basis of the observed ratio between the spontaneous emission with and without lasing and taking some account of the single pass gain.

Comparison of performance of Helium & Neon

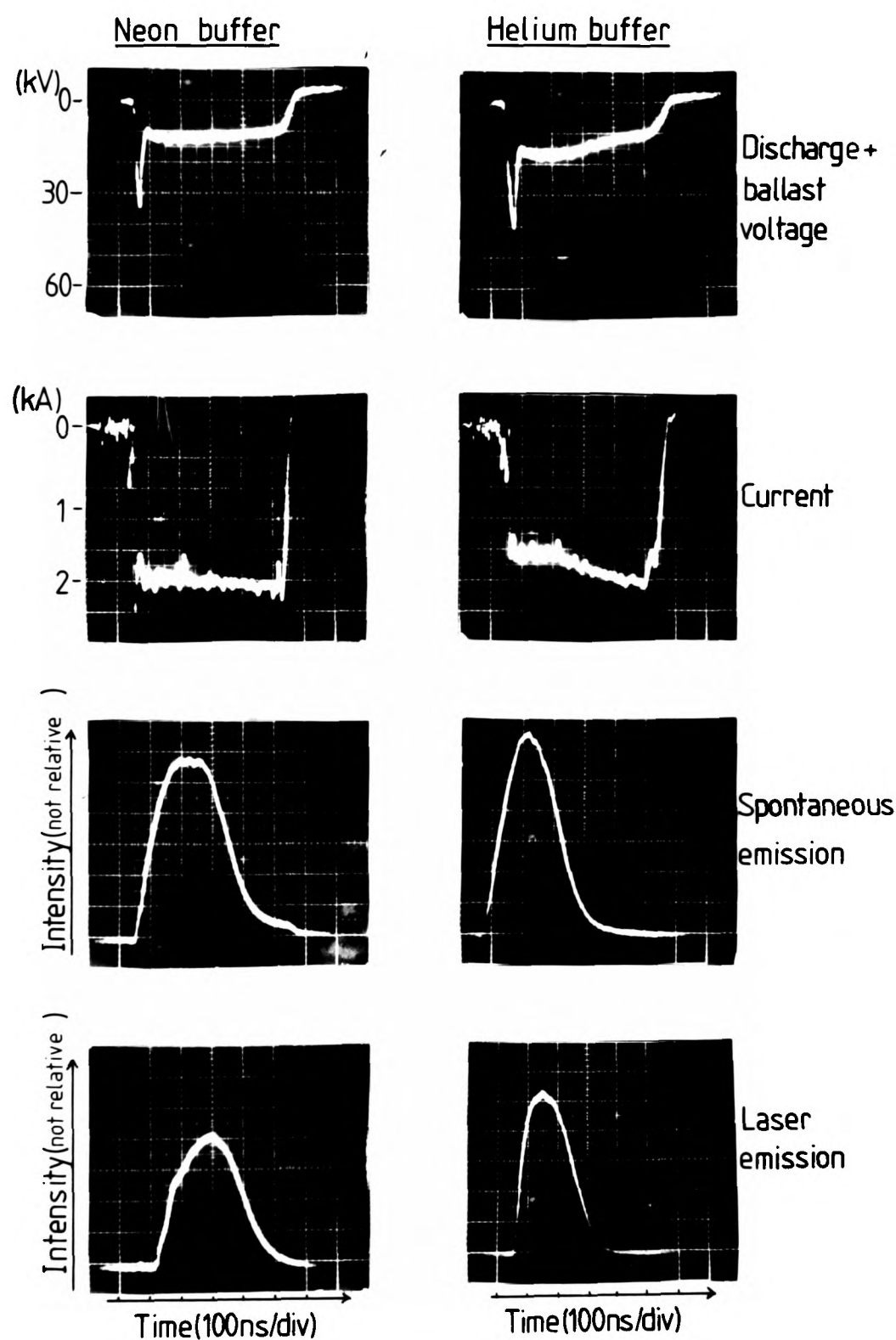


PLATE 4.4: Comparison of temporal evolution of various parameters with either Helium or Neon as the buffer gas. See table 4.2 for experimental details.

related to a higher voltage and lower current (but not low enough to offset the greater voltage) than in neon.

In an effort to quantify basic differences in the discharge characteristics of helium and neon, studies were made of voltage and current waveforms in partial mixtures of the two gases. Plate 4.5 contains voltage and current waveforms from discharges containing helium alone, neon alone, helium with 25 mbar xenon, and neon with 25 mbar xenon all at an overall pressure of 2.33 atmospheres. Plate 4.5 contains other interesting features, apart from the difference in 'running' voltage and current between helium and neon: discharge initiation (the time it takes the current to reach its running value) in neon is considerably faster than in helium both with and without xenon. In the mixture containing xenon, the current risetime is too fast to be resolved on an oscilloscope having a risetime of the order of 1 ns.

4.6.3 Studies of the effect of varying the composition of the laser gas mixture on the XeCl^* emission - discussion:

(a) Discussion of the effects of varying the xenon partial pressure:

The explanation for the observed saturation in laser output upon increasing the xenon partial pressure above 25 mbar must necessarily take account of the effect of such a change on the electron temperature of the discharge. As was discussed in chapter 2, the impedance of the discharge driver circuit will constrain the value of the total current and therefore constrain the electron density (given that the discharge is stable and uniformly fills the volume). The electric field across the discharge gap (and therefore the electron temperature) will be determined by the need to sustain ionization at a sufficient rate to keep the total current constant in the face of electron loss processes. If we assume that, once the discharge has been initiated in the LPD laser, ionization by multi-step processes is dominant over direct ionization from the ground state, it is possible to write down two (somewhat over-simplified)

Comparison of VI characteristics of discharges of various gas mixtures not containing HCl in the LPD laser

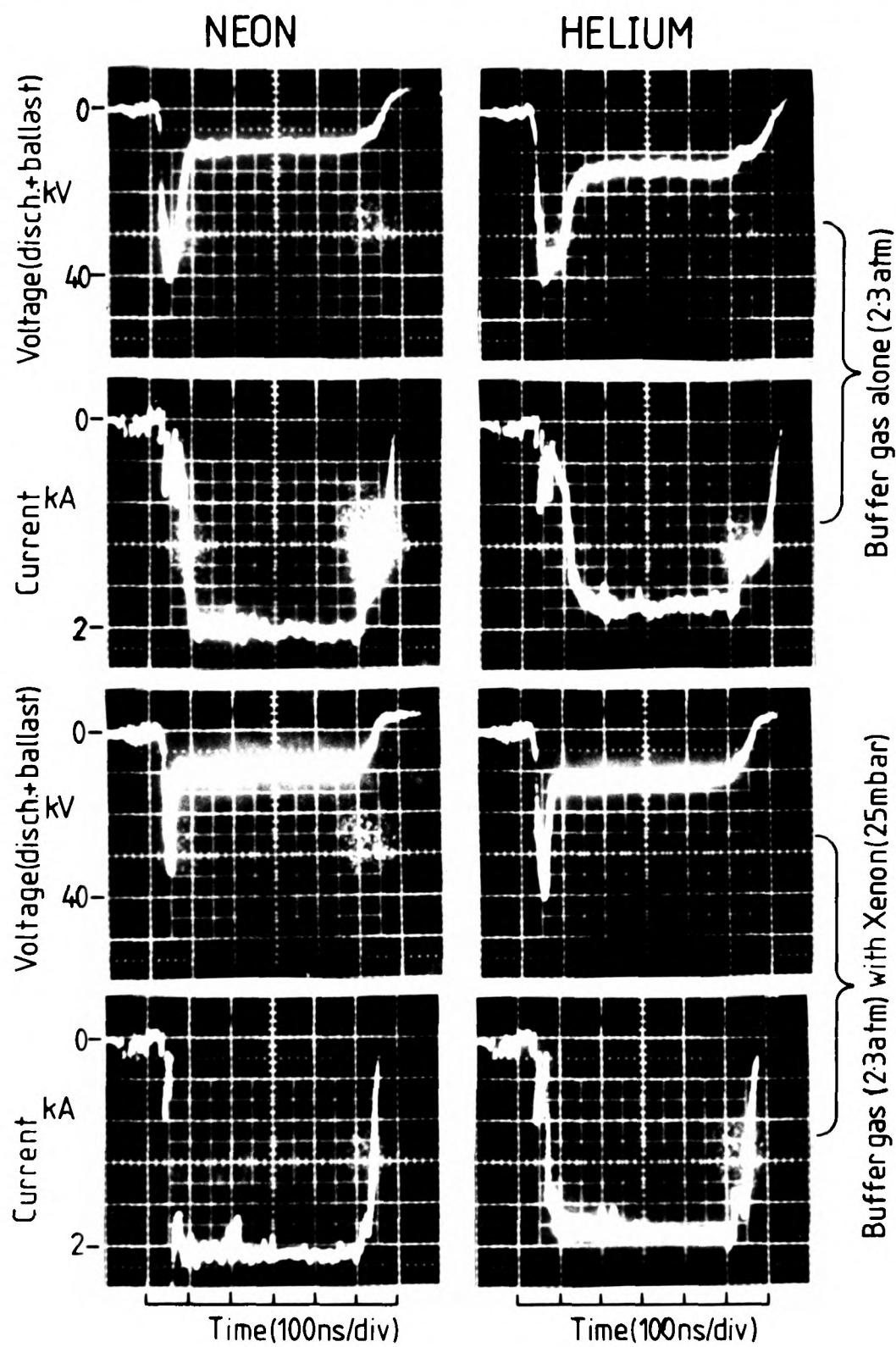


PLATE 4.5

rate equations governing the formation of both the XeI excited state atoms, Xe^* , and the xenon ions, Xe^+ :

$$\frac{d}{dt}[\text{Xe}^*] = k_1 n_e [\text{Xe}] - k_2 n_e [\text{Xe}^*] - k_3 [\text{HCl}] [\text{Xe}^*] = 0 \quad (4.17),$$

and

$$\frac{d}{dt}[\text{Xe}^+] = k_2 n_e [\text{Xe}^*] - k_4 [\text{Cl}^-] [\text{Xe}^+] [\text{M}] = 0 \quad (4.18),$$

where k_1 is the rate constant for excitation of xenon to excited states, k_2 is the rate constant for ionization of excited-state xenon, k_3 is the rate constant for XeCl^* formation through the 'neutral' channel (eq.1.2), k_4 is that for XeCl^* formation through the ion channel (eq.1.1), and M is a third body. In the steady-state regime (the plateau of power input after discharge initiation) the time derivative of the Xe^* and Xe^+ populations may be set equal to zero: a balance exists between ionization and loss of electrons to maintain a steady-state current. If we demand, quite reasonably, that overall charge neutrality exists within the discharge,

$$[\text{Positive ions}] = [\text{electrons}] + [\text{negative ions}] \quad (4.19).$$

The majority of negative ions in the discharge are Cl^- ions and their rate of formation can be taken as constant in this steady-state regime.[†] As stated before, the electron density is effectively constrained by the current that can be delivered by the power source. The question which remains then is: what proportion of the total positive ion population can be accounted for by xenon ions? Table 4.3 contains a list of some of the species which would readily be expected to be present in a 'normal' gas mixture (along with their ionization potentials and initial number densities). It is evident that xenon has the lowest ionization potential and that the neutral species of this element has a significantly greater number density than species having similar potentials: the probability of formation of a xenon ion, at the 1-2 eV electron temperatures typical of discharges such as these, will be higher than the probability of formation of any other ion. Xenon ions would therefore be expected to dominate the positive ion population. However, the relative proportion of xenon ions to

[†] With the small excursions in electron temperature we will be considering - see Kearsley[1980].

Table 4.3
Ionization potentials and typical number densities

Species	Ionization Potential(eV)	Initial partial pressure(mbar)	Number density(cm ⁻³)
Xe	12.08	25	6.6 x 10 ¹⁷
Cl	12.952	-	*6.6 x 10 ¹⁶
Cl ₂	13.2	-	*3.3 x 10 ¹⁶
H	13.527	-	*6.6 x 10 ¹⁶
HCl	13.8	2.5	6.6 x 10 ¹⁶
H ₂	15.6	-	*3.3 x 10 ¹⁶
Ne	21.47	2300	6.3 x 10 ¹⁹
or He	24.46	2300	6.3 x 10 ¹⁹

* Absolute maximum only: assumes total dissociation of HCl

the total positive ion population will be dependent on the ground state xenon density: at a given electron temperature, the discharge will find it more difficult to sustain ionization with a small ground state xenon density than with a high ground state density. The electron temperature must rise to increase the frequency of ionization events: ionization of species other than xenon will become more likely and xenon ions will make up a smaller proportion of the total positive ion population. Initially then, as the xenon ground state density is increased,†the formation of XeCl^{*} will be enhanced through both the ion and neutral channels. However, as the xenon ground state density is increased further still, the xenon ion number density will saturate as it approaches the total positive ion density constrained via eq.4.19: the loss of Xe⁺ via the formation of XeCl^{*} through the ion channel will also saturate since the other two quantities([Cl⁻] and n_e) will remain relatively constant. The product k₂[Xe^{*}] will have to be constant for eq.4.19 to remain valid in this situation. Any further increases in the xenon ground state density will be dealt with by a further decrease in the electron temperature with a consequent decrease in the formation rates k₁ and k₂. Data published by Levin et al.[1981]

† With a consequent drop in the electron energy required to sustain ionization.

indicates that the change in the rate k_1 is an order of magnitude greater than that in the rate k_2 for a change in electron temperature of 1 eV. This being the case, the XeI excited state population, $[\text{Xe}^*]$, though increasing slightly to offset the decrease in k_2 , will not significantly enhance the formation of XeCl^* through the neutral channel.

(b) Discussion of the effects of varying the HCl partial pressure:

At first sight, from the theory of formation of XeCl^* through both the neutral and ion channels, increased XeCl^* formation would be expected with an increased concentration of HCl. In particular, Kearsley[1980] concluded that, under the conditions present in his laser, the formation of Cl^- via dissociative electron attachment (eq.1.3) is the rate limiting step in the formation of XeCl^* through the ion channel. The formation rate of Cl^- will be given by:

$$\frac{d}{dt}[\text{Cl}^-] = n_e \cdot [\text{HCl}] \cdot \sigma(\epsilon) \cdot v(\epsilon) \cdot f(\epsilon) \cdot d\epsilon \quad (4.20),$$

where $\sigma(\epsilon)$ is the cross-section for electron attachment, $v(\epsilon)$ is the velocity, $f(\epsilon)$ is the probability distribution, of electrons of energy ' ϵ ' (frequently taken to be Maxwellian). Naively then, the formation rate of Cl^- might be expected to increase with the partial pressure of HCl. However, as can be seen from figure 4.13, both the electron density and the electron energy distribution are affected by the HCl partial pressure: the electron density is decreased (lower current and assuming constant current cross-section) and the electron energy increased (higher voltage) - the overall effect may well be one of an increased Cl^- population and therefore an increased XeCl^* population. In fact, it is reasonable to assume that the reason that the current is lower in the presence of HCl is entirely due to the lower mobility, and hence current carrying capability, of the Cl^- - the proportion of negative ions in comparison to electrons having risen considerably in the face of a loss of electrons through dissociative attachment to the HCl. The voltage is consequently higher in order to sustain the discharge in the face of this loss.

In order to explain the sharp transitions in both the voltage and current, one could conjecture, at least for the smaller HCl partial pressures (<3 mbar) where the change is not so dramatic, that progressive depletion of HCl within

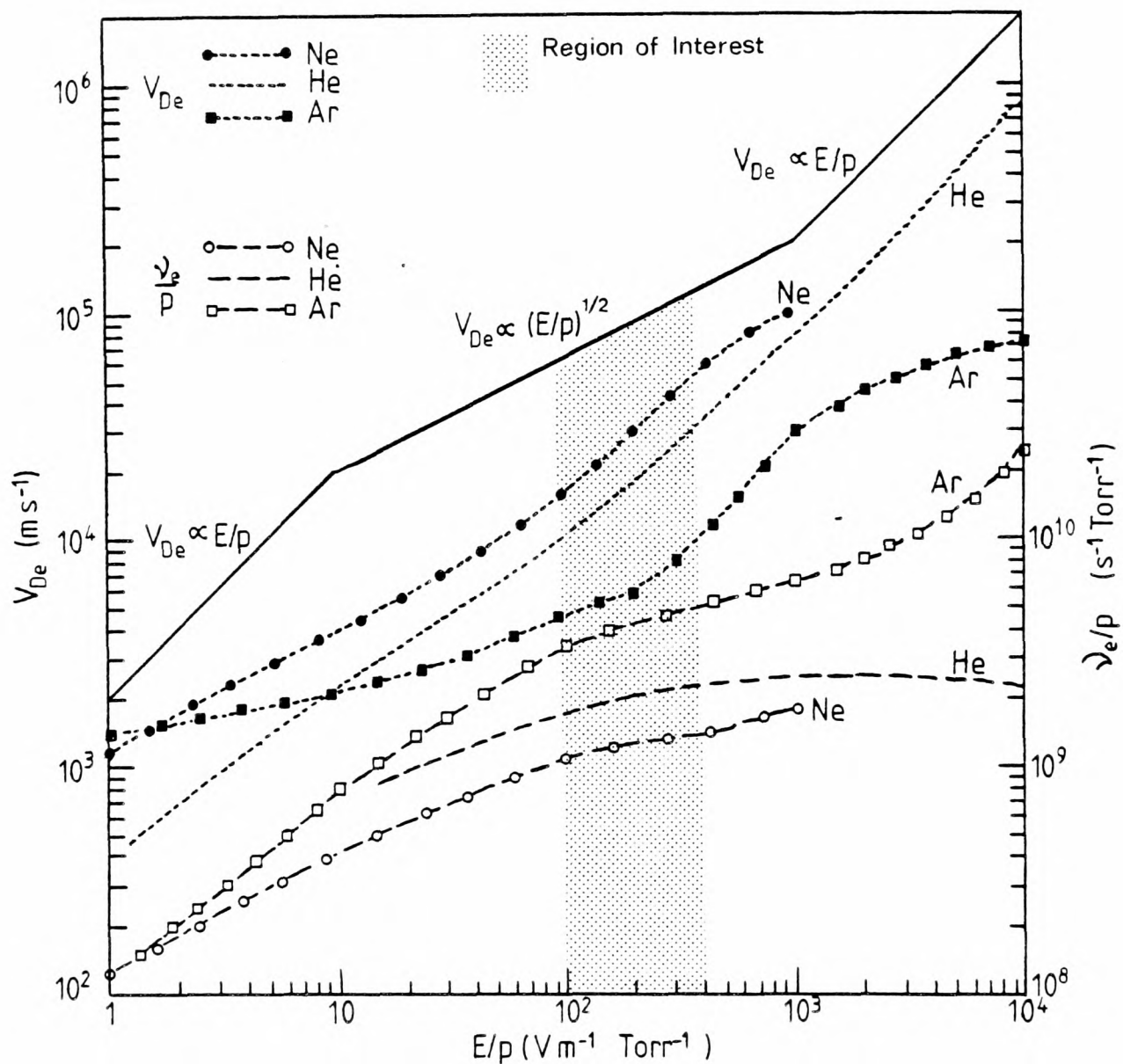


FIGURE 4.15: Measured drift velocities, V_{De} , of He, Ne, & Ar as a function of reduced field, E/p , and the corresponding derived reduced frequency for momentum transfer, ν_{De} , defined by $eE/m\nu_{De}p$. Also shown for comparison are lines corresponding to a constant collision frequency, $V_{De} \propto E/p$ (this is a copy of fig.1.2 in Franklin[1976])

the active volume is entirely responsible for the premature termination of the XeCl^* emission. As noted above, the lower 'running' current in the initial stages can be attributed to the greater rate of loss of electrons due to the dissociative attachment of HCl ; if the current then returns to a value typical of a discharge containing no HCl , it is tempting to conclude that there is no longer any HCl in the volume. However, in the light of the observed behaviour of discharges with a high HCl partial pressure, it is difficult to believe that this can be the complete explanation. If it were, one would expect, all other things being equal, that it would take longer for the HCl to be depleted if there was a higher initial concentration. Further discussion of this point will be deferred till a later stage.

(c) Estimation of the electron density in He and Ne buffered gas mixtures:

Before proceeding to the discussion of the observed effects of variation of the type and pressure of the buffer gas on the XeCl^* emission, it is worthwhile to investigate the effect that changing the type of buffer gas would have on the electron density.

The total discharge current, I , is given by the product of the cross-sectional area through which current flows, A , and the current density, j . The current density itself is given the product of the electronic charge, ' e ', the electron density, n_e , and the magnitude of the electron drift velocity, v_{drift} , which is dependent upon E/p . i.e.

$$I = n_e \cdot A \cdot e \cdot v_{\text{drift}} \quad (4.21).$$

Table 4.4 contains estimated values for the electron density in 'normal' helium and neon buffered laser gas mixtures at a time of ~ 200 ns after discharge initiation derived via eq.4.21. Also shown are the values which were used in arriving at the estimates: the voltage and current shown for neon were taken from figure 4.13, those for helium were taken from similar data (not presented here); values of the drift velocity were obtained from figure 4.15 using values

E/p rather than the temperature independent quantity E/N is used here merely because it was used by Franklin[1976] from which figure 4.15 is taken.

Table 4.4
Estimation of electron density for helium and neon buffered
laser gas mixtures via equation 4.21

	neon	helium
Voltage (at 200ns)	5000±5%	7500±5%
Current (at 200ns)	2000±5%	1600±5%
E/p (Vm ⁻¹ torr ⁻¹)	~130±5%	~190±5%
Electron Drift Velocity (cm/s x 10 ⁶)	1.7±10%	1.7±10%
Current-flow cross-section (cm ²)	11±50%	11±50%
Electron Density (cm ⁻³ x 10 ¹⁴)	6.4±65%	5.2±65%

of E/p which were calculated upon the assumption[†] of a uniform electric field, E, across the discharge gap, d, whence:

E/p = V/(d x p)

(4.22),

where V is the voltage across the gap in volts, p is the total pressure (1770 torr), and d is 2.2 cm. The large uncertainty quoted for the electron density is mainly a result of the uncertainty in the cross-sectional area through which current flow occurs. This is a point which is discussed later in terms of discharge constriction: although the discharge length of 38 cm used is accurate for times up to ~200 ns, the thickness of the discharge varies not only across the gap, but may actually vary in cross sectional area from the moment of discharge initiation according to the discharge parameters.[¶] For the specific case of a 'normal' laser gas mixture treated here the uncertainty in the electron density would be more fairly estimated at ~25%.

(d) Discussion of the effects of varying the total buffer gas pressure and differences between He and Ne:

It can be argued that the effect of increasing the pressure of neon must be to reduce the electron drift velocity by a reduction in E/p. If the discharge is to sustain the same current, either the field, E, must increase to restore the value of E/p or the electron density must increase.

[†] It is very difficult to justify any assertion regarding the electric field distribution in the high pressure discharge. All that can be said is this approximation provides an estimate of the value of E/p which is as plausible as any other.

[¶] See plate 4.9 and figure 5.28: a value of 3 mm was used in these calculations.

An increase in voltage was observed up to pressures of 1700 mbar, whereas the current remained relatively constant: this voltage increase accounts for the slight increase in total power deposition seen in fig.4.14(b).

It is likely that the increase in pressure is not totally offset by a sufficiently large increase in voltage to maintain the electron drift velocity at its (higher) low pressure values. In this case, an increase in electron density would be required to maintain the level of current with a consequent increase in XeCl^* formation via both the ion and neutral channels.

This argument, in conjunction with the knowledge that the spontaneous emission traces presented in figure 4.14(a) are affected by single-pass gain, serves to explain the increase in XeCl^* output at 2400 mbar compared to that at 1700 mbar when, at the same time, the total power deposition in the discharge is observed to fall. The drop in power deposition can be accounted for by an observed drop in the voltage across the discharge gap (the current remains virtually the same): E/p and the electron drift velocity will have also decreased. The shortfall in total current flow, which would otherwise result from this, is not made up simply by an increase in the electron density. Apart from anything else, the rate constants for ionization are much more sensitive to changes in E/p than v_{drift} : a decrease in E/p would reduce the rate of ionization and hence serve to reduce the electron density. The solution to the problem lies in the discharge volume which is pumped: visual observation of the discharge along the optic axis indicated a tendency towards more diffuse (larger volume) discharges as the total pressure was decreased. It must be assumed that the fractional reduction in the cross-sectional area for current flow was sufficient to provide an overall increase in the electron density per unit volume. In this way the total number of XeCl^* molecules along the (now thinner) optic axis would be greater than it was at lower total pressures. The single pass gain would therefore be higher resulting in a higher extraction efficiency and an increase in the observed output.

The reason for the observed difference in performance of helium and neon as buffer gases can possibly be attributed to a higher electron drift velocity

in neon than in helium (or for that matter argon) at a given E/p (figure 4.15). This implies that, given two discharges one neon buffered and the other helium buffered, a higher electron density will be required in the helium buffered mixture to sustain the same current. Conversely, given a particular electron density, a higher E/p (higher voltage) will be required in the discharge whose bulk gas has the lower electron drift velocity. In practice, the result is a compromise between both current and voltage: a slightly lower current, but higher voltage for helium than for neon. Plate 4.5 illustrates this point very well.

The difference in current risetime in helium and neon buffered mixtures could well result in a difference in the initial discharge uniformity achieved with the two buffer gases. Results presented later in this chapter will show conclusively that discharge uniformity is one of the primary factors responsible for the difference in the duration of 308 nm emission between helium and neon buffered discharges. However, discussion of the reason for this difference will be left until section 7.2.3.

4.6.4 Gas lifetime:

Although gas-fill lifetime extension is a major technical problem which limits the performance (and usefulness in certain applications) of RGH lasers, no study of the problem was made by this author. This was not only because the problem was not directly related to generation of long duration laser pulses, but also because other members of the laser group at the Clarendon laboratory were engaged upon such investigations. The effects of gas lifetime on the LPD laser were essentially the same as were observed for the prototype laser. Studies of this problem for the rare gas fluorides have been reported by Gower et al.[1980] and by McKee et al.[1980] for the rare gas chlorides.

It is worthwhile to make a few comments on materials compatibility however. The highly corrosive nature of the halogen donor gases means that very few, if any, materials are totally compatible from the point of view of extended gas-lifetime. Studies of gas-lifetime with various materials by other workers at the Clarendon Laboratory would suggest that the best materials to use for all

LPD laser: pulse duration vs. ballast resistance

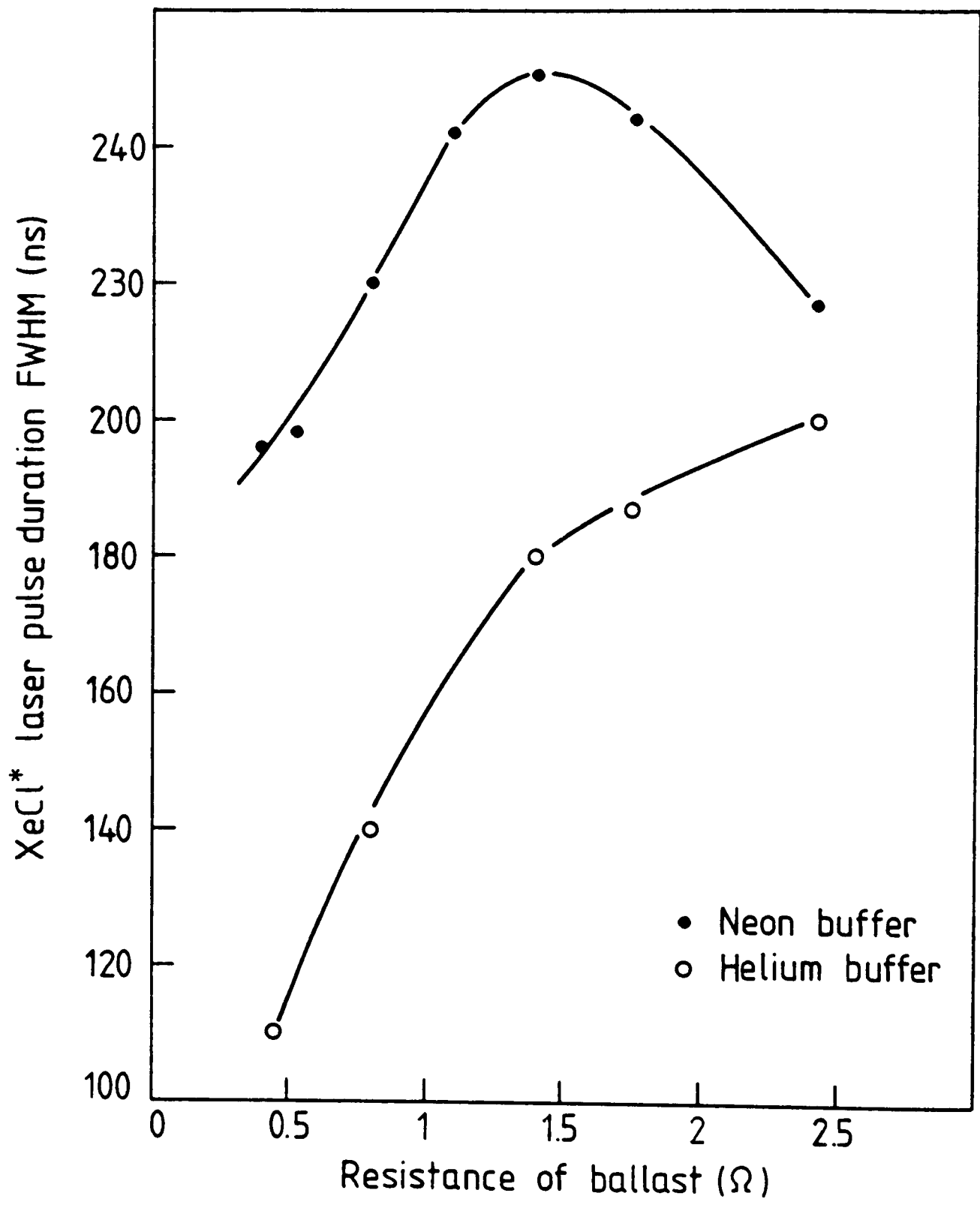


FIGURE 4.16

RGH systems are nickel and fluorinated plastics (such as PTFE). The useful lifetime of a single gas-fill in the LPD laser was quite short (~1.5 hours with the large volume attached). This would indicate that epoxy-glassfibre is far from ideal in terms of gas compatibility and that its use should be avoided. The belief that laser performance is degraded by a build up of large molecular species which absorb both laser and preionizing radiations has been confirmed by the success achieved in extending gas lifetime by the use of cryogenic traps to remove these species. Such traps are now produced commercially by Oxford Lasers Ltd.

4.6.5 The effect of varying the ballast resistance:

(a) Results:

The variation of the laser pulse duration (FWHM) was monitored as a function of the equivalent series resistance presented to the circuit by the ballasted bath. This was done for both helium and neon buffered gas mixtures for ballast resistances in the range 0.5–2.5 ohm: the results are presented in figure 4.16. An optimum ballast resistance of ~1.5 ohm was found for neon buffered discharges. The dependence of laser pulse duration and energy on the ballast resistance was much stronger in the case of helium buffered discharges in the 0.5 to 1.5 ballast resistance range. This dependence was reflected in the ‘running’ voltage and current of the discharge (measured in the plateau region ~200 ns after discharge initiation). Values of voltage and current from a helium buffered discharge for ballast resistances of 0.5,1,and 1.5 ohm are presented in Table 4.5. The laser pulse energy was found to peak at ballast resistance values of ~1.5 ohm for both helium and neon.

Unlike the prototype laser, variation of the ballast resistance in the LPD

Table 4.5
‘Running’ voltage and current in a helium buffered XeCl* discharge
vs. ballast resistance

Ballast Resistance(ohm)	Tot.Volts (kV)	Discharge Voltage (kV)	Current (A)	Laser Pulse (ns) at FWHM
0.5	2.5	1.3	2300	110
1	4	1.8	2200	150
1.5	10	7	2000	200

laser had little, if any, measurable effect on voltage or current risetimes, nor did it have any effect on the shape of the traces of power deposition into the discharge volume in time.

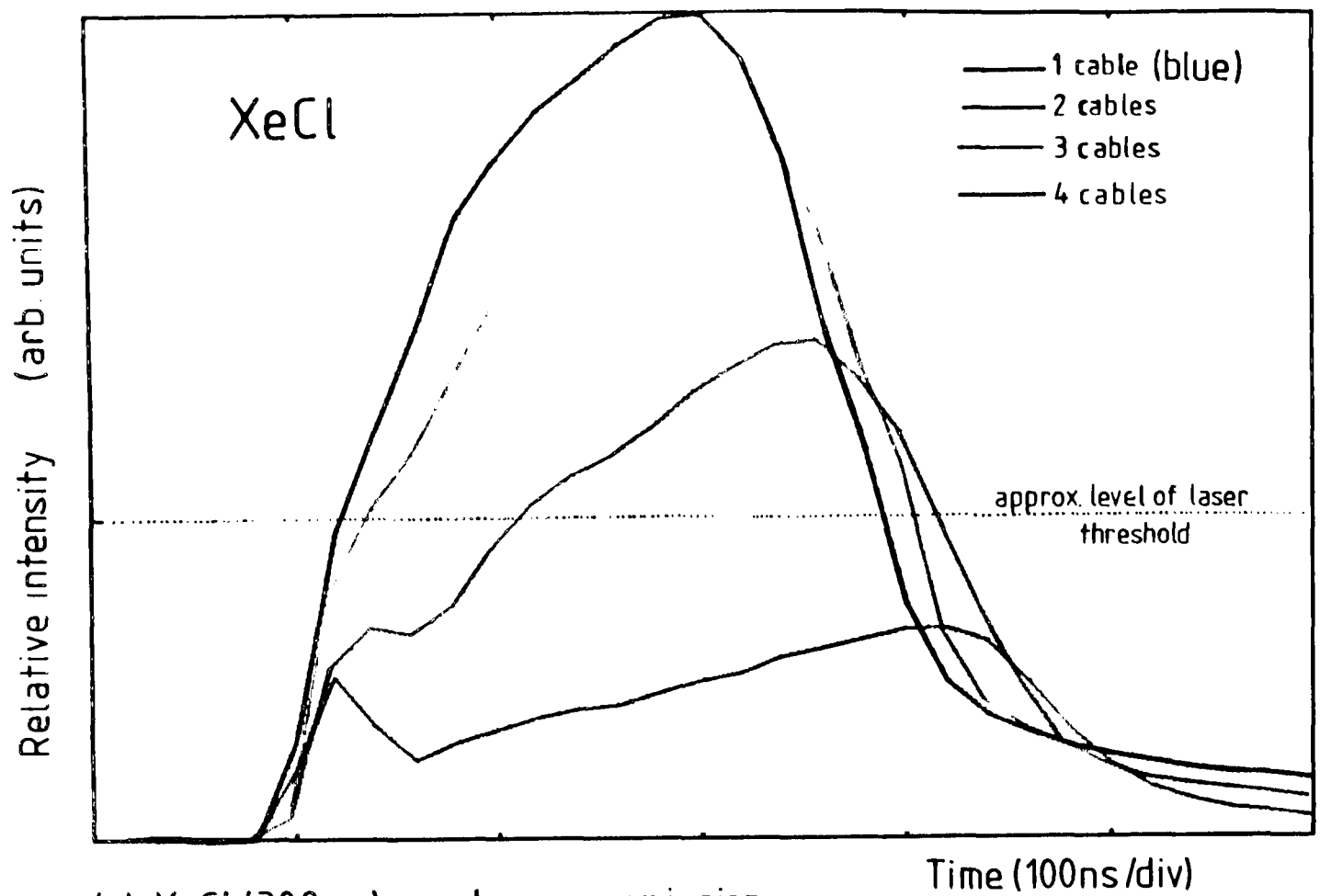
Direct visual observation of the discharge uniformity through the sidelight window of the LPD laser revealed that at low ballast resistances the discharge consisted mainly of individual columnar discharges associated with each pin of the ballasted electrode. As the ballast resistance was increased the discharge became more diffuse - filling out the regions between the pins. The discharge uniformity in the case of the neon buffered discharge was better than in the helium buffered discharge at all values of ballast resistance but particularly so when the ballast resistance was low. This difference in discharge uniformity was strongly reflected in the laser performance for the two gases (figure 4.16).[†] The LPD laser was also tested with a nominally zero ballast resistance by filling the electrolyte reservoir with Mercury: no lasing was observed and the discharge was observed to consist of a few, randomly-situated, bright arcs. This was not really a fair test of the efficacy of resistive stabilization since the discrete pin electrode would have exacerbated discharge instability due to field enhancement at the edges of each pin.

(b) Discussion:

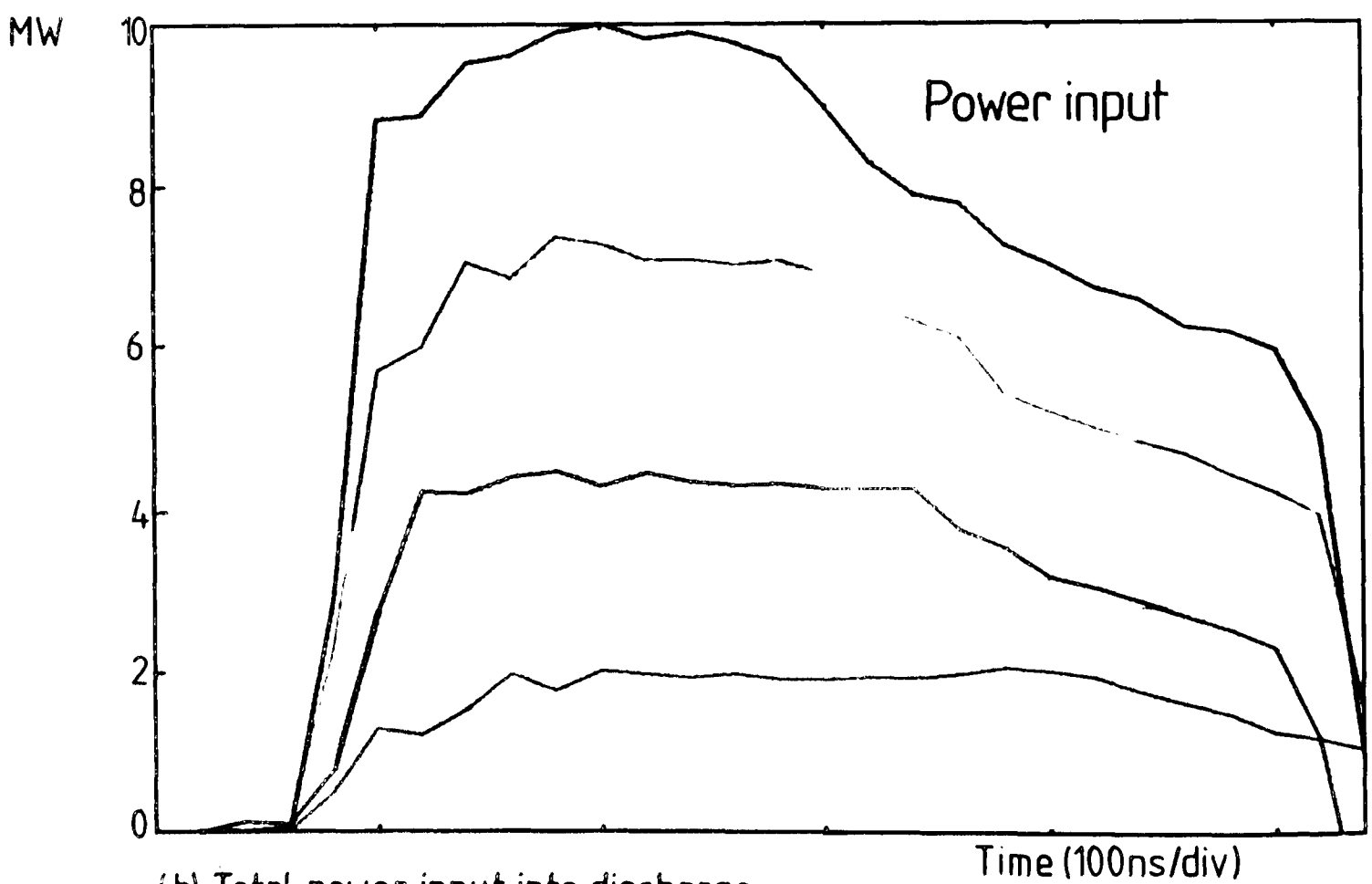
It is possible to infer the effect of increased ballast resistance merely by examination of table 4.5: the large drop in the discharge voltage at low ballast resistance values must be the result of a much smaller discharge volume. This was confirmed by the results of direct visual observation of the discharge uniformity.

The stabilizing influence of the ballast resistance is manifest not only in the prevention of 'hard' arcs, but also in some mechanism whereby the total discharge volume is enhanced. The peaking of laser pulse duration in the case

[†] It must be emphasized that the change in discharge uniformity when altering the neon buffer gas pressure (section 4.6.2(c)) from 1700 mbar to 2400 mbar was negligible in comparison to the changes observed when the ballast resistance was altered.



(a) XeCl (308nm) spontaneous emission



(b) Total power input into discharge

FIGURE 4.17 : Spontaneous XeCl emission for different total input powers into a Neon buffered discharge.
(60ns resolution in time base)

of a neon buffered discharge seems to be the result of a compromise between the uniformity of the discharge and a reduction in the total power delivered to the volume - both of which are a consequence of increasing the ballast resistance. The higher ballast resistance required to maximize the laser pulse duration in a helium buffered discharge is indicative of the lower stability of these discharges. These points will be discussed further in section 7.2.2.

4.6.6 The effect of varying power deposition in the discharge:

Although the laser pulse energy was found to increase monotonically (though sub-linearly) with increasing power input to the discharge (PFNs consisting of up to ten cables in parallel were tested), the power input for maximum laser pulse duration was found to be optimized at quite a low level. This is illustrated in figure 4.17(a), which shows the temporal evolution of XeCl^* spontaneous emission for discharges driven by PFNs consisting of from one to four cables connected in parallel. The evolution of the instantaneous power delivered to the discharge by each of these PFNs is given in figure 4.17(b). It should be noted that the major contribution to the increased power deposition as the number of cables was increased was an increase in the total current supplied by the PFN as a result of its lower impedance. The discharge voltage was approximately constant in comparison to the increase in total current flow.

The feature common to all the parametric studies described so far - premature pulse termination with a corresponding drop in the instantaneous power input - reappears in this investigation as well. As expected, the peak intensity of XeCl^* emission increases with increasing input power. However, the time elapsed between discharge initiation and the time at which pulse termination is evident steadily decreases with increased input power. The level of laser threshold (see section 4.6.2(c)) indicated on fig.4.17(a) gives a good indication of what was observed in lasing: pulse energy and duration for four- and three-cable PFNs was very similar, the only difference being the later appearance of the laser pulse in the case of the three-cable PFN. The cavity build-up time was even longer in the case of a two-cable PFN and both the laser pulse duration (FWHM) and energy were curtailed. No lasing was observed in the

case of a one-cable PFN.

The termination of the XeCl^* spontaneous emission in the case of one- and two-cable PFNs cannot definitely be attributed to pulse termination from this data alone since the 'termination' occurs quite near the end of the first 'double transit' of the PFNs. A one-cable PFN with a double-transit time of 1000 ns was tested to verify explicitly that the XeCl^* emission terminated not because the end of the PFN pulse had been reached but because it also suffered from the same premature termination effects as at higher input power levels. The results of this test are presented in the next chapter (figure 5.19) and they show conclusively that, even in the case of the modest total instantaneous power delivery of 2MW of a one-cable PFN, pulse 'termination' occurs.

4.6.7 Preionization:

The importance of preionization of the discharge volume to the initiation of uniform, stable discharges was outlined in chapter 2. To date, no extensive parametric study of preionization in relation to RGH lasers has been reported in the literature, though Armandillo et al.[1982] have noted that the output from a Nitrogen C-state laser increases as the energy input into a preionization array is increased. Unfortunately, because of the electrolyte ejection problems noted earlier in this chapter, the total input power to the preionization array could not be increased. However, the fact that discharge instabilities did not appear in the LPD laser until a late phase of the discharge - well after initiation - indicates that the preionization was sufficient, and that it is unlikely that increased preionization intensity would have had any bearing on the eventual output.

The one preionization parameter which was studied was the effect of the preionization delay time on the laser pulse duration. The optimum delay between the peak of light emission from the preionization array and the onset of the main discharge current pulse was found to be $40 \text{ ns} \pm 20 \text{ ns}$. This delay time would place the peak of emission almost precisely at the time that the spark gap of the PFN began to conduct. As was mentioned earlier, the delay time could be altered by changing the length of a coaxial cable which was connected between

Preionization Timing

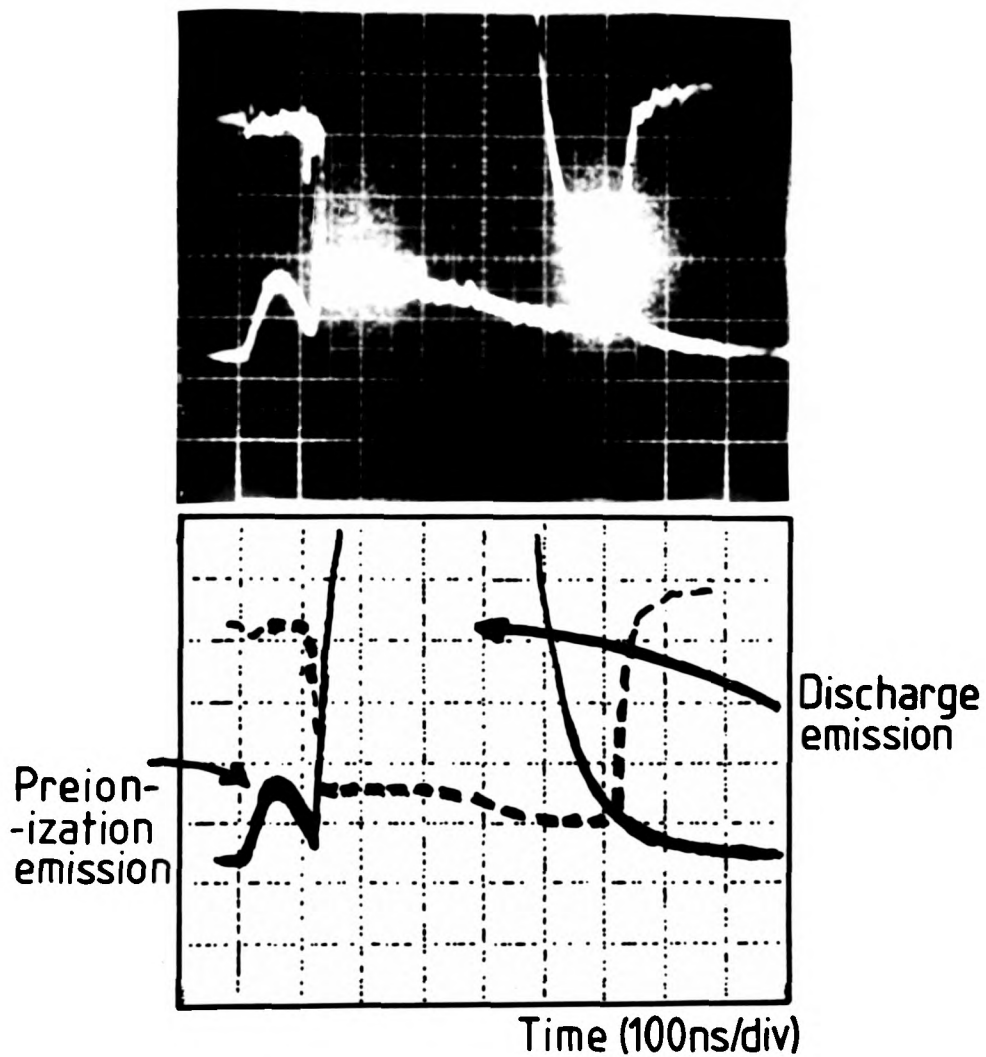


PLATE 4.6: Light emission from the preionization array (heavy line) and main discharge (off scale) with current pulse (dashed line) overlaid for time reference.

the preionizer circuit and the trigger pin of the main spark gap. Plate 4.6 shows the emission of the preionization array followed by the emission from the main discharge (off scale). A typical discharge current pulse is overlaid (negative-going) and provides a time reference.

4.6.8 The spectral profile of the LPD laser output:

A brief investigation was carried out to determine if there was any significant spectral line narrowing in the laser spectrum of the LPD laser as a result of its long pulse duration. Its output was compared with that of a short (0.5m gain length) 'standard' design XeCl* laser which had a pulse duration of ~20 ns FWHM. The spectral tuning characteristics of this 'standard' laser were determined by Dr.A.J.Andrews at the Clarendon Laboratory and his data will be compared with those obtained from the LPD laser.

(a) The fluorescence spectrum:

The XeCl* fluorescence spectrum is quite broad (~2nm) and, unlike the rather structureless fluorescence spectra of the krypton and argon halides, contains four distinct features: the wavelengths of these peaks and the assignments of the transitions which give rise to them are listed in Table 4.6. This difference in the structure of the spectra is merely a result of the fact that XeCl has a slightly bound lower level whereas the halides of all lighter rare gases have a repulsive ground state. Although it should be possible to tune the laser output over a very wide range of the fluorescence spectrum, in practice this is very difficult to achieve: the degree of wavelength discrimination required to inhibit lasing from one of the 'peaks' cannot usually

Table 4.6
Principal emission features in the XeCl (B-X) laser spectrum

Peak	Transition (v'-v'')	Wavelength (nm)	Franck-Condon Factor (x10 ³)
1	0 - 3	308.459	190
2	0 - 2	308.210	249
3	0 - 1	307.961	226
4	0 - 0	307.704	104

Source: Tellinghuisen et al.[1976]
Isotopic splitting (³⁵Cl and ³⁷Cl) does not become evident until the v' =2, 3, and 4 bands at shorter wavelengths with the bands of the less abundant (24.5%) heavier isotope occurring at slightly longer wavelengths.

Gain-narrowed XeCl* fluorescence spectra

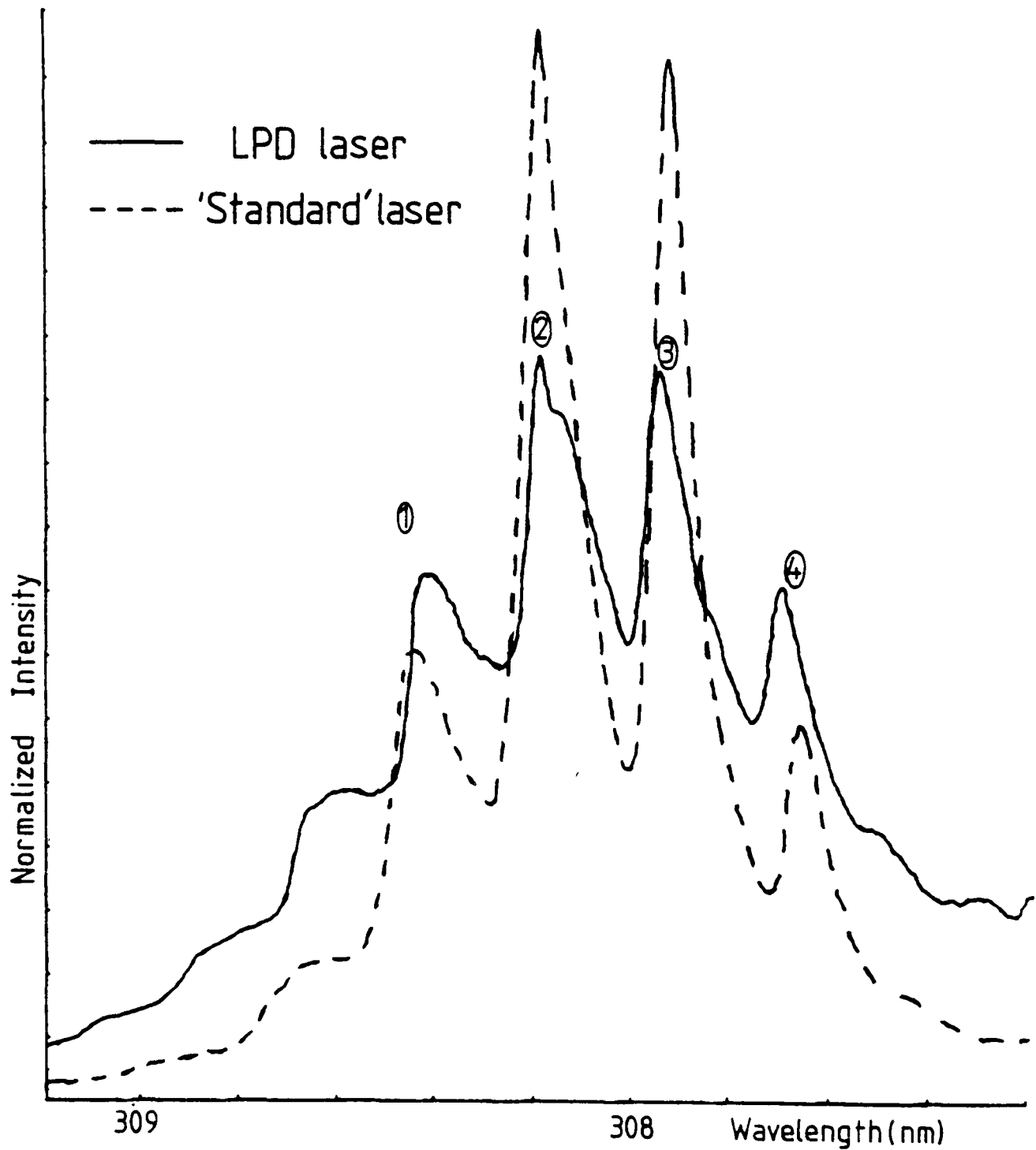


FIGURE 4.18 : Comparison of 'endlight' fluorescence spectra from the LPD laser and a 'standard' XeCl laser. See table 4.5 for identity of numbered peaks.

be achieved.

Time-integrated 'endlight' (i.e. along the optic axis) fluorescence spectra from both the LPD laser and the standard laser are presented in figure 4.18. The integrated intensity of each spectrum has been normalized for comparison and the numbers on the 'peaks' correspond to those in table 4.6. Both of these spectra are distorted due to single pass gain along the optic axis and significant gain narrowing,[†] in comparison to the LPD laser, is evident in the 'standard' device. The net gain (i.e. the gain above losses such as transient absorption) of the 'standard' laser was of the order of $10\% \text{cm}^{-1}$, that of the LPD laser (on the basis that lasing was observed with a 25% reflectivity output coupling mirror but not with a 10% reflectivity output coupler) was $\sim 5\% \text{cm}^{-1}$.

(b) Lasing:

The time-averaged, untuned, laser output from the LPD device (cavity as in table 4.2) was not significantly narrower than the output from the standard laser: the cavity of which was a nominal 100% total reflector and a 10% reflectivity output coupler. The reason for this was that the benefit obtained in line narrowing due an increased laser pulse duration (eq.1.4) had to be offset against a lower degree of gain narrowing in the LPD laser. Had it been possible to carry out time-resolved, rather than time-averaged, studies of

[†] Gain narrowing can be understood by reexamination of Beer's Law (eq.4.15). The part of the absorption coefficient k_ν due to the XeCl^* molecules becomes negative when the population per state of the upper level, N_2/g_2 , is greater than that per state in the lower laser level, N_1/g_1 :

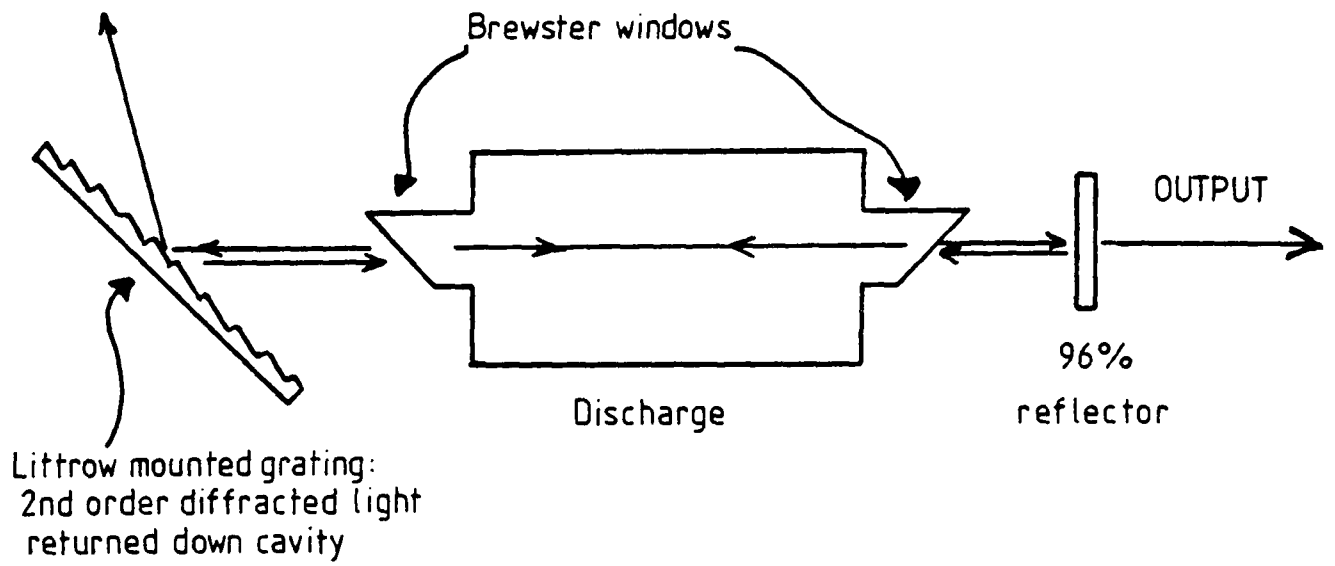
$$k_\nu(\text{XeCl}^*) = \sigma_\nu(N_1 - (g_1/g_2)N_2) \quad (4.23),$$

where g_1 and g_2 are statistical weights dependent upon the degeneracy of each energy level, and the optical cross section per atom, σ_ν , is dependent on a function determined by the dominant line broadening mechanism:

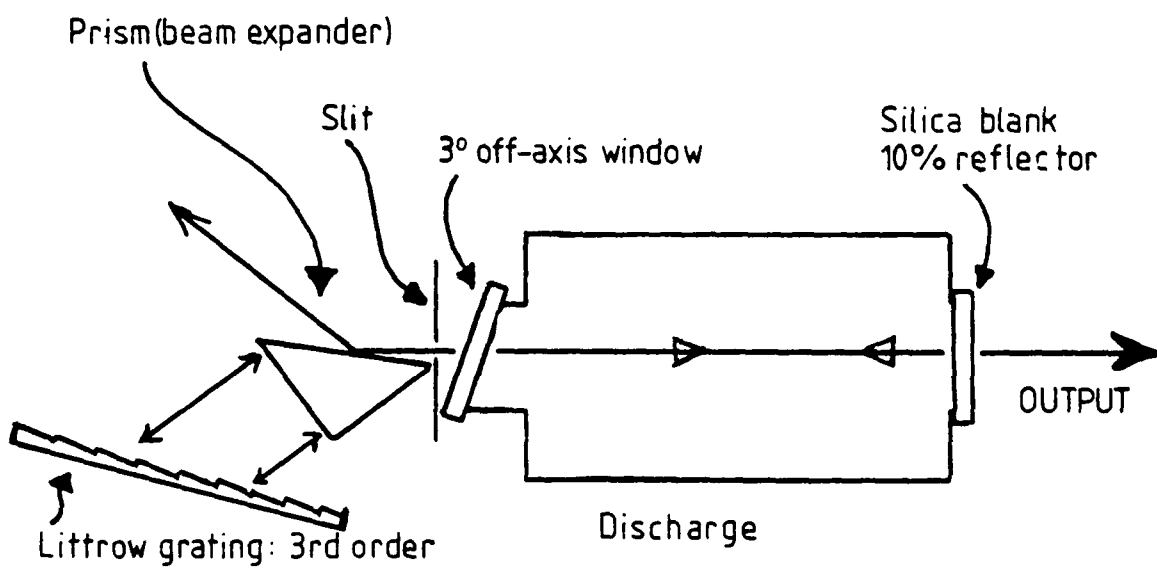
$$\sigma_\nu \propto g(\nu - \nu_0) \quad (4.24).$$

Gain, at a particular frequency ν , will occur when the (negative) $k_\nu(\text{XeCl})$ is greater than the combined (positive) absorption coefficients of all other processes. Since the gain coefficient is a frequency dependent quantity, the intensity at line centre, $I(\nu_0)$, is not only greater than that at some frequency $\delta\nu$ away from line centre, $I(\nu_0 - \delta\nu)$, but it will grow faster as a function of distance. The overall effect will be an apparently narrower spectral profile.

The difference in the degree of line narrowing of the respective 'endlight' fluorescence spectra of the 'standard' and LPD lasers is simply a consequence of the higher (~ 10 times) specific input power, and slightly longer gain length, of the 'standard' device. The larger upper level population thus created increases the magnitude of the (negative) $k_\nu(\text{XeCl})$ (eq.4.24).



(a) Littrow mounted grating alone (used with LPD laser)



(b) Littrow grating with prism beam expansion

FIGURE 4-19: Tuning arrangement schematic

spectral emission it is likely that a narrower laser line-width would have been observed at times late in the pulse.

Comparison of the tuned output of the two lasers revealed that the linewidth of the LPD laser was comparable (at ~ 0.06 nm FWHM when tuned on the (0 - 2) transition) to that of the 'standard' laser although tuning optics of lower dispersive power were fitted to the LPD laser. A simple Littrow mounted grating arrangement (figure 4.19(a)) was used for the LPD laser with the grating reflecting the 'second order' diffracted light back into the cavity. In contrast, a '3rd order' Littrow mounted grating with prism beam expansion was used for the 'standard' laser (figure 4.19(b)). Tuning arrangements with greater dispersion, but higher losses, such as 'grazing incidence' gratings were not investigated because the combination of the (relatively) low gain and the short gain length of the LPD laser would almost certainly not have permitted laser oscillation. Tuned, time-averaged, laser spectra from the LPD laser are shown in figure 4.20 (a,b,c, and d) to provide an idea of the degree of selectivity which could be obtained against the other 'peaks'. In each case the laser was tuned for the maximum discrimination against the other 'peaks' in the spectrum which still allowed lasing on the peak of interest; the intensities in the figure are not relative. Also, in all but figures 4.20 (a) and (d), the linewidth indicated is very badly affected by instrument resolution (~ 0.025 nm).

In conclusion, some evidence of line narrowing was present in the LPD laser. However, the low gain, coupled with a short gain length, of the device meant that the relative improvement in linewidth over a 'standard' XeCl^* laser was not as great as might have been expected just from its pulse length. Any further development of long pulse duration RGH lasers would have to include a longer gain length to achieve worthwhile improvements in linewidth: increasing the gain by increasing the input power is not a viable solution in the light of the results presented in section 4.6.5.

4.7 DISCHARGE UNIFORMITY:

Until now, discussion of discharge uniformity has been couched in very qualitative terms. However, as will become apparent in the rest of this chapter

LPD laser: tuned laser spectra

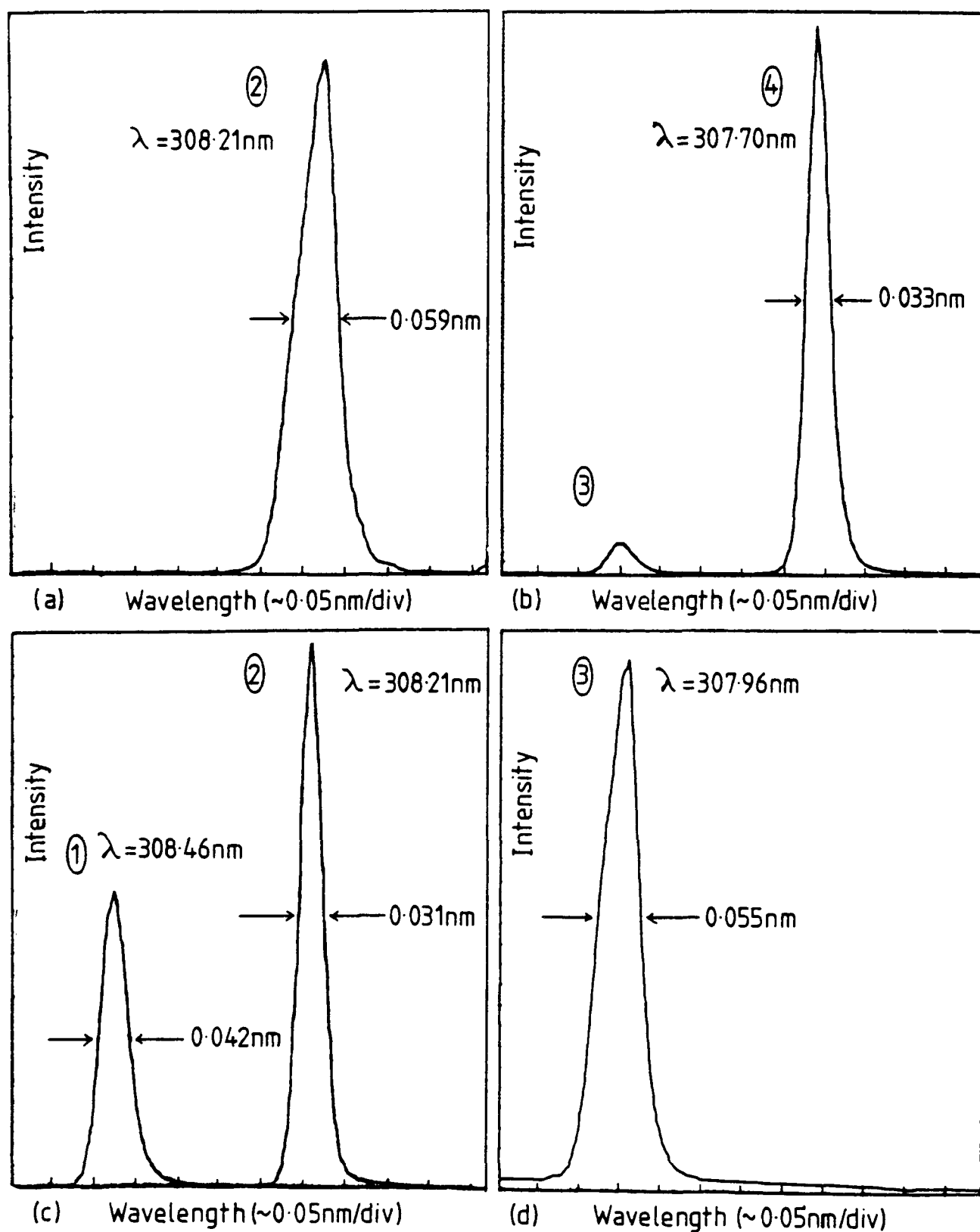


FIGURE 4.20: Tuned laser output from the LPD laser. A simple Littrow mounted grating was used (fig 4.19(a)). Instrument resolution was only 0.03nm. See Table 4.5 for identity of peaks.

and the next, a degradation in discharge uniformity was found to be the common factor in the termination of both the spontaneous and stimulated XeCl^* emission, and in the unexplained transition to lower total input powers to the discharge at the time of termination. Although valuable information concerning discharge uniformity could be gained by visual observation through the sidelight window this gave only a time-integrated view of what was occurring. Time resolved data of discharge uniformity would be able to give a much clearer view of the evolution of spatial non-uniformities.

4.7.1 Time-resolved observations of discharge uniformity:

The production of time resolved images of an event which lasts only 500 ns is not an easy thing to accomplish. Fortunately, it was possible to borrow a fast 'framing' camera (Hadland Imacon series 700).[†] This type of camera can produce images of transient events as a series of 10 ns exposure 'frames' with an 'interframe' time of 50 ns (i.e. the time elapsed between the initiation of one 10 ns frame and the initiation of the frame immediately following it). This is achieved, in simple terms, by allowing the image of a (luminous) transient event to be formed (via imaging optics) on a very sensitive photocathode. A gated H.V. pulse accelerates the photo-electrons emitted from the cathode onto a phosphor screen: the duration of this gated pulse determines the exposure time. Deflecting plates, situated between the photocathode and the phosphor screen, are switched between frames to direct each 10ns frame to a different region of the phosphor screen. A photograph of the images produced on the screen thus provides a permanent record of the time-resolved spatial uniformity of the event.

The camera which was available could not be used at its minimum (10 ns) exposure time because the emission from the discharge was not sufficiently bright (in sidelight) to enable a sufficiently magnified image to be recorded at this speed. The reason for this was mainly due to the large (~3 m) distance between the camera and the discharge. It was not considered wise to place the

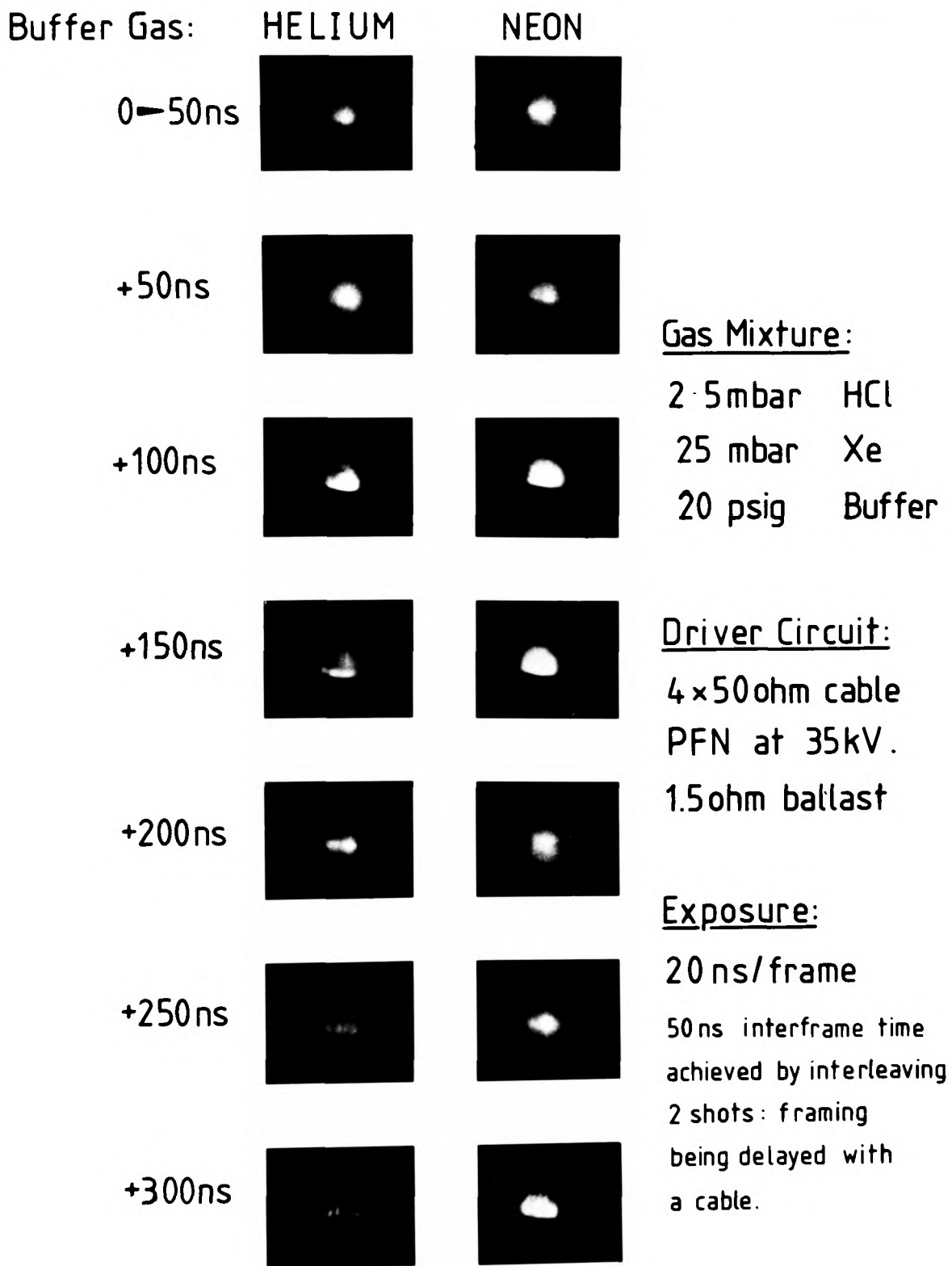
[†] The author wishes to thank Dr.A.Walker of the Culham Laboratory for the use of the framing camera and John Hadland Photonics Ltd. for the loan of an appropriate UV-transmitting objective.

camera inside the 'Faraday-cage laboratory' where it would be subjected to high voltage transients - especially since the camera was on loan. It was therefore placed in an adjoining laboratory and an image of the discharge in sidelight was transmitted to it via a system of mirrors and a silica 'field' lens (to provide a degree of magnification). In this configuration the intensity of 'visible' emission from the discharge was insufficient for effective operation of the camera: a UV objective had to be fitted to increase the total intensity of available light in this magnified image. The poor quality of the silica field lens which was available served to introduce spherical aberration into the resultant images - but not so badly that a recognisable image of the discharge was not formed when a 20 ns exposure time was employed. The camera was triggered using a fast-rising voltage pulse from the spark gap of the preionization circuit and the resultant jitter was of the order of 20 ns.

Plate 4.7 shows a comparison of the temporal evolution of the spatial uniformity of the LPD laser discharge (through the sidelight window) when helium and neon were used as the buffer gas. The optic axis is in the plane of the paper and goes from left to right; the direction of the transverse electric field is also in the plane of the paper and goes from bottom to top. Each folio of photographs (one for He, one for Ne) is the result of combining the images from the time histories of two discharge events: a 50 ns delay cable was introduced into the camera triggering line to obtain the images corresponding to 50, 150, and 250 ns after discharge initiation. Fluctuations in the overall intensity of neighbouring images should therefore be ignored. There is an uncertainty of ± 25 ns in the timing of the initial frame with respect to discharge initiation. The quality of reproduction in plate 4.7 is poor because it is a 'twice-removed' reproduction of the original polaroid images: features that were visible on the originals may not, therefore, be so apparent.

The spatial uniformity of both the helium and neon buffered discharges appears to be quite good in the first three frames of plate 4.7 (up to ~ 100 ns). The helium buffered discharge departs from this general uniformity at ~ 150 ns, whereas the neon discharge does not appear to change until ~ 250 ns

Framing Camera History of XeCl Discharge



(though there is slight evidence of non-uniformity at 200 ns). The uniformity of both discharges becomes progressively worse as time elapses beyond this point. Frames for times later than 300ns are not shown because the intensity of emission from the discharge was too low to enable an image to be recorded.

The change in uniformity would seem to be best described as a constriction of the discharge volume into individual columnar discharges associated with each pin of the ballasted electrode. In each case the time of onset of this constriction correlates well with the observed time of onset of laser pulse termination and with the fall in instantaneous power deposition into the discharge. It should be stressed that these 'columnar discharges' are certainly not fully-developed arcs. The appearance of an arc column in the discharge is altogether different. This is shown to good effect in plate 4.8 which is a time resolved folio of a discharge which was deliberately induced to arc by disconnecting the preionization circuit. Also, the individual columnar discharges of plate 4.7 are not the striations reported by Emeleus and Wolsey[1970] for discharges at low pressure in electronegative gases: these columnar discharges run parallel to the electric field whereas the striations reported by Emeleus and Wolsey run perpendicular to the electric field. One further point is that almost the entire intensity in the images of plate 4.7 is due to the 308 nm XeCl^* spontaneous emission rather than any other emitting species.

4.7.2 Time-integrated observations of discharge uniformity:

Photographs of the time-integrated spatial uniformity of the discharge as a function of the HCl partial pressure of the gas mixture were taken to provide a permanent record of what was observed visually. The reason for choosing the concentration of HCl as the variable was that this parameter had the most marked effect on the spatial uniformity of the discharge.

Open-shutter photographs of single discharge events were taken both through the sidelight window and along the optic axis. Photographs of discharges containing four representative HCl partial pressures are presented in plate 4.9. The anode and (ballasted) cathode are indicated. These photographs

Temporal evolution of a forced arc in LPD laser

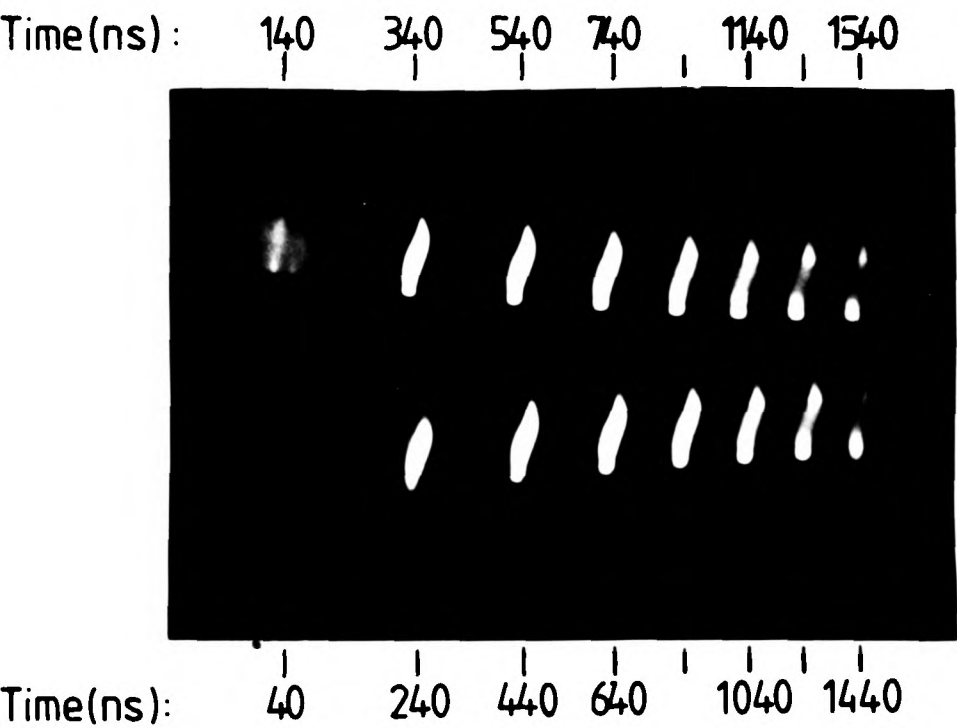


PLATE 4.8: Temporal evolution of a 'forced' arc in the LPD laser. Individual frame exposure of 20ns.

record the time-integrated intensity of 'visible' emission only: any 308 nm emission would have been filtered out by the glass lens of the camera. Care was taken to align the camera along the optic axis so as not to record an image which showed the discharge to be thicker than it really was. However, there has been some unavoidable artificial thickening of the discharge width in these photographs.

Plate 4.9 shows, rather dramatically, the destabilizing influence of HCl on the discharge. This is very much a result of the electronegative, rather than molecular, nature of the gas. The LPD laser was found to operate as a short-pulse duration Nitrogen C-state laser (337nm - kinetically terminated) and the discharge uniformity was then excellent. Returning to plate 4.9, it can be seen that the columnar discharges associated with each ballasted pin dominate the discharge at HCl partial pressures of 3.5 mbar and 10 mbar. The discharge becomes progressively more diffuse at lower HCl pressures until in the absence of HCl (not shown) the discharge fills the entire discharge gap as seen in sidelight.

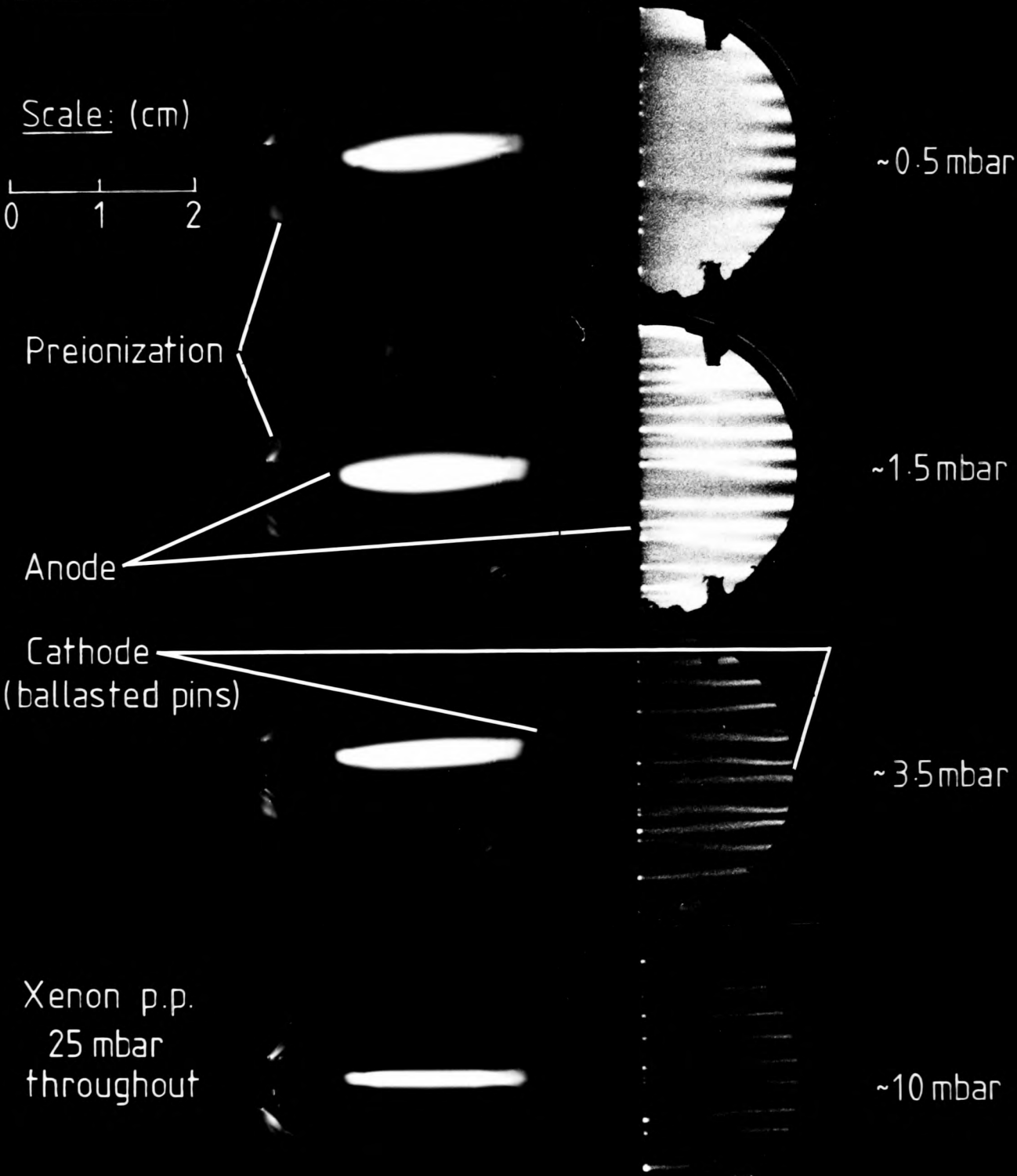
The discharge uniformity at the usual 2.5 mbar partial pressure for lasing was more akin to that shown for a 1.5 mbar HCl partial pressure than that shown for a 3.5 mbar partial pressure. The decanting volume was used in this experiment and it is likely that the 3.5 mbar pressure indicated is an underestimate - by as much as 1 mbar.

Although giving a time-integrated view of discharge uniformity vs. HCl partial pressure, plate 4.9 provides an analogue for the evolution of the spatial uniformity of the discharge in time: the low HCl partial pressure photographs correspond to the uniformity early in the discharge and the high pressures correspond to the lack of uniformity in the later stages. Unlike the intensity of the images produced in the time-resolved study, the intensity in these images was due mainly to visible light (UV transmitting optics were not used). It is shown in section 5.3 that the dominant emission in the later stages of each discharge is from a xenon ion transition and that in the early stages is from a xenon neutral transition. The onset of emission from the ion

DISCHARGE UNIFORMITY versus
HCL Partial Pressure

Viewing
Direction: Along Optic Axis ■ Transverse

Scale: (cm)
0 1 2



Total Pressure: 2.5 bar (Ne buffer)

transition becomes earlier as the HCl concentration is increased. There is obviously a very close connection between the observation of the constricted columnar discharge and the onset emission from the ion transition. It is in this sense that it can be claimed that plate 4.9 provides an insight into the temporal evolution of the discharge.

4.8 PULSE TERMINATION:

It is quite a simple matter, with the benefit of hindsight, to identify the major cause of the laser pulse termination: massive reduction in the discharge volume. However, there is no obvious explanation for why discharge constriction should occur. Some initial insight into the problem might be gained by examination of a proposed mechanism for pulse termination which was being discussed in the Clarendon Laser Group before the evidence for discharge volume constriction became overwhelming.

Given a constant discharge volume there is no obvious reason why the production of XeCl^* should not proceed at a time independent rate provided that the primary reactants are replenished at a rate equal to that at which they are consumed. For instance, it is unlikely that the population of xenon ions, $[\text{Xe}^+]$, is depleted on the basis of the overall charge neutrality argument put forward in section 4.6.2(a). Depletion of $[\text{Xe}]$ from which the ions (or the excited states which form the ions) are formed can also be discounted: not only on the basis of the high initial number density ($\sim 5 \times 10^{17}/\text{cm}^3$) but also because any xenon ions lost through the formation of XeCl^* become available for reuse as neutral xenon almost immediately after the emission of the UV photon.[†]

On the other hand, depletion of $[\text{HCl}]$, which supplies 'fuel' for the formation of XeCl^* through both the ion and neutral channels, is quite

[†] The lifetime of the XeCl ground state molecule is very short since its binding energy is only of the order of the thermal energy of the gas at room temperature (Tellinghuisen[1976]).

plausible.[†]

Replenishment of HCl within the active volume by diffusion from the surrounding gas volume will be insignificant on the time scales of interest. The average displacement, $\bar{x}$, of an individual particle in a gas in one co-ordinate, is given by

$$\bar{x} = \sqrt{2Dt} \quad (4.25),$$

where 't' is the time in seconds and 'D' is the mutual diffusion coefficient of the gas. Diffusion coefficients for HCl in helium or neon could not be found. However, if we provide a generously large alternative - such as Argon (which has a similar mass to HCl but would have a smaller collisional cross-section) in helium - then the point can still be made. Present[1958] gives D for He-Ar as $1.4 \text{ cm}^2/\text{s}$ at atmospheric pressure and a temperature of 418 K. At a pressure of 2.33 atmospheres, $D = 0.6 \text{ cm}^2/\text{s}$, because it is inversely proportional to the pressure of the gas. For the duration of the 500 ns discharge pulse, the mean distance travelled by the 'HCl' will not be greater than

$$\bar{x} = \sqrt{2 \times 0.6 \times 500 \times 10^{-9}} = 8 \mu\text{m}$$

which is very small even compared to the thickness of a constricted column ($\sim 0.5 \text{ mm}$). Replenishment of HCl by diffusion on the time scales of an individual pulse can therefore be discounted.

On the basis that HCl, once dissociated, is effectively lost from the system, an estimate of the scale of depletion of HCl can be made: one 308 nm photon corresponds to one 'deceased' XeCl^* molecule which, in turn, corresponds to the disappearance of one HCl molecule. The energy of a 308 nm photon is

[†]The initial population of HCl is an order of magnitude lower than that of xenon. The rates for dissociative electron attachment and for vibrational excitation of HCl are strongly dependent on $[\text{HCl}(v=0)]$: Cl^- is required for XeCl^* formation via the ion channel and vibrationally excited HCl is required for a large part of the formation of XeCl^* through the neutral channel in the case of the low-energy XeI excited state atoms - a reaction which is otherwise slightly endothermic (the low-energy XeI excited states account for a significant proportion ($\sim 5\%$) of the total xenon excited state population - Kearsley[1980]). The rate of reformation of HCl from neutral Chlorine atoms (the product of the decay of the XeCl ground state) and atomic Hydrogen is on the millisecond time scale so, once used, an HCl is effectively lost from the system. Neutral atomic Chlorine itself may be able to take part in XeCl^* formation but this would be significantly slower than if HCl were available because the cross section for electron attachment to Cl is ~ 10 times smaller than that for HCl.

$\sim 6.5 \times 10^{-19} \text{ J}$. The total laser pulse energy in plate 4.3 was $\sim 2.5 \text{ mJ}$: the optical cavity consisted of 3° off-axis windows and plane-plane mirrors with nominal reflectivities of 99.6% and 96% (soft dielectric - so loss due to absorption was possibly high). Each face of each 3° window will give rise to a loss of $\sim 4\%$ per pass (8% double-transit) - which is equivalent to the loss to the intracavity flux due to the 96% reflectivity output coupler. The total loss of laser photons in a double transit of the cavity (assuming 100% reflection at the far end and no transient loss) will be nine times the observed laser output (4 faces of 3° windows $\times 2$ + loss due to output). Therefore the specific density of laser photons extracted from the discharge is

$$\frac{9 \times (\text{pulse energy})}{\text{Volume} \times (\text{photon energy})} = \frac{(9 \times 2.5 \times 10^{-3})}{(15 \times 6.5 \times 10^{-19})} = 2.3 \times 10^{15} \text{ photons/cm}^3$$

which assumes that the active volume over the 500ns duration of the discharge pulse is a constant 15 cm^3 (which blatantly is not true). The ratio of integrated intensities of the spontaneous emission with and without lasing is $\sim 1:2$ (from plate 4.3). Therefore, the specific density of XeCl^* molecules produced per cm^3 (taking into account processes such as electron quenching but not passive or transient absorption of photons) in the 500 ns is $\sim 6.9 \times 10^{15}$. This represents some 10% depletion of the initial HCl population - a very generous underestimate considering the assumptions used. Depletion of HCl in a constricted discharge region would be enhanced because of the higher electron density.

In conclusion, it would seem that pulse termination is likely to be the result of two closely linked factors: HCl depletion in the active volume and discharge constriction. However discussion of a mechanism which could give rise to discharge volume constriction on the basis of HCl depletion will be deferred until section 7.2.2. The proposal that HCl depletion is, in some manner, responsible for constriction is consistent with the observation (section 4.6.2(b)) that relatively minor changes in the initial HCl partial pressure have a strong influence on the behaviour of the discharge.

4.9 SUMMARY:

The major objective of this study was achieved with the construction of a long laser pulse duration, discharge-excited XeCl^* laser. This was done by employing a distributed resistance electrode in the stabilization of the discharge. Some of the principal experimental results presented in this chapter have been published elsewhere (Hogan et al.[1981]).

The inclusion of good diagnostic facilities at the design stage enabled extensive parametric characterization of the laser system. This has permitted constriction of the total discharge volume coupled with the depletion of the halogen donor gas, HCl , within that volume, to be identified as the cause of the premature termination of both spontaneous and stimulated emission at the $\text{XeCl}(\text{B-X})$ transition wavelength of 308 nm. Work described in the next two chapters provides further evidence for this laser pulse duration limitation mechanism for discharge excited XeCl^* lasers. A proposal for the actual mechanism responsible for constriction of the discharge volume will be put forward in chapter 7.

C H A P T E R F I V E

Time-resolved measurements of emission and absorption in the LPD laser: excited-state xenon populations and other factors

5.1 INTRODUCTION:

5.1.1 Aims:

Time-resolved measurements of emission and absorption in the LPD laser provided a further means of characterizing the behaviour of the device. Since many of the experiments described below were carried out before the full extent of discharge constriction was known, much of what had been initially hoped to be achieved with these measurements could not be realised. In particular, measurements of xenon excited state populations in the period after constriction had occurred were not reliable: accurate knowledge of the length of the discharge path length is required for absolute population measurements via absorption.

Three distinct experiments are described in this chapter. However, it is difficult to discuss the results of each in isolation since conclusions about the possible cause of a particular effect in one experiment are arrived at, in many instances, in the light of observations from one of the others. Consequently, although each experiment will be discussed separately, frequent reference may be made to other sections. The three experiments were:

- (1) Measurements of time-resolved absorption of ultraviolet radiation at selected wavelengths (i.e. those that were readily available) by the LPD laser discharge: short duration ($\sim 20\text{ns}$) laser pulses (193, 249, 337, and 353nm) were passed through the LPD laser discharge at different times during the (500ns) discharge pulse.
- (2) Measurements of time-resolved emission from the LPD laser discharge from

~300nm to ~600nm: concentrating particularly on xenon neutral and ion emission lines.

- (3) Time-resolved measurements of the absorption of short-duration, high-intensity, broad spectral-band, visible radiation by the LPD laser discharge: the 460-490nm fluorescence from a laser pumped dye was passed through the LPD laser discharge at different times during the discharge pulse.

Initially it was hoped that the first experiment would provide information concerning the ground-state HCl population - to determine if depletion of HCl was a primary cause of laser pulse termination. In the event this was not possible (see below). However, an indication of the degree of transient loss in the discharge at the laser wavelength was obtained. The emission measurements were to provide conclusive evidence of discharge constriction and an estimate of the electron density in the discharge: this estimate could then be compared with that obtained via eq.4.21.[†] The final experiment was to be used to obtain a measure of populations of xenon excited states as a function of various discharge parameters (e.g. gas mixture composition) including time. These results could then be compared with those obtained by Kearsley[1980].

5.1.2 Theory:

It is possible to measure populations of excited state species via emission from and absorption by a plasma. For the relatively simple case of line emission from an optically thin, homogeneous slab of plasma, the spontaneous radiation is emitted in all directions, and the energy emitted per unit solid angle per second by unit area, $B(\nu_o)$, is given by:

$$B(\nu_o) = \frac{1}{4\pi} A_{21} N_2 \delta x h \nu_o \quad (5.1),$$

where $A_{21} \cdot N_2 \cdot \delta x$ is the number of spontaneous transitions per second in a slab of unit cross-section and path length δx , N_2 is the number density of the upper state, and A_{21} is the spontaneous transition probability. Consequently, if it

[†] The importance of having a reliable estimate of the electron density lies in the fact that it provides an upper bound estimate limit of the xenon ion population (via the charge neutrality condition) and thus the contribution to XeCl^* formation from the ion channel (eq.1.1).

is wished to determine the population, N_2 , absolutely, a measurement of $B(\nu_0)$ is required (assuming A_{21} is known). The normal practice is to compare the source with a standard source, with due precautions over solid angles, equivalent light paths, apertures, etc. - which is by no means trivial (Thorne[1974]). However, if the experimental arrangement is unaltered and the emitting volume remains constant, comparison of the relative intensities of emission can provide a sensitive monitor of the relative population of a particular level either in time or under different discharge conditions.

The population in an energy level, N_1 , from which absorption takes place, assuming a comparatively small upper level population, is given by:

$$N_1 = \frac{4\epsilon_0 mc}{e^2 f_{12}} \int k_\nu d\nu \quad (5.2),$$

where e , m , and c have their normal meanings, and f_{12} is the oscillator strength for the transition which is related to the transition probability, A_{21} by:

$$f_{12} = \frac{g_2}{g_1} \frac{\epsilon_0 mc^3}{2\pi e^2 \nu^2} A_{21} \quad (5.3),$$

where g_1 and g_2 are the statistical weights (degeneracy) of the levels. The value of $\int k_\nu d\nu$ is determined experimentally. It is arrived at through the integration of the appropriate transformation of Beer's Law (eq.4.15):

$$\int k_\nu d\nu = \frac{1}{\ell} \int \ln\left(\frac{I_\nu(0)}{I_\nu(\ell)}\right) d\nu \quad (5.4),$$

which is merely the area contained by the logarithm of the normalized absorption over the full width of the absorption line.

In comparison to emission studies, measurement of populations by absorption is relatively simple in RGH discharges. A significant factor in favour of using absorption to determine the xenon excited state populations under the conditions present in RGH discharges is that the absorption features are generally very broad[†] (0.05-0.25 nm): this enables $\int k_\nu d\nu$ to be determined directly.

[†] The broadening is due to the Stark effect as a result of the high electron densities required to operate a discharge-excited RGH laser. When the absorption feature is well resolved 'equivalent width' and 'curve of growth' methods (Thorne[1974]) are not required. These methods were used by Kearsley[1980] for some xenon lines because he did not find it practicable to examine all the xenon absorption features under high resolution. In this study only three xenon neutral absorption features were examined and all were examined with sufficient resolution that instrument broadening was not significant.

A major source of error in the determination of energy level populations via either eq.5.1 or eq.5.2 is the value of transition probability, A_{21} , which is used. As is discussed by Kearsley[1980], the transition probabilities for only very few xenon transitions have been determined experimentally: for most transitions A_{21} has only been determined theoretically. Furthermore, most experimental measurements are of poor reliability - especially those done before ~1970. In those cases where experimental and theoretical values of the transition probability are known, they differ by as much as 40%.

The population, N , of a species can equivalently be determined simply through:

$$N = k_{\nu} / \sigma_{\nu} \quad (5.5),$$

if the absorption cross-section at a particular frequency, σ_{ν} , is known.

5.1.3 Hydrogen Chloride:

The initial impetus for wanting to measure transient absorptions in the ultraviolet was the hope that the time evolution of the ground state HCl population could be monitored. The calculation performed at the end of chapter 4 indicated that depletion of HCl could be responsible (at least partly) for laser pulse termination.

HCl has absorption features in the infrared (IR), associated with its molecular structure, and it exhibits broad-band (~250-180 nm) absorption in the ultraviolet(UV) (see Liefson[1928] and Vodar[1948]) due to transitions to excited electronic states of the molecule. Under low resolution, the fundamental IR absorption band of HCl appears to be continuous between ~3-4 μm with a peak at ~3.5 μm : at first sight the 3.39 μm He-Ne laser transition would seem to provide an ideal probe source for absorption. However, under high resolution, this fundamental absorption is seen to have considerable fine structure (Herzberg[1950]). The width to spacing ratio of these features could be expected to be similar to those of Hydrogen Fluoride (HF) at these pressures. Eberhardt and Pryor[1981] have obtained data pertaining to the broadening of the infra-red absorption spectrum of HF in the presence of a 'foreign' gas. Assuming that HCl has an infra-red absorption cross-section similar to HF, the absorption at line centre due to one of these features would

be complete (i.e. no transmission) for a 2.5 mbar partial pressure along the 40 cm absorption path presented by the LPD laser:

$\sigma_v = 2 \times 10^{-17} \text{ cm}^2$ and $[\text{HCl}] = 6 \times 10^{16} \text{ cm}^{-3}$ (2.5 mbar) which implies (from eq. 5.5) that $k_v = 1.2 \text{ cm}^{-1}$ and $\ell k_v = 40 \times 1.2 = 48$. Substituting this into equation 4.15, the ratio of transmitted to initial intensity through the LPD laser would be:

$$I(\text{trans})/I(0) = e^{-48} = 10^{-21},$$

which is clearly too much absorption to obtain a meaningful measurement.[†]

This approach is further complicated by the need to obtain a narrow band light source of sufficient intensity which emits at the line centre of one of the absorption peaks. If line broadening of the HCl IR absorption features has a similar functional dependence on 'foreign' gas pressure to that of HF then the ratio of linewidth to line-spacing would be ~1:1000. Furthermore, the frequency of line centre is also pressure dependent and there is no accurate method of predicting the magnitude of the shift. Some element of luck would therefore be required to find a readily available laser source with an emission feature overlapping one of these absorption peaks. Monitoring the temporal evolution of the population of vibrationally excited HCl in the IR would be almost impossible mainly because of a lack of appropriate detectors at these wavelengths. Although fast IR detectors are available, there is a considerable trade-off between response and sensitivity - typically IR laser radiation would be required for reasonable sensitivity at (say) 100 ns response times. An attempt was made to monitor HCl absorption in the IR with a 3.39 μm He-Ne laser without success. Since no other infrared source of reasonable intensity was available this approach was abandoned.

Measurement of HCl absorption in the UV region of the spectrum also proved to be unsuccessful. An experiment was performed to determine the passive absorption by HCl of ArF (193 nm) laser radiation (20 ns duration) at different pressures (no other gas present). The 193 nm radiation was passed down the

[†] This is especially true when one considers, with the benefit of hindsight, that the amount of depletion along the total absorption path will be smaller than the depletion within the constricted columnar discharges, i.e. there are large 'unpumped' regions between each columnar discharge.

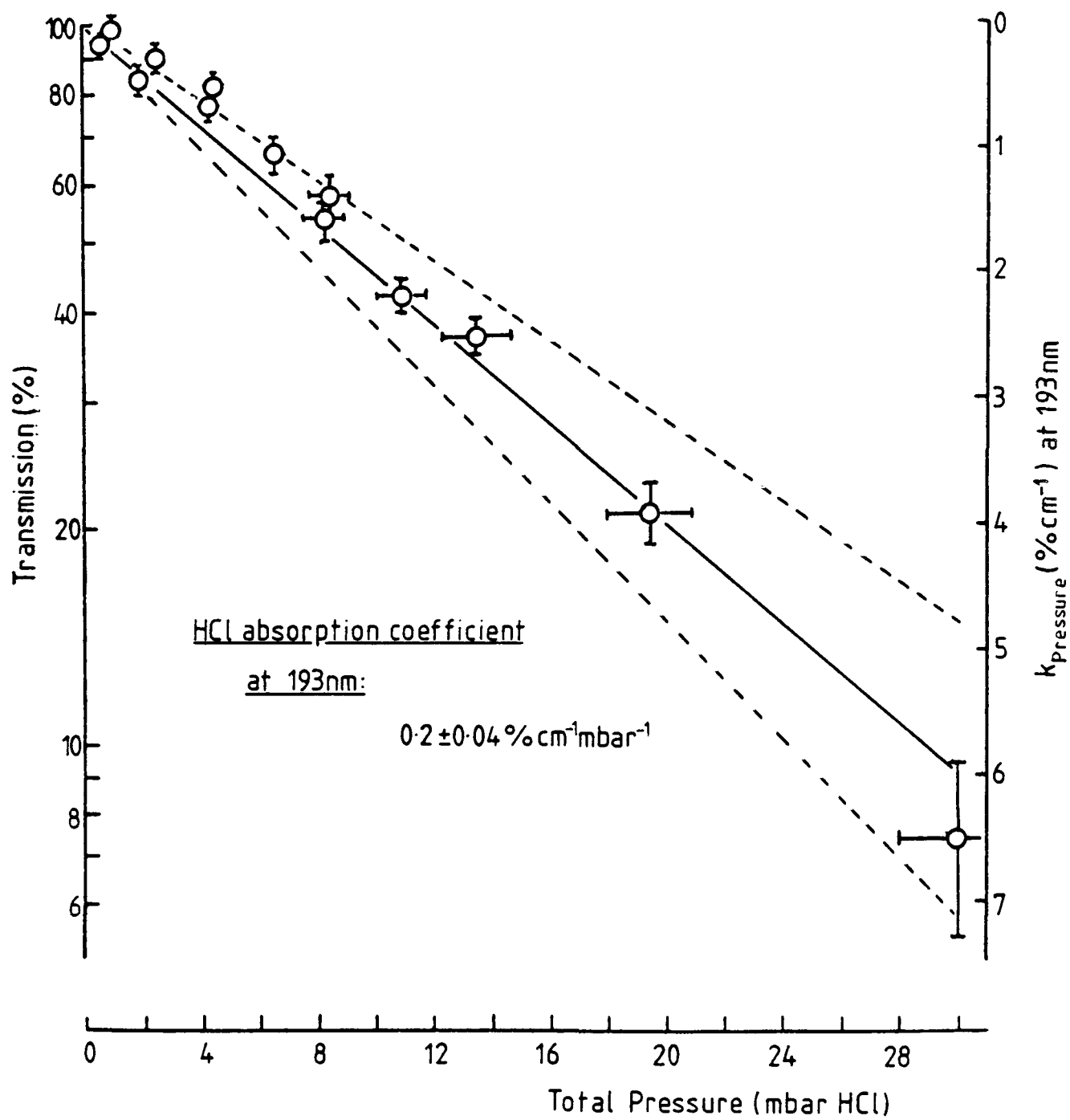


FIGURE 5.1 : Transmission of 193nm (ArF) laser radiation through a 40cm absorption path containing HCl at different pressures.

length of the LPD laser in the absence of a discharge and was detected with a fast, UV-sensitive photodiode. The peak height of the signal appearing on a fast storage oscilloscope was recorded with the laser tube evacuated (to determine the $I(0)$ intensity) and with various partial pressures of HCl present. It was assumed that $I(0)$ did not alter significantly over the course of the half hour experiment. The results are presented in figure 5.1. The error bars for pressure are a consequence of the difficulty in reading the semi-logarithmic scale of the Leybold pressure gauge; those errors in the percentage transmission are the result of poor shot-to-shot reproducibility in the probe laser output (only one shot was taken at each pressure point). This accounts for the large ($\sim 20\%$) error in the HCl absorption coefficient, $0.2 \pm 0.04 \text{ cm}^{-1} \text{ mbar}^{-1}$ at 193 nm, derived from the gradients of the lines indicated on the figure.

The total absorption along the (40cm) absorption path at a HCl partial pressure of 2.5 mbar (typical for lasing) was no more than 10%. When the absorption was monitored during a discharge event however, rather than the expected decrease in absorption (as would be expected if the HCl was being depleted), a very large increase in absorption was observed. Transient absorbing species dominate absorption in the LPD laser at 193nm.

5.2 TRANSIENT ABSORPTIONS IN THE ULTRAVIOLET:

5.2.1 Experimental details:

The experimental arrangement which was used for measuring transient absorptions in the UV is set out in figure 5.2. An 'Oxford Lasers KX10'[†] RGH laser provided short duration (~ 20 ns) laser pulses at various UV wavelengths (see Table 5.1) depending on the gas mixture with which it was filled. This light was used to probe the LPD laser discharge for absorption at different times during the 500 ns excitation pulse of the LPD laser. Low jitter (~ 5 ns) triggering of the LPD discharge and the probe laser was achieved by employing a

[†]The author wishes to thank Oxford Lasers Limited for the loan of this device for the absorption experiments described in this chapter.

Transient Absorptions at 193nm, 249nm, & 351nm : Experimental Arrangement

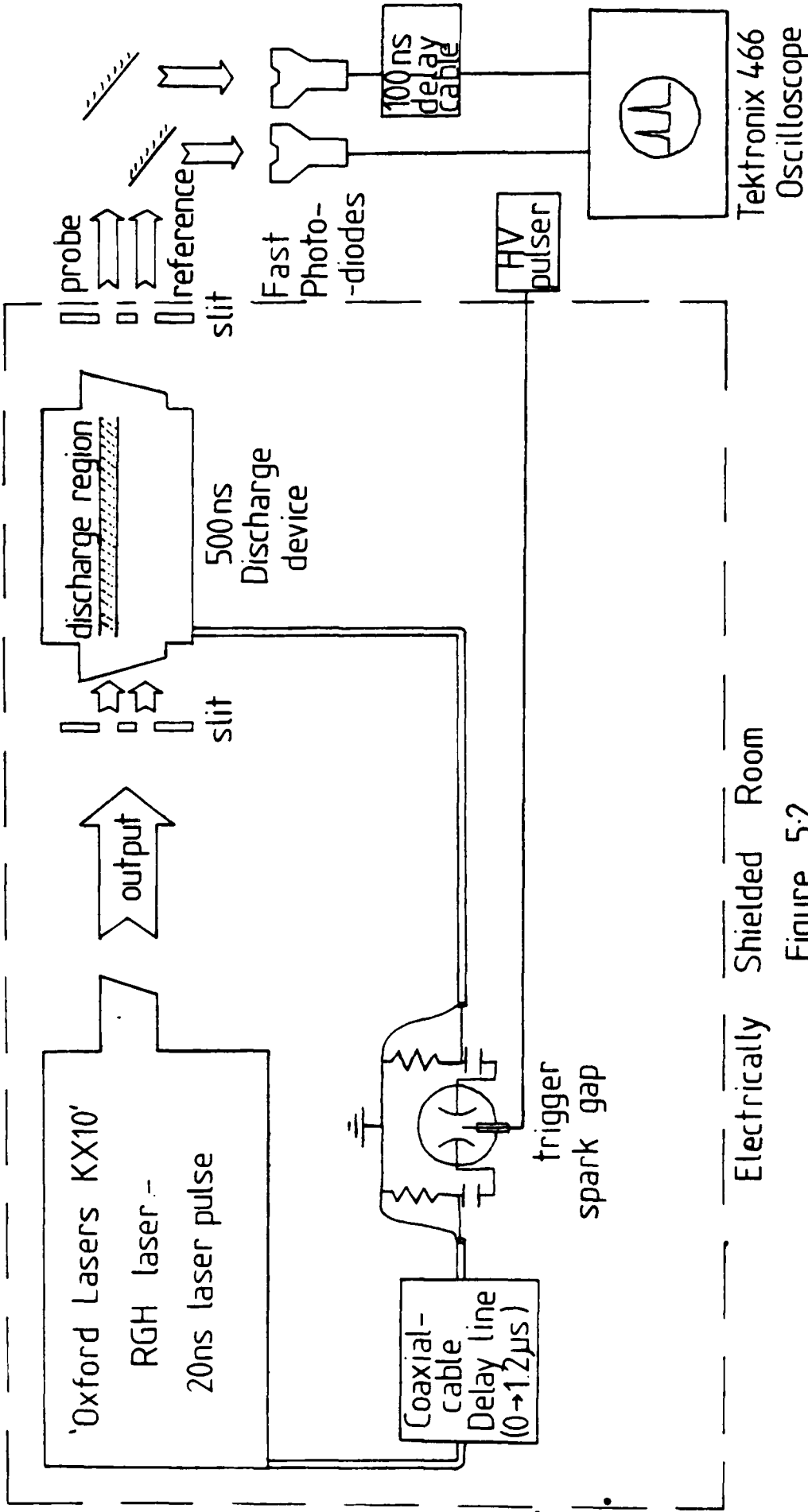


Figure 5.2

Table 5.1
UV laser wavelengths used for the measurement of transient absorptions

Laser	Wavelength (nm)	Wavenumbers (cm ⁻¹ x 10 ⁴)	Photon Energy (J x 10 ⁻¹⁹)	Photon Energy (eV)
XeF*	353	2.83	5.63	3.52
N ₂	337	2.97	5.90	3.68
XeCl*	308	3.25	6.46	4.03
KrF*	249	4.02	7.99	4.99
ArF*	193	3.25	10.3	6.43

'master' spark gap to provide fast-rising HV trigger pulses, via delay cables of adjustable length, to the main spark gap of each device.

Pairs of slits were placed at each end of the absorption cell (the LPD laser): each pair consisted of two similar (1.5 mm x 2 cm) slits separated by ~1.5 cm. The top slit at each end defined a path through the discharge region of the absorption cell for the probe radiation; the lower slit at each end of the absorption cell defined a path through a similar length of passive gas for a reference beam.[†] The slits were placed in the correct position by aligning them such that lasing was observed from the absorption cell when the slits were in an intra-cavity position. The reference beam was used to reduce problems associated with the shot-to-shot reproducibility of the KX10 laser and the build-up of passive absorbing species within the absorption cell over the duration of a 'run'.

The separate probe and reference beams emerging from the far end of the absorption cell were passed through a porthole in the Faraday cage laboratory and onto two fast UV sensitive photodiodes (one for each beam). The pulses were directed into a fast storage oscilloscope and their peak heights were measured. Since a dual-beam oscilloscope was not available, a 100 ns coaxial-cable delay line was introduced between the photodiode which detected the probe pulse and

[†] The KX10 laser was at a sufficient distance from the absorption cell that the beam had diverged to cover both slits in each pair. Initially, it was assumed that the relative intensities of light from different parts of the beam remained constant during the course of an experiment, however it was soon found that there was a consistent (~10%) decrease in the ratio of the intensity of the reference to probe intensity over the course of an experiment. Consequently, the relative intensities of the two beams were checked (without discharge) before and after each 'run'. Each data point was subsequently multiplied by a factor which corresponded to a linear change in this ratio with time.

Transient Absorptions at 193nm, 249nm, & 351nm : Experimental Arrangement

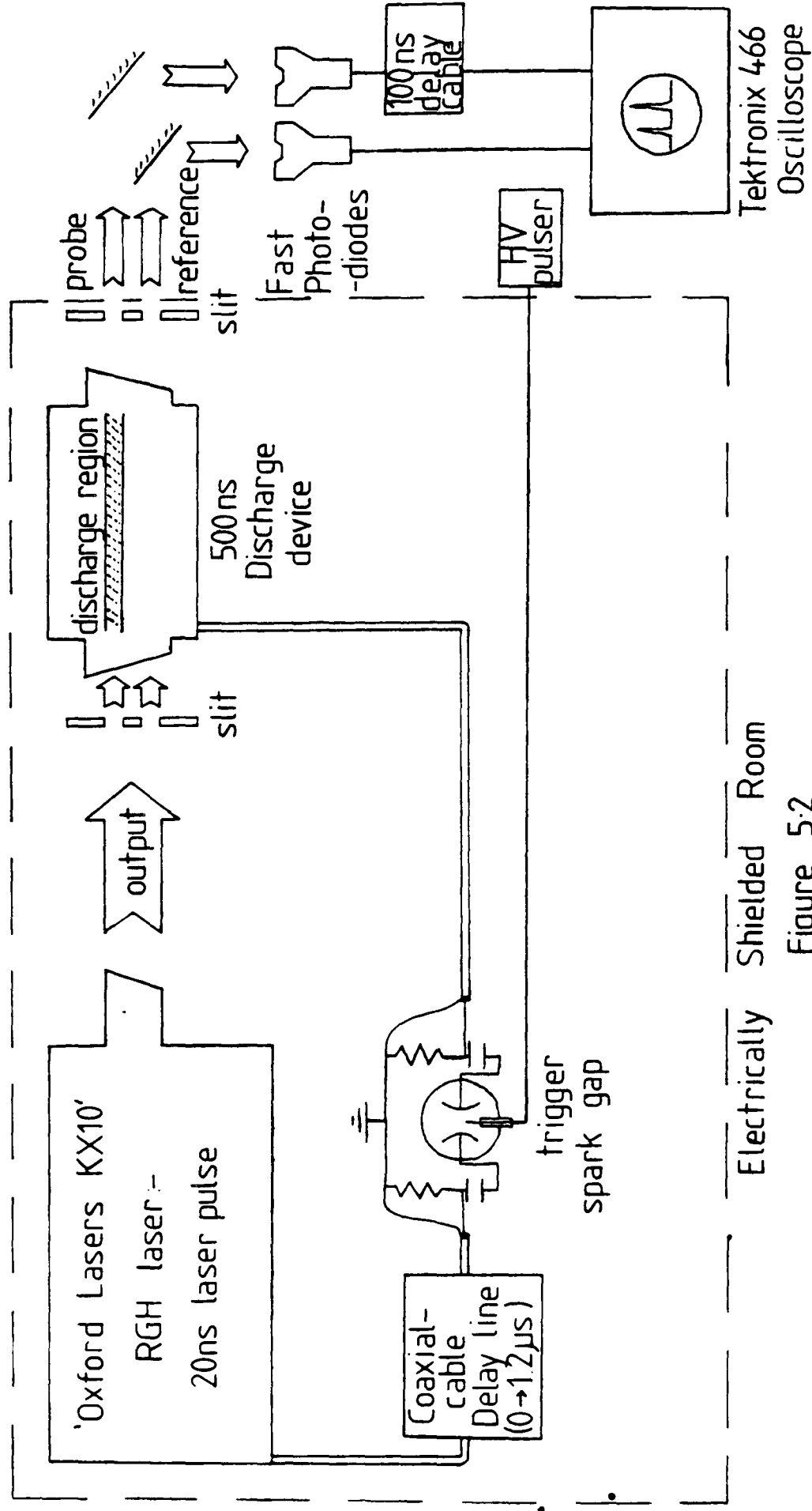


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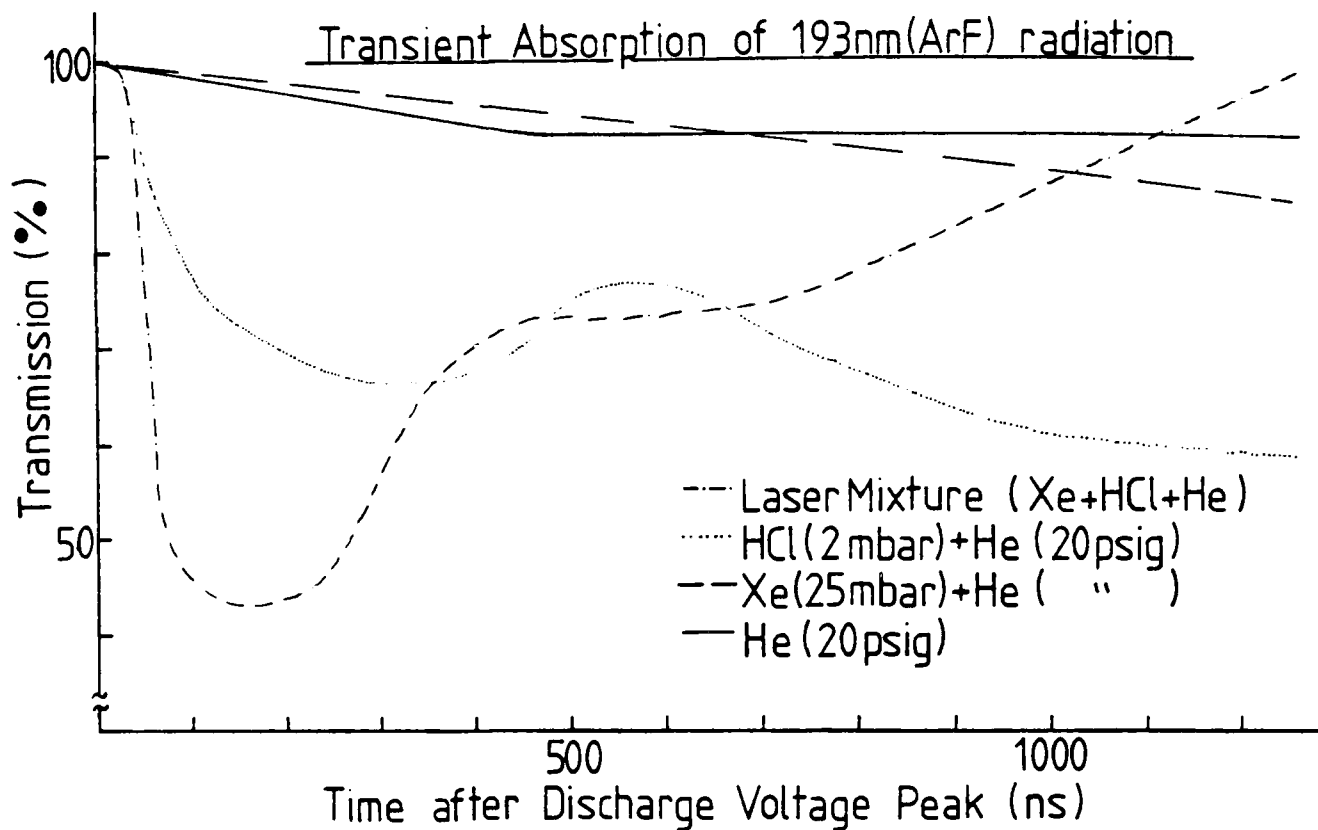


Figure 5.3

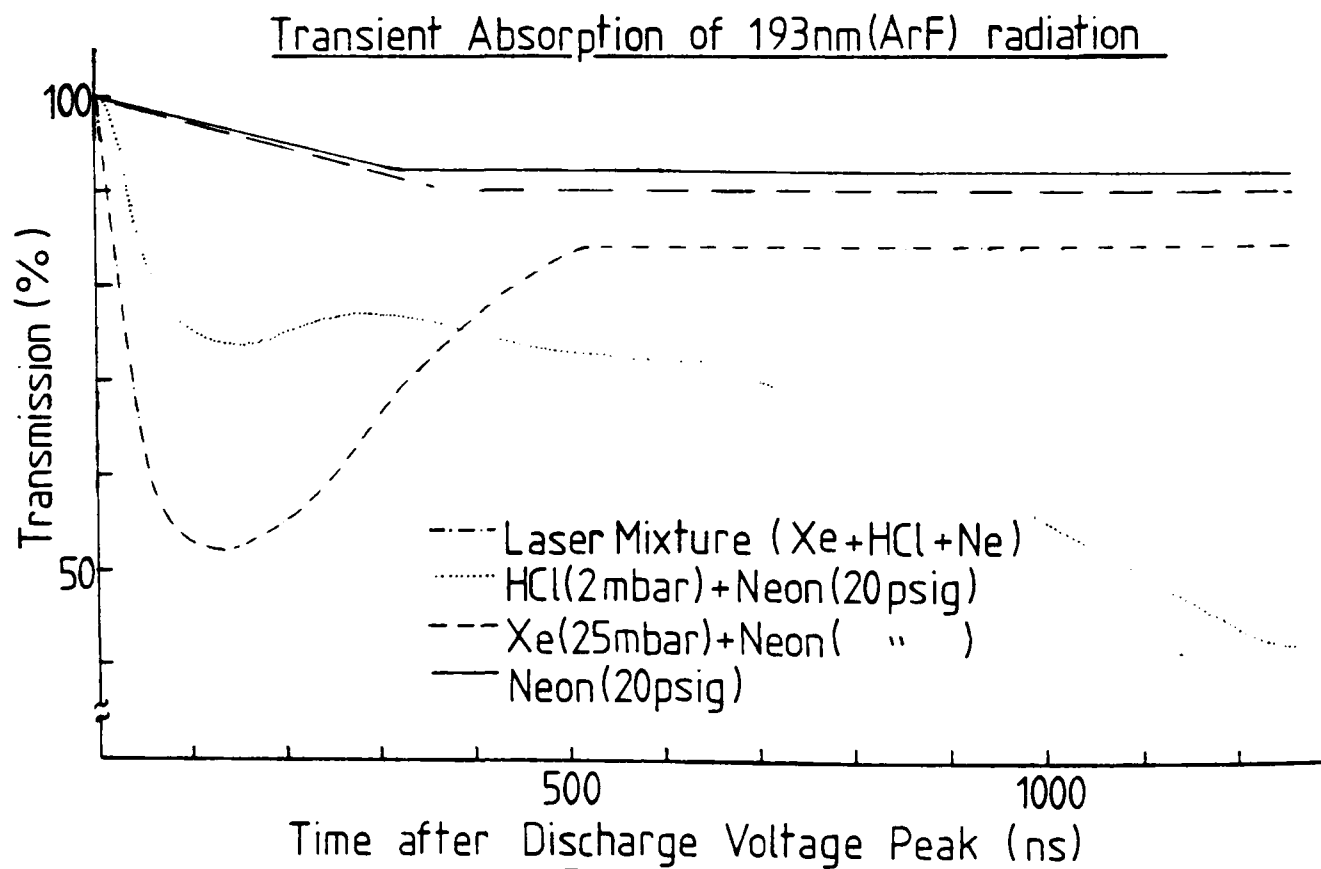


Figure 5.4

the oscilloscope to separate the two signals. It was confirmed that this cable delay did not alter the measured peak height of the probe pulse within the accuracy to which it could be read on the oscilloscope.

5.2.2 Results:

The results of this experiment are presented in six figures: 5.3 through to 5.8. In the case of transient absorption of ArF^* radiation, each data point was arrived at by taking the mean of ten shots; the mean of five shots was obtained at all other wavelengths. The root mean square deviation from the mean of each data point gives an indication of the shot-to-shot reproducibility of the LPD laser discharge as an absorber at that time. The peak height of the probe signal was divided by the peak height of the reference signal for each shot taken. The normalized (percentage) transmission was obtained by dividing the mean of peak/reference at a particular time by the (adjusted:see footnote p.94) mean ratio of peak to reference in the absence of a discharge in the absorption cell.

Figures 5.3 and 5.4 show the absorption of ArF^* radiation by the LPD laser discharge with time for various combinations of gases: in one case neon is the major constituent, in the other helium is the major constituent. Error bars are not included in these figures but it can be assumed that the magnitude of the error is similar to that shown in figures 5.6, 5.7 and 5.8. The most striking feature of these results is the very large increase in absorption in those discharges containing HCl : as noted in the previous section, if it had been hoped to measure a depletion of HCl ground state molecules then a decrease in total absorption (i.e transmission > 100%) would have been expected.

A clearer view of the functional dependence of this absorption with wavelength can be obtained by examination of figures 5.5 (helium buffered laser mixture), 5.6 ($\text{HCl} + \text{He}$), 5.7 (neon buffered laser mixture), and 5.8 ($\text{HCl} + \text{Ne}$). The absorption of $\text{N}_2(\text{C-state})$ laser radiation was measured only in the case of a helium buffered laser mixture. The magnitude of the increase in absorption in $\text{HCl} + \text{He}$ and $\text{HCl} + \text{Ne}$ gas mixtures between discharge initiation and ~ 100 ns and up to 500ns becomes progressively smaller at longer wavelengths. In helium and

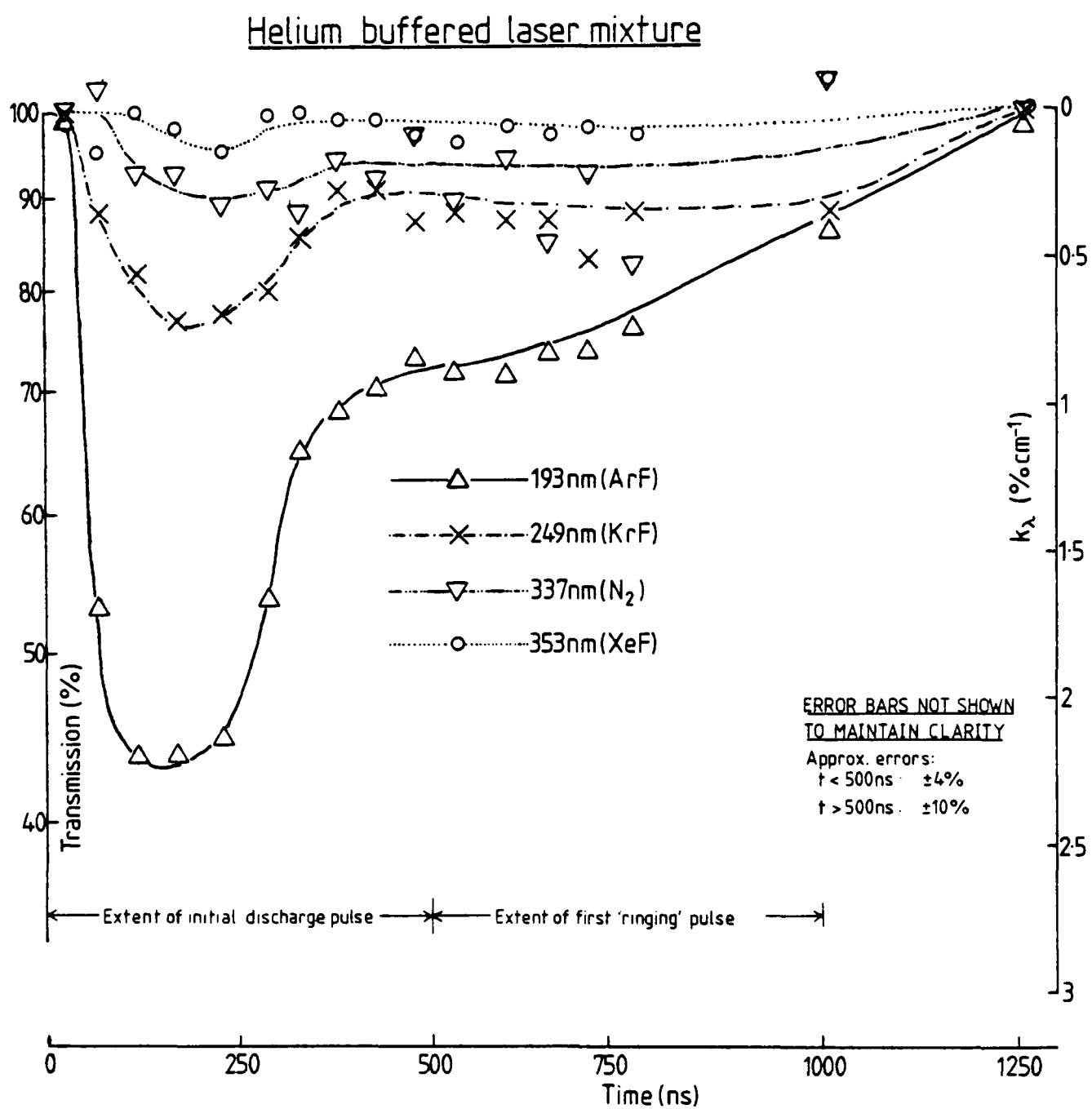


Figure 5.5: Transient absorptions

neon buffered laser gas-mixtures this general feature is retained. However there are significant differences:

- (1) the absorption shows a marked decrease in magnitude at ~ 250 ns whereas it is comparatively flat in those mixtures not containing xenon.
- (2) a smooth decay of absorption after 500ns is observed in the case of mixtures containing xenon whereas the absorption shows a general increase in magnitude and deterioration in shot-to-shot reproducibility when xenon is not present.

5.2.3 Discussion:

(a) General:

As will be discussed further in the next chapter, transient loss is possibly the single largest unknown which affects laser performance. The correct interpretation of experimental measurements of transient loss is therefore vital to the understanding of the laser operation.

The marked decrease in absorption at ~ 250 ns in discharges containing all three of the gases required for laser action can be accounted for by discharge constriction. However, this observation by itself is not sufficient evidence for concluding that constriction is occurring. For example, if absorption by XeCl^* (leading to photodissociation) were the cause of the absorption, the proposed explanation of laser pulse termination would depend on an argument which is dangerously circular. What can be said is that this observation is entirely consistent with the idea that discharge constriction is responsible both for laser pulse termination and the fall in absorption late in the discharge.

The temporal evolution of the absorption at times greater than 500 ns (i.e. after the initial discharge pulse) is rather erratic: a decay in the magnitude of the absorption is seen in discharges containing all three gases, whereas a large increase is observed in those discharges not containing xenon. Power continues to be delivered to the discharge at these late times as a consequence of voltage and current 'ringing' in the circuit (refer to fig.4.11). The deterioration in shot-to-shot reproducibility is indicative of a

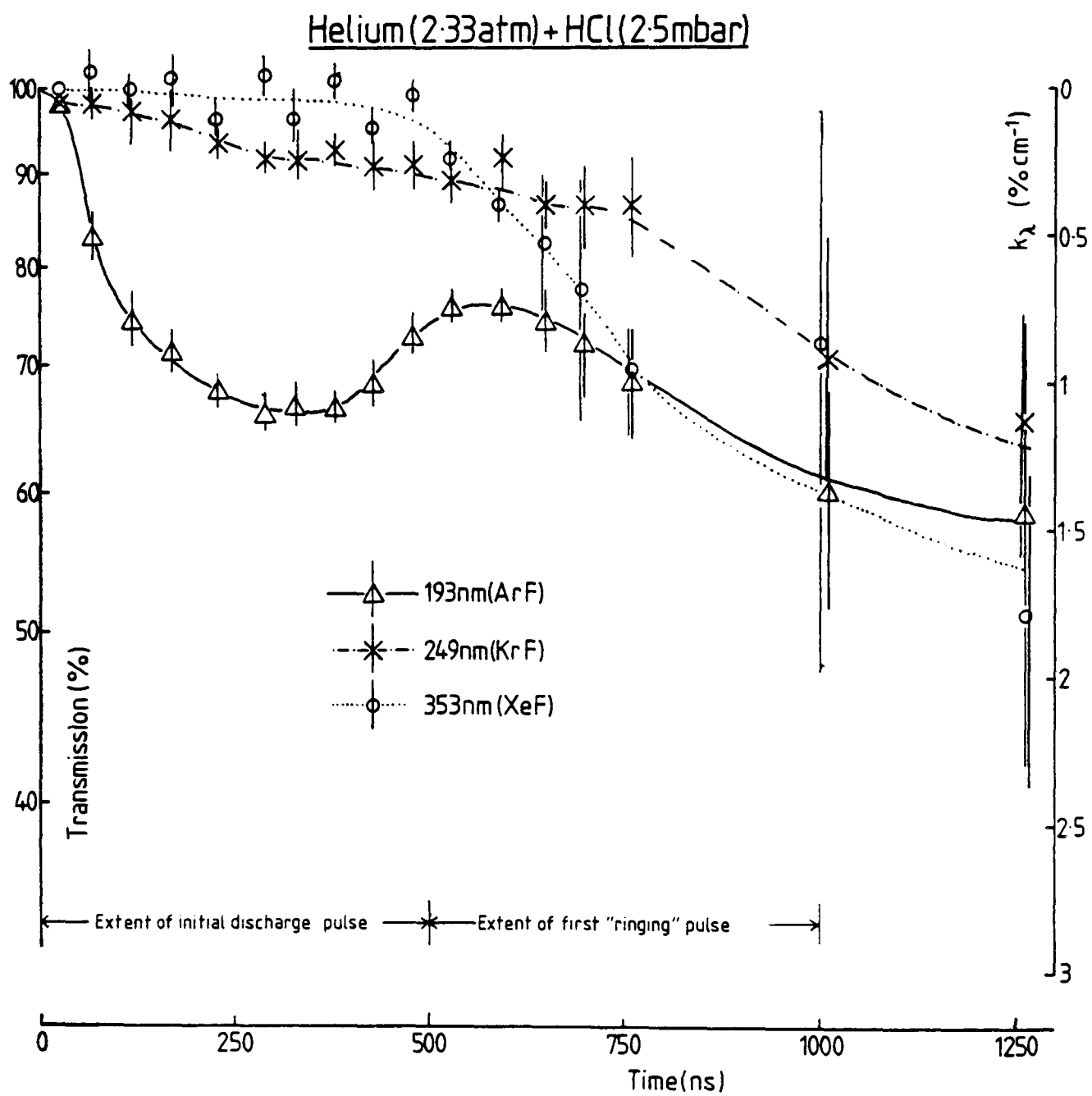


Figure 5.6: Transient absorptions

less stable discharge: the much larger variations present for discharges not containing xenon would imply that they are inherently less stable (though any explanation offered as to why this might be the case could only be conjecture at the moment). It may well be suggested that in the absence of xenon there is a significant increase in the Cl^- population and that this is responsible. However two factors militate against this explanation:

- (1) The absorption at 353 nm increases in a similar manner to that at shorter wavelengths: the energy of a 353 nm photon is below the photodetachment threshold of Cl^- .
- (2) Destruction of Cl^- through formation of RGH molecules should proceed at a similar rate whether or not xenon is present: the XeCl^* and NeCl^* formation rates are very nearly the same (Levin[1981]). No net increase in the Cl^- population would be expected on this basis.

The explanation may lie in the degree of constriction which occurs in the two cases (i.e. with or without xenon). If the xenon-free discharge constricts more strongly one could well imagine that the columnar discharges have a 'lensing' effect[†] on the probe radiation - an effect which is perhaps not so marked in the discharges containing xenon. The absorption would be more apparent than real in this regime since a part of the probe radiation that might otherwise have been transmitted through the exit slit would have been diverged by this series of 'lenses'.

At the moment, no definite answer to the question of what causes the transient absorption can be given: nor can one species be ruled out (or included) in the contribution to absorption in a laser gas mixture on the basis of observations in partial mixtures. For example, it was observed that the population of xenon metastable atoms increased by a factor of ~ 4 over that measured for a gas mixture containing no HCl when the (normal) 2.5 mbar of HCl was present (section 5.4). It is instructive, however, to attempt to identify

[†]Lensing due to inhomogeneous heating of the background buffer gas causing major perturbations in gas density and hence refractive index.

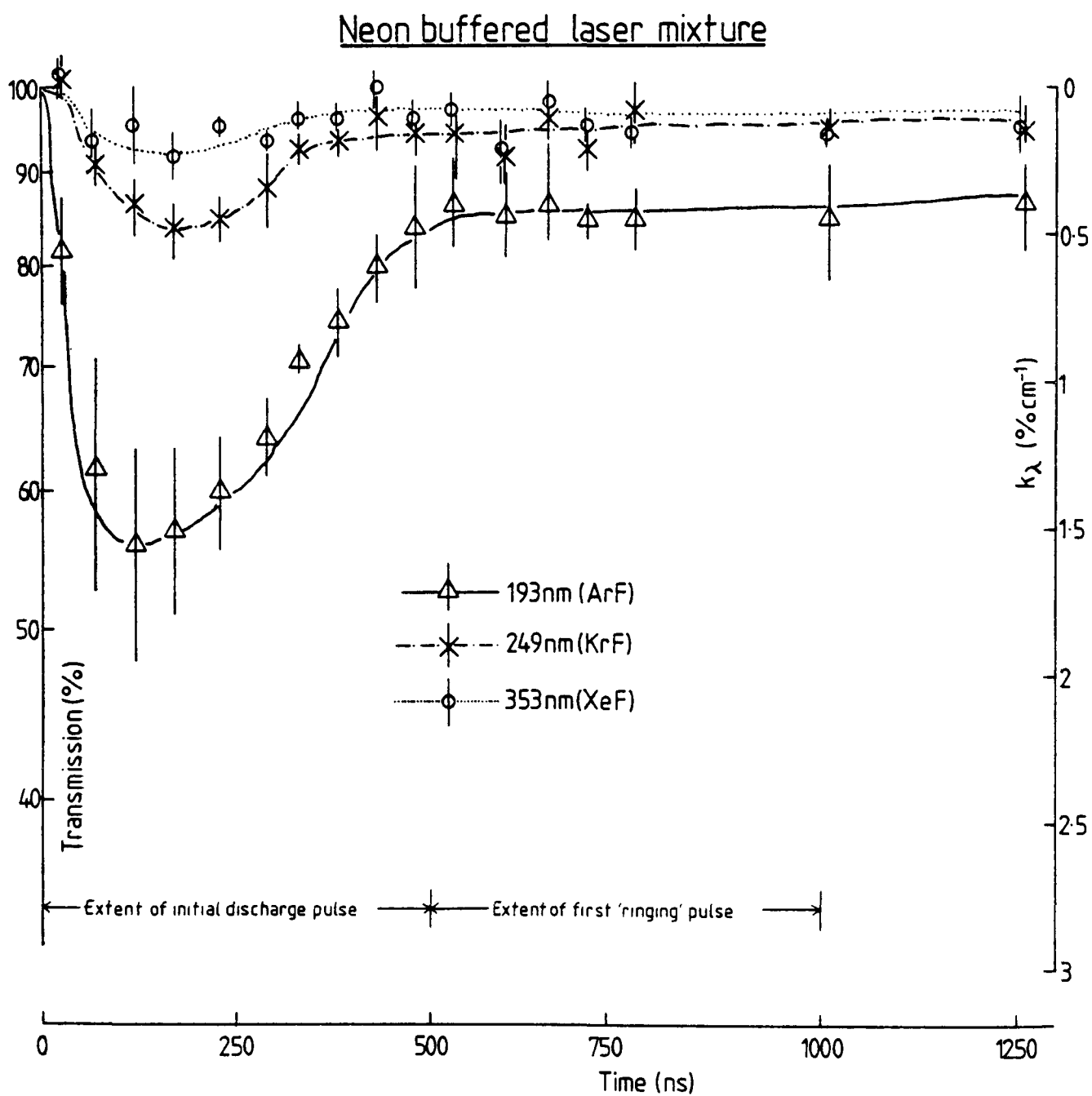


Figure 5.7 : Transient absorptions

Table 5.2
Photodissociation, photodetachment and photoionization cross sections
at selected UV wavelengths

Wavelength(nm)	353	337	308	249	193
Species	Cross section (cm ² x 10 ⁻¹⁸)				
CsCl(a,e)	0	0	0	8.5	12
CsCl(b,e)	0	0	0	12	17.5
Cl ⁻ *	0	5	11.5	14	19
Xe*	0*	0*	0.05*	0*	0.02*
Ne*	0*	0*	0*	0.01*	0.06*
Xe ₂ *	?	?	14*	~8*	?
Xe ₂ ⁺ (c)	48*	?	16*	0.003*	?
Xe ₂ ⁺ (d)	34*	?	18*	0.041*	?
Ne ₂ ⁺ (c)	0.96*	?	7.4*	23*	?
Ne ₂ ⁺ (d)	2.5*	?	8.6*	21*	?

? =value not known; * =theoretical; a =1900 K; b =800 K; c =300 K; d =600 K.

CsCl(a): Mandl[1977]; CsCl(b): Davidovits and Broadhead[1967];
Cl⁻: Mandl[1976]; Xe* (metastable): McCann and Flannery[1977];
Ne* (metastable): McDaniel[1978] vol.2 p.658;
Xe₂* (metastable): ibid. p.713;
Xe₂⁺ and Ne₂⁺: Wadt et al.[1977](theoretical values));
(e) CsCl is taken as an analogue to XeCl*;
CsCl binding energy = 4.532 eV and XeCl* binding energy = 4.537 eV;
Cl⁻ electron affinity = 3.613 eV; Xe* ionization potential = 3.84 eV.

the major candidates and rule out some possibilities.†

A short list of likely candidates as species responsible for the transient absorption would include Cl⁻ (electron photodetachment), xenon atoms in an excited state (photoionization), XeCl* (photodissociation), and heavy ions and neutrals such as Xe₂⁺ (photodissociation). Values for the absorption cross-sections of some of these species are given in table 5.2. Those given for the XeI excited state atoms, Ne₂⁺, and Xe₂⁺, are theoretical values: very little experimental work on the absorption of these species in the UV has been done but very broad-band (200-300 nm wide) emission and absorption spectra are predicted. At present, G.J.Hirst of the Clarendon Laboratory is undertaking a study to identify the species responsible for the broad band emission and absor-

† It would be meaningless to estimate populations at times later than ~200ns (made on the basis of eq.5.5 and using values of k from figures 5.5-5.8) since it was assumed that the absorption path length was 38 cm throughout. The framing camera study presented in chapter 4 showed that the discharge path length is time dependent after ~200ns.

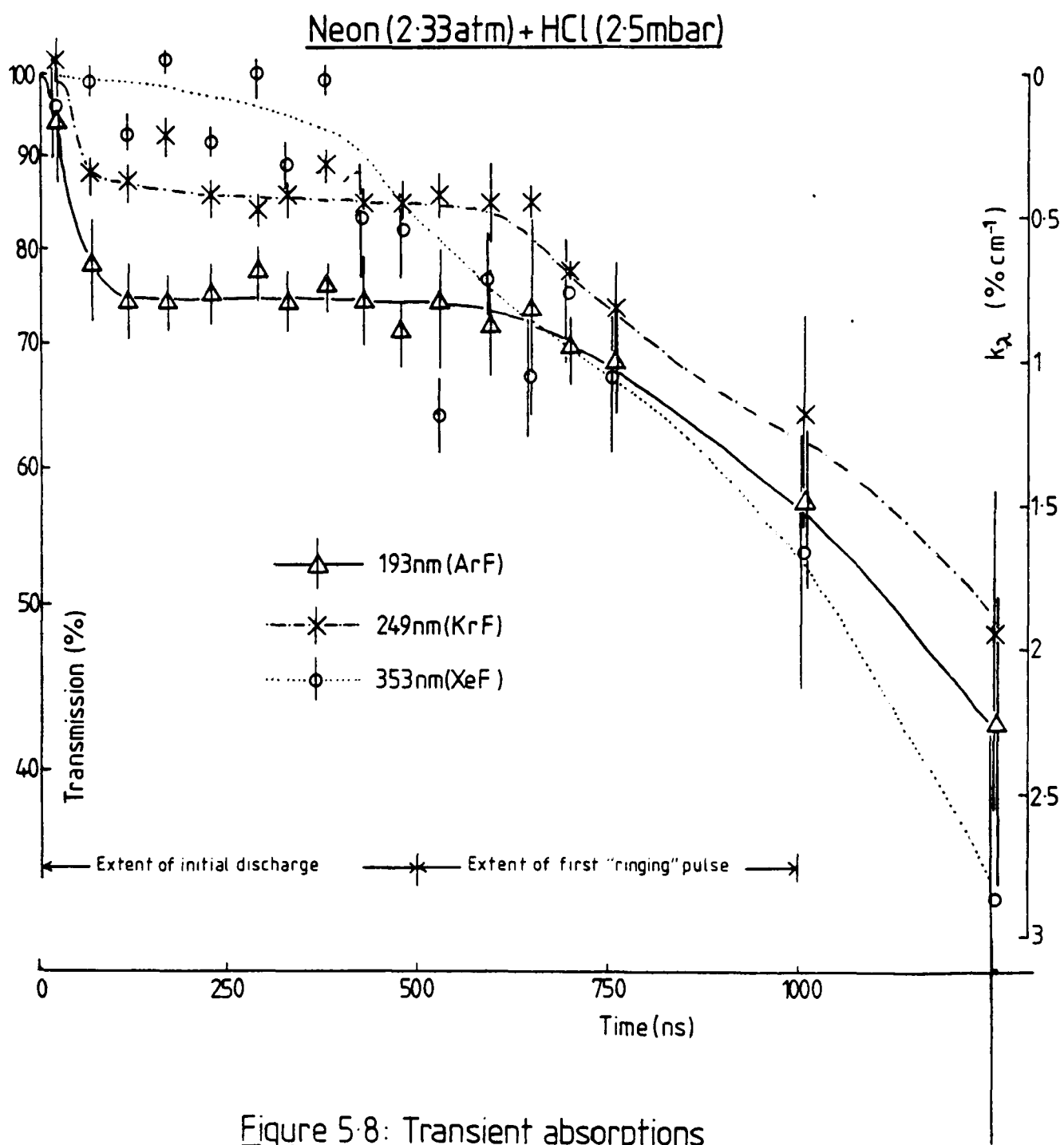


Figure 5.8: Transient absorptions

-ption which was observed both in this work and in that done by Kearsley[1980]. Photodissociation data for XeCl^* are not available; as was mentioned in chapter 1, however, each excited RGH molecule is structurally very similar to the alkali halide which follows it in the Periodic Table. It could be expected, therefore, that the cross sections for photodissociation of CsCl closely resemble those of XeCl^* .[†]

If the magnitude of the contribution of the species listed in table 5.2 to the total observed transient absorption is to be assessed, an estimate of the population of these species at the time of interest must be made.

(b) Estimation of various populations:

The quasi-steady state XeCl^* population can be estimated with good reliability because the results presented in figures 5.5 and 5.7 give a clear indication of the magnitude of the loss due to transient absorptions. There is absolutely no reason to suppose that the background level of absorption at 308 nm departs from the smooth variation in wavelength in the figures. The magnitude of the absorption could realistically be expected to lie between that at 337 nm and 249 nm. A value of k_v of $0.4\% \text{cm}^{-1}$ will be used in the following calculations.

In a quasi-steady state regime the population of XeCl^* molecules, $[\text{XeCl}^*]$, will be given by the rate of production of XeCl^* per unit volume, $d/dt(\text{XeCl}^*)_{\text{cr}}$, multiplied by the lifetime, τ , of XeCl^* against destruction by all processes:

$$[\text{XeCl}^*] = \frac{d}{dt} (\text{XeCl}^*)_{\text{cr}} \times \tau \quad (5.6).$$

$d/dt(\text{XeCl}^*)_{\text{cr}}$ may be obtained from the photon extraction efficiency per cm^3 from

[†]The XeCl^* binding energy is 4.537 eV (Tellinghuisen et al.[1976]) and that of CsCl is 4.532 eV (Tsu-Min and Riley[1980]), so although the peak XeCl^* absorption might be slightly further into the UV, the difference is likely to be negligible. The papers by Mulliken[1970], Wadt[1981], Michels et al.[1979], and Lee et al.[1979] will provide the reader with an introduction to absorption by other heavy rare-gas ions and neutrals for which no UV absorption data is available.

the active (discharge) volume.[†] Assuming that a 15 cm^3 discharge volume[¶] produces the 5 mJ laser pulse of 250 ns FWHM duration (which was regularly observed), a value of 1.3 kW/cm^3 is obtained for the output power extracted per cm^3 of discharge volume through the 4% transmitting output mirror. In order to obtain the total number of photons produced per cm^3 , it is necessary to find what fraction of the total loss of photons this 'useful' output represents. The percentage loss due to transient absorption over a double transit of the cavity ($2 \times 38 = 76 \text{ cm}$ discharge path) can be estimated by assuming that losses at the Brewster angle windows and at the mirrors are only a small perturbation. The fractional transmission of 308 nm photons is:

$$(1 - e^{-k_v \ell}) = 0.74$$

In other words 26% of the total photons which are obtained as stimulated emission will be lost to transient absorption in one double transit of the cavity. The total loss will be 30% in a double transit (adding the 4% output), and thus the 'useful' output represents $4/30 = 0.13$ of the total power generated as photons, i.e. Total power extracted $= 1.3/0.13 = 10 \text{ kW/cm}^3$. In terms of 308 nm photons, the total power extraction $= 1.6 \times 10^{22} \text{ photons cm}^{-3} \text{ s}^{-1}$. The production rate of $\text{XeCl}^*/\text{cm}^3$ is obtained by taking into account loss of XeCl^* due to processes such as quenching by electrons. Spontaneous emission of 308 nm photons will represent a fixed (very small) proportion of these losses. The ratio of the peak intensity sidelight spontaneous emission in the absence of lasing to that in the presence of lasing will therefore give an idea of the magnitude of the total (non-photon emitting) loss processes. This ratio is 1:2 (from plate 4.3):

[†] The subscript 'cr' will be used to distinguish between a rate of creation and a net production rate - which in the quasi-steady state would be zero.

[¶] This figure is arrived at by multiplying the total discharge length (38 cm) by the discharge gap (2.2 cm) and the diameter of an individual ballasted screw (0.18 cm: the discharge thickness). As can be seen from plate 4.9, the total length of discharge may be smaller than 38 cm since the discharge does not necessarily fill the gaps between the individual ballasted screws. This must be balanced against the observation that the 'thickness' of the discharge is, generally, not constant across the discharge gap: it tends to be thicker towards the centre. A 15 cm^3 volume therefore represents a best estimate which could be up to a factor of two in error (see figure 5.28 for definition of 'thickness').

$$d/dt(\text{XeCl}^*)_{\text{cr}} = 2 \times 1.6 \times 10^{22} = 3.2 \times 10^{22} \text{ cm}^{-3} \text{ s}^{-1}.$$

The lifetime of XeCl^* against destruction is the inverse of the rate of destruction of XeCl^* by all processes: this will simply be the rate coefficient for destruction of XeCl^* by stimulated emission multiplied by the ratio of spontaneous emission in the absence to that in the presence of lasing. The rate of stimulated emission is given by the product of the intra-cavity energy flux, ϕ_{ic} , and the cross section for stimulated emission, σ_{stim} :

$$\frac{d}{dt} (\text{Stimulated Emission}) = \phi_{\text{ic}} \times \sigma_{\text{stim}} \quad (5.7).$$

The intra-cavity flux, ϕ_{ic} , is proportional to the output flux, ϕ_{output} , and the reflectivity to transmission ratio of the output coupler, $T = 24$, multiplied by 2 (to account for counter-propagating photons reflected by the 100% reflector). The value of ϕ_{output} is given by the peak laser power, P_{peak} , divided by the cross-sectional area of the laser beam, $A_{\text{beam}} = 0.7 \text{ cm}^2$, so,

$$\phi_{\text{ic}} = \frac{P_{\text{peak}}}{A_{\text{beam}}} \times 2T = 2.1 \times 10^{24} \text{ cm}^{-2} \text{ s}^{-1} \quad (5.8).$$

Levin[1981] gives σ_{stim} as $2.5 \times 10^{-16} \text{ cm}^{-2}$. Combining this and the result of eq.5.8 and substituting into eq.5.7, a stimulated emission rate of $5.4 \times 10^8 \text{ s}^{-1}$ is obtained. Multiplying this rate by the ratio of stimulated emission in the absence to that in the presence of lasing and reveals that

$$\tau = 0.9 \text{ ns}$$

Substitution of τ and $d/dt(\text{XeCl}^*)_{\text{cr}}$ into eq.5.6 produces an estimated XeCl^* population:

$$[\text{XeCl}^*] = 2.8 \times 10^{13} \text{ cm}^{-3}$$

An upper limit for the population of Cl^- ions can be estimated[†] using the following relation:

$$\frac{d}{dt} (\text{XeCl}^*)_{\text{cr}} = k[\text{Cl}^-][\text{Xe}^+] \quad (5.9),$$

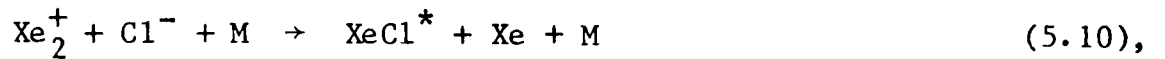
where k is the saturated rate coefficient[‡] for the reaction and has a value of $2 \times 10^6 \text{ cm}^3 \text{ s}^{-1}$ (Levin[1981]). The xenon ion density, $[\text{Xe}^+]$, will not be less than the electron density (section 4.6.3(a)) which was calculated to be $\sim 6.4 \times 10^{14} \text{ cm}^{-3}$

[†]Assuming XeCl^* formation is dominated by the ion channel (eq.1.1)

[‡]The buffer gas density is sufficiently high that the rate is independent of it.

(section 4.6.3(c)). Substitution of these values into eq.5.9 yields an upper limit of $2.5 \times 10^{13} \text{ cm}^{-3}$ on the Cl^- density.

An upper limit on the Xe_2^+ density could be arrived at if it was assumed that the lifetime against destruction was similar to that of Xe^+ . This would be reasonable if the dominant Xe_2^+ loss process is via XeCl^* formation since the rate of formation of XeCl^* via the dimer-ion channel,



where M is a third body, can be expected (Levin[1981]) to be just as fast as the simpler reaction (eq.1.1). The formation of Xe_2^+ via the (dominant) reaction



is given by the rate equation

$$\frac{d}{dt} (\text{Xe}_2^+)_{\text{cr}} = k[\text{Xe}^+][\text{Xe}][\text{Ne}] \quad (5.12).$$

In this case $k = 1.5 \times 10^{-31} \text{ cm}^6 \text{ s}^{-1}$. Substituting the [Ne] and [Xe] from table 4.3 and putting $[\text{Xe}^+] = n_e$ in eq.5.12, a rate of formation of Xe_2^+ of $4 \times 10^{21} \text{ cm}^{-3} \text{ s}^{-1}$ is obtained. If the above assumption is correct, the lifetime of Xe_2^+ (and Xe^+) against destruction, τ_D , can be found by dividing the Xe^+ density by the rate of formation of XeCl^* ,

$$\tau_D = 6.4 \times 10^{14} / 3.2 \times 10^{22} = 20 \text{ ns, thus,}$$

$$[\text{Xe}_2^+] = 4 \times 10^{21} \times 20 \times 10^{-9} = 8 \times 10^{13} \text{ cm}^{-3}$$

Neon dimer ions would not be expected to be present in any significant quantity since the precursor (Ne^+) density will be low as a result of its high ionization potential (section 4.6.2(a)). The results presented in section 5.4 indicate a peak Xe^* density in the $6s[3/2]$ levels of $\sim 9 \times 10^{13} \text{ cm}^{-3}$. Xenon dimer excited state densities cannot be readily estimated.

It is very important to regard all of these estimates only as order of magnitude guesses in the light of the assumptions used in arriving at them. Furthermore, some of the data, such as the electron density, used in the calculations are subject to large uncertainties themselves. A summary of the estimates made here is presented in table 5.3. Values for the same quantities for the high pump density, short pulse duration XeCl^* laser studied by Kearsley[1980] are included for comparison.

Table 5.3

Summary of estimates of various populations and rates, including comparison with those estimated by Kearsley[1980]

	This study	Kearsley
Electron density (n_e) (cm^{-3}) (= Xe^+ density)	6.4×10^{14}	3.5×10^{15}
Intracavity flux ($\text{cm}^{-2} \text{s}^{-1}$)	2.1×10^{24}	3.1×10^{24}
Transient loss absorption coefficient ($\% \text{cm}^{-1}$)	0.4	?*
Transient loss in one cavity round trip (%)	26	?*
Ratio of peak spontaneous emission without lasing to that with lasing	2:1	2:1
Rate of stimulated emission (s^{-1})	5.4×10^8	6.3×10^8
Laser photon emission rate ($\text{cm}^{-3} \text{s}^{-1}$)	1.6×10^{22}	2×10^{22}
XeCl^* lifetime (ns)	0.9	0.8
XeCl^* production rate ($\text{cm}^{-3} \text{s}^{-1}$)	3.2×10^{22}	4×10^{22}
XeCl^* density (cm^{-3})	2.8×10^{13}	3.2×10^{13}
Cl^- lifetime (ns)	0.8	0.17
Cl^- density (cm^{-3})	2.5×10^{13}	6.5×10^{12}
Xe_2^+ and Xe^+ lifetime (ns)	20	?
Xe_2^+ density (cm^{-3})	8×10^{13}	?
Xe_2^* ($6s[3/2]_2$) density (cm^{-3})	5.6×10^{13}	(a) 5.6×10^{13}
Xe_2^* ($6s[3/2]_1$) density (cm^{-3})	3.5×10^{13}	(a) 9.6×10^{13}
Xe^* (total) density (cm^{-3})	4×10^{14}	4×10^{14}

* (a) A_{21} values adjusted to those used here.
Kearsley did not estimate the effects of transient loss

(c) Comments regarding the source of the transient absorption:

The estimates for the densities of the various species presented in table 5.3 are all far too small to account for the observed transient absorptions. Xe_2^+ could possibly account for the observed absorption at 353 nm in neon and helium buffered laser mixtures, but its trend towards a diminishing cross section at shorter wavelengths makes it an unlikely candidate for any significant contribution at, say, 193 nm. Even if the populations of Cl^- and XeCl^* have been grossly underestimated, the comparatively slow functional dependence of their cross sections with wavelength rule them out as being responsible for the large increase in total absorption between 193 and 249 nm in laser gas mixtures.

There are, of course, many other species which could be present in the discharge. It is likely that no single species is responsible, but rather that the total absorption is made up of small contributions from many species. At the moment, the only conclusion we can reach is this: it is not known what

Experimental Details:

Acquisition of Time Resolved Discharge Emission

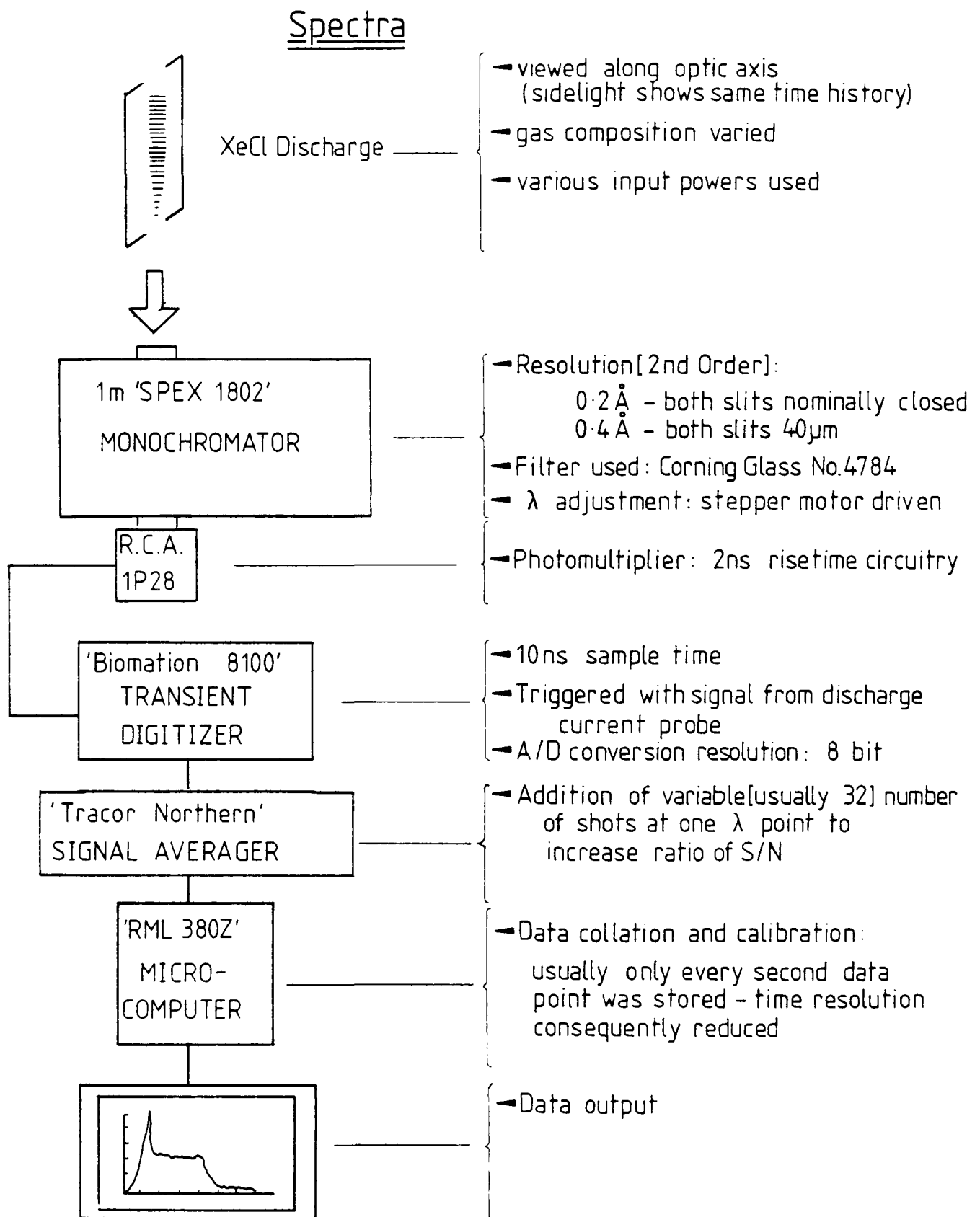


FIGURE 5.9

causes the absorption - the problem requires further investigation. However, the absorption is undoubtedly real - it is not an experimental artifact.

5.3 TIME-RESOLVED MEASUREMENTS OF EMISSION AND ABSORPTION IN THE VISIBLE- EXPERIMENTAL DETAILS AND RESULTS:

5.3.1 Experimental details - emission measurements:

Figure 5.9 sets out in block diagram form the apparatus which was used in the time-resolved examination of emission in the LPD laser. In all but a few cases - where a comparison of absorption effects was to be made - the emission from the LPD laser was viewed in 'endlight' (i.e. along the optic axis). This led to a slightly undesirable effect when emission at 308 nm was to be monitored,[†] but it was essential for observations at other wavelengths because of their comparatively low intensity of emission when viewed in 'sidelight'.

In all these experiments the mirrors forming the laser cavity were removed and the Brewster angle windows were replaced by 3° off-axis silica windows. Unlike the previous experiment, no slits were placed around the the discharge. The unimpeded emission from the discharge was passed through a porthole in the screened room and imaged by a silica lens on the entrance slit of a 1 m Cerny-Turner monochromator (Spex model 1802). The horizontal stripe image of the discharge formed on the entrance slit in such a way that (essentially) only the light emanating from the discharge region midway between the two electrodes of the LPD laser was sampled - discussion of the consequences of this action is left until section 5.4. The monochromator was invariably used in 2nd order to maximise the spectral resolution. The instrument resolution was checked by observation of the 313.3 nm doublet and 546.1 nm spectral lines from a Mercury discharge lamp. The monochromator could resolve 0.02 nm with both the entrance and the exit slit at minimum opening and 0.04 nm with both slits open by a further ~40 μm. In order to prevent higher order UV emission appearing when emission in the visible was being monitored an appropriate filter was placed

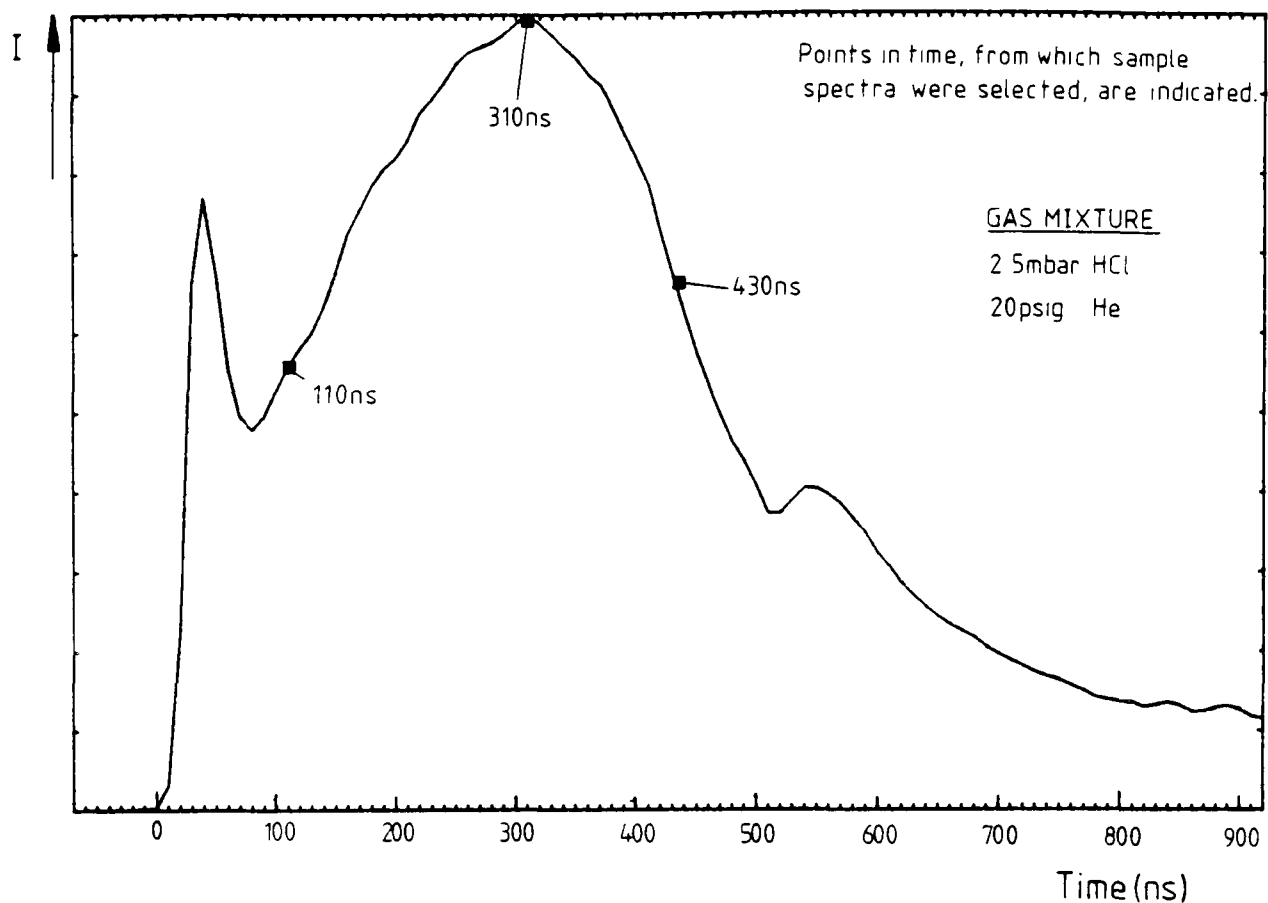
[†] It was mentioned in chapter 4 that single pass gain along the optic axis had some effect on the temporal evolution of the intensity of 308 nm emission: no significant problems in interpretation arise from this.

over the monochromator entrance slit (Corning glass no.4784). The signal was detected at the exit slit by a photomultiplier (RCA type 1P28) with specially modified circuitry for a fast signal risetime ($\sim 2\text{ns}$). Approximately 600 V was applied to the dynode array for the detection of 308 nm radiation, whereas up to 2000 V was used at other wavelengths. A stepper-motor drive was attached to the monochromator to allow the wavelength setting to be increased by 0.01 nm steps reproducibly when a spectral feature was to be scanned.

The signals from the photomultiplier were fed directly into a transient digitizer (Biomation model 8100). The minimum temporal resolution of this device was $\sim 50\text{ ns}$ (i.e. a delta function would appear as five points each separated by 10ns). Initially, problems were encountered in triggering the digitizer with respect to the discharge. A long ($\sim 1\text{ }\mu\text{s}$) coaxial cable delay was inserted between the photomultiplier and digitizer to overcome this problem, and it effectively decreased the resolution to $\sim 100\text{ ns}$. Fortunately this situation did not persist: the 'pre-trigger' facility on the digitiser was finally made to operate. Some of the results presented in this section were subject to this reduced temporal resolution.

In particular, all the data used to obtain the three-dimensional plots in figures 5.11, 5.12, and 5.13, is affected in this way - as is that in figure 5.19. The only notable effect is that the sharp peak in emission at discharge initiation from the xenon line at 467.123nm is curtailed: the overall trend in intensity variation is not distorted. The same cable also introduced some extra apparent broadening of the spectral FWHM of the hydrogen Balmer beta line. This emission feature was to be used to provide a check on the value of electron density obtained via eq.4.21. All other data has a resolution of at least 50 ns. Some voltage and current curves (those not presented in plates) were collected with the digitizer also. This was done merely by connecting the Rogowski coil or voltage divider output to the digitizer and sending the digitized data to a signal averager as was done for the photomultiplier signal. The breadth (in time) of the voltage spikes at discharge initiation (fig.4.13(b)) gives an indication of the best temporal resolution achieved.

(a) Time evolution of H_{β} at line centre - 4 cable PFN



(b) H_{β} spectrum at various points in time during discharge: 4 cable PFN

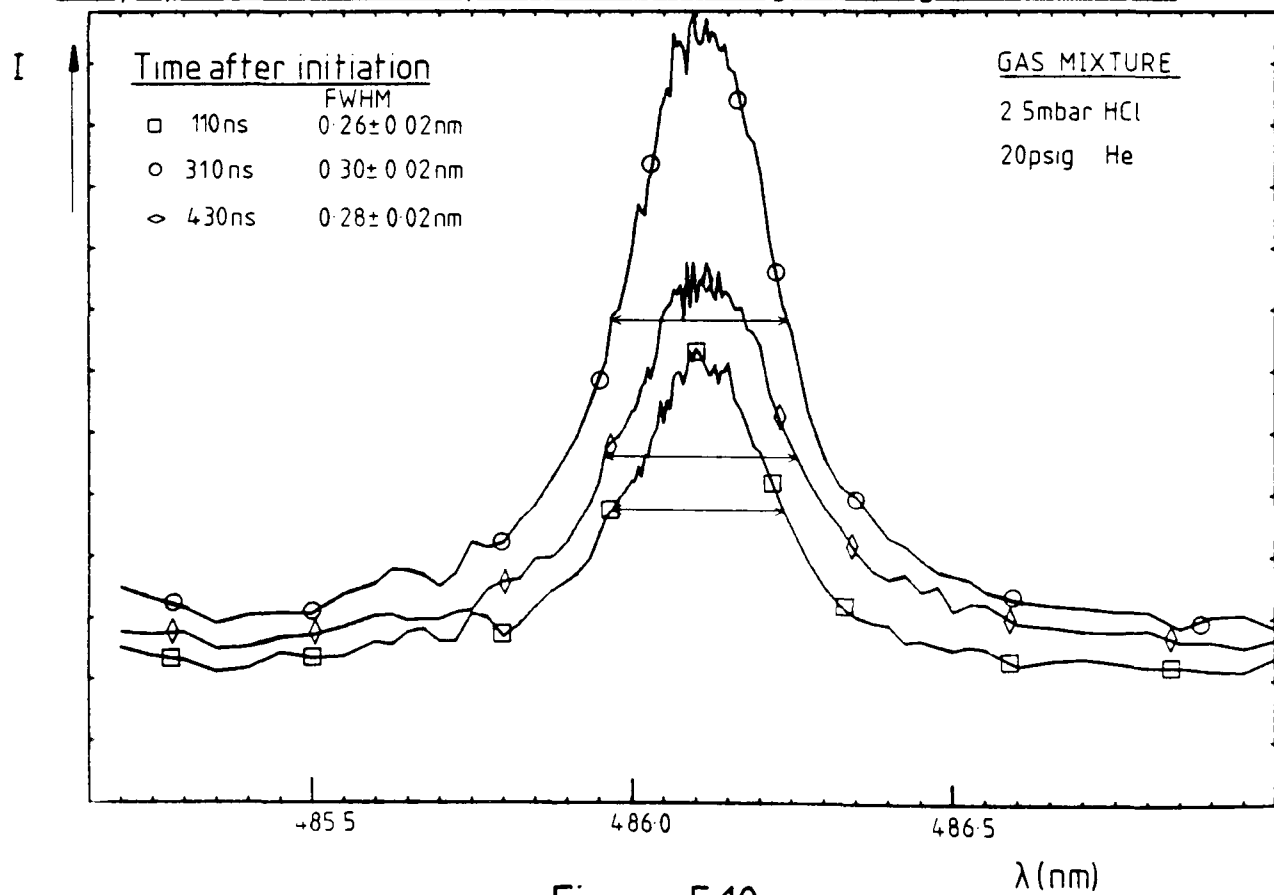


Figure 5.10

Data from the digitizer was sent to a signal averager (Tracor Northern NS-570A) before subsequent transferral to a microcomputer (Research Machines 380Z) and permanent storage on floppy disks. This was done for two reasons: firstly, the signal-to-noise quality of the signal could be monitored as the data for one particular wavelength point was collected (usually the signal from 32 discharge events at one wavelength gave a good quality signal); secondly, the LPD laser occasionally misfired and there was no method of detecting if the data was the result of a misfire unless it was monitored directly. A full spectral profile of an emission feature consisted of some 1000 data points. Graphics software developed by the author and based on RML Algol was used to produce the three-dimensional plots of intensity as a function of wavelength and time which are presented here. It was considered sufficient to monitor only the temporal evolution of a feature at line centre once one full spectral profile had been obtained: the two-dimensional plots of XeI, XeII, and XeCl* emission vs. time are for line centre (308 nm for XeCl*). The curves of power input into the discharge (figs.4.12(b),4.14(b) and 4.17(b)) and that of the actual voltage drop (fig.4.13(b)) across the discharge (as opposed to across the discharge and ballast resistance) vs. time were calculated by subtracting the contribution due to the ballast ($I \times R_{\text{ballast}}$) at each time point.

5.3.2 Results - emission measurements:

(a) Observation of hydrogen balmer beta and XeII emission:

The method used by Kearsley[1980] to determine the electron density of his XeCl* discharge was to measure the spectral FWHM of the hydrogen Balmer beta (486.1 nm) line. This method is very well known and the reader is referred to Griem[1964] for a detailed discussion of the theory. The equation governing the relationship between the spectral FWHM of Balmer beta emission and electron density, n_e is:

$$n_e = C(n_e, T) \Delta\lambda^{3/2} \quad (5.13),$$

where $\Delta\lambda$ is the spectral FWHM of the line in Å, and $C(n_e, T)$ is only a slowly varying function of n_e . Kearsley was unable, for reasons discussed in section 4.2.1, to make a measurement of the total current flow in his discharge and

Time Resolved Emission Spectrum from a XeCl Laser Gas Mixture Discharge

(Helium buffer)

Xe II - 486.254 nm ($7s^4P_{5/2} \rightarrow 6p^4P_{5/2}^o$)

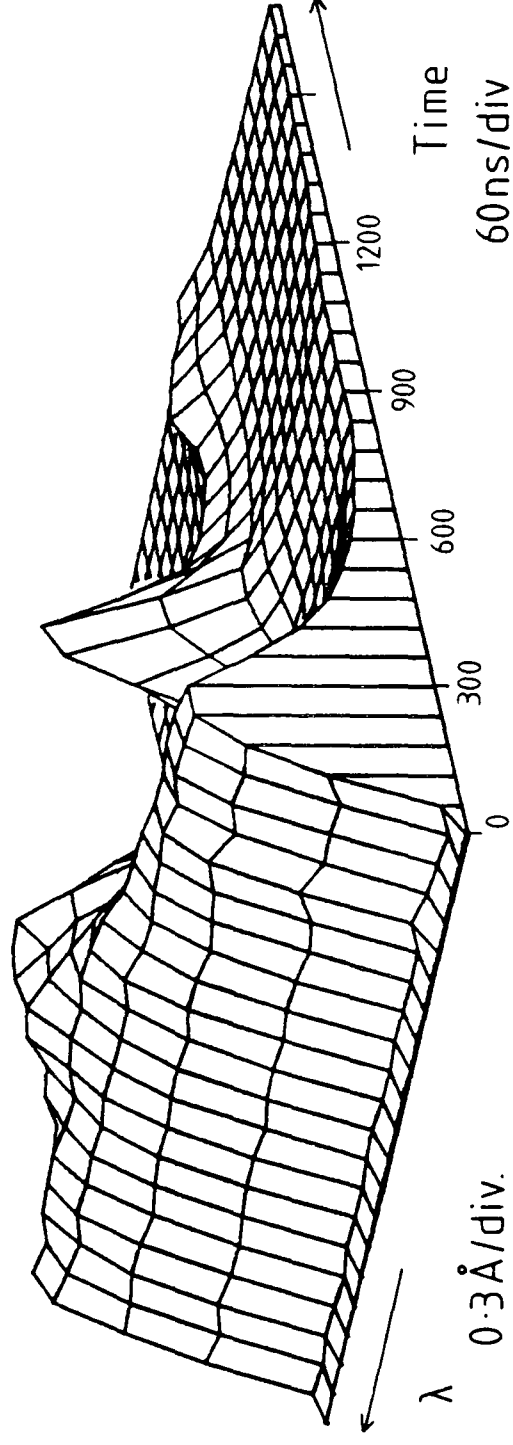


Figure 5.11

arrive at the electron density through equation 4.21.[†] His measurement of Balmer beta could only be performed in a discharge not containing xenon because of the presence of broad background emission. It was hoped that with the use of the signal averager it would be possible to extract the Balmer beta emission from this background in a full laser mixture. A check was made on Balmer-beta for a discharge containing 2.5 mbar of HCl and 2.33 atm of He (as Kearsley had done): the data is presented in figure 5.10 (helium buffer). The spectral profile of the line - as sampled at three times during the discharge pulse - is presented in the figure, as is the temporal evolution of intensity at line centre. As was noted in the preceeding sub-section, the poor temporal resolution of the data collection apparatus at this stage resulted in very little difference being detected in the spectral FWHM at different times during the discharge. A mean spectral FWHM value of $0.28 \pm 0.02 \text{ nm}^\ddagger$ is the only useful information that can be extracted from this data. Insertion of this value into eq.5.6, using $C(n_e, T) = 3.6 \times 10^{14}$ (Griem[1964]), yields an electron density of $1.8 \times 10^{15} \text{ cm}^{-3}$ which is in reasonable agreement with that obtained for a full laser mixture via eq.4.21. It was found that Balmer-beta could not be seen in a full laser mixture. This was due in part to the presence of broad-band background emission. However, it was also found that another very intense feature was present. This emission feature, which was present in all discharge mixtures containing xenon, was some 10 times more intense the Balmer beta emission. Balmer beta emission was detectable only in those gas mixtures which did ^{NOT} contain xenon.

The feature was identified as the 486.254 nm xenon ion line ($7s^4p_{5/2} -$

[†] Hydrogen balmer beta is used because the broadening is due to the linear ($\propto E$) - as opposed to the quadratic ($\propto E^2$) - Stark effect. The wavefunctions of hydrogen are known well theoretically and a measurement of electron density by this method can be correct to within 30%.

[‡] This error only represents the uncertainty in measuring the FWHM from figure 5.10. It does not include the broadening due to the poor temporal response of the apparatus at that stage. It was not considered worthwhile to repeat the measurement since it could not be performed in a full laser gas mixture. The estimate of electron density presented in chapter 4 - and obtained via eq.4.23 - was considered to be a far more reliable indicator.

Time Resolved Xenon Ion & Neutral Emission Spectra
from a XeCl Laser Gas Mixture
 (Helium buffer)

Xe I - 467.123nm ($7p[5/2]_3 \rightarrow 6s[3/2]_2$)

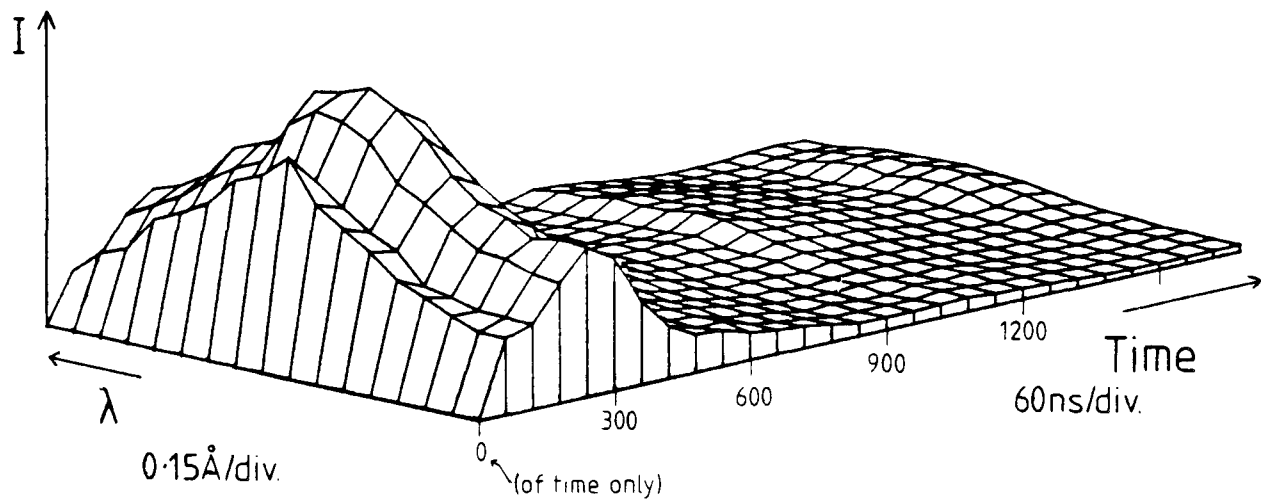


Figure 5.13

Xe II - 484.433nm ($6p^4D_{7/2}^{\circ} \rightarrow 6s^4P_{5/2}$)

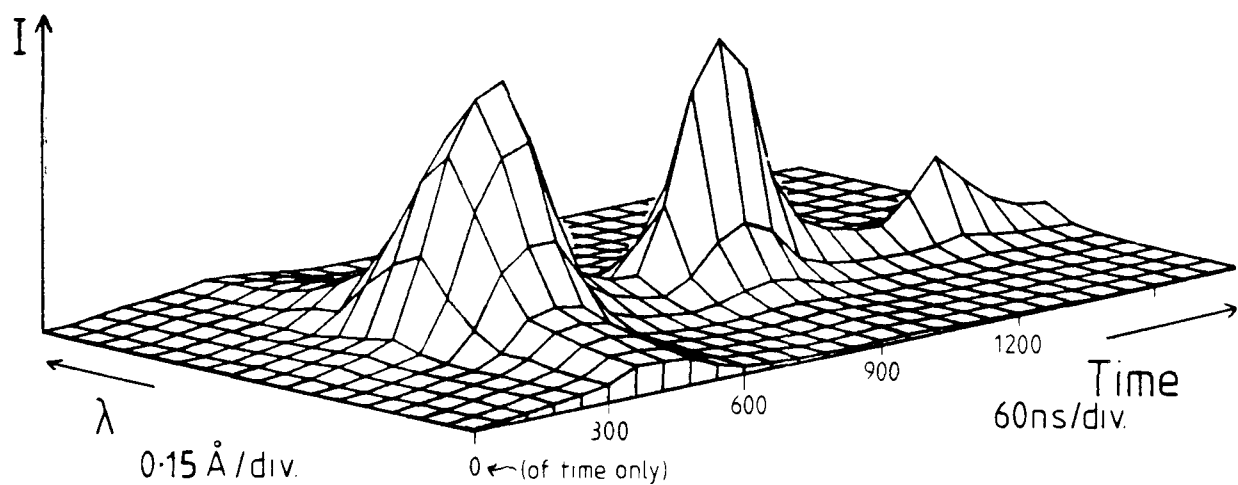


Figure 5.12

$6p^4p_{5/2}^o$). Its intensity as a function of wavelength (around line centre) and time is shown in figure 5.11 (helium buffer). A broad-band background emission obscures its 'wings' during the major part of the initial discharge pulse, however it shows a curious 'ringing' in time with a period corresponding to the double transit time of the cable-PFN.

In order to check that this transition was not a special case, emission from the nearby XeII transition at 484.433nm ($6p^4D_{7/2}^o - 6s^4p_{5/2}$) was observed. The intensity of this line was greater than any feature - other than XeCl* emission - in the range 300-600 nm: it is shown in figure 5.12, as a function of wavelength and time in a helium buffered gas mixture (in neon buffered mixtures the emission in the initial pulse was similar but there was comparatively little emission in the 'ringing' pulses). The broad-band background in the initial discharge pulse is still present but is totally dwarfed by the intensity of emission at 484.433 nm. Its temporal evolution is very similar to that of the 486.254 nm transition. In this case, however, it is clear that the ion emission is most intense in the later stages of the initial discharge pulse and in the later stages of the subsequent 'ringing' pulses (see fig.4.11).

A very significant and obvious feature is that emission from xenon neutrals was totally complementary to the ion emission: most intense in the initial stages of the discharge pulse and falling as the XeII emission rose. XeI emission in the 'ringing pulses' was weak in both helium and neon buffered mixtures and limited to the point in time at which the voltage and current changed polarity.

The XeI transition at 467.123nm ($7p[5/2]_3 - 6s[3/2]_2$) is a good representative example and it is shown in figure 5.13 (helium buffer). This line was also observed in sidelight and exhibited the same temporal behaviour. This rather unexpected occurrence of strong emission from a high-lying[†] ion energy level indicated that a dramatic change was taking place in the discharge at late times. Consequently, the behaviour of the emissions from the XeI(467nm)

[†] The lower level of the 484.433 nm line is some 12 eV above the ion ground state: see figure 5.14.

Xe II Term Diagram (G.S. to $7s^4P$ level)

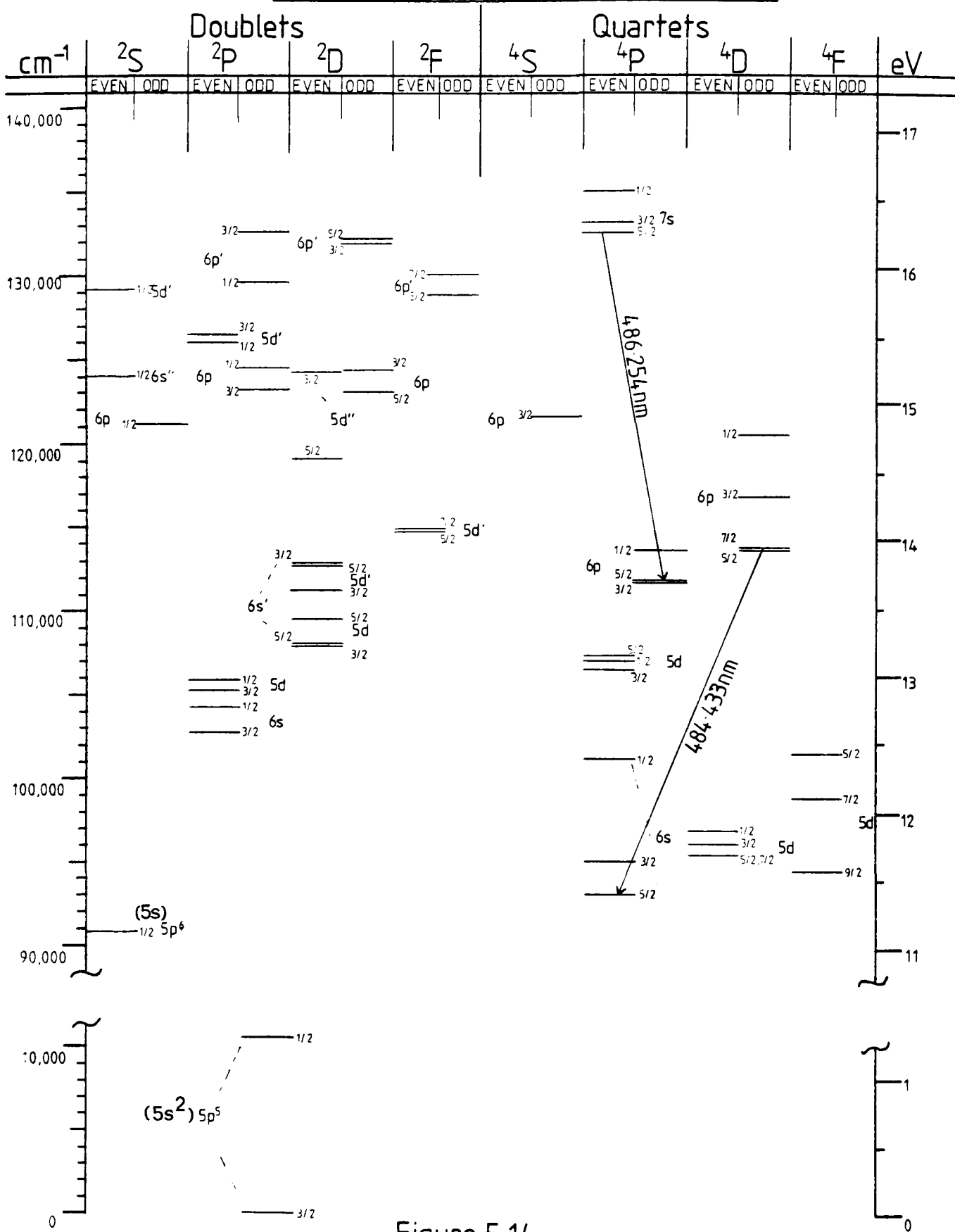


Figure 5.14

and XeII(484 nm) transitions (as representative examples) were studied to identify their functional dependence on different discharge parameters.

(b) The parametric dependence of XeI-467.123 nm and XeII-484.433 nm emission:

Results of studies of the dependence of emission from the XeI-467.123 nm and the XeII-484.433 nm transitions on different discharge parameters are presented in this section. Except for (1) below, only the temporal evolution at line centre was studied. The investigations undertaken included:

- (1) Emission around 484 nm as a function of time and wavelength in a discharge which, except for the absence of HCl, was run under the same conditions as for a 'normal' laser discharge: figure 5.15 (helium buffer)
- (2) Variation of the HCl partial pressure: figure 5.16 (cf. figures 4.12 and 4.13).
- (3) Variation of the neon buffer gas pressure: figure 5.17 (cf. figure 4.14).
- (4) Variation of the total input power by changing the number of cables in the PFN: figure 5.18 (cf. figure 4.17).
- (5) Temporal evolution of XeI, XeII, XeCl^* , and the discharge voltage and current, for a discharge driven by a 1-cable, 1 μs pulse duration, PFN: figure 5.19.

Unless otherwise stated all other parameters were the same as set out in table 4.2 for a neon buffered laser gas mixture. Figures indicated for comparison show XeCl^* emission under the same conditions.

Figure 5.15 shows that in the absence of HCl, the ion emission is barely detectable: emission in the region is dominated by the XeI-484.329 nm ($7p[3/2]_2 - 6s[3/2]_1$) transition. The XeI transitions which have been referred to are shown on the XeI term diagram (figure 5.20 : back folio of thesis).

A more comprehensive view of the effect of HCl on XeI and XeII emission is provided by figure 5.16. XeII emission initiates earlier as the HCl partial pressure increased - XeI emission, on the other hand, begins to terminate at precisely the time that XeII emission is increasing. The behaviour of XeCl^* emission (figure 4.12(a)) mirrors the XeI emission. The relative intensities of

Time Resolved Emission Spectrum around 484.4 nm from a Xe (25mbar) plus He(20psig) discharge

XeI (foreground) – 484.329 nm [7p[3/2]₂ → 6s[3/2]₁]
 XeII (in wings) – 484.433nm [6p⁴D_{7/2}^o → 6s⁴P_{5/2}]

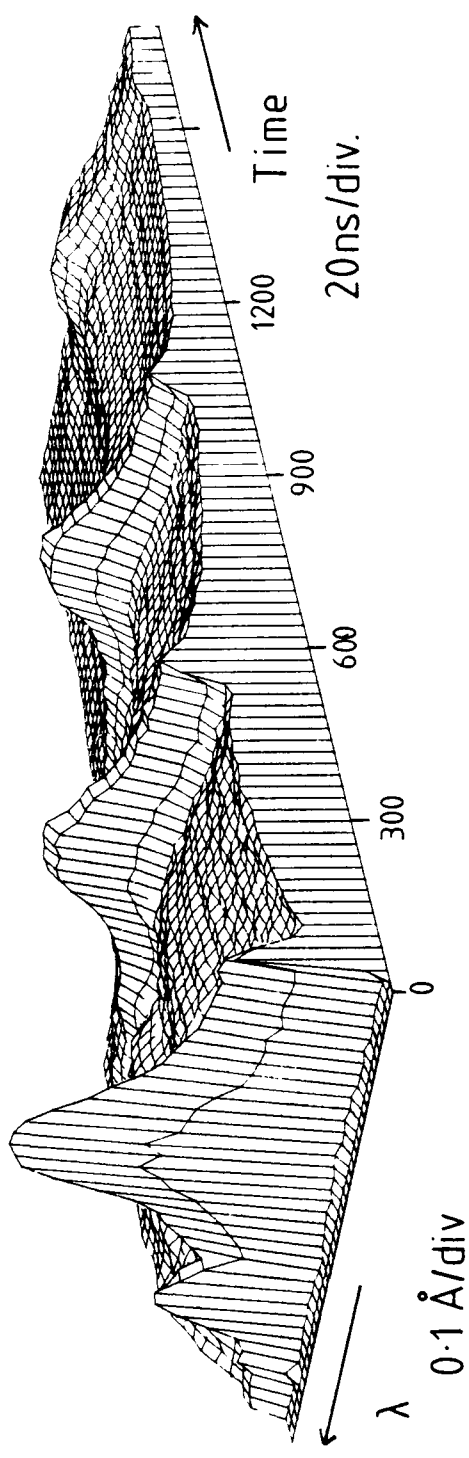
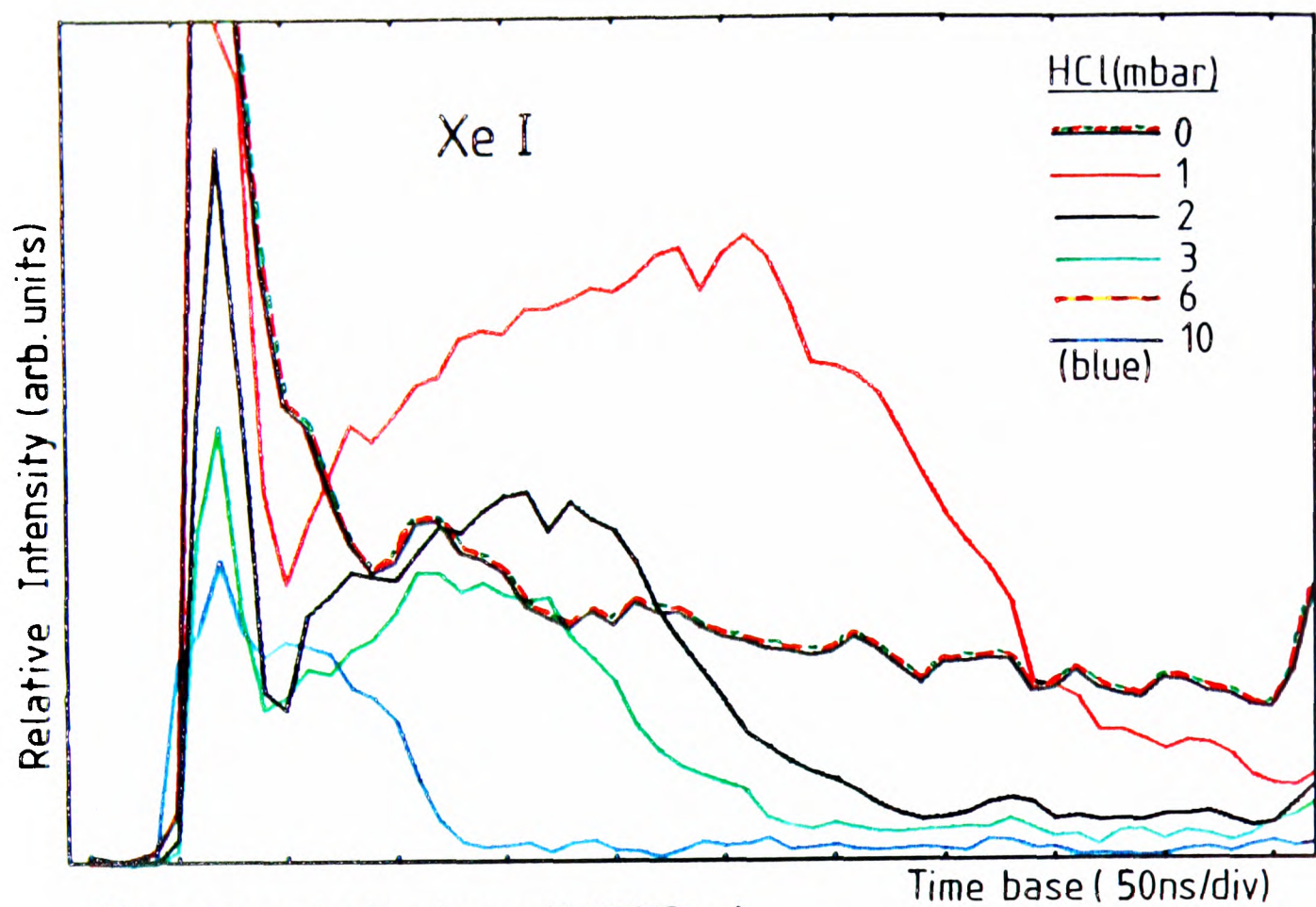
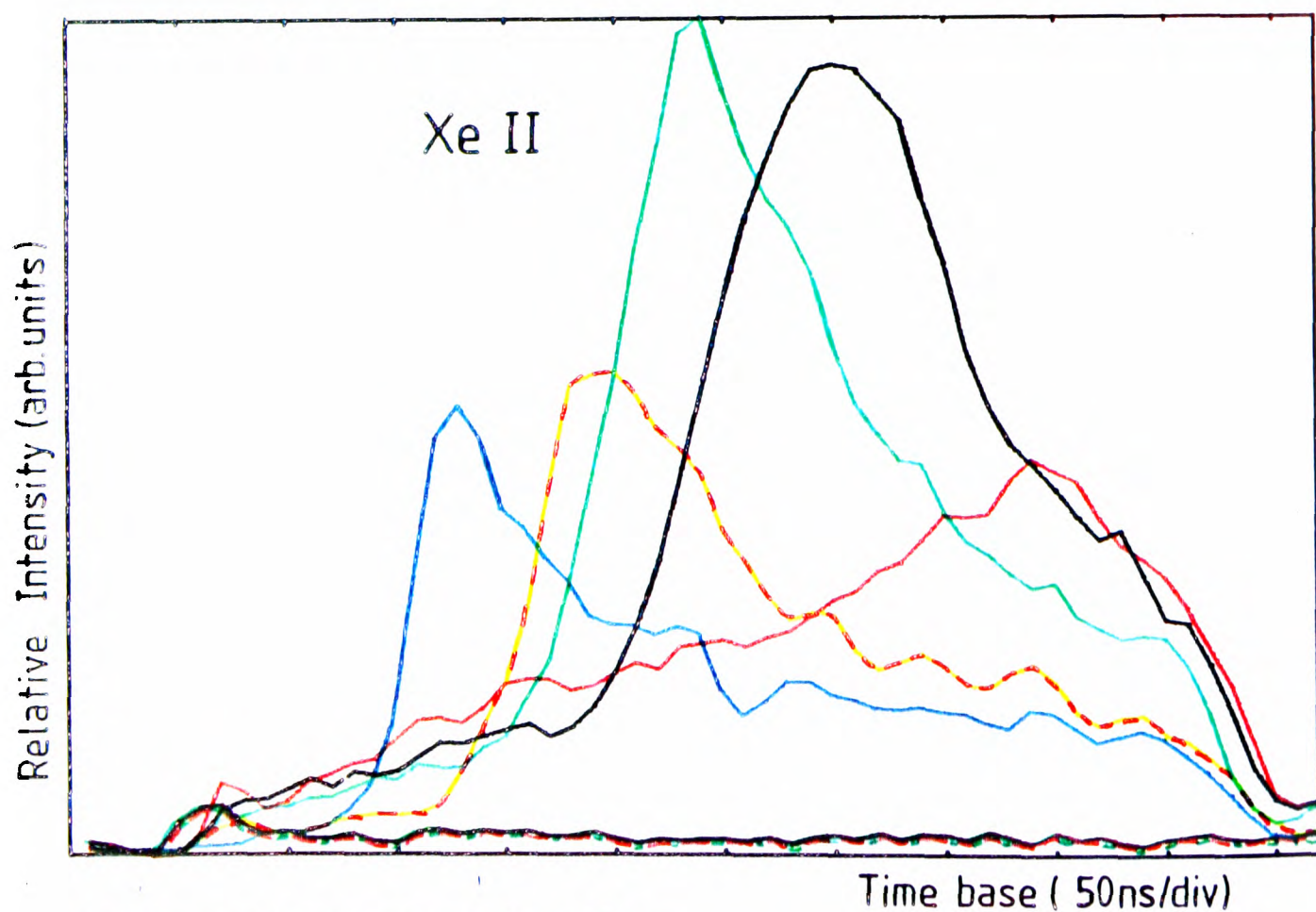


Figure 5.15



(a) Xenon neutral emission (467.123nm)



(b) Xenon Ion emission (484.433nm)

FIGURE 5.16: Xenon ion and neutral emission vs. HCl partial pressure in a Neon buffered discharge (cf. fig. 4.13)

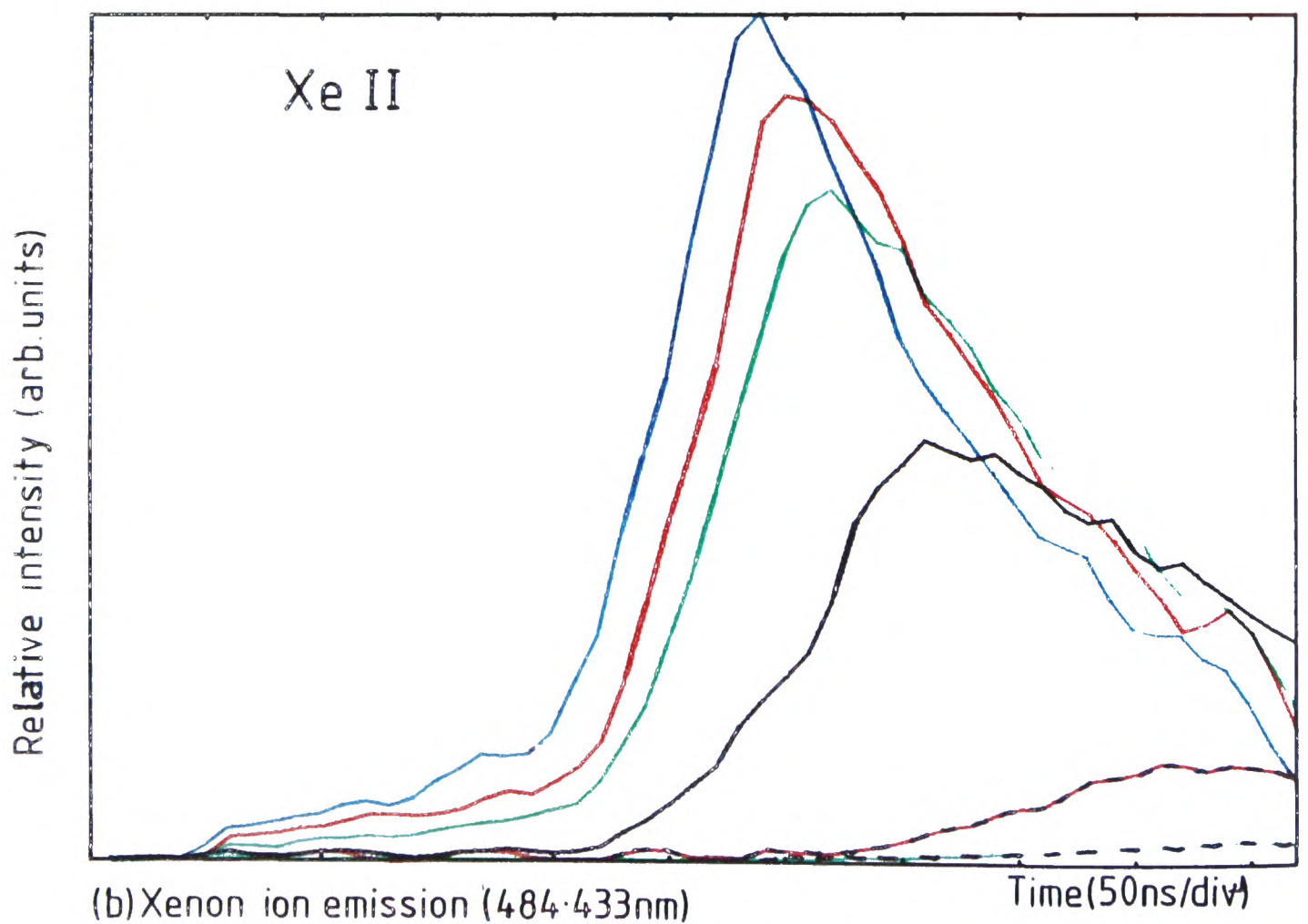
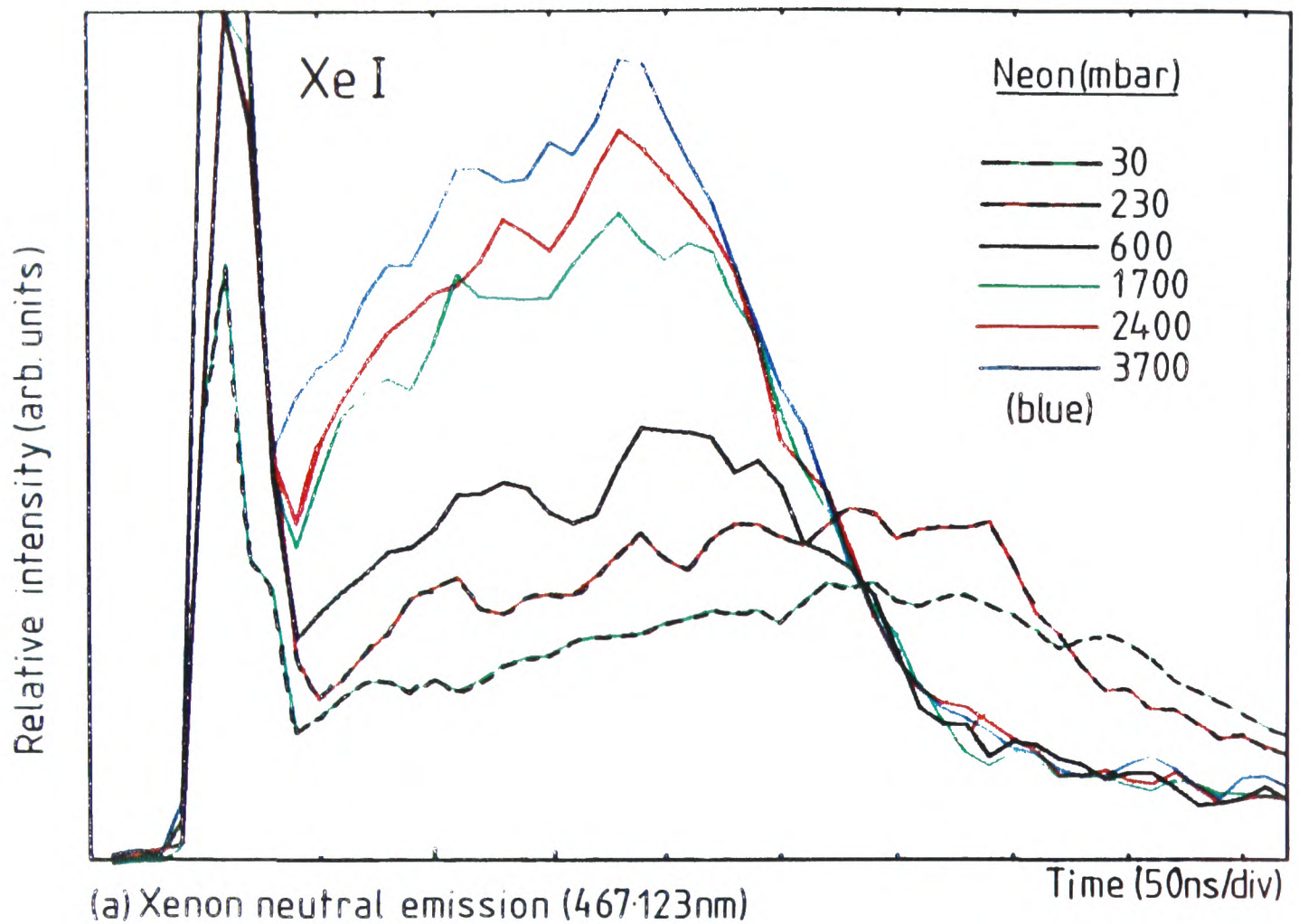
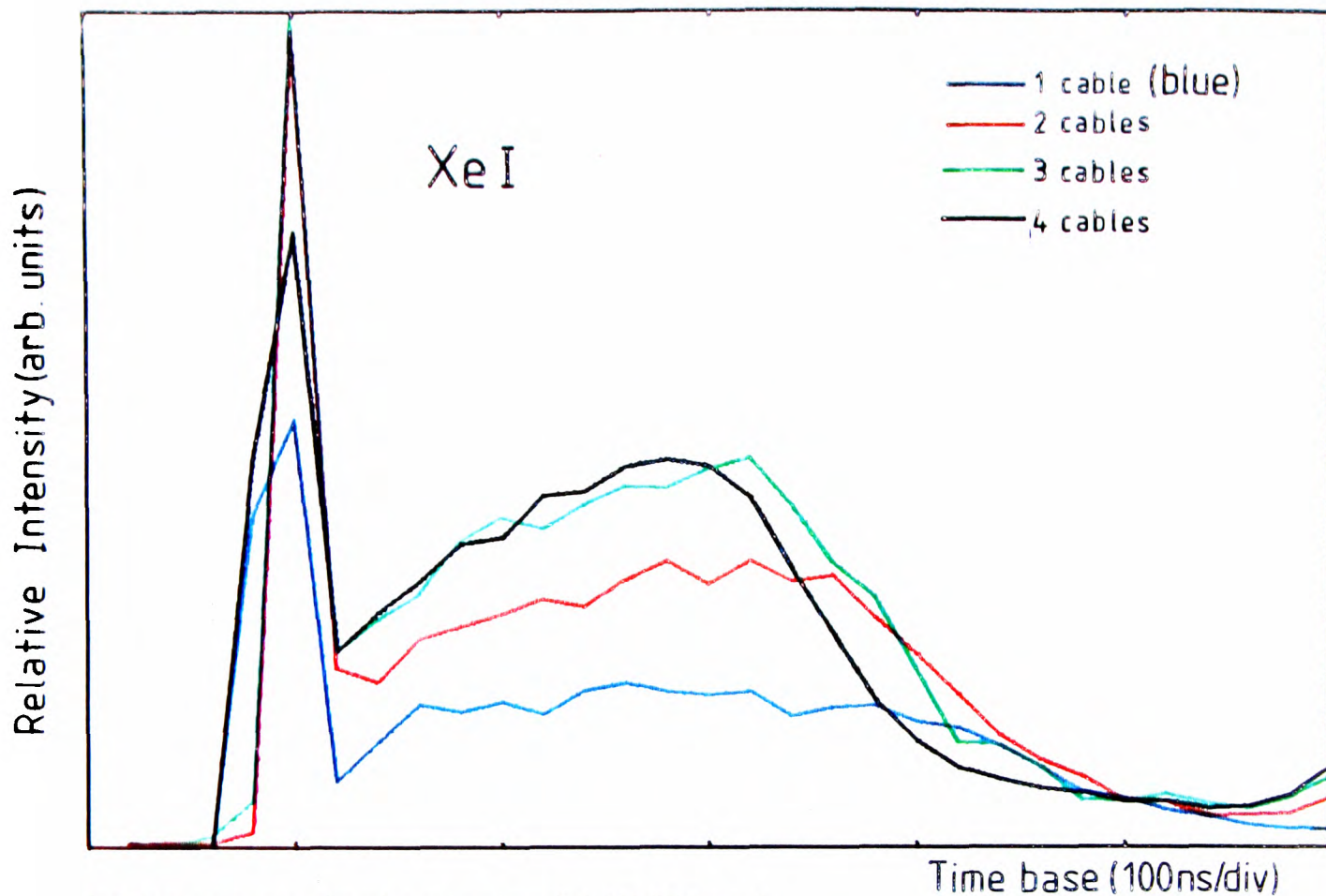
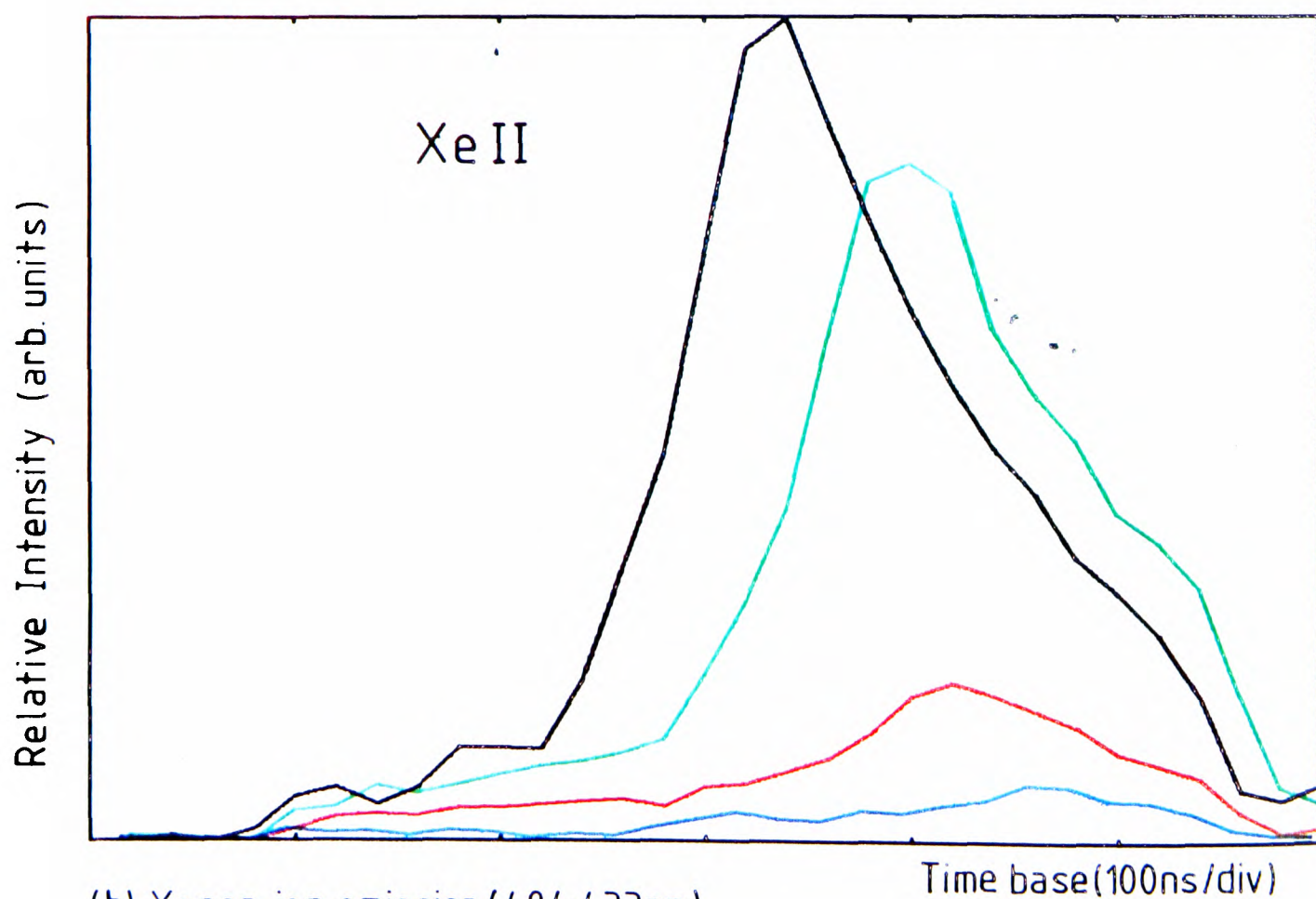


FIGURE 5.17 : Xenon ion and neutral emission vs. Neon buffer gas partial pressure. See figure 4.14 also.
(~30 ns time resolution)



(a) Xenon neutral emission (467.123nm)



(b) Xenon ion emission (484.433nm)

FIGURE 5.18 : Xenon ion and neutral emission vs. different input powers (see fig.4.17(b)) into a Neon buffered discharge.
 Scaling factor: $\text{Int(b)} = \text{Int(a)} \times 1.13$
 (60ns resolution in time base)

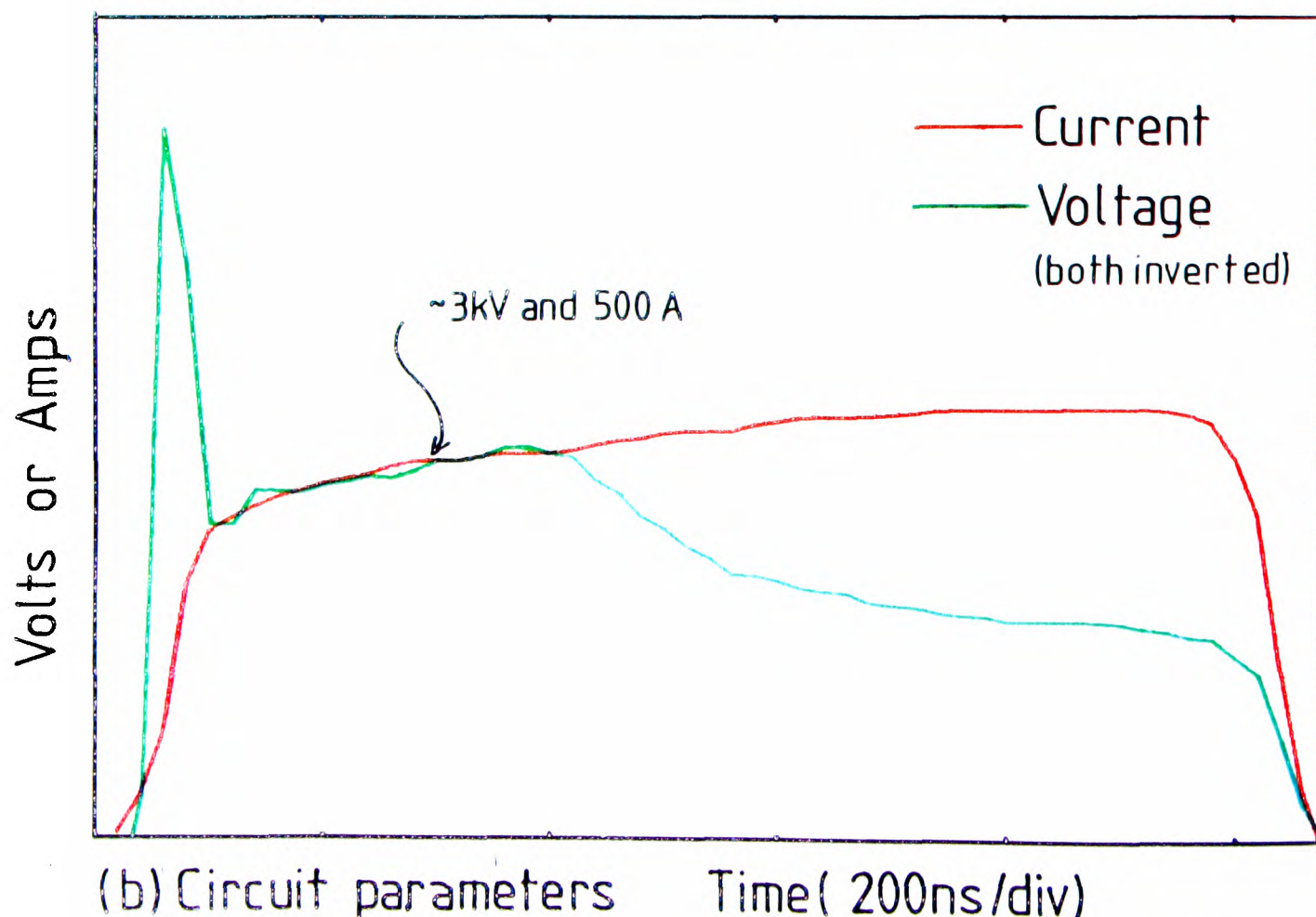
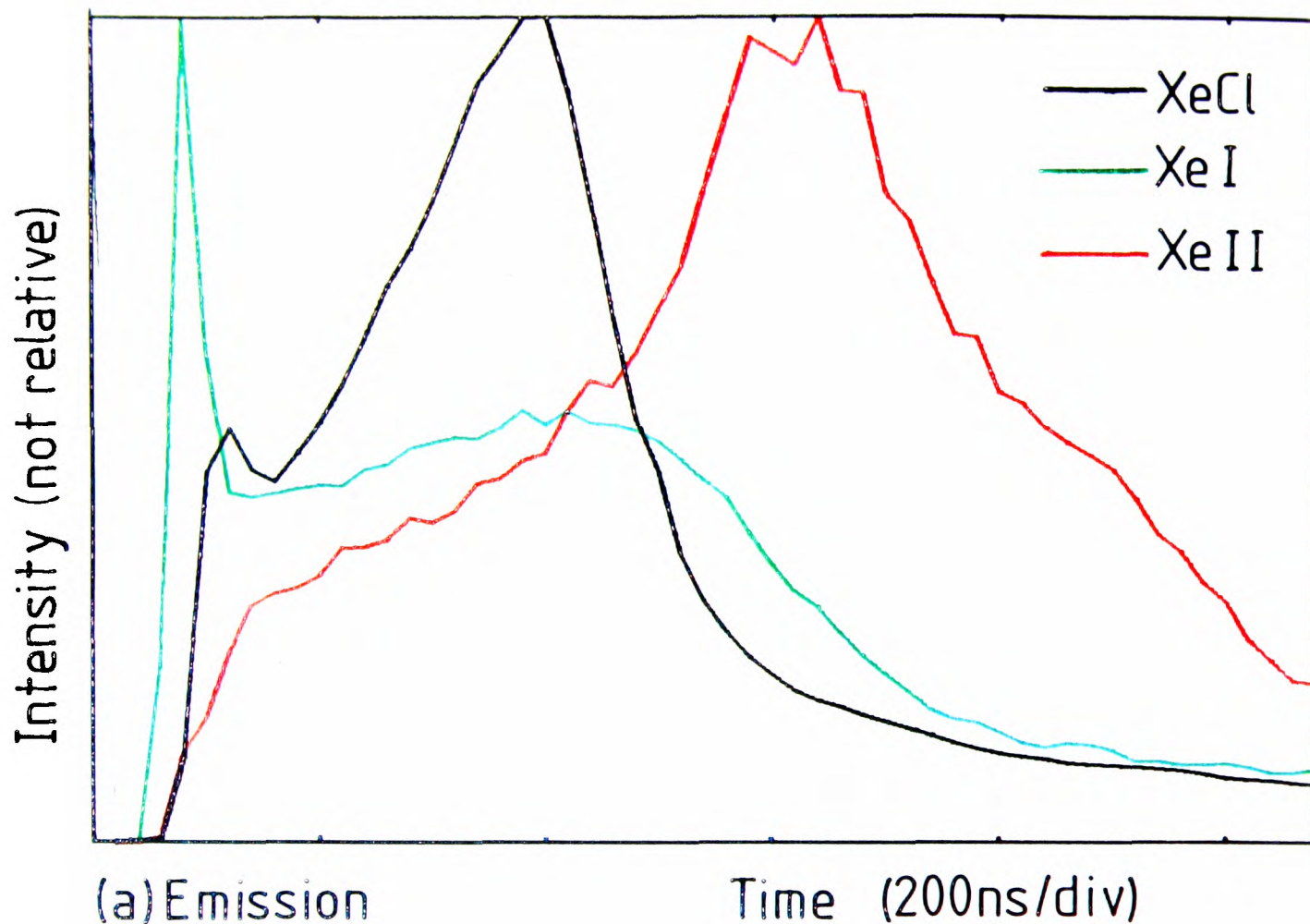


FIGURE 5-19 : Temporal evolution of various parameters when the discharge was driven with a $1\mu\text{s}$, one-cable PFN. Temporal response of recording apparatus was very poor: $\sim 120\text{ns}$.

emission of each of these species do not, however, have such a simple relationship to the HCl partial pressure: XeI emission increases up to 1 mbar HCl, then decreases, and both XeII and XeCl* emission peak at ~3 mbar.

Variation of the neon buffer gas pressure has little effect on the XeI and XeII emission in comparison to that caused by variation of the HCl partial pressure (figure 5.17). Although there is a general trend of increasing XeI and XeII emission with Ne pressure, there appears to be a sharp transition in the nature of the discharge between 200 mbar and 600 mbar: XeI emission, from discharges with a total pressure of 600 mbar or more, terminates well before that observed at lower pressures.

The total discharge current affects the XeI and XeII emission in the manner illustrated in figure 5.18. There is a tendency towards earlier termination of XeI and earlier onset of XeII emission as the total discharge current is increased. In the case of a 1-cable PFN, there was doubt as to whether or not XeI termination and XeII onset occur merely as a result of reaching the end of the discharge pulse. For this reason, studies were conducted with a PFN of longer pulse duration. Examination of figure 5.19 - emission from a discharge driven by a 1 μ s duration, 1-cable PFN - reveals that XeI termination and XeII onset arise in the same manner (but at a later time) as they arise in discharges with higher total currents and with shorter durations.

One further point to be made is that a brief scan of the entire emission spectrum between 300 and 600 nm indicated that xenon ion and neutral transitions were the only ones present: emission due to transitions in helium and neon was too weak to be observed above the broad-band background emission - if such emission was present at all.

5.3.3 Experimental details- absorption measurements:

The experimental arrangement used to measure the time resolved populations of various Xenon excited neutral and ion energy levels is shown in figure 5.21. For the sake of clarity the description of this arrangement will be separated into three parts: the absorption apparatus (contained within the electrically shielded room), the detection apparatus (situated outside the shielded room),

Populations of Xenon excited states by absorption: Experimental Details

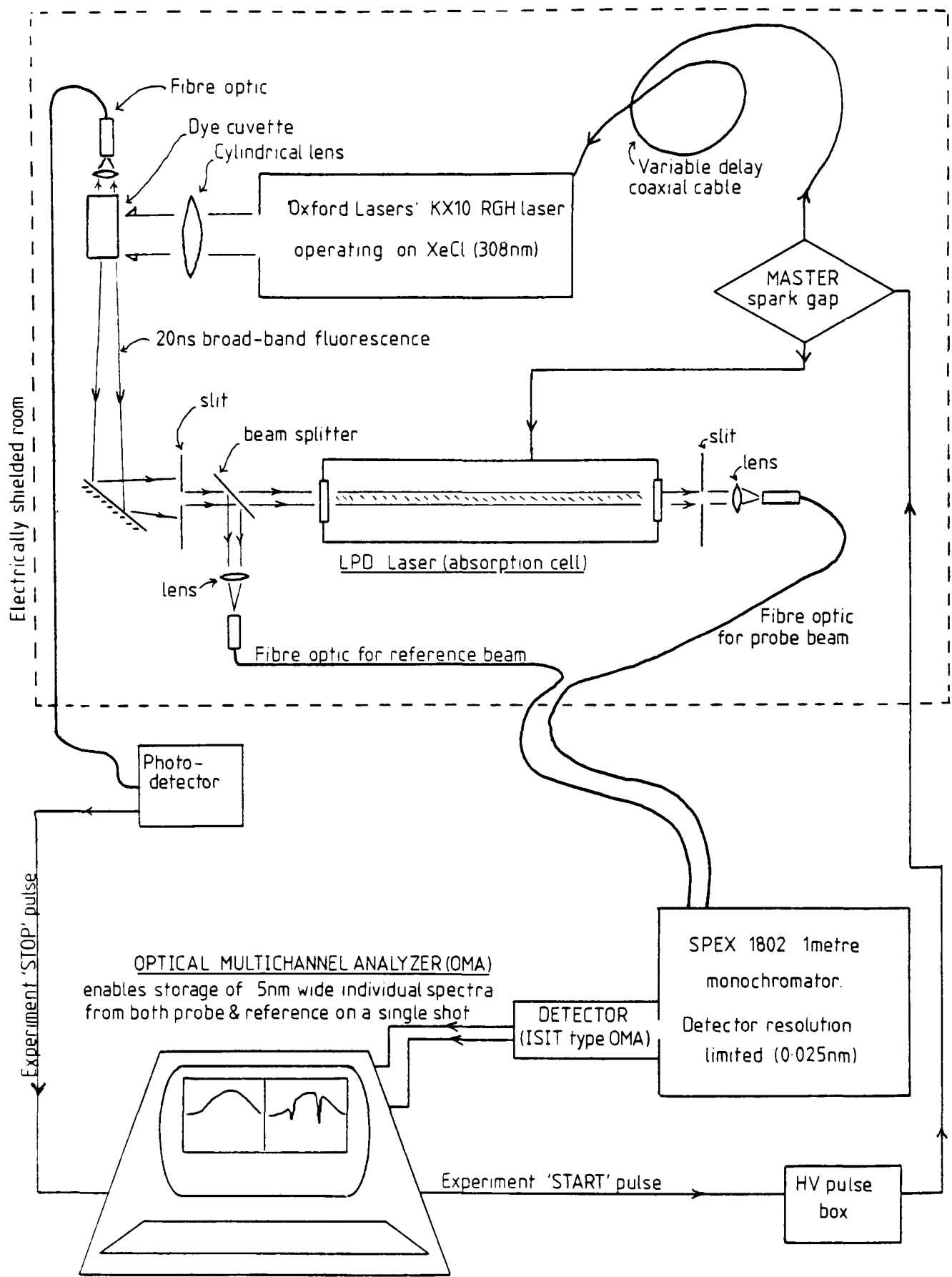


FIGURE 5.21

and data analysis.

(a) The absorption apparatus:

The main components of the absorption apparatus were: the absorption cell (LPD laser) and a source of high intensity, short duration, broad band, visible radiation. Temporally resolved absorption spectra were obtained using a similar method to that employed for transient absorptions in the UV: trigger pulses from a 'master' spark gap were sent via a variable length coaxial delay line to the respective 'slave' gaps of the LPD discharge and a short duration (~ 15 ns) XeCl^* laser (Oxford Lasers KX10) which provided the short (15 ns) UV pulse to optically pump a dye cell.

One type of dye - Coumarin 1 (7-diethylamino-4-methyl coumarin) dissolved in methanol - was used exclusively since all the features of interest lay within its fluorescence spectrum (440-500 nm). The criterion used to judge the optimum concentration of the dye in the solvent was that it be sufficiently low that amplified spontaneous emission did not cause narrowing of the dye fluorescence spectrum. Broad band fluorescence of the dye was desirable because, using the detection apparatus described below, an entire absorption profile could be obtained from a single discharge event. The intensity of dye emission was approximately two orders of magnitude greater than emission from the LPD discharge in the spectral region of interest. The dye cuvette had to be tilted to stop its end faces acting as a laser cavity.

The fluorescence from the dye was directed through the LPD discharge using the same two slits employed in the UV transient absorption measurements. In this case however, the reference beam was obtained by using a beam splitter to divert some of the fluorescence intensity into a fibre-optic cable after it had passed through the first slit. Problems with variation in the spatial intensity profile of the fluorescence were thereby eliminated. The probe radiation was also collected in a fibre optic cable before being passed out of the electrically shielded room for detection. The transmission characteristics of the (plastic) fibre-optic were such that it was not necessary to provide an additional filter to remove 308 nm emission from the LPD discharge.

(b) The detection apparatus:

The key piece of equipment in this experiment was a microprocessor controlled optical multichannel analyser ('OMA' - Princeton Applied Research Corporation model no.1215 processor with a model no. 1254 SIT detector head incorporating a UV scintillator). The importance of this device lay in the speed at which an entire absorption spectrum could be recorded. A summary of the principles of operation of an OMA detector head is included as Appendix 5. To understand the precise details of its operation, however, is not vital to this discussion. It is important only to appreciate that this device made it possible to record, in a single discharge event, a 500 point intensity profile of both the probe and the reference signals as a function of wavelength. The digitised data could then be analysed to obtain $\int k_{\lambda} d\lambda$ using the OMA software.

The OMA detector head was mounted directly onto a monochromator (SPEX model 1802) in the exit focal plane centred on the position normally occupied by the exit slit: the spectral region which could be scanned was determined by the dispersion of the monochromator at the wavelength of interest. Spectra were always recorded in the second order of the grating. The dispersion at a wavelength of ~ 450 nm was such that a 5 nm wide spectrum could be recorded on a single shot. The resolution of individual spectral features was limited by the diode spacing of the OMA detector head to ~ 0.025 nm.

Individual spectra from both the probe and the reference beams were collected simultaneously from one discharge event. This was done by using a microprocessor software facility which enabled the detector head to be subdivided into two (or more) horizontal light sensitive areas (tracks) - one above the other. The ends of the fibre-optic cables carrying the probe and reference beams were brought into direct contact with the entrance slit of the monochromator using a purpose designed clamp. Since these ends were separated on the entrance slit by ~ 1 cm, they formed separate spectra in the plane of the detector head. It was a simple matter to arrange that the OMA tracks were in a position to detect these spectra.

The OMA microprocessor was used to automate the entire experiment. An

Absorption spectrum: XeI 467.123nm

at 100ns after discharge initiation in a laser mixture

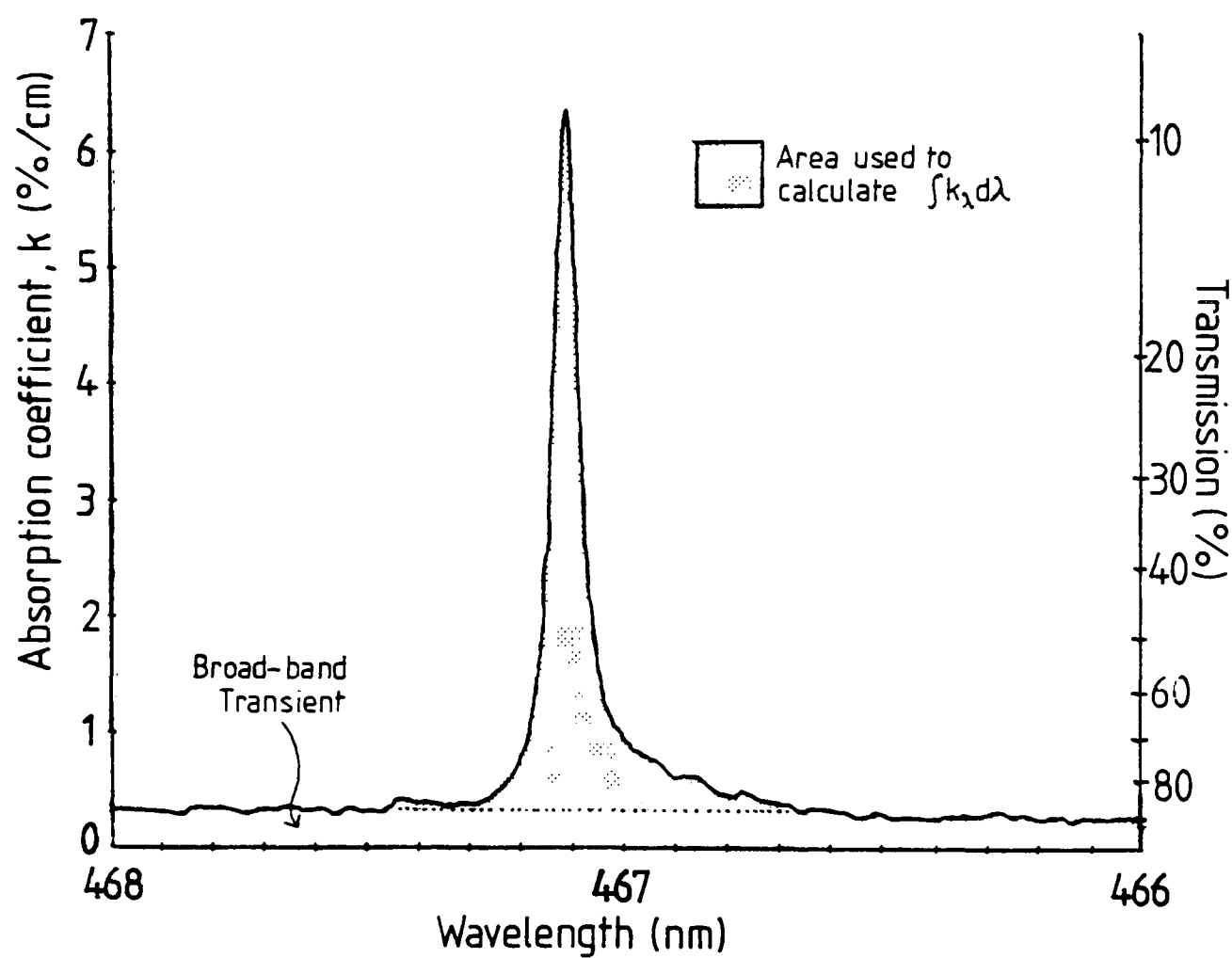


Figure 5.22

'experiment-start' pulse from the OMA switched a HV pulser box which subsequently triggered the master spark gap. The processor commenced the scanning of the detector head $\sim 1 \mu\text{s}$ after receiving an 'experiment-stop' pulse from a photodetector (as shown in figure 5.21).

(c) Data analysis:

In addition to automating the data collection process, the OMA microprocessor was used to perform all the data analysis.

At each parametric data point,[†] the spectral profiles of each of approximately twenty shots were collected. Each reference profile was divided by the corresponding probe profile before averaging. The mean ratio of reference to probe, thus obtained, required normalisation because a difference existed in the absolute intensities of the probe and reference beams in the absence of an LPD discharge. A spectral normalisation curve was obtained for each parametric data point. Each normalisation curve was an average of two reference/probe intensities ratios which were taken:

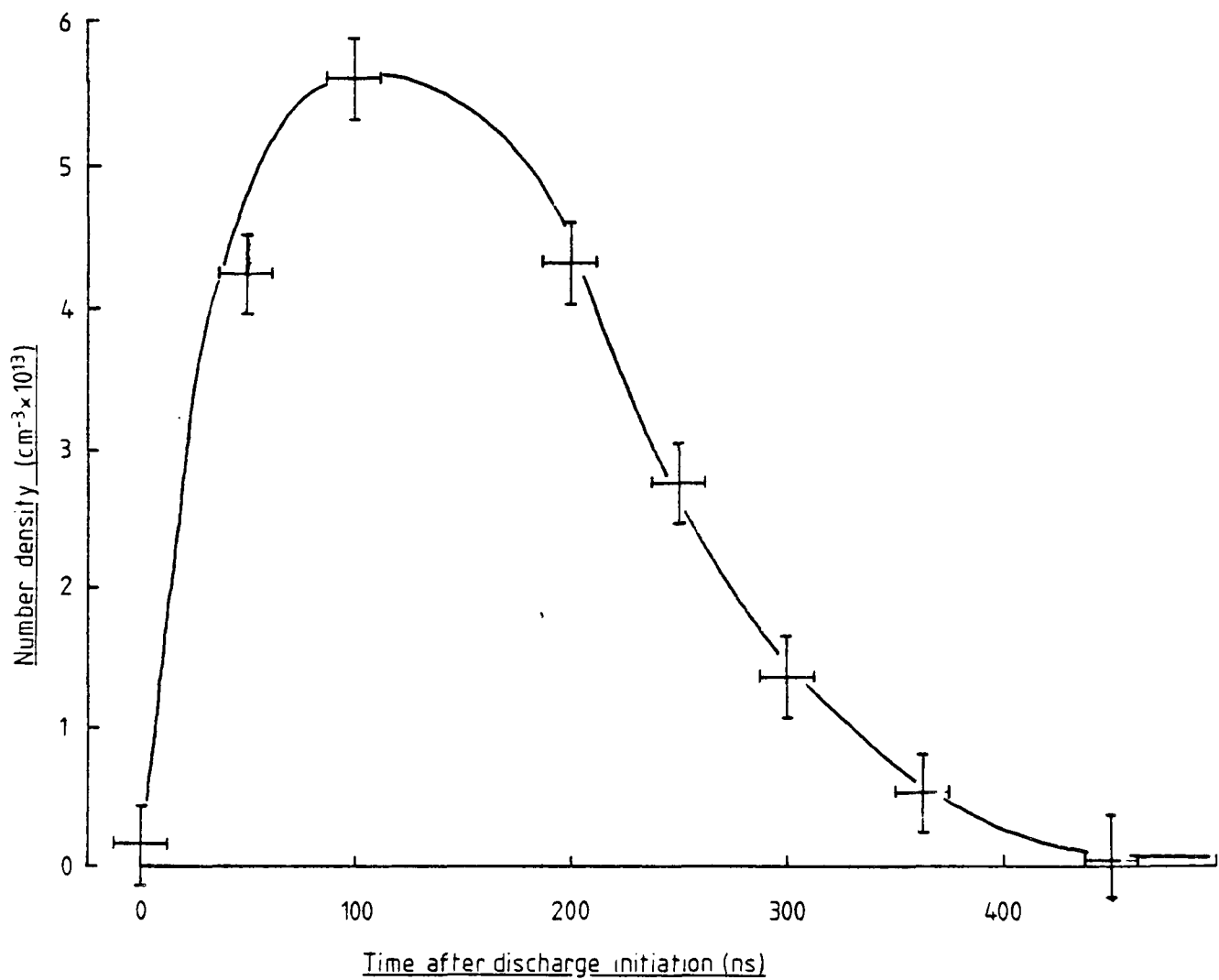
- (1) at the beginning and end of each '20 shot' data collection run
- (2) in the absence of an LPD discharge.

Often, there was a detectable difference between the normalisation ratio taken at the beginning and that taken at the end of a run. Since the average ratio was used for normalisation, this difference made itself evident in the final absorption spectra by introducing a shift into the baseline (100% transmission). The magnitude of the shift was never more than 1% at any particular wavelength, but it varied across the spectrum. The error due to this was insignificant, but accounts for what might otherwise be interpreted as gain (101% transmission) at various wavelengths in some of the absorption spectra.

Curves of k_λ were obtained by taking the natural logarithm of each (normalised) mean spectral intensity ratio and dividing by the 38 cm absorption path length. The value of $\int k_\lambda d\lambda$ for each parametric data point was obtained

[†] At one time during the discharge or at one particular HCl partial pressure.

XeI - $6s[3/2]_2$ (metastable) population vs. time



A constant 38cm absorption path was assumed here. The measurement was performed in a normal laser gas mixture (see table 4.2). A transition probability of $1.07 \times 10^6 \text{ s}^{-1}$ was used.

Figure 5.23

using an OMA integrating function which summed the magnitudes of k_λ over an operator-specified number of wavelength channels. The number of channels chosen depended upon where the wings of an absorption feature were judged to terminate. As was the case in Kearsley's excited state xenon population measurements, a transient broad band background absorption was found to be present. The typical absorption spectrum in figure 5.22 shows how this absorption was excluded from the determination of $\int k_\lambda d\lambda$.

5.3.4 Results - absorption measurements:

Results of the following measurements are presented in this section:

- (1) The time-resolved population of the $6s[3/2]_2$ XeI (metastable) level through absorption by the $6s[3/2]_2 - 7p[5/2]_3$ (467.123 nm) transition in a normal laser gas mixture: figure 5.23.
- (2) The time-resolved population of the $6s[3/2]_1$ XeI (resonance) level through absorption by the $6s[3/2]_1 - 7p[3/2]_1$ (482.971 nm) and the $6s[3/2]_1 - 7p[3/2]_2$ (484.329 nm) transitions in a normal laser gas mixture: figure 5.24.
- (3) The time-resolved population of the $6s^4P_{5/2}$ XeII level[†] through absorption by the $6s^4P_{5/2} - 6p^4D_{7/2}^0$ (484.433 nm) transition in a normal laser gas mixture: figure 5.25.
- (4) Dependence of the population in (1) above on HCl partial pressure at a time of ~100 ns after discharge initiation: figure 5.26.
- (5) Ditto (4), dependence on total discharge current (and therefore electron density): figure 5.27.

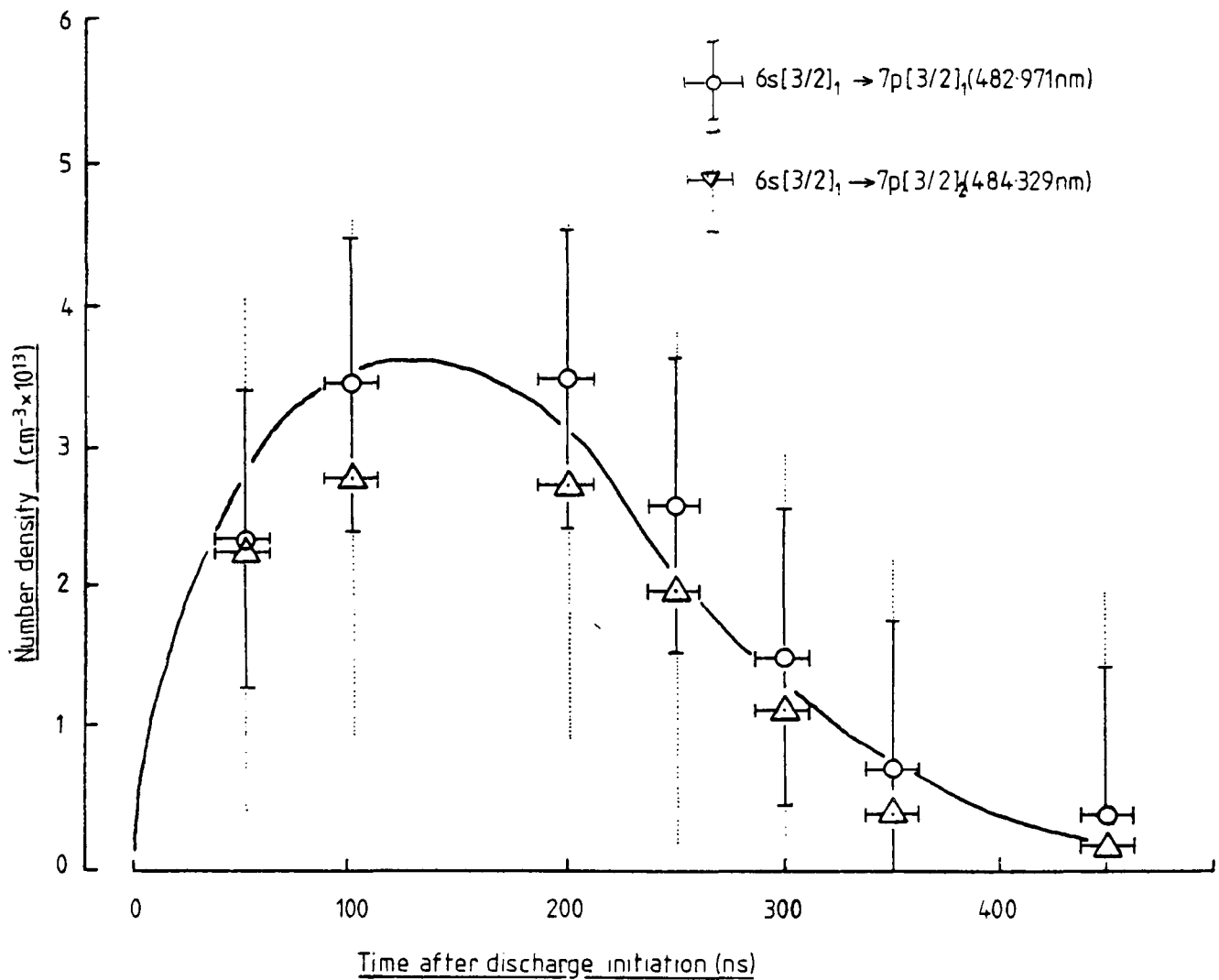
Except where stated, the values of all parameters were as set out in table 4.2.

(a) General comments:

It must be stressed that the number densities in figures 5.23-5.27 were calculated on the basis of a constant 38 cm absorption path length: the ramifications of this assumption will be made clear later. The error bars only provide an indication of the uncertainty in the measurement of $\int k_\lambda d\lambda$ from the

[†] Transition from this level to the ion ground state is forbidden by the $\Delta S=0$ rule, but L-S coupling is a poor description in XeII.

XeI- $6s[3/2]_1$ (resonance) population vs. time



A constant 38 cm absorption path was assumed here. The measurement was performed in a normal laser gas mixture (see table 4.2). The following transition probabilities were used: $4.9 \times 10^5 \text{ s}^{-1}$ (482.9nm) & $1.6 \times 10^5 \text{ s}^{-1}$ (484.3nm).

Figure 5.24

absorption spectra - they do not include uncertainties in transition probabilities (A_{21}). The measurement of $\int k_{\lambda} d\lambda$ for the XeI(484.329 nm) and XeII(484.433 nm) transitions was subject to larger errors not only because the magnitude of the absorption was less than the others, but also because there was some overlap of these features in their wings. The k_{λ} spectra used to obtain each data point are included as an appendix (6).

Equations 5.2-5.4 are expressed in terms of frequency, however, the quantity which is measured directly is $\int k_{\lambda} d\lambda$. An expression for the number density in terms of wavelength can be found using the relation $\lambda\nu = c$. Combining this with eqns.5.2 and 5.3 an expression is found for the number density of the lower level, N_1 , in terms of A_{21} and the wavelength, λ , of the transition:

$$N_1 \approx \frac{g_1}{g_2} \frac{8\pi c}{\lambda^4 A_{21}} \int_{\text{line}} k_{\lambda} d\lambda \tag{5.14},$$

in which it is assumed that λ does not vary significantly over the absorption feature and that the upper level population is small. Table 5.4 contains data used in the calculation of the various Xe populations at the time of peak absorption via eq.5.14. The values of A_{21} used for the three XeI transitions

Table 5.4
Data used for calculation of peak XeI and XeII number densities

Data	Energy level and wavelength of transition(nm)			
	XeI (metastable)	XeI (resonance)		XeII
Time of Peak (ns after initiation)	100	200	100	300
$\lambda(m \times 10^{-9})$	467.123	482.971	484.329	484.433
$\lambda^4(m^4 \times 10^{-27})$	47.6	54.4	55.0	55.1
g_1/g_2	5/7	3/3	3/5	6/8
$\int k_{\lambda} d\lambda \ (x10^{-11})$	52.6	12.4	5.5	1.2
$A_{21} \ (s^{-1}x10^6)$	1.07 ^a	0.49 ^a	0.16 ^a	172 ^b
$N_1 \ (cm^{-3})$ (Kearsley) ^c	5.6x10 ¹³ (5.6x10 ¹³)	3.5x10 ¹³ (9.4x10 ¹³)	2.8x10 ¹³ (8.0x10 ¹³)	7.2x10 ⁹

$$8 \pi c = 7.54x10^9 ms^{-1}$$

(a) Experimental value - Richou[1980];

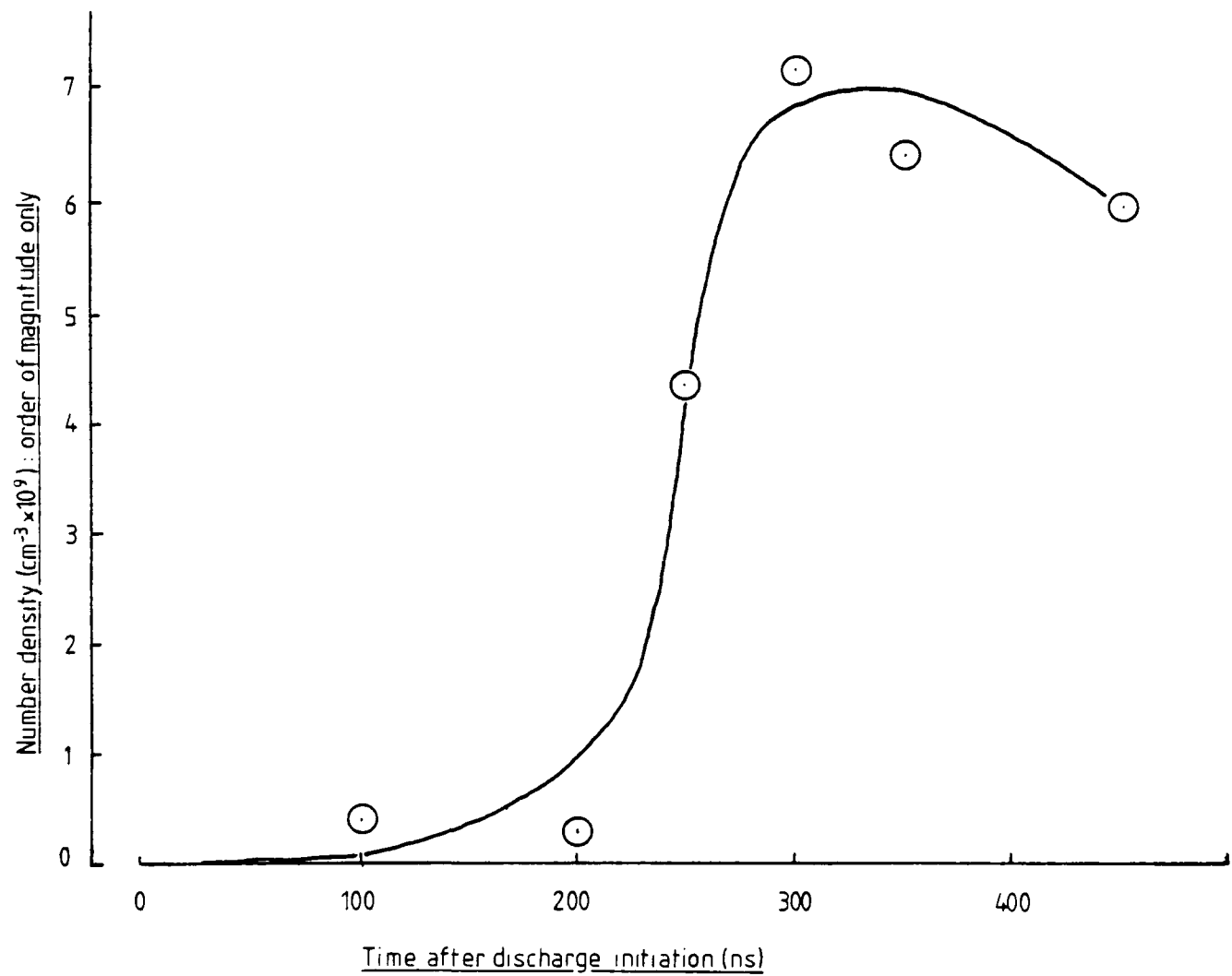
(b) Inverse of the upper level lifetime (5.8 ns) - Mohamed and King[1979];

(c) Kearsley[1980]: values have been adjusted here to include same A_{21} values;

These data points are for a normal laser gas mixture and a 4-cable PFN.

They correspond to the maximum observed absorption as a function of time under the stated conditions. A constant 38 cm absorption path length was assumed.

XeII - 6s $^4P_{5/2}$ population vs. time



A constant 38cm absorption path was assumed. The measurement was performed in a normal laser gas mixture (see table 4-2). The inverse of the upper level lifetime ($6p^4D_{7/2}^{\circ} : 1.72 \times 10^8 s^{-1}$) was used for the transition probability.

Figure 5-25

are the most recently available experimental values (Richou[1979]). The transition probability quoted for the XeII transition is merely the inverse of the upper level ($6p^4D_{7/2}^0$) lifetime measured by Mohamed and King[1980]. Since its use contains the implicit assumption that radiative decay occurs only to the $6s^4P_{5/2}$ level, this value will represent an upper limit on A_{21} , and therefore a lower limit on the calculated number density of the $6s^4P_{5/2}$ level.

(b) Specific features of the results:

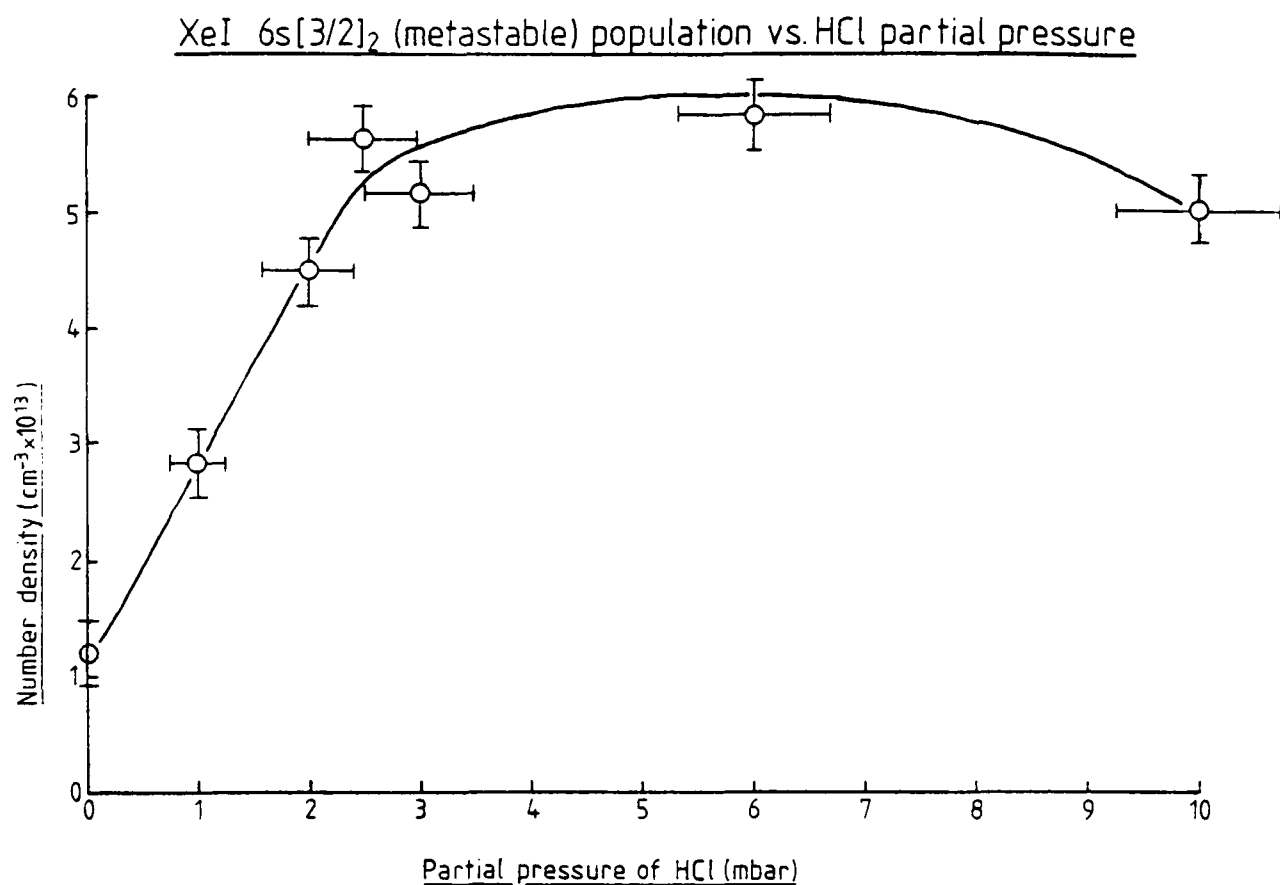
The temporal evolution of both the XeI(metastable) and XeII populations, in figures 5.23-5.25, show a marked similarity to the temporal evolution of the corresponding emission features presented in section 5.3.2 . The XeI metastable population exhibits a sharp rise after discharge initiation but then begins to fall after ~200 ns has elapsed. The behaviour of the population of the XeI resonance level with time is, within experimental error, exactly the same.

On the other hand, the XeII- $6s^4P_{5/2}$ number density does not rise until late in the 500 ns discharge pulse, and it remains high until pulse termination.

The dependence of the XeI metastable population on the HCl partial pressure (figure 5.26) would appear to go against this trend of a correlation between upper and lower level populations.[†] In figure 5.16(a) (at $t \approx 100$ ns), the intensity of emission decreases steadily as the HCl partial pressure is increased; in apparent contradiction, the XeI number density, measured in absorption, increases monotonically in the 0-2.5 mbar range and then reaches a plateau.

Large increases in the total discharge current (achieved by changing the number of cables in the PFN) have a comparatively small effect on the XeI metastable population (figure 5.27). It would appear that the discharge is operating close to a point at which the population becomes independent of the electron density. For purposes of comparison, the population measured by Kearsley[1980] is also included in figure 5.27. The electron density in the discharge used by Kearsley was ~5 times greater than that present in the LPD discharge when the total LPD laser discharge current was 2000 A.

[†] Upper level population of a particular transition as monitored in emission; lower level population as monitored in absorption.



All data points taken at a time of 100ns after discharge initiation. All other parameters as in table 4.2. Compare with figure 5.23. Constant absorption path assumed (38cm).

Figure 5.26

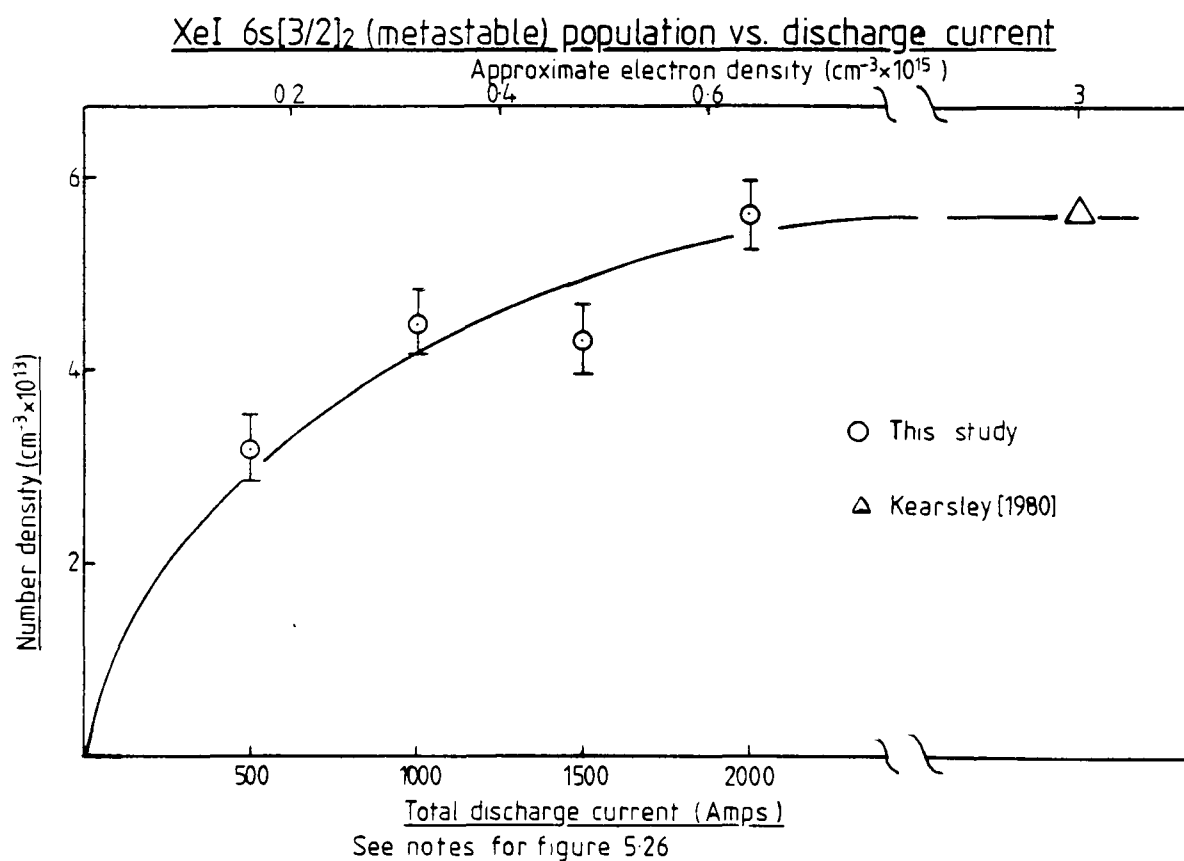


Figure 5.27

It also interesting to note that the ratio of the measured number densities of the metastable to resonance levels is very close to that which would be expected if they were 'well mixed' via electron impact. The ratio of the degeneracies of the metastable to resonance levels is $5:3 = 1.67$. At 100 ns after discharge initiation in a normal laser gas mixture, the ratio of the measured populations (metastable:resonance) is 1.6 when the resonance level is measured via the 482.971 nm transition, and is 2.0 when it is measured via the 484.329 nm transition.[†]

5.4 TIME-RESOLVED MEASUREMENTS OF EMISSION AND ABSORPTION IN THE

VISIBLE - DISCUSSION:

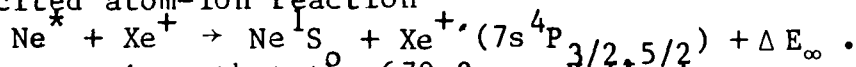
5.4.1 The occurrence of XeII emission as a proof of discharge constriction:

The presence of the XeII emission was a key factor in arriving at the conclusion that discharge constriction was the major cause of laser pulse termination.

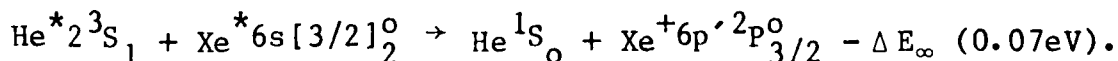
Excitation to the high-lying upper levels (26 and 28.5 eV above the xenon neutral ground state) of the observed ion transitions will be dominated by electron impact.[¶] Given this fact, it could be postulated that single-step

[†] It would be expected that the ratio obtained via the latter measurement of the resonance level would be high because, as mentioned above, the measurement of $\int k_{\lambda} d\lambda$ was affected by the proximity of the absorption line of the XeII transition.

[¶] For completeness it is noted that there are indeed two alternative mechanisms which could populate the upper level of the 486.254 nm transition by atom-atom collisions. Willett[1974](p.420) notes that selective excitation of the upper level 572.855 nm and 609.529 nm XeII laser transitions occurs in a Xe-Ne mixture via the excited atom-ion reaction



Willett also mentions that the 670.2 nm XeII laser upper level is possibly subject to selective excitation via the atom-atom excitation transfer reaction



It is conceivable that the $6p^2\text{P}$ XeII levels are well mixed via electron collisions with the $7s^4\text{P}$ levels. A pathway for populating the upper level of the XeII transition shown in fig.5.11. (and that of the XeII transition in figure 5.12, if the $6p^4\text{P}$ and $6p^4\text{D}$ are also well mixed - E is ~ 0.34 eV), other than via electron impact, is therefore possible in either helium or neon buffered mixtures.

However, although any such explanation can be dismissed on grounds of numerical estimates of rates, it can equally well be rejected on the basis of simple experimental observation.

ionization from the neutral ground state is responsible. However, this proposal can be rejected by the following simple argument: under the same discharge conditions and at the same time, little, if any, emission is detectable from either neon or helium excited state transitions. This is despite the fact that:

- (1) The ground state density of the buffer gas is two orders of magnitude greater than the ground state xenon density.
- (2) The energy required for ionization, let alone excitation of He and Ne is less (1.5 eV in helium and 4.5 eV in neon) than that required for single-step excitation to the XeII- $6p^4D$ levels.
- (3) If excitation and ionization in a single step were responsible, the required increase in the mean electron energy, ϵ , must occur at the same late stage of the discharge pulse at which the XeII emission rises. The observed drop in the discharge voltage at this time points, in contradiction, to a decrease in ϵ .

The only plausible process by which excitation might occur is multi-step excitation via the XeII ground state.

Recalling now the discussion in subsection 2.2.2, it can be deduced from eq.2.4 that, in the case of multi-step excitation via the ion ground state, the population in the excited state will have at the very least a linear dependence on the electron density. This arises because the ground state XeII density is dependent only on the electron density (charge neutrality). Thus, even in the case of $d_j \gg A'_j$, where the n_e terms in both the numerator and denominator of eq.2.4 cancel, an n_e dependent term still remains.

This linear dependence on n_e represents the minimum case: that of one 13-14 eV step from the XeII ground state to the $6p^4D$ levels. It is very likely that excitation will be influenced considerably, and perhaps dominated, by processes involving excitation of the ion in more than one step.

For example, excitation could proceed from the $(5s^2)5p^5\ ^2P_{1/2}^0$ state via the $(5s)6s\ ^4P_{5/2}$ state (~ 10 eV jump) and then to the $6p^4D$ levels (~ 2.5 eV jump). The sharp dependence of the rate of excitation by electron impact on the threshold energy for excitation, ϵ_{ij} , is the reason that processes involving

more than one excitation step from the ion ground state could compete effectively with single step excitation from the ion ground state.

Let us now compare the efficiency of excitation via these two schemes. For the case of a Maxwellian electron energy distribution and a step function excitation cross section, Q_{ij} , starting at ϵ_{ij} (T_e in eV), the rate coefficient for excitation, S_{ij} , is given by

$$S_{ij} \propto T_e^{1/2} \left(1 + \frac{\epsilon_{ij}}{T_e}\right) \exp\left(-\frac{\epsilon_{ij}}{T_e}\right) \quad (5.15)$$

(see e.g. Green and Webb[1975]). The difference in the S_{ij} 's (of the one-step process and the first step of the multiple-step process in the above example), brought about solely by the difference in threshold energies, can be found by inserting the appropriate ϵ_{ij} 's into eq.5.15. For electron temperatures of 1.5 eV and 2 eV, either of which could be expected to be typical of this type of discharge,

$$\frac{S_{ij}(10\text{eV})}{S_{ij}(14\text{eV})} \approx 10 \frac{Q_o(10\text{eV})}{Q_o(14\text{eV})} \quad (T_e = 1.5 \text{ eV})$$

$$\frac{S_{ij}(10\text{eV})}{S_{ij}(14\text{eV})} \approx 5 \frac{Q_o(10\text{eV})}{Q_o(14\text{eV})} \quad (T_e = 2 \text{ eV}),$$

where the Q_o 's are the magnitudes of the cross section above threshold. It is conceivable that the peak value of the cross section for single step excitation of the ion ground state is larger than those for multiple collision excitation; it is likely, however, that they are similar considering the similarity of the change in electronic configuration required.

The above calculation indicates that the rate of excitation for the 10 eV jump in the postulated multiple step excitation process is ~10 times greater than that of single step excitation from the ion ground state solely on the basis of the difference in the threshold energy for the excitation. A comparison of single step excitation from the ion ground state with the second step (~2.5 eV) of the multiple step process, on the same basis, indicates a rate which is ~600 times greater. However, this does not take account of the much smaller lower state number density which would be expected in the second step: It may well be that the proportions of the excited state XeII density

contributed by the single step and the multiple step process are similar.

The dependence of the combined excitation rate to the upper level of the observed XeII transitions on the electron density, in the case of multiple step excitation from the ion ground state, will be given by:

$$S_{ij} \propto n_e^{(k+1)} \quad (5.16),$$

where k is the number of collisions involved, and it is assumed that the populations of the intermediate levels are not saturated with respect to the ion ground state. Any intermediate state which is saturated with respect to the ion ground state will reduce the exponent of n_e in eq.5.16.

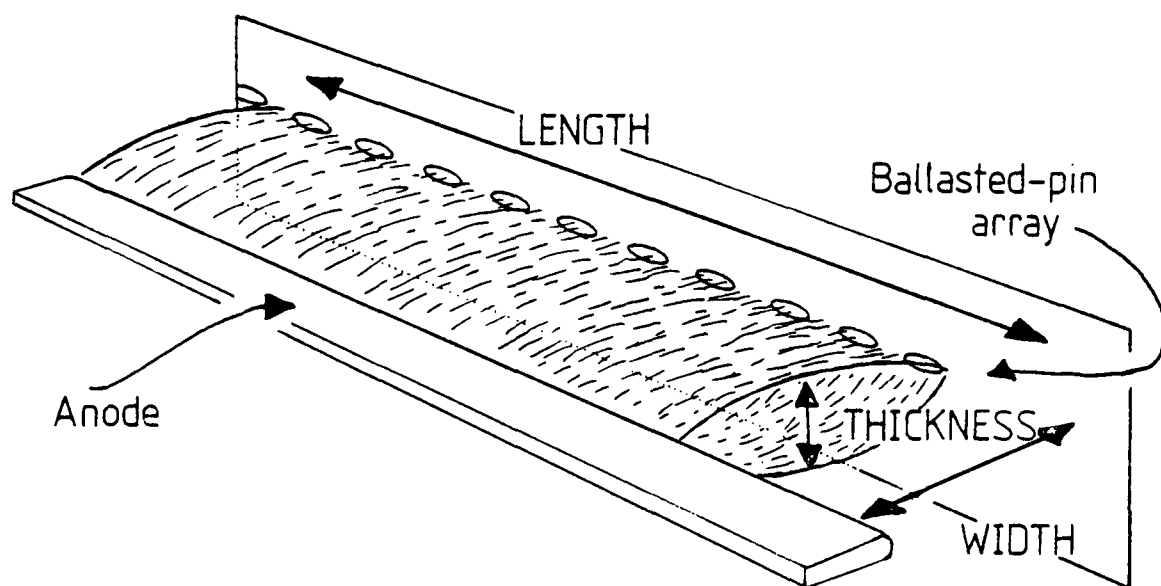
The conclusion which is arrived at, then, is that the observed XeII emission is brought about by many step excitation of xenon neutral atoms via the ion ground state as a direct result of an increase in the electron density. The number density of the upper level of the 484.433 nm transition will be at the very least linearly dependent on the electron density and possibly (assuming two steps from XeII ground state) have a cubic dependence on n_e .

The implications of a requirement for an increase in the electron density on the macroscopic parameters of the discharge becomes evident by re-examination of eq.4.21:

$$n_e = \frac{I}{eAv_{\text{drift}}} \quad (5.17).$$

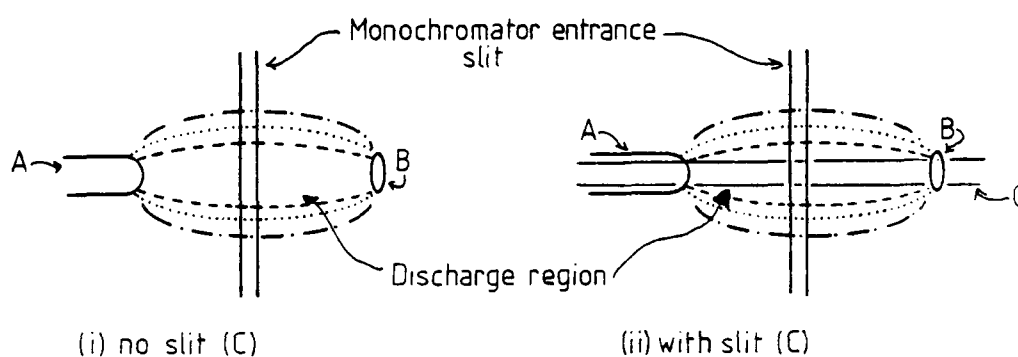
In the case of a normal (2-3 mbar HCl) gas mixture, the change in the discharge voltage and current over the entire 500 ns is small - 5% at most. An increase in electron density at constant gas density can therefore only be accounted for by a reduction in the cross sectional area for current flow, A , i.e. discharge constriction.

The apparent drop in the XeI excited state density, particularly the metastable and resonance levels, is consistent with this conclusion. As was shown in section 5.3.4(b), these levels are saturated with respect to the electron density - their number densities will therefore be a function of the electron temperature alone. The electron temperature would be expected to fall with the observed drop in discharge voltage and a consequent fall in the population would therefore be expected. As is discussed in the next section,



(a) Terms used to describe the dimensions of the LPD discharge

A = anode ; B = cathode; C = 'sampled-cross-section-limiting' slit around discharge



Discharge thickness	Cross sectional area of emission detected	
	(i) no slit	(ii) with slit

.....		

(b) How variation in discharge thickness could affect the measurement of relative intensities of emission

Figure 5.28

however, the dominant cause of the drop in number density indicated in figures 5.23 and 5.24 is a result of the assumption of constant absorption path length.

5.4.2 The effect of discharge constriction on number density measurements:

Discharge volume constriction will affect not only the number density of a level through collisional excitation and de-excitation, but will also affect the relationship between the number density and the parameters which are used to quantify it experimentally. For example, in the measurements of number densities via absorption as a function of time, the integrated absorption coefficient as a function of time, $\int k_{\lambda}(t)d\lambda$, must be known (eq.5.14). The measured quantity, however, is the product of $\int k_{\lambda}(t)d\lambda$ and ℓ , the length of absorber through which the probe radiation must pass. If ℓ is time-dependent (if the volume between each ballasted pin is not pumped at times late in the discharge - section 4.7) and this dependence is not included in the calculation, not even the relative magnitudes of the number density at various times may be accurately compared. If the discharge path length, at a particular point in time, has a functional dependence on some other parameter (such as the HCl partial pressure), 'number densities' - calculated on the basis of a constant ℓ - can only be compared if this fact is borne in mind.

Constriction of the volume of the transverse discharge will not take place in just one dimension: although the discharge width, defined by the separation of the transverse electrodes, will remain constant, both the thickness and length of the discharge may vary (see figure 5.28(a)). The absorption measurements are affected by constriction in one dimension only (the length) because the slits at each end of the discharge ensure that the sampled 'thickness' does not change. On the other hand, the emission measurements presented here were subject to changes in both the thickness and length of the discharge: not only because volume defining slits were not used, but also because the monochromator sampled the emission (mainly) from a slice of discharge parallel to the 'thickness' dimension of the discharge (see figure 5.28(b)).

It is useful at this stage to differentiate between:

(1) Spatial constriction of the discharge which occurs at some time during the discharge excitation pulse, which will be called Temporal Volume Constriction or TVC, and,

(2) Changes in the volume occupied by the discharge - just after initiation of the discharge excitation pulse but before TVC has set in - brought about by differences in initial discharge parameters (such as gas pressure). This will be called Static Parametric Contraction or SPC.

The analogous effect to SPC, in the CW longitudinal rare-gas discharge, is the contraction of the positive column observed upon increasing the pressure (Massey and Cannon[1965]).

5.4.3 Interpretation of the parametric variation of XeI and XeII emission and absorption in the light of discharge constriction:

The dependence of XeCl^* (fig.4.12), XeI, and XeII (fig.5.16), emission on the partial pressure of HCl is now more readily understood. The time of onset of XeII emission is indicative of the onset of TVC. Thus, from figure 5.16(b), it can be concluded that HCl is almost exclusively responsible for TVC: there is no XeII emission in the absence of HCl and, as the partial pressure of HCl is increased, the time of onset of XeII emission moves steadily closer to the time of discharge initiation. Reduction in the XeCl^* and XeI emissions, voltage, and total input power, and the increase in total discharge current, all occur at the same time that XeII emission is seen to increase. The detailed reasons why XeII emission should decrease before the end of the discharge pulse can only be conjectured at the moment. It is clear, however, that once the discharge voltage has completed its transition to its lower 'running' value, the combined effects of a lower mean electron energy and the reduced discharge volume might well overcome the tendency for the (now larger) electron density to increase XeII excitation.

The investigations of the dependence of the $\text{XeI-6s}[3/2]_2$ population on HCl partial pressure (figure 5.26) and the total discharge current (figure 5.27) by absorption were carried out at a time of ~ 100 ns after discharge initiation. From the preceding discussion, it is evident from figure 5.16 that TVC has not

taken place by this time - even in gas mixtures containing up to 10 mbar HCl. Although this precaution was taken, the relative intensities of emission from the upper level do not show the same dependence on HCl partial pressure as that shown by the population of the lower level measured by absorption: absorption measurements show an initial increase in the lower level population up to 2.5 mbar HCl before a plateau is reached; on the other hand, although there is an increase in the total emission from the upper level up to 1 mbar HCl, the dominant trend is a drastic decrease in total emission as the HCl partial pressure is increased. This is in spite of the fact that, in other respects, changes in both emission from the upper and absorption by the lower level of a particular transition appear to maintain a close correspondence.[†]

The number densities of the Xenon neutral levels, in this pre-TVC regime, would be expected to increase as the HCl partial pressure is increased: a higher mean electron energy (a higher discharge voltage is observed) is required by the discharge to sustain ionization in the face of higher electron loss due to an increased rate of electron attachment to the HCl. In both emission and absorption an increase is observed initially.

Both the decrease in total emission from the $7p[5/2]_3$ level and the 'plateauing' of the $6s[3/2]_2$ number density, as measured in absorption, can be explained in terms of SPC: an increase in the HCl partial pressure reduces the volume occupied by the discharge from the moment that it is formed. The difference between absorption and emission is merely a result of the way in which each measurement was performed.

The behaviour of the $6s[3/2]_2$ number density as measured is the simplest to explain: the provision of slits made the absorption measurement, to good approximation, dependent upon the discharge length alone. The transition from a

[†] It has been found by Mr.G.J.Hirst, who has been performing further absorption measurements on the LPD laser, that there is a sharp peak in the XeI lower level population corresponding (in time) to the peak in emission from the upper level at discharge initiation: it occurs between the nominal zero time delay point and the 50 ns delay point of figure 5.23. A larger population in these levels would be expected close to discharge initiation because the discharge voltage is high at this point in time: a high E/N implies a high electron temperature and therefore a greater rate of excitation.

monotonically increasing to a constant absorption indicates the point at which SPC begins to reduce the absorption path length: that is, the point at which the discharge no longer fills the volume between each screw of the ballasted array. A plateau is reached because, although the absorption path continues to decrease as a result of SPC, the increase in number density within the constricted columnar discharges counteracts the effect of the reduced absorption path.

In the absence of HCl, the thickness of the discharge far exceeded the screw-to-screw spacing of the ballasted electrode: SPC could therefore proceed up to 2.5 mbar HCl by reducing the discharge thickness alone and avoiding detection in the absorption measurements. This was not the case in the measurements of the relative intensities of emission because, as mentioned in the preceding sub-section, this measurement was affected by changes in both the discharge thickness and the discharge length. It is contended that SPC occurs whenever HCl is present: the increase in emission at a 1 mbar HCl partial pressure is merely the result of an increased upper level density overcoming the effects of a reduced emitting volume. The continual decrease in emission above this partial pressure reflects a larger overall decrease in the emitting volume in comparison to the increase in the number density of the upper level of the particular transition.

It is now clear why there is a very critical optimum HCl partial pressure required for laser action - at least from the point of view of peak power output. A sensitive balance must be struck between:

- (1) The advantages of an increased production rate of XeCl^* through the provision of more of the halogen precursor species (HCl for the 'excited' channel and Cl^- for the ion channel) and, initially, a higher electron density and Xe^+ density as a result of volume contraction, and,
- (2) The disadvantage of a decreased XeCl^* production volume (and gain length) brought about by SPC.[†]

[†] The reader is reminded that the optimum HCl partial pressure for maximum pulse duration and energy in the LPD laser was 2.5 mbar, which value corresponds very well to the transition point in figure 5.26. It should also be noted that the output from 'conventional' short pulse duration discharge-excited XeCl^* lasers is also very sensitive to HCl partial pressure and always lies within 1 or 2 mbar of this value.

These considerations provide a plausible and simpler explanation for the critical dependence on HCl partial pressure of the laser output in short pulse duration, discharge excited systems. This critical dependence, which has been reported widely in the literature, has been attributed to, amongst other things, increased quenching of the upper laser level by HCl (Maeda et al.[1982]). In the specific case of laser pulse duration extension, a shortening of the time before onset of TVC, and therefore pulse termination, would seem to be a consequence of stronger SPC brought about by the increase in halogen donor concentration.

5.4.4 Production of XeCl^* via the ion channel vs. that via the neutral channel:

The measurements of the absolute number densities of the XeI resonance ($6s[3/2]_1$) and metastable ($6s[3/2]_2$) levels presented here cannot be used to estimate the total XeI excited state density any more accurately than was done by Kearsley[1980].

There is close agreement between the present measurements and those of Kearsley on the peak $6s[3/2]_2$ population. Figure 5.27 clearly illustrates that this population is effectively saturated with respect to the electron density. A higher population for Kearsley's device, despite its higher current density, would therefore not be expected. The resonance level population measured by Kearsley was, unaccountably, almost twice as large as his measurement of the metastable level, whereas the ratio of the two populations, as measured here, was very close to the ratio of the degeneracies of these two levels. It is possible that there was not sufficient time in Kearsley's device for these two levels to be well mixed via electron impact. Even though there is a difference in the measured resonance level population, the errors involved in the measurement of $\int k_\lambda d\lambda$ and the uncertainties in the experimental A_{21} values would make it a rather fruitless exercise to try to make a new estimate of the total excited state population.

For the purpose of estimating the possible contribution to XeCl^* production via the neutral channel (eq.1.2) in the LPD laser, there is one

significant difference to the approximations used by Kearsley; since the duration of the excitation pulse is considerably longer a large proportion of HCl molecules could be expected to be vibrationally excited. One might expect, as a consequence of this, that the true XeI metastable level could contribute to XeCl^* production. The rate of production of XeCl^* via the neutral channel,

$$\frac{d}{dt} (\text{XeCl}^*)_{\text{cr}}^{\text{n.ch.}} = k[\text{Xe}^*][\text{HCl}] \quad (5.18),$$

assuming that all excited state xenon neutrals take part and the rate coefficient for the reaction is $5.6 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ (Levin[1981]), will be

$$d/dt(\text{XeCl}^*)_{\text{cr}}^{\text{n.ch.}} = (5 \times 10^{-10}) \times (4 \times 10^{14}) \times (6.6 \times 10^{16}) = 1.3 \times 10^{22} \text{ cm}^{-3} \text{ s}^{-1}.$$

In section 5.2 it was estimated that an XeCl^* production rate of $3.2 \times 10^{22} \text{ cm}^{-3} \text{ s}^{-1}$ would be required to account for the observed photon emission. Formation of XeCl^* via the neutral channel can, therefore, only account for one third of the required XeCl^* formation - even with the most generous estimates for the precursor densities. This is entirely consistent with Kearsley's conclusions. The XeCl^* production rate estimated by Kearsley was too low because transient absorption at the laser wavelength was not taken into account. This is the reason why the XeCl^* production rates compared in table 5.3 are so similar.

There are indications from other data, however, that this estimated upper limit on the contribution made by the neutral channel is an extremely generous one:

(1) In figure 4.17, a fourfold increase in the electron density brings about a similar increase in the XeCl^* emission (the total current is increased by increasing the number of cables in the PFN). The metastable population (figure 5.27) and XeI emission (figure 5.18) show only a factor of two increase when the electron density is increased fourfold. There is no plausible mechanism by which XeCl^* production could be expected to have a super-linear dependence on the total XeI excited state population.

(2) At a total discharge current of 500 A (1-cable PFN), the XeCl^* emission shown in figure 4.17 has a definite peak at discharge initiation. This

feature becomes less pronounced as the total discharge current is increased. The metastable level (and probably the resonance level) population and that of more highly excited XeI states (as measured in emission) also show a peak at discharge initiation (see footnote on p.123). It would be reasonable to assume that the peak in XeCl^* emission which is visible for a 1-cable PFN discharge is the result of XeCl^* production via the excited channel.

If the peak referred to in (2) above is the result of metastable excitation, the fact that the XeI excited state population is approximately five times larger at this point in time than it is anywhere else would indicate a neutral channel contribution more of the order of 5%, rather than 33%, for a discharge driven by a 4-cable PFN.

No attempt will be made here to explain the cause of the broadband visible absorption (and emission) which was present in these experiments since nothing further can be added to the comments made by Kearsley[1980]. As noted in section 5.2, Mr.G.Hirst of the Clarendon Laboratory is presently investigating this phenomenon.

5.5 SUMMARY:

Three experiments were described in this chapter: time-resolved transient absorption at various UV wavelengths; the temporal and parametric evolution of emission from XeI and XeII transitions in the visible; the measurement, via absorption, of the temporal and parametric evolution of the populations of the $\text{XeI-6s}[3/2]_{1,2}$ levels and of the $\text{XeII-6s}^4\text{P}_{5/2}$ level.

The value of the peak transient absorption at the laser wavelength of 308 nm, which was arrived at by measuring time-resolved absorption at other UV laser wavelengths, was estimated to be of the order of $0.4\% \text{cm}^{-1}$. The measurements of the temporal evolution of transient absorption are consistent with the conclusion reached in chapter 4 that an increase in the density of transient absorption species cannot possibly be responsible for the termination of the LPD laser pulse. The source of the transient absorption could not be

identified.

It was concluded that the occurrence of emission from high-lying XeII transitions, at the time of termination of XeCl^* fluorescence, proved beyond doubt that temporal constriction of the discharge volume (TVC) was primarily responsible for laser pulse termination. Furthermore, it was found that the presence of HCl in the discharge is entirely responsible for the development of TVC. Examination of the dependence of XeI emission and the number density of the $\text{XeI-6s}[3/2]_1$ level on the partial pressure of HCl revealed that increasing the HCl partial pressure progressively restricts the volume occupied by the discharge from the time of discharge initiation. It is believed that this static contraction of the discharge volume (SPC) is responsible for the sensitivity of the LPD laser output to HCl partial pressure. An indication of why SPC and TVC should occur is provided in section 7.2.2.

Comparison of the measured peak population of the $\text{XeI-6s}[3/2]_2$ (metastable) level in the LPD laser with that in Kearsley's [1980] device showed that it is saturated with respect to the electron density.

It was concluded that an estimate of the total XeI excited state population on the basis of the measurements presented here would be no more accurate than the (generous) $4 \times 10^{14} \text{ cm}^{-3}$ estimated by Kearsley. A calculation of the neutral channel XeCl^* (eq.1.2) production rate, on the basis of this population, was ~ 3 times smaller than that required to account for the observed 308 nm emission. Other data indicated that the actual contribution to XeCl^* production via the neutral channel was at most $\sim 5\%$.

C H A P T E R S I X

A semi-empirical kinetic model of the LPD XeCl^{*} laser

6.1 INTRODUCTION:

Before proceeding to the discussion of specific details, it is worthwhile to outline why we were motivated to develop a numerical model of the kinetics of the LPD laser and what we expected to achieve by that effort.

The philosophy underpinning the use of computer models should be to test[†] numerically whether a particular physical theory can satisfactorily predict the observed behaviour of a complex system. Whilst some systems can be simulated adequately - perhaps even uniquely - by such codes, numerical modelling requires a knowledge of the system which may not be available in the detail required. In the specific case of the modelling of plasmas or discharges, this limitation arises because of several interrelated factors, the most fundamental of which are concerned with the assumptions that must be made regarding the electron density and the distribution of electron energies. Since electron interaction processes dominate the evolution of a discharge any assumption concerning the electrons themselves will permeate the entire kinetic model. In addition, if collisional cross sections are not known accurately,[‡] one could be justifiably surprised if an exact replica of an experimental laser output pulse could not be produced with judicious, yet reasonable, adjustment of the various rate coefficients.

At the time of writing, there have been no adequate attempts to model the discharge excited XeCl^{*} laser system. The model described recently by Maeda et al.[1982] adds little to the work of Levin et al.[1981] with respect to long

[†]They themselves should not be used as a priori evidence.

[‡]Or have to be guessed - Levin et al.[1981] include ~75 kinetic reactions in their model of an e-beam pumped XeCl^{*} system.

pulse discharge excited systems because it fails to address, amongst other things, the fundamental difference between e-beam excitation and self-sustained discharge excitation: the problem of maintaining constant the volume occupied by the discharge for the duration of the excitation pulse. Given this and the fact that Maeda et al.[1982] assume (correctly) that the dominant formation channel for XeCl^* is via the three body ion-ion recombination reaction (eq.1.1), it is not at all surprising that their predicted XeCl^* laser output should follow the evolution of the discharge current.

The aims of the kinetic model described here were, in comparison with computer models based on a solution of the steady-state Boltzmann equation, very conservative: at no point was it planned that the model should be applicable to systems other than the LPD laser. The model was developed upon semi-empirical lines: any quantity that could be measured experimentally was constrained to have that value within the model.

The most fundamental approximation was that the temporal evolution of the electron density, and through that the Xe^+ density (via an assumption of charge neutrality on a local basis), could be extracted from the experimental voltage and current waveforms via

$$n_e = \frac{I}{eAv_{\text{drift}}} \quad (5.17).$$

This assumption obviated the need both:

- (1) to have a precise knowledge of the cross sections of those processes most strongly dependent on the electron energy distribution
- (2) to include an equation to model the electrical circuit.

Leaving aside considerations of the amount of time and effort that would have been required to develop a full Boltzmann code, it was believed that constraining those quantities which were known experimentally, and consequently reducing the number of parametric 'knobs' which could be 'turned', would enable a more reliable insight into the pulse termination mechanism to be obtained.

The dynamic number densities of the various species were predicted via step-wise integration of combined production and loss rates over a time

interval, Δt , which was small in comparison to the total electrical pulse duration. The instantaneous rates of production and loss of a species, 'X', were calculated on the basis of

$$\frac{dX}{dt} = k \cdot [B] \cdot [C] \quad (6.1),$$

where 'k' is the rate coefficient for the reaction, obtained from published data, and [B] and [C] are the number densities of the reactants at the end of the previous iteration. The sum of a number of such dX/dt terms (for different production and loss reactions) multiplied by Δt gave the change in X at the time of interest.

The structure of the model was to be subjected to a continual process of evolution as our thoughts on the mechanism responsible for the premature termination of the laser pulse were modified in the light of new experimental measurements on the LPD laser. The question to be asked at each stage of development was: given what we hypothesize about the mechanisms of pulse length limitation through the available experimental evidence - mechanisms included implicitly in the model through the set of approximations chosen for it - is it possible to predict termination of the laser output and spontaneous emission at 308 nm before the end of the discharge current pulse? If the answer was 'yes', a unique solution would not necessarily have been found because it was likely that further judicious adjustment of uncertain rate coefficients etc. would provide an equally plausible prediction of laser output. If the answer was 'no', even after adjustment of the rate coefficients over the full range of plausible values had been carried out, it would be reasonably certain that a fundamental ingredient was missing from the hypothesized pulse limitation mechanism.

In summary, it is emphasized that the purpose of this model was not, and is not, to provide a complete picture of the kinetic evolution of discharge excited XeCl^* lasers. The role it played as an 'aid to thought' - in the identification of those quantities to which laser output is extremely sensitive - justifies its inclusion here.

Table 6.1
Reactions taken account of in the discharge model

			Rate or Cross Section	Reference
<u>Electron Collisions</u>				
6.2	$\text{Xe} + e$	$\rightarrow \text{Xe}^+ + 2e$	not used explicitly -	
6.3	$\text{Xe}^+ + e$	$\rightarrow \text{Xe}$	charge neutrality	
6.4	$\text{Xe}^* + e$	$\rightarrow \text{Xe}^+ + 2e$	See Table 6.2	(1)
6.5	$\text{Xe}^* + e$	$\rightarrow \text{Xe}^* + e$	See Table 6.2	(1)
6.6	$\text{Xe}^* + e$	$\rightarrow \text{Xe} + e$	See Table 6.2	(1)
6.7	$\text{HCl} + e$	$\rightarrow \text{H} + \text{Cl}^-$	$(1-6) \times 10^{-10} \text{ cm}^3/\text{s}$	(2)
6.8	$\text{Cl}^- + e$	$\rightarrow \text{Cl} + 2e$	$1 \times 10^{-18} \text{ cm}^3/\text{s}$	(3)
<u>Reformation of HCl</u>				
6.9	$\text{H} + \text{Cl}^-$	$\rightarrow \text{HCl} + e$	$9.6 \times 10^{-10} \text{ cm}^3/\text{s}$	(4)
6.10	$\text{H} + \text{Cl}$	$\rightarrow \text{HCl}$	$6.0 \times 10^{-14} \text{ cm}^3/\text{s}$	(5)
<u>XeCl* formation</u>				
6.11	$\text{Xe}^+ + \text{Cl}^- + \text{M}$	$\rightarrow \text{XeCl}^* + \text{M}$	$2 \times 10^{-6} \text{ cm}^3/\text{s}$	(6)
			(where M is a third body, this rate assumes saturation with respect to third body density)	
6.12	$\text{Xe}^* + \text{HCl}$	$\rightarrow \text{XeCl}^* + \text{H}$	$2 \times 10^{-10} \text{ cm}^3/\text{s}$	(1)
<u>Formation of other molecules</u>				
6.13	$\text{M}^+ + \text{Cl}^- + \text{Ne}$	$\rightarrow \text{MCl}^* + \text{Ne}$	As for (6.11) above	
			(where M is a +ve ion other than Xe^+)	
6.14	$\text{XeCl}^* + \text{Xe} + \text{Ne}$	$\rightarrow \text{Products}$	$1.2 \times 10^{-30} \text{ cm}^6/\text{s}$	(7)
<u>Emission and absorption at the laser wavelength</u>				
6.15	XeCl^*	$\rightarrow \text{XeCl} + h\nu$	$6 \times 10^7 \text{ s}^{-1}$	(1)
6.16	$\text{XeCl}^* + \phi$	$\rightarrow \text{XeCl} + 2\phi$	$2.5 \times 10^{-16} \text{ cm}^2$	(1)
6.17	$\text{Xe}^* + \phi$	$\rightarrow \text{Xe}^+ + e$	$6.2 \times 10^{-20} \text{ cm}^2$	(8)
6.18	$\text{Cl}^- + \phi$	$\rightarrow \text{Cl} + e$	$2.7 \times 10^{-17} \text{ cm}^2$	(9)
6.19	$\text{M} + \phi$	$\rightarrow \text{Products}$	(through experimental transient loss)	
			(where M is any other absorbing specie and ϕ is a 308nm laser photon)	
<u>Quenching of the upper laser level</u>				
6.20	$\text{XeCl}^* + e$	$\rightarrow \text{Xe} + \text{Cl} + e$	$6 \times 10^{-7} \text{ cm}^3/\text{s}$	(1)
6.21	$\text{XeCl}^* + \text{Xe}$	$\rightarrow 2\text{Xe} + \text{Cl}$	$6 \times 10^{-11} \text{ cm}^3/\text{s}$	(1 & 10)
6.22	$\text{XeCl}^* + \text{HCl}$	$\rightarrow \text{Xe} + \text{Cl} + \text{HCl}$	$1.4 \times 10^{-9} \text{ cm}^3/\text{s}$	(1 & 10)
6.23	$\text{XeCl}^* + \text{Ne}$	$\rightarrow \text{Xe} + \text{Cl} + \text{Ne}$	$1 \times 10^{-12} \text{ cm}^3/\text{s}$	(1 & 10)

References

- (1) see e.g. Levin[1981] (2) Nighan and Brown[1980] (3) Esaulov[1980]
 (4) Zwier[1980] (5) Pilling[1975](see text) (6) Flannery[1978]
 (7) Rokni[1977] (8) McCann and Flannery[1977] (9) Mandl[1976](a)
 (10) see e.g. Maeda[1982]

Numerical model of XeCl discharge: FLOW CHART

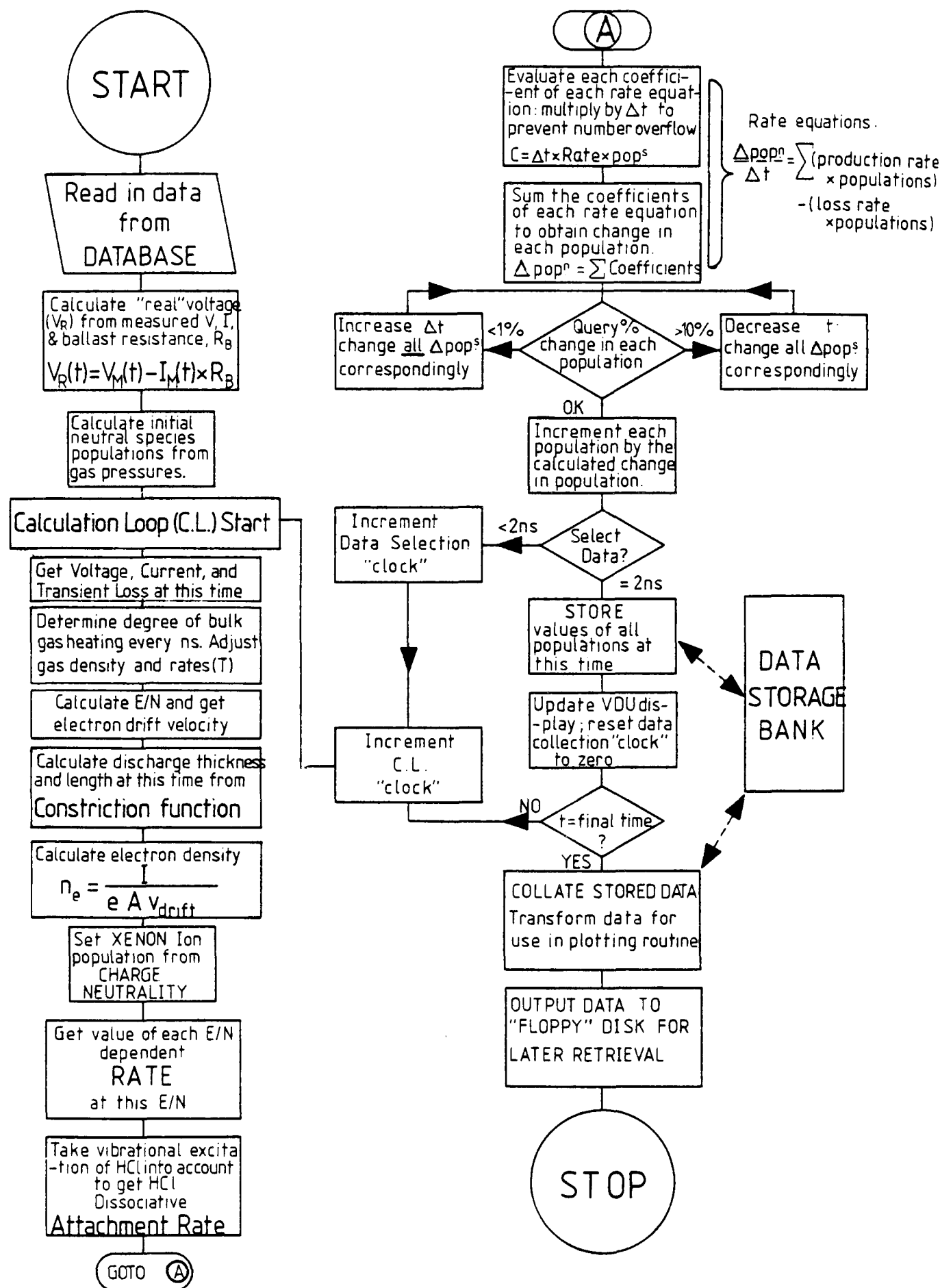


FIGURE 6.1

6.2 THE STRUCTURE OF THE MODEL:

This section provides an overall view of the structure of the model. The constraints (or approximations) initially applied to the model remained basically unaltered through its evolution - from the time of its conception to the finished version presented here. They are listed, without discussion, in the sub-section which follows. Table 6.1 contains a summary of all the reactions considered in the model together with the respective rate coefficients and cross sections used in the final version. It should be noted that, although in most cases the references cited do quote the rate coefficient used here, there are instances where, for various reasons, adjustments have been made. Where a discrepancy does seem to occur the reader is referred to body of the text for an explanation.

Immediately following subsection 6.2.1, there is a discussion of the methods by which the number densities of particular species were determined. A description of how the effects of gas heating and discharge constriction were treated is presented in subsection 6.2.3.

The computer programme itself will not be discussed in detail, though reference may be made to certain aspects of it. A logic flow chart of the programme, containing its essential details, is presented in figure 6.1. A listing of the programme (without input/output procedures), together with descriptions of the various programme variables is contained in appendix 2.

A Z80 microprocessor-based microcomputer (Research Machines(RML) type 380Z) was used to perform the calculations. The programme was written in a RML implemented version of Algol 60. Although in the initial stages of development the limited speed and accuracy of this 8-bit machine could be tolerated, these factors played a major role in restricting the degree to which the model could be evolved.

6.2.1 Initial approximations:

(1) E/N as a function of time was determined by assuming a uniform electric field across the discharge gap i.e. $E(t) = V(t)/d$, where 'V' was the measured discharge voltage as a function of time and 'd' was the anode-cathode

separation. The electron transport properties of the buffer gas were assumed to be dominant (see also inclusion of bulk gas heating).

- (2) The electron drift velocity as a function of time was determined using published data of v_{drift} vs. E/N (see figure 4.15) for Neon.
- (3) The electron density was constrained via eq.5.17 using the experimental value of the discharge current at the time of interest, the electron drift velocity corresponding to the measured discharge voltage (see (2) above), and the time-dependent cross-sectional area for current flow (see notes on inclusion of constriction below).
- (4) The Xenon ion density was obtained by introducing the constraint of local charge neutrality, i.e. $[Xe^+] = n_e + [Cl^-]$ (see note below on other positive ions).
- (5) Heavy species ($Xe_2, Cl_2, H_2, Xe_2^*, Xe_2^+$ etc.) or ions other than Xe^+ and Cl^- were not included explicitly, in certain cases such formation was included implicitly by adjustment of rate coefficients.
- (6) Loss of distributed photon density due to transient absorptions was included as experimental data in the photon density rate equation. However, photodetachment of Cl^- and photoionisation of Xe^* were included as loss terms in their respective rate equations using published data for the cross sections for these reactions.
- (7) No explicit account was taken of differences between the different excited states of Xenon neutral atoms.
- (8) All $XeCl^*$ production was assumed to result in B-state molecules. The ground state $XeCl$ population was assumed to be so small (due to fast collisional dissociation) that it could be ignored.
- (9) The starting point in time for the model was that at which the voltage across the discharge gap peaked. All transient species were assumed to have a zero population at this time. The effect of preionization was only included in terms of the initial density of electrons available at the time of initial breakdown (guessed to be of the order of 10^9 cm^{-3} - but the predictions of the model were not sensitive to this value).

6.2.2 Calculation of the various populations:

(a) General:

All calculations were performed by considering a 'typical' 1cm^3 volume. The integration time step, Δt , was varied in such a way that, during a particular iteration, the percentage change in the number density of the most rapidly changing species (whether positive or negative) with respect to that of the previous iteration was never greater than 10% nor less than 1% (except in the first nanosecond where Δt was set at 1 ps).

The number densities of the following species were determined as a function of time: Ne, Xe, Xe^* , Xe^+ , n_e , HCl, Cl, H, Cl^- , XeCl^* . In the initial stages of the development of the model, the number density of neon was held constant, later it was given a functional dependence on the gas temperature, as were all other species, to take account of localised gas expansion. The number densities of Xe and HCl were determined at the beginning of each iteration loop by subtracting from their initial density the number densities of all Xe and Cl containing transient species respectively:

$$[\text{Xe}](t) = [\text{Xe}](0) - [\text{Xe}^+] - [\text{Xe}^*] - [\text{XeCl}^*] \quad (6.24);$$

$$[\text{HCl}](t) = [\text{HCl}](0) - [\text{Cl}^-] - [\text{Cl}] - [\text{XeCl}^*] \quad (6.25).$$

(b) Xenon ions:

Excitation of xenon ions was ignored. The xenon ion density was determined at the beginning of each iteration loop by demanding charge neutrality on a local basis,

$$[\text{Xe}^+] = f (n_e + [\text{Cl}^-]) \quad (6.26),$$

where 'f' is a number between 0 and 1. The adjustable fraction, 'f', was included so that the effect of changing the proportion of positive ions other than Xe^+ could be monitored. Usually, however, 'f' was chosen to be equal to one.[†] When 'f' was chosen to be less than one, the other positive ions, M^+ , which were therefore implied to be present, and whose number density was given by:

[†] As was discussed in section 4.6.2, this is reasonable when one considers that xenon has the lowest ionization potential of any of the species within the discharge.

$$[M^+] = (1-f)(n_e + [Cl^-]) \quad (6.27),$$

were assumed to take part in ion-ion recombination (eq.6.13) with Cl^- . The rate coefficient for formation of these (unidentified) molecular species was taken as being the same as that for $XeCl^*$ formation via eq.6.11.

Although a considerable population of Xe_2^+ might arise during the course of an excitation pulse, it was assumed - as is done by both Levin[1981] and Maeda[1982] - that this population would also give rise to $XeCl^*$ formation via an ion-ion recombination reaction with a similar rate coefficient to that for the 'normal' ion channel (eq.6.11). This is not an entirely satisfactory assumption because the reaction does not require a third-body (the extra xenon atom would take away the excess energy) and, consequently, the reaction rate coefficient could well be significantly higher†

Any contribution to the transient absorption at the laser wavelength - due to the presence of Xe_2^+ or for that matter other heavy species - is taken account of by using experimentally measured values of the absorption in the photon density equation.

(c) Xenon excited state atoms:

The number density of xenon excited state atoms was determined using the rate equation:

$$\begin{aligned} d/dt(Xe^*) = K_1.[Xe].n_e - (K_2.[Xe^*].n_e + K_3[Xe^*].n_e + K_4.[Xe^*].[photons] \\ + K_5.[Xe^*].[HCl]) \end{aligned} \quad (6.28).$$

K_1 , K_2 , and K_3 are the E/N dependent rate coefficients for electron impact excitation to Xe^* (eq.6.4), de-excitation from Xe^* (eq.6.5), and ionization from Xe^* (eq.6.6), respectively. These rates were determined as a function of E/N in the manner described in table 6.2. K_4 is the rate coefficient for photoionization of Xe^* (eq.6.17) which is the product of the cross-section for the reaction and the speed of light. K_5 is the rate coefficient for formation of $XeCl^*$ via the neutral channel (eq.1.2 now renumbered to eq.6.12). Since no

† Fortunately, the rate of production of $XeCl^*$ via reactions requiring the participation of Cl^- is almost totally independent of their rate coefficients under a large range of conditions - a conclusion arrived at by Kearsley[1980]. This point will be discussed in greater detail below.

differentiation was made between degrees of excitation of neutral xenon, and because the density of vibrationally excited HCl was not calculated explicitly, an 'average' rate coefficient for XeCl^* production via the various neutral channels (see eg. Levin[1982] Table IV 'neutral formation') was used. Even if the largest rate coefficient quoted by Levin[1982] had been used, formation of XeCl^* through this channel would have been small in comparison to formation via the ion channel.

Table 6.2
E/N Dependent Rate Coefficients

T_e (Helium) (eV)	E/N (V-cm ² x 10 ⁻¹⁷)	$\text{Xe} + e \rightarrow \text{Xe}^*$ (cm ³ /s)	$\text{Xe}^* + e \rightarrow \text{Xe}$ (cm ³ /s)	$\text{Xe}^* + e \rightarrow \text{Xe}^+$ (cm ³ /s)
		(A)	(B)	(C)
	0	1 x 10 ⁻¹³	1 x 10 ⁻¹⁰	1 x 10 ⁻¹²
1	3.5	3 x 10 ⁻¹²	4 x 10 ⁻⁹	1.6 x 10 ⁻⁹
3	7	8 x 10 ⁻¹¹	8 x 10 ⁻⁹	8.5 x 10 ⁻⁹
4	10	1.5 x 10 ⁻¹⁰	9 x 10 ⁻⁹	1.4 x 10 ⁻⁸
6	15	8 x 10 ⁻¹⁰	1 x 10 ⁻⁸	2 x 10 ⁻⁸
8	25	8 x 10 ⁻¹⁰	1 x 10 ⁻⁸	2 x 10 ⁻⁸

These rates were calculated using the expressions given by Levin[1981]:

(A) Rate = 1.20 x 10⁻⁸ x T_e^{0.72} x exp(-8.36/T_e)

(B) Rate = 3.47 x 10⁻⁸ x T_e^{0.72} x exp(-12.13/T_e)

(C) Rate = 2.85 x 10⁻⁸ x T_e^{0.71} x exp(-3.27/T_e)

E/N was related to the electron temperature, T_e, using the data given by McDaniel[1976] (II pp.717) for helium.

(d) Chlorine ions (Cl⁻):

The number density of Cl⁻ was determined using the rate equation:

d/dt(Cl⁻) = K₆(t) · [HCl] · n_e - K₇ · [Cl⁻] · [Xe⁺] - K₇ · [Cl⁻] · [M⁺]

-K₈ · [Cl⁻] · n_e - K₉ · [Cl⁻] · [H] - K₁₀ · [Cl⁻] · [photons]

(6.29):

K₆ is the rate coefficient for dissociative electron attachment to HCl (eq.6.7); K₇ is the rate of formation of XeCl^* via the 'ion' channel (eq.6.11); M⁺ is the number density of positive ions which are not xenon (eq.6.13: see (b) above); K₈ is the rate coefficient for electron detachment from Cl⁻ via electron impact (eq.6.8); K₉ is the rate coefficient for reformation of HCl from Cl⁻ and H

(eq.6.9); K_{10} is the rate coefficient for photodetachment of an electron from Cl^- (eq.6.18).

A crucial shortcoming of this model was the manner in which the rate coefficient for dissociative electron attachment to HCl was treated. There has been considerable discussion in the literature as to the degree of enhancement of this rate coefficient resulting from the production of vibrationally excited HCl. Measurements by Allan and Wong[1981] indicate that the rate coefficient for $\text{HCl}(v=1)$ is some 38 times larger than for $\text{HCl}(v=0)$. Nighan and Brown[1980] were the first to propose that vibrational excitation of HCl plays a major role in enhancing the formation of $\text{XeCl}^*(\text{B-state})$ via the ion channel through increased Cl^- formation. Levin et al.[1981] agreed with this interpretation on the basis of predictions by their computer model. Recent measurements of vibrational excitation of HCl in conditions typical (sic) of e-beam pumped XeCl^* lasers by Center et al.[1982] contradict this conclusion however. Center et al. propose that the maximum 10% excitation to $\text{HCl}(v=1)$ that they observe is far too small to justify the tenfold increase in the rate coefficient predicted both by Nighan and Brown[1980] and by Levin et al.[1981]. Maeda et al.[1982] include vibrational excitation of HCl in their rate equation system but appear to prefer not to discuss what effect it has on the rate of production of Cl^- .

A calculation of the functional dependence of the rate of dissociative electron attachment to HCl on the mean electron energy (and therefore E/N), which was presented by Kearsley[1980], indicated that in the region of interest (1-3eV) the change in the rate coefficient is small. The dependence of this rate on E/N was therefore ignored in the model presented here.

Allowance was made for the growth of a $\text{HCl}(v>0)$ population by forcing the net rate coefficient to increase linearly from the $\text{HCl}(v=0)$ value at the time of discharge initiation for a specified time interval, $\Delta\tau$ (usually 50 ns); at the end of this time interval, it was assumed that the ratio of $[\text{HCl}(v=0)]:[\text{HCl}(v>0)]$ would have stabilized to some equilibrium value. This assumption was implemented by holding the net rate coefficient constant at some

multiple, ' $1+m$ ', of that for $\text{HCl}(v=0)$, i.e.

$$K_6 = \text{Coeff}(\text{HCl}(v=0)) \text{ at } t=0,$$

$$K_6 = \text{Coeff}(\text{HCl}(v=0))(1 + (t \times m)/\Delta\tau) \text{ for } 0 < t < \Delta\tau,$$

$$K_6 = \text{Coeff}(\text{HCl}(v=0))(1+m) \text{ for } t \geq \Delta\tau$$

An value of ' m ' equal to 6 was used in the final version of the model; this was equivalent to assuming that $\text{HCl}(v>0)$ made up some 16% of the total HCl population. In comparison to other models this is quite a modest estimate of the total vibrational excitation.

Four Cl^- loss processes were included in the model: electron detachment via electron impact or photoabsorption, recombination with H to reform HCl , and recombination with positive ions (usually Xenon see (b) above).

Rate coefficients for electron detachment from Cl^- via impact with another species are small in comparison to the other loss processes as a result of the large threshold energy required (~ 3.6 eV). Mandl[1976](a) and Champion and Doverspike[1976](a) have both reported measurements of rates of electron detachment from Cl^- via impact with rare gas atoms. Although they disagree with each other as to the interpretation of their results (Champion and Doverspike[1976](b) and Mandl[1976](b)), even the largest rates quoted are too small to be concerned with here. Peart et al.[1976] have reported measurements of electron impact detachment cross sections for a number of electronegative species including F^- . Esaulov[1980] shows that Peart's measurements can be described with a simple classical scaling law. Since F^- and Cl^- have similar electron affinities it was assumed that their respective cross sections for this reaction would also be similar. Any estimate of a cross section for this reaction, however, had to take account of the fact that, in the LPD laser, the mean electron energy would be well below the threshold for electron impact detachment. An estimated rate coefficient of $10^{-18} \text{ cm}^3 \text{ s}^{-1}$ was used.

Photodetachment of Cl^- was discussed in chapter 5. The rate coefficient for reformation of HCl from H and Cl^- is very fast (Zwiler[1980]). Since this reaction does not result in the loss of HCl from the system, as is effectively the case in other Cl^- loss processes, the effect of this reaction will merely be

to slow the net production rate of Cl^- from HCl .

(e) XeCl:

Ground state XeCl was assumed to dissociate so rapidly that its population could be ignored. All excited state XeCl produced was assumed to be channelled into the B-state (upper level of the UV (308nm) laser transition). The number density of XeCl^* was determined with the rate equation:

$$\begin{aligned} d/dt(\text{XeCl}^*) = & K_7[\text{Cl}^-][\text{Xe}^+] + K_5[\text{Xe}^*][\text{HCl}] - (\sigma \cdot c \cdot \ell_g / \ell_c) [\text{photons}][\text{XeCl}^*] \\ & - A[\text{XeCl}^*] - K_{11}[\text{XeCl}^*][\text{Xe}] - K_{12}[\text{XeCl}^*][\text{Ne}] - K_{13}[\text{XeCl}^*][\text{HCl}] \\ & - K_{14}[\text{XeCl}^*] \cdot n_e - K_{15}[\text{XeCl}^*][\text{Ne}][\text{Xe}] \quad (6.30) \end{aligned}$$

K_7 is the (saturated) three-body ion-ion recombination coefficient (eq.6.11); K_5 is the excited channel rate coefficient (eq.6.12); A is the lifetime of the $\text{XeCl}(\text{B-state})$ (eq.6.15); σ is the stimulated optical cross section, 'c' is the speed of light, and ℓ_g and ℓ_c are the gain and cavity lengths respectively (eq.6.16: see notes on constriction below); [photons] is the stimulated photon density distributed through the cavity; K_{11} , K_{12} , K_{13} , and K_{14} are the rate coefficients for quenching of XeCl^* by xenon (eq.6.21), neon (eq.6.23), HCl (eq.6.22), and electrons (eq.6.20), respectively. K_{15} is the rate coefficient for three-body quenching processes (eq.6.14): Xe_2Cl^* formation for example.

Except for quenching of XeCl^* by electrons, the rate coefficients K_{11} - K_{14} were taken directly from either Levin[1981] or Maeda[1982]. The reader is referred to these two papers for the original sources. The coefficient for quenching by electrons was increased by a factor of two over that quoted by Levin[1982]. The reason for this will be discussed in section 6.4 below.

The rates used for the three body ion-ion recombination reaction (eq.6.11) were effective two body rates, i.e. it was assumed that the number density of the buffer was effectively constant for the duration of the electrical pulse. Although, as is discussed below, a reduction in the buffer gas density of ~12% may be expected when localised gas heating is taken into account, the virtual independence of the XeCl^* production rate on the rate coefficient in the regime of interest did not make it worthwhile to revert to the use of three body rates

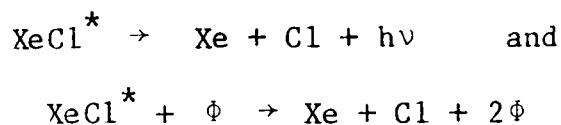
in the later stages of development of the code.

(f) Neutral chlorine:

The neutral chlorine number density was monitored via the rate equation:

$$\begin{aligned} d/dt(\text{Cl}) = & A \cdot [\text{XeCl}^*] + (\sigma \cdot c \cdot \ell_g / \ell_c) [\text{photons}] [\text{XeCl}^*] + K_{10} [\text{Cl}^-] [\text{photons}] \\ & + [\text{XeCl}^*] (K_{11} [\text{Xe}] + K_{12} [\text{Ne}] + K_{13} [\text{HCl}] + K_{14} n_e + K_{15} [\text{Ne}] [\text{Xe}]) \\ & K_8 [\text{Cl}^-] n_e + K_7 (1-f) (n_e + [\text{Cl}^-]) [\text{Cl}^-] - K_{16} [\text{H}] [\text{Cl}] \quad (6.31). \end{aligned}$$

K_{16} is the rate coefficient for reformation of HCl from H and Cl which was taken to be four orders of magnitude smaller than that for reformation of HCl from H and Cl^- . This is a very generous overestimate. The only data which could be found on reformation of HCl from neutrals dealt with diatomic-atomic reactions (e.g. $\text{Cl}_2 + \text{H}$ or $\text{H}_2 + \text{Cl}$). One could expect these reactions to be faster than the atomic-atomic reaction (eq.6.10) since the atomic-atomic reaction will require the presence of a third body. Pilling[1975] gives a rate for $\text{Cl} + \text{H}_2$ forming HCl of $\sim 6 \times 10^{-17} \text{ cm}^3 \text{ s}^{-1}$. A value of $6 \times 10^{-14} \text{ cm}^3 \text{ s}^{-1}$ was used here to illustrate that reformation of HCl is insignificant - even when the HCl reformation rate is increased by three orders of magnitude above that for the more likely diatomic-atomic reaction. The first eight terms of eq.6.31 are the loss terms for XeCl^* in eq.6.30. The term containing 'f' results from ion-ion recombination of Cl^- with positive ions other than xenon (see (b) above). The inclusion of the spontaneous and stimulated emission terms in eq.6.31 indicates that equations 6.15 and 6.16 were not used explicitly, rather the overall reactions including the consequent dissociation of XeCl were used:



(g) Laser photon density:

The density of stimulated laser photons, distributed through the laser cavity, was calculated via the rate equation:

$$\begin{aligned} d/dt(\text{photons}) = & k \cdot A \cdot [\text{XeCl}^*] + (\sigma \cdot c \cdot \ell_g / \ell_c) [\text{photons}] [\text{XeCl}^*] \\ & - (c/2\ell_c) \cdot \ln(1/R) \cdot [\text{photons}] - (c \cdot \ell_g / \ell_c) \cdot \gamma(t) \cdot [\text{photons}] \quad (6.32). \end{aligned}$$

The first term represents the fraction of spontaneously emitted 308 nm photons which have directions within the solid angle of the laser cavity, where k is a

fraction which was estimated[†] as 1.5×10^{-4} . Stimulated emission is taken account of by the second term in eq.6.32 where σ , c , ℓ_g , and ℓ_c , have the same meaning as in (e) above (ℓ_g was time-dependent when constriction was included - see below). Laser output is a loss term from the intra-cavity photon flux and is included via the third term in eq.6.32, where R is the reflectivity (96%) of the output coupling mirror; the factor of one half in this term takes account of the fact that only those photons travelling towards the output coupling mirror are available for output (see eg.Yariv[1971] pp.98). No account was taken of losses due to diffraction or absorption by the dielectric mirror coatings. The mirror at the non-output end of the cavity was assumed to have a reflectivity of 100% (the actual value of its reflectivity was not so different as to make any significant difference to the results). The final term in equation 6.32 takes account of all transient absorption in the laser cavity: $\gamma(t)$ is the transient loss cm^{-1} within the gain region and its value was determined using the experimental results presented in chapter 5 (see figure 5.7). The fraction ℓ_g/ℓ_c converts the loss term into one which is evenly distributed through the cavity rather than concentrated in the discharge region. The value of ℓ_g in the transient loss term was maintained even during the modelling of constriction since the effect of discharge constriction was already implicit in the experimental values of $\gamma(t)$ used.

The laser output power as a function of time was determined using the following equation:

$$\text{Power}(t) = \frac{1}{2} \cdot \frac{h \cdot c}{\lambda} \cdot c \cdot \ln(1/R) \cdot (w_{\text{disch}} \cdot t_{\text{disch}}(t)) \cdot [\text{photons}(t)] \quad (6.33),$$

where 'h' is Planck's constant, c is the speed of light, λ is the laser wavelength (308 nm), w_{disch} is the discharge width, and t_{disch} is the time-dependent discharge thickness. The symbol $[\text{photons}(t)]$ represents the instantaneous photon density distributed through the cavity, changes in which are determined via eq.6.32.

[†] Again this is a value which does not affect the result of the calculation in any significant way.

6.2.3 Gas Heating:

The level of specific power input to the gas volume in the LPD laser, though perhaps an order of magnitude smaller than typical of short pulse duration discharge excited RGH lasers, was still large enough for gas heating effects to be considered. Localised expansion of the gas, with a consequent decrease in the total gas number density, and the dependence of the ion channel (eq.6.11) rate coefficient on temperature (see chapter 1) were taken into account.

The change in temperature, ΔT , for a specific input energy, ΔQ , in a 1 cm^3 volume was calculated via:

$$\Delta T = \frac{A_a}{N \cdot c_v} \cdot \Delta Q \quad (6.34),$$

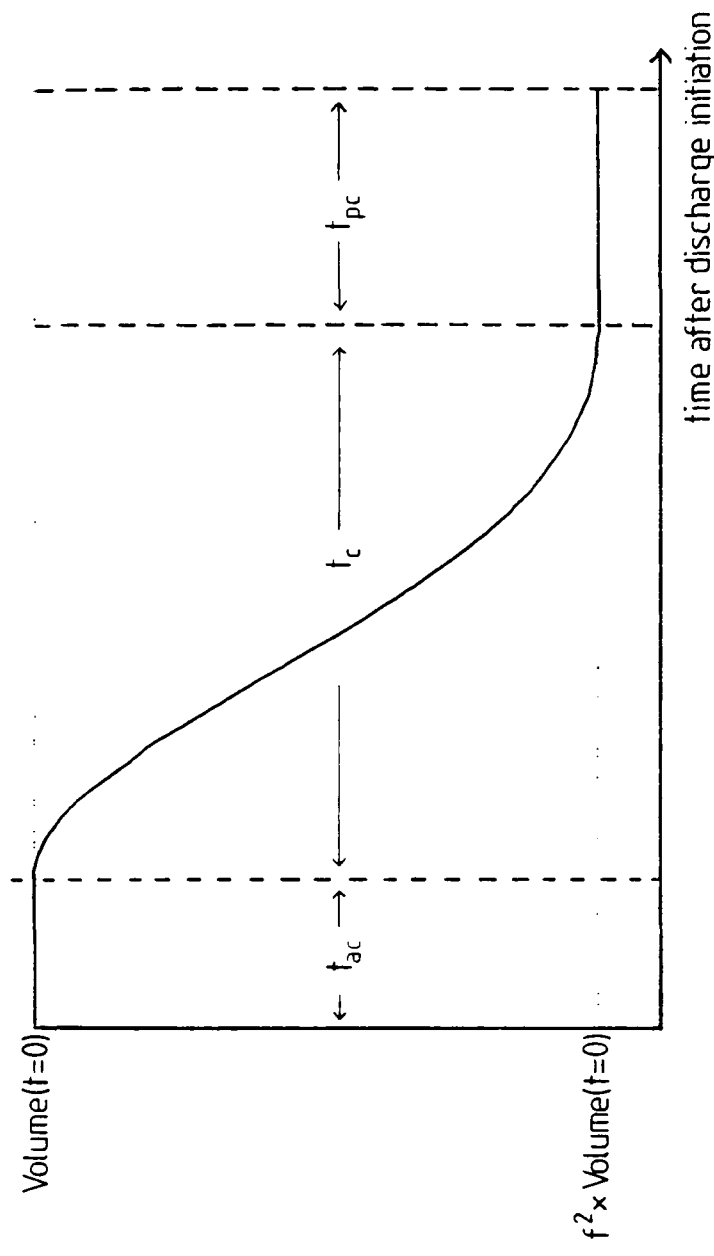
where A_a is Avagadro's number, N is the total gas number density at this time, and c_v is the molar specific heat (at constant volume) of an ideal monatomic gas. The specific input energy over a time interval Δt was calculated using:

$$\Delta Q = f \cdot \frac{V(t) \cdot I(t) \cdot \Delta t}{\text{volume}} \quad (6.35),$$

where $V(t)$ and $I(t)$ are the instantaneous experimental discharge voltage and current respectively and 'f' is the fraction of the instantaneous input power which is taken to contribute to gas heating. When constriction was included in the model this volume was time dependent; the effective increase in specific energy input as a consequence of constriction resulted in a prediction of a faster rise in temperature. It was assumed that ~95% of the total input power would result in heating of the bulk gas, i.e. $f = 0.95$.

Although the rise in temperature of the bulk gas within the discharge volume was assumed to occur instantaneously, expansion of the gas was taken to proceed at a rate consistent with an expansion velocity lower than the speed of sound. As was the case for the calculations of the various populations, the problem was treated on a constant volume (1 cm^3) basis. The ambient pressure was assumed to be constant: a increase in temperature would therefore give rise to a decrease in the total number density within the 'typical' volume. The ratio of

Constriction in the computer model



$$\text{Volume}(t) = \text{Thickness}(t) \times \text{Length}(t) \times \text{Width}(t \text{ independent})$$

For $\text{time} < t_{ac}$: $\text{thickness}(t) = \text{thickness}(t=0)$; $\text{length}(t) = \text{length}(t=0)$.

For $\text{time} > t_{ac} + t_c$: $\text{thickness}(t) = f \times \text{thickness}(t=0)$; $\text{length}(t) = f \times \text{length}(t=0)$,

For $t < \text{time} < t_{ac} + t_c$: $\text{thickness}(t) = A(f, t) \times \text{thickness}(t=0)$; $\text{length}(t) = A(f, t) \times \text{length}(t=0)$,

where $0 < f < 1$ and

$$A(f, t) = 1 - (1 - f) \times \sin^2\left(\frac{\pi}{2} \cdot \frac{(\text{time} - t_{ac})}{t_c}\right)$$

Figure 62

the total number density at some time, $N(t)$, to that at discharge initiation, $N(0)$, was determined using the following relation:

$$\frac{N(t)}{N(0)} = \frac{T(0)}{T(t)} + (1 - \frac{T(0)}{T(t)}) \exp(\frac{t}{\psi}) \quad (6.36),$$

where $T(t)$ and $T(0)$ are the temperatures of the gas at time 't' and at discharge initiation, respectively, and ψ is the time by which the gas is assumed to have reached its fully relaxed density (taken as 1 μ s). All species were assumed to have identical rates of expansion and were thus all multiplied by the fraction calculated in eq.6.36. The temperature dependence of the ion channel rate coefficient was a parameter which was changed between 'runs' of the model to gauge its effect.

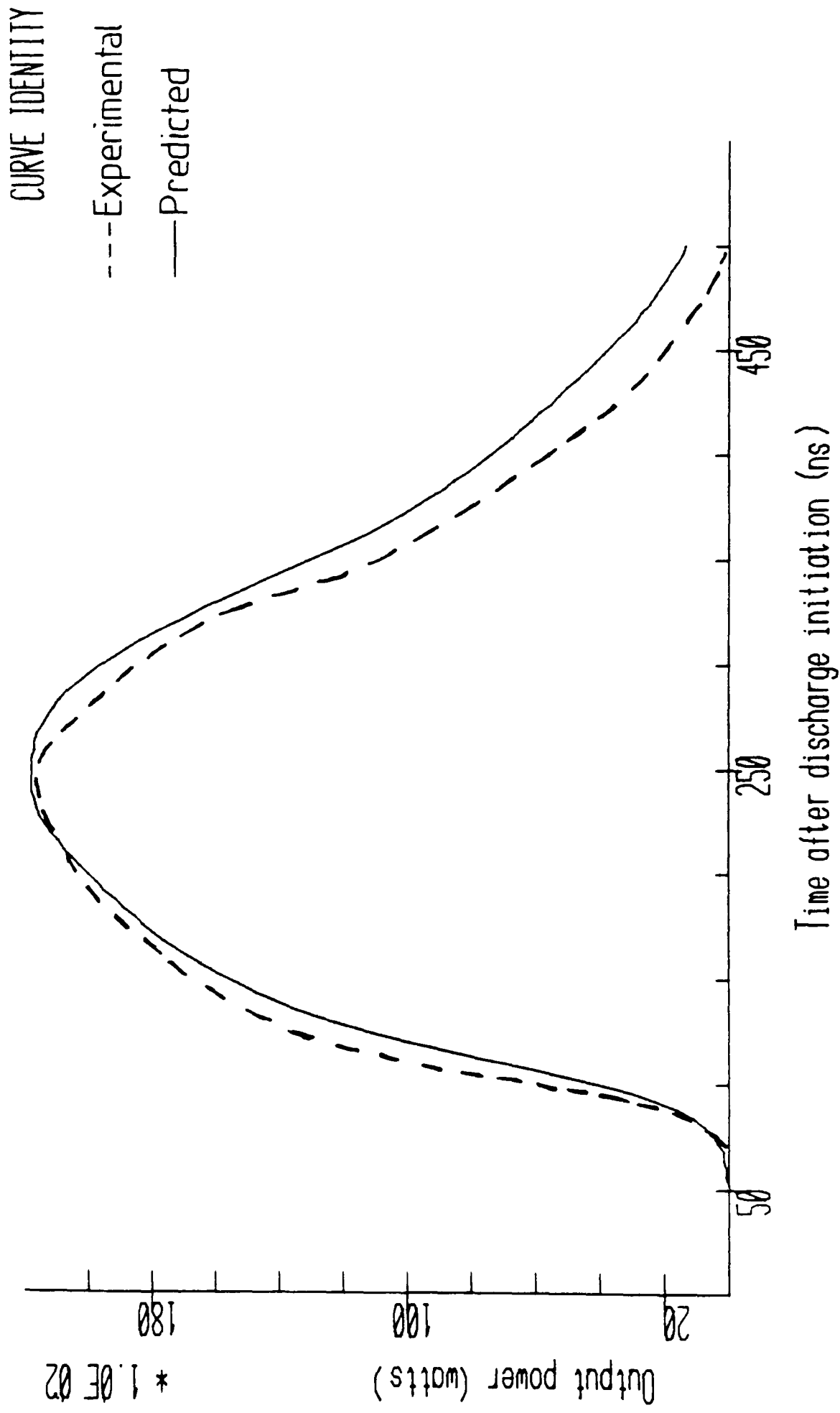
6.2.4 Temporal Volume Constriction:

Temporal constriction of the discharge volume was included in the model in the manner indicated in figure 6.2. The electrical pulse was divided into three temporal blocks: an ante-constriction phase, a constriction phase, and a post-constriction phase of durations t_{ac} , t_c , and t_{pc} , respectively. During the ante-constriction phase the discharge volume maintained its initial value (discharge length of 38 cm, discharge width of 2.2 cm and discharge thickness of 3 mm). In the post constriction phase the volume was taken to be some fraction, f^2 , of its initial value (usually 0.1). The volume was given a $\sin^4$ dependence in the constriction phase (see equation for $A(f,t)$ in fig.6.2) so that a relatively smooth transition would result: the actual choice of the functional dependence was quite arbitrary.

The lack of precise experimental data on the temporal evolution of the discharge volume meant that the values of the variables t_{ac} , t_c , t_{pc} , and 'f', had to be guessed. Plates 4.7 and 4.9 did, however, provide a reasonable indication of the values which should be assigned. Needless to say, many different values of these parameters were tried in the model in order to gauge their effect.

The effects of the constriction function pervaded the entire model. At the base level, it had a profound effect on the calculated electron density by decreasing the cross sectional area available for current flow ('A' in

FIGURE 6.3: Laser output - experimental and predicted



eq.5.17)). All those production and loss rates dependent upon the electron density were consequently altered. The predicted laser output power was affected not only by the reduction in stimulated emission (as a result of a shorter gain length) but also by the reduction in the cross sectional area of the output beam ($w_{\text{disch}} \times t_{\text{disch}}$ in eq.6.33). The bulk temperature of the gas was also closely linked to the constriction function because of the increase forced upon the specific input power, ΔQ .

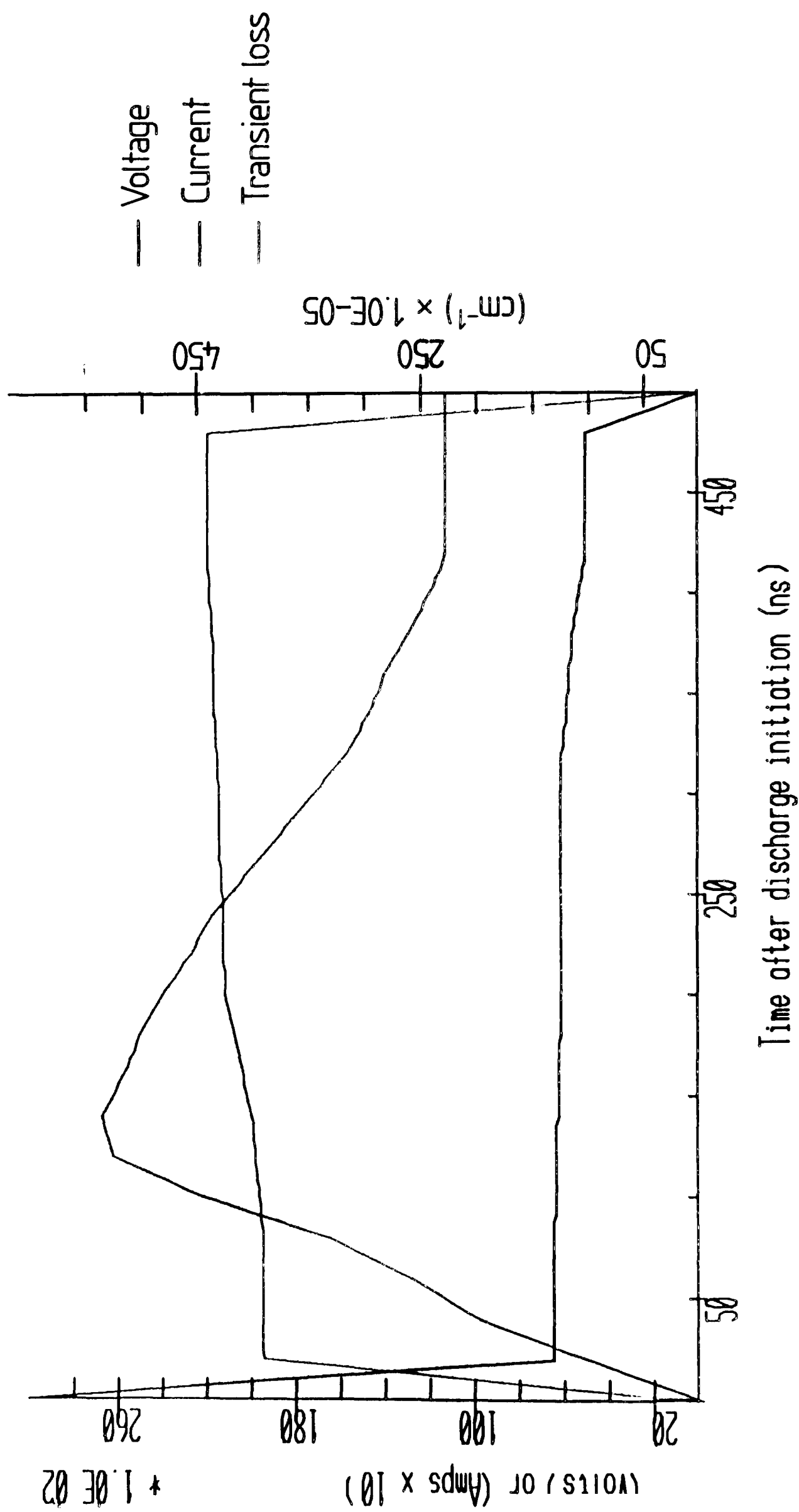
6.3 RESULTS:

The results from only a small number of the total number of 'runs' performed with the model will be presented here. As a starting point, the results from the 'run' which produced a predicted laser output curve most like the experimental curves actually observed will be presented. Following this, various other results, obtained by changing the value of just one parameter from its value in the 'ideal' case, will be presented.

Table 6.3
Parameter values used to obtain best fit to experimental laser output

<u>Quantity</u>	<u>Value</u>	<u>Equations</u>
ℓ_c , length of the optical cavity	62 cm	6.30-6.33
ℓ_g , length of discharge region (gain length)	38 cm	6.30-6.33
w_{disch} , the discharge gap	2.2 cm	5.16, 6.33
t_{disch} , the discharge thickness	3 mm	5.16, 6.33
t_{ac} , duration of ante-constriction discharge	75 ns	fig.6.2
t_c , duration of constriction phase	300 ns	fig.6.2
$f^c \times f$, the fraction of initial discharge volume (post constriction)	0.1	fig.6.2
$T(0)$, the gas temperature at discharge initiation	300 K	6.36
f , the fraction of total energy input which results in gas heating	0.95	6.35
Ψ , the relaxation time for full expansion of the bulk gas because of heating	1 μ s	6.36
Dependence of the three-body ion-ion recombination rate coefficient on temperature	T^{-1}	6.31
R , the reflectivity of the output coupling mirror	0.96	6.32, 6.33
k , the fraction of Spont. Emission along optic axis	1.5×10^{-4}	6.32
The initial total gas pressure	2.33 atm.	
The initial partial pressure of Xenon	25 mbar	
The initial partial pressure of HCl	2.5 mbar	
The initial electron density (preionisation - guess)	$5 \times 10^9 \text{ cm}^{-3}$	
The time after discharge initiation at which electron dissociative attachment peaks	50 ns	6.29
m , the multiplication factor for the initial electron dissociative attachment rate coefficient	5	6.29
f , the fraction of positive ions due to Xenon	1	6.26, 6.27

FIGURE 6.4: Experimental data input - voltage, current and transient loss



6.3.1 'Best fit' predictions:

Figure 6.3 shows a 'best' prediction laser output curve from the model (solid line) together with a typical experimental curve (dashed line: taken from the neon laser mix curve in plate 4.4). The values of those parameters not shown (or not explicit) in tables 6.1 and 6.2 are presented in table 6.3.

Figure 6.4 contains the three quantities input as experimental data: the discharge voltage, the discharge current, and the transient loss. The shape of the transient loss curve is the same as that for loss at 193 nm (figure 5.7) but the actual values used are consistent with the level of absorption at 249 nm. The reason for this was that the larger total absorption at 193 nm was thought to provide better signal-to-noise quality than the absorption measured at longer wavelengths. The somewhat arbitrary adjustment of the transient loss (within the range available via experimental error) will be discussed further below. It should be noted, however, that the predicted laser power was extremely sensitive to adjustment of the transient loss: an alteration of $0.1\% \text{cm}^{-1}$ could (depending on the point in time at which the change was made) produce a change of $\sim 25\%$ in the predicted instantaneous power output.

Figures 6.5-6.8 show the temporal evolution of the various neutral species (fig.6.5), electrons, ions and excited state species (fig.6.6), the active volume, E/N ($1 \text{ Td (Townsend)} = 10^{-17} \text{ Vcm}^2$), the bulk gas temperature, and the three body ion-ion recombination rate coefficient (fig.6.7), and the total number of spontaneous emissions per second into 4π at the laser wavelength in the presence and absence of lasing (fig.6.8). To obtain the correct value of any of these quantities divide the axis scale by the scaling factor shown for each parameter in the legends.

The reduction in the neon and xenon number densities (fig.6.5) with time is due entirely to the inclusion of the (guessed) gas expansion function resulting from localised heating of the gas. The number density after 500 ns is some 70% of the initial value. The reduction in density of HCl over the duration of the excitation pulse is far greater than this - the final density is only some 25% of the initial density within the active discharge volume.

FIGURE 6.5: Predicted temporal evolution of neutral species

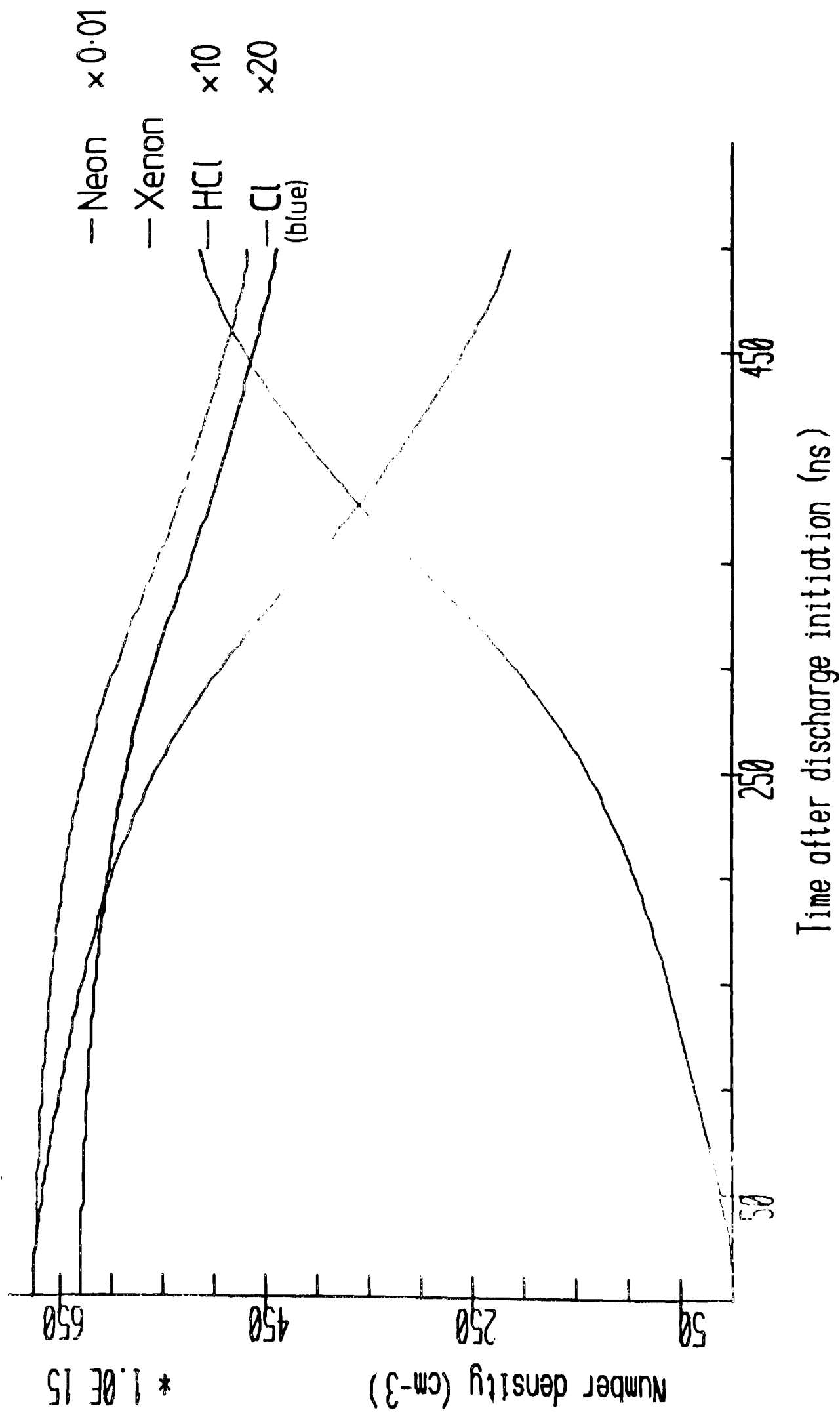


FIGURE 6.6: Predicted number densities of ions and excited state species

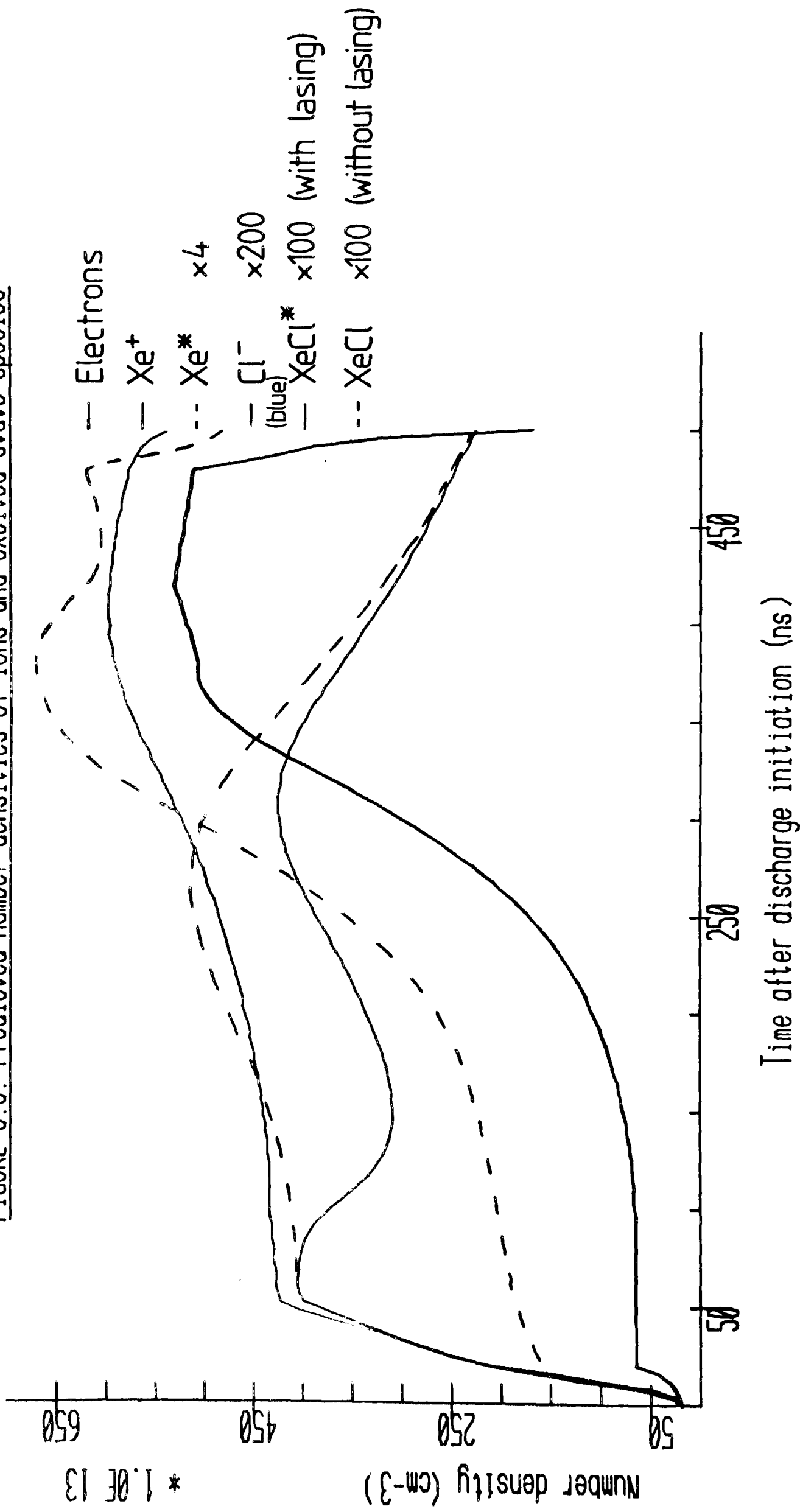
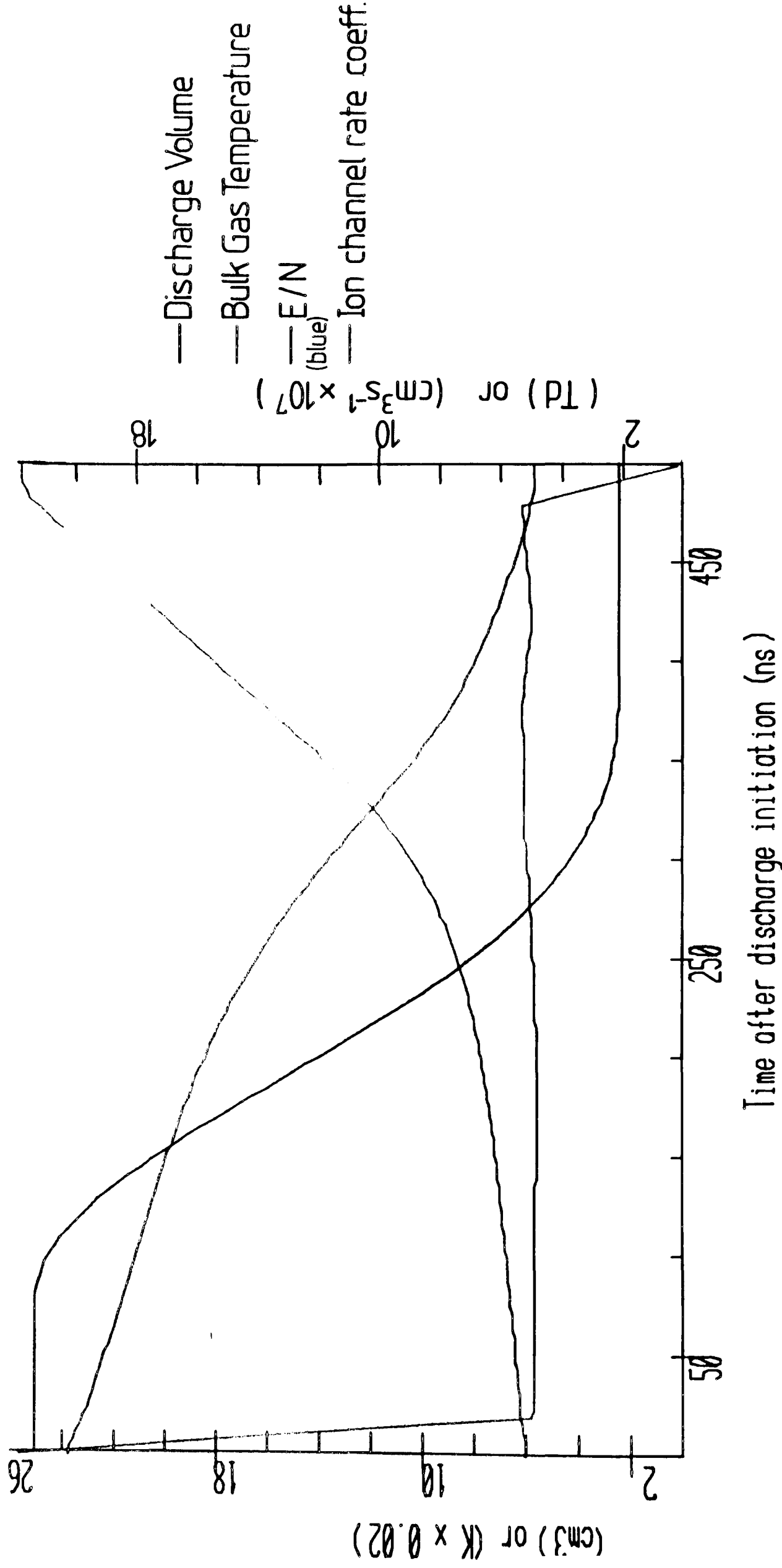


FIGURE 6.7: Temporal evolution of various quantities in the 'best fit' case



The electron and Xe^+ densities (fig.6.6) are almost identical and initially have the same shape as the current. Both quantities begin to increase as a consequence of the reduced cross section for current flow brought about by the (hypothesized) constriction function (cf. Volume(t) fig.6.7) until reaching a value an order of magnitude larger in the final stages of the excitation pulse. The predicted density of XeI excited state atoms (Xe^* in fig.6.6) in the pre-constriction regime agrees well with the estimate of $4 \times 10^{14} \text{ cm}^{-3}$ arrived at in chapter 5. Although in the later stages of the pulse the predicted density increases beyond this value, it should be remembered that the results presented in chapter 5 were themselves affected by constriction and the specific densities presented here are for 1 cm^3 of the active volume as a function of time.

The evolution of the XeCl^* density in the presence and absence of lasing, though showing the expected 'burn-down' due to lasing, bears only a slight resemblance to the experimentally observed spontaneous emission shown in plate 4.3. To be fair, the experimental curves are affected by constriction of the volume from which emission takes place. Multiplication of the rate of spontaneous emission by the time dependent volume in an effort to reproduce the experimental pulse (fig.6.8) does not, however, produce curves which correspond much better to those in plate 4.3.

The rates of production of XeCl^* via the neutral (eq.6.12) and the ion (eq.6.11) channels are compared in figure 6.9. The neutral channel represents between ~5-~25% of total XeCl production which agrees well with the estimate put forward in chapter 5. It is interesting to note that the rate of production via the ion channel has increased by almost an order of magnitude in the later stages of the excitation pulse. This occurs despite the fact that the rate coefficient (fig.6.7) for the reaction has decreased by a factor of three at this time (as a result of its guessed inverse dependence on the bulk gas temperature). This point will be discussed further below.

6.3.2 The effects of varying certain parameters:

The values of the parameters used to obtain the 'best fit' result presented in the previous sub-section are by no means unique; it is certain that

FIGURE 6.8: Predicted spontaneous emission - with and without lasing

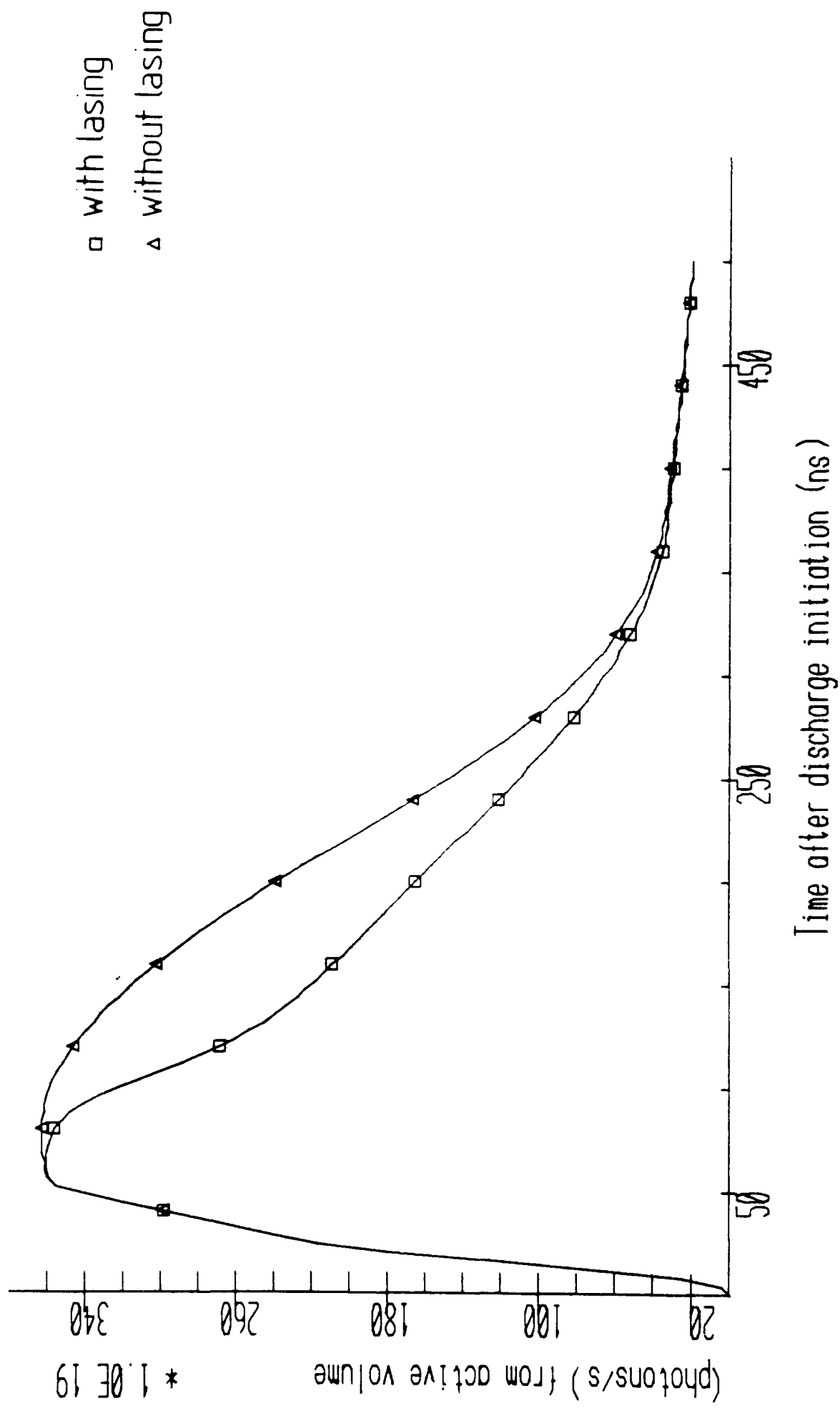


FIGURE 6-9. Comparison of XeCl^* production via the 'ion' and 'excited' channels

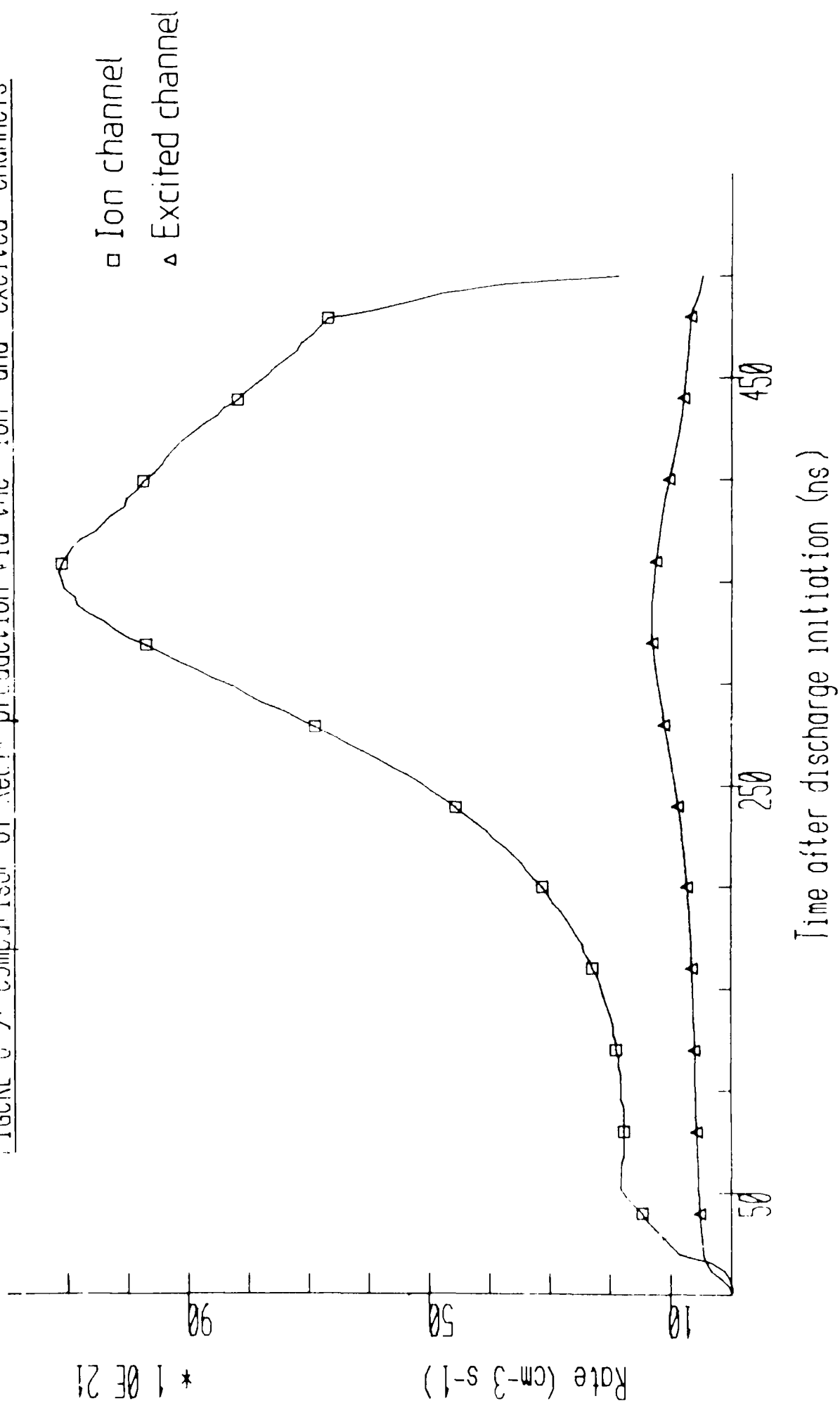
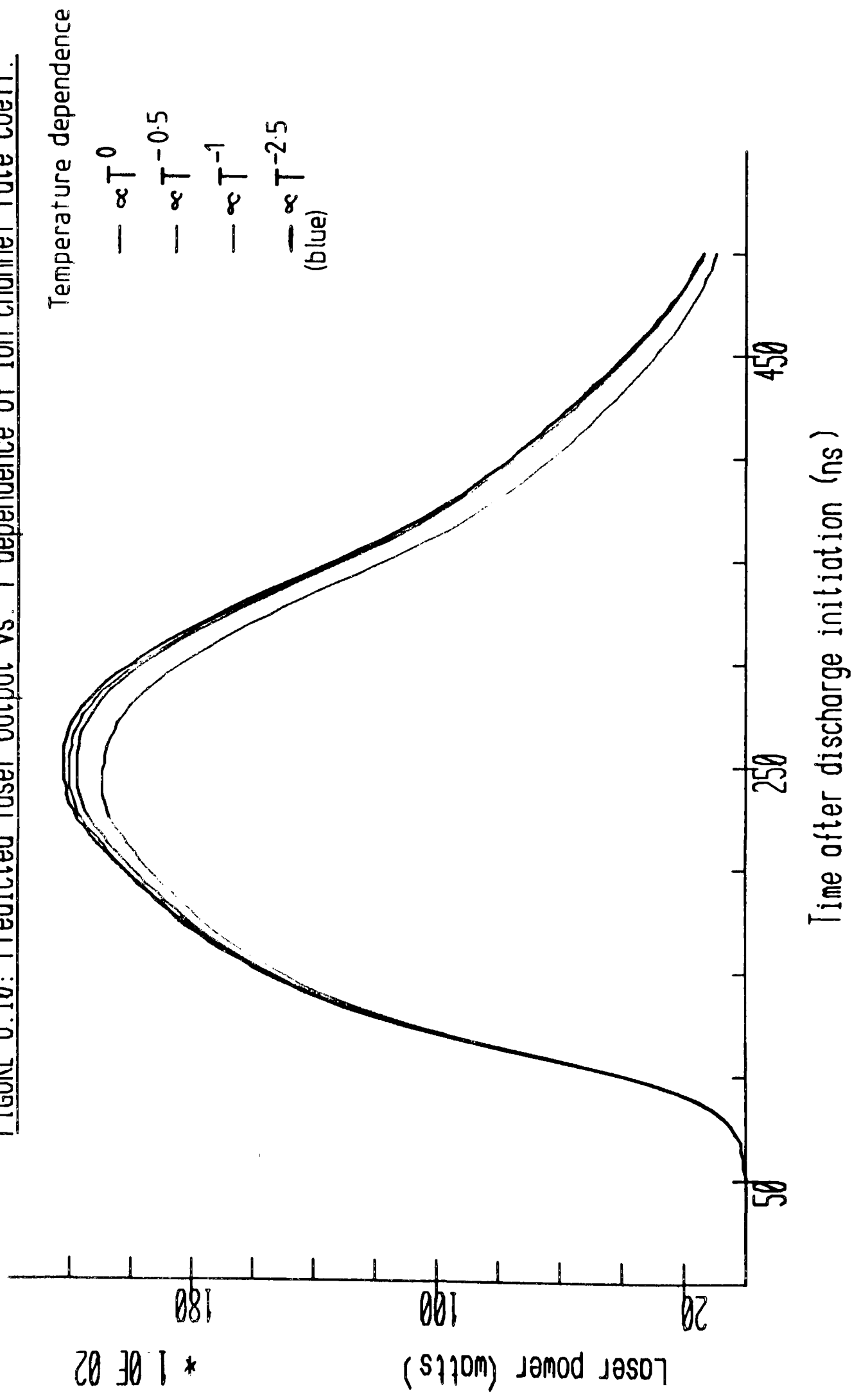


FIGURE 6.10: Predicted laser output vs. T dependence of ion channel rate coeff.



another equally plausible set of values could have produced a predicted laser output pulse which agreed well with that which was observed experimentally. This is not to say, however, that no useful information can be extracted from the model. In this sub-section results will be presented which illustrate the effect that changing certain parameters in the model has on the predicted laser output.

(a) The effect of varying the temperature dependence of the ion channel rate coefficient on the laser output:

Figure 6.10 shows the variation of the predicted laser output for ion channel rate coefficients having a T^0 , $T^{-1/2}$, T^{-1} , and $T^{-5/2}$ dependence on the bulk gas temperature: the effect is negligible. The rate coefficients and the rates of production are shown in figure 6.11. As was the case for the predicted laser output, the rate of production of XeCl^* is virtually independent of the rate coefficient over a range of approximately three orders of magnitude. This insensitivity of the rate of formation of XeCl^* to its rate coefficient was first proposed by Kearsley[1980], however the degree of this independence had not been realised (Kearsley[1982]). Kearsley argued that the rate of formation of Cl^- was the limiting step in the ion channel - indeed the curves for the rate of production of Cl^- by dissociative electron attachment to HCl were found here to be almost identical to those for XeCl^* production via the ion channel. Only in the case of an ion channel rate coefficient with a $T^{-5/2}$ dependence was any difference between the two rates discernible.

The predicted variation of the instantaneous Cl^- number density with changes in the temperature dependence of the ion channel rate coefficient (figure 6.12) uncovers another shortcoming of the model. The increased absorption at the laser wavelength as a result of photodetachment is not taken account of in the laser output equation since it was assumed that the experimental transient loss would include losses via this process. The effect of a higher Cl^- number density at times late in the excitation pulse is not reflected in the corresponding predicted laser output curve (fig.6.10: in the case where the ion channel rate coefficient is proportional to $T^{-5/2}$).

FIGURE 6.10: Predicted laser output vs. T dependence of ion channel rate coeff.

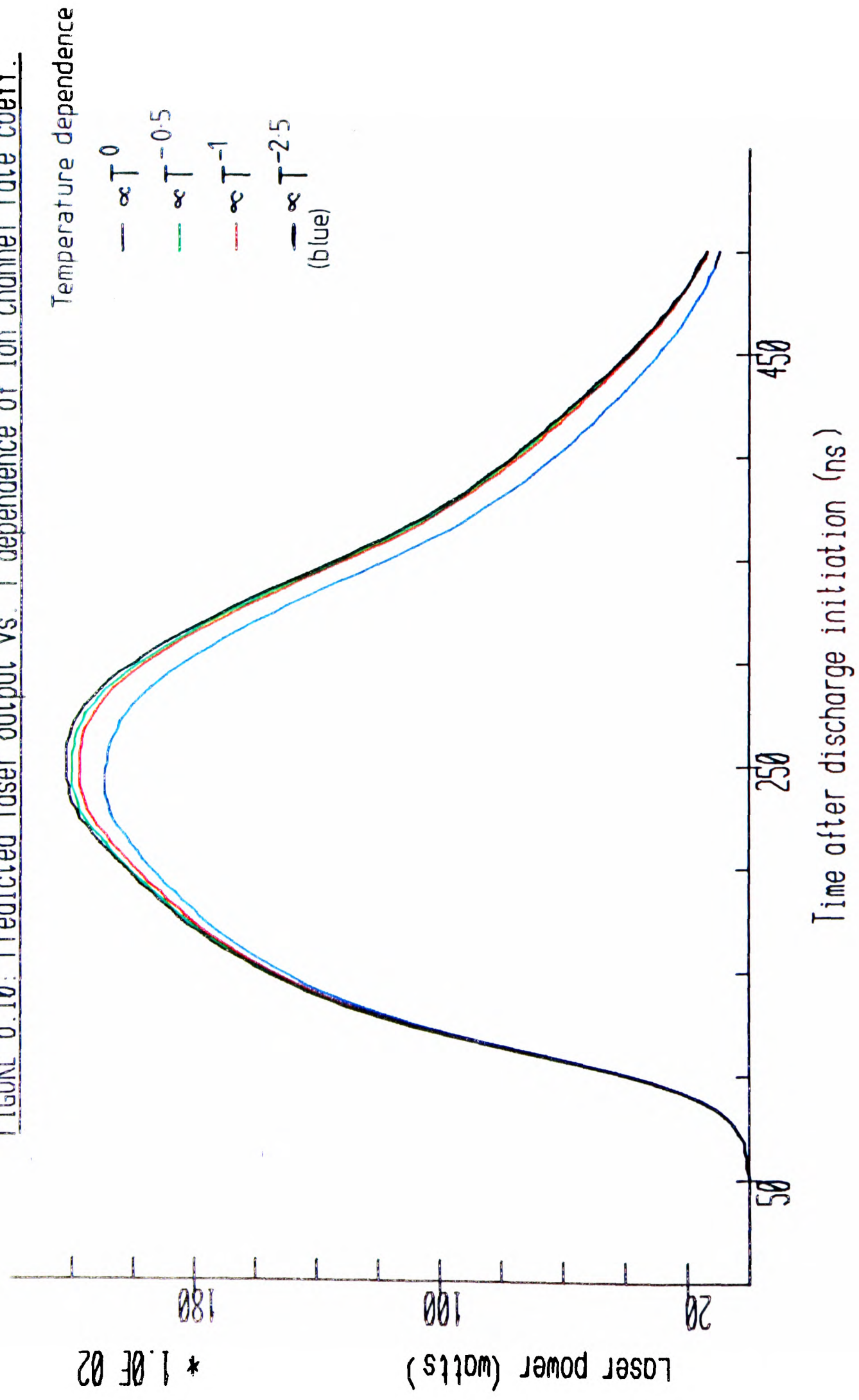


FIGURE 6.11: Rate of production of XeCl^* vs. ion channel rate coefficient

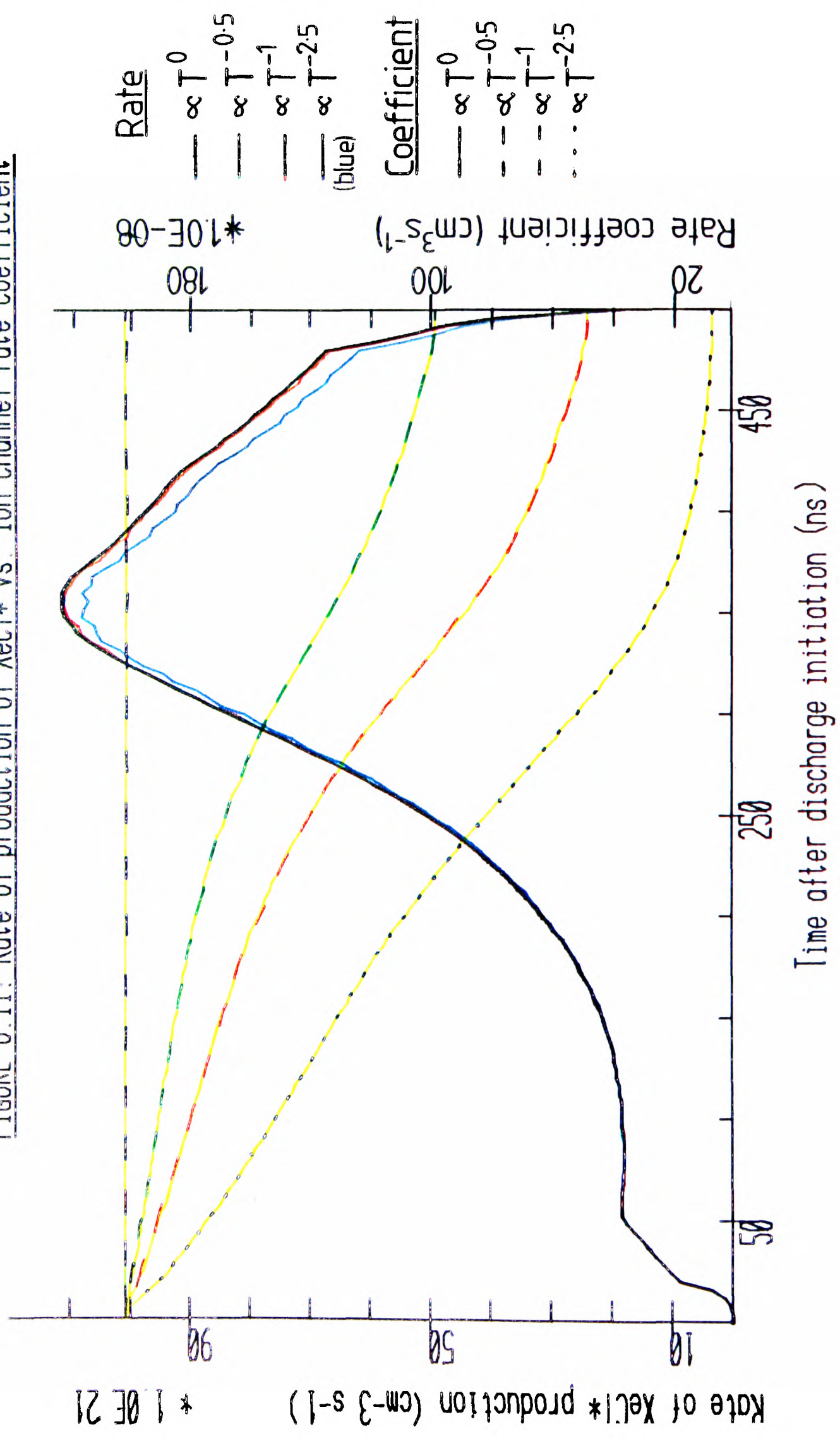


FIGURE 6.12: Cl- population vs. ion channel rate coefficient (cf. fig. 6.11)

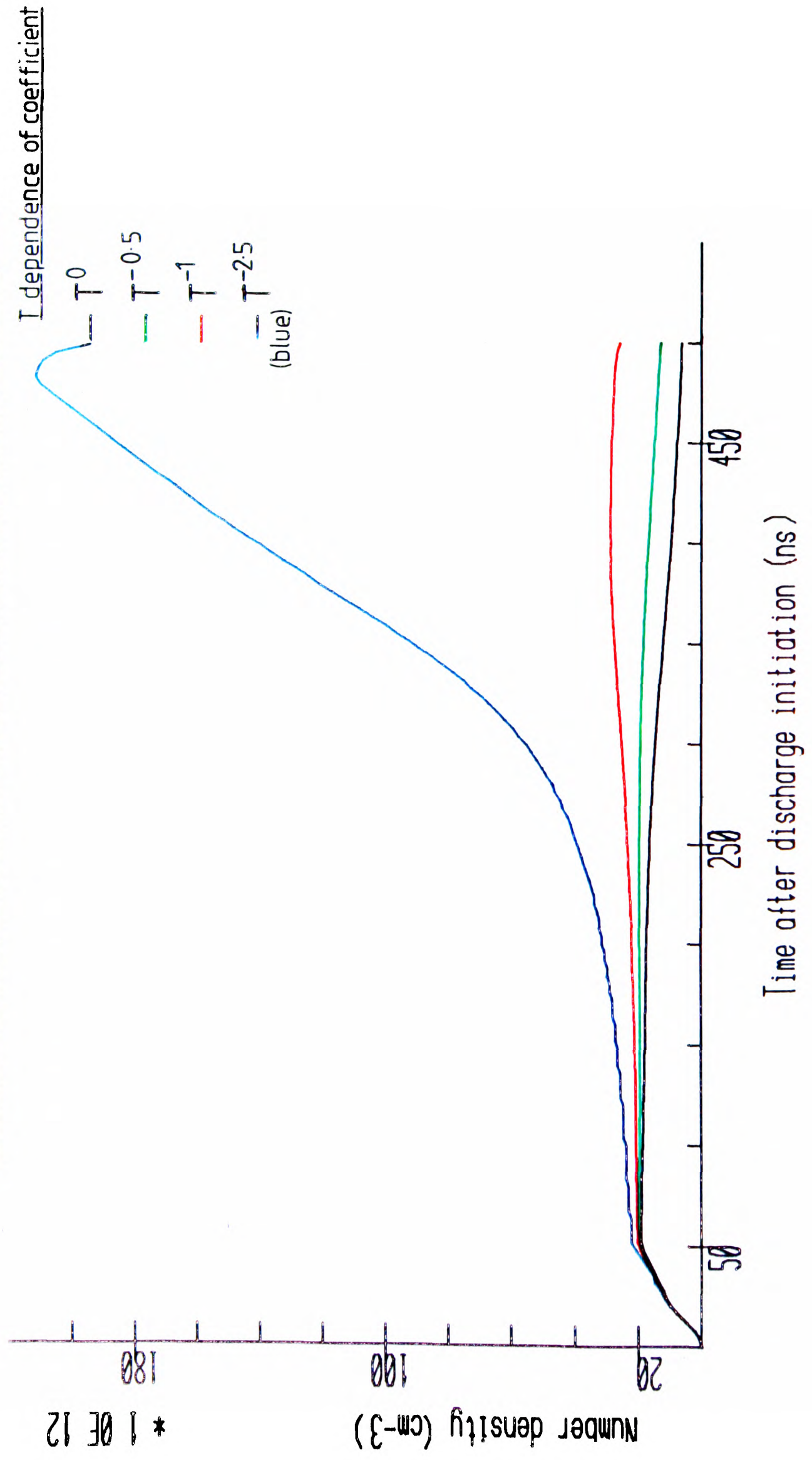
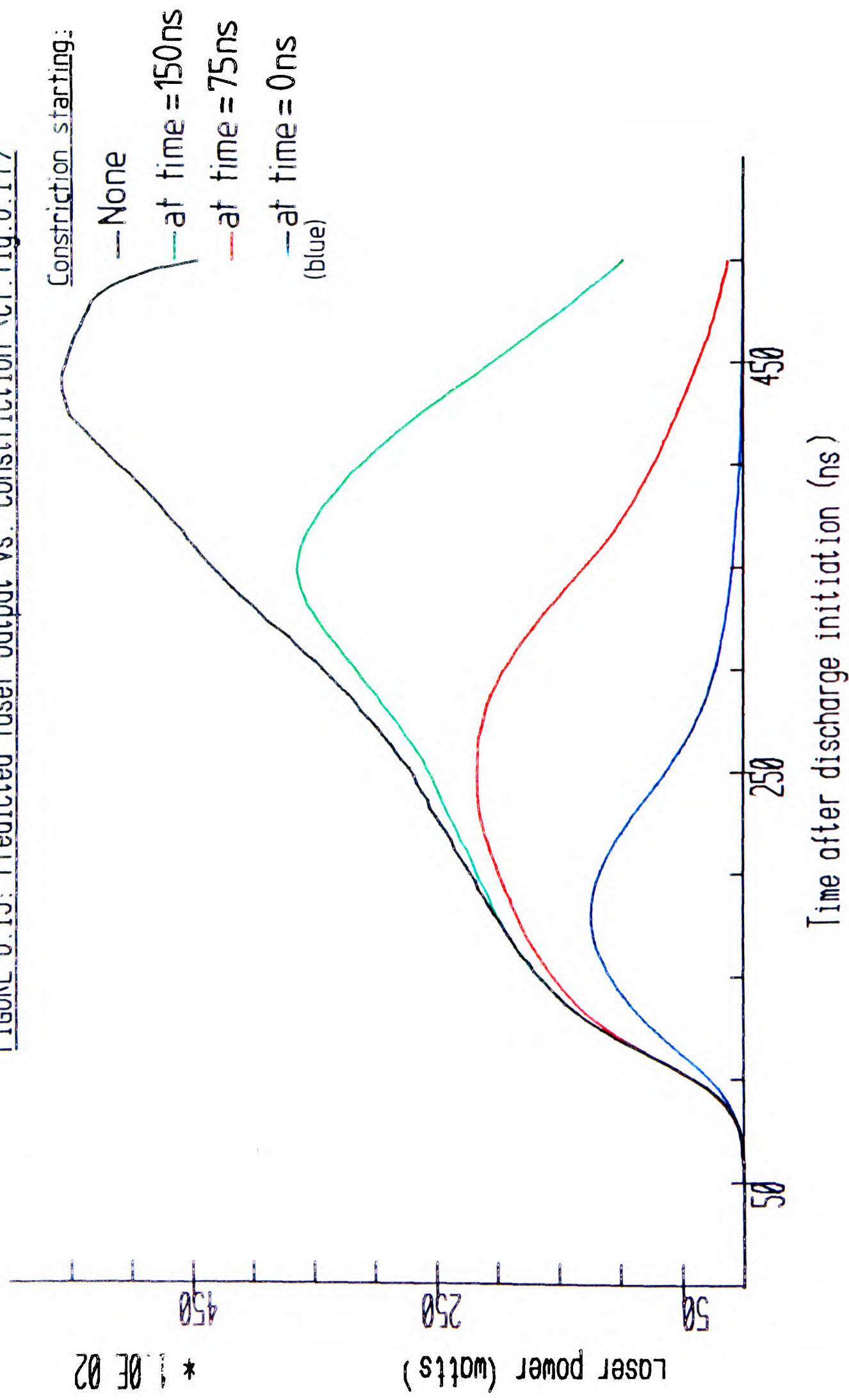


FIGURE 6.13: Predicted laser output vs. constriction (cf. fig. 6.14)



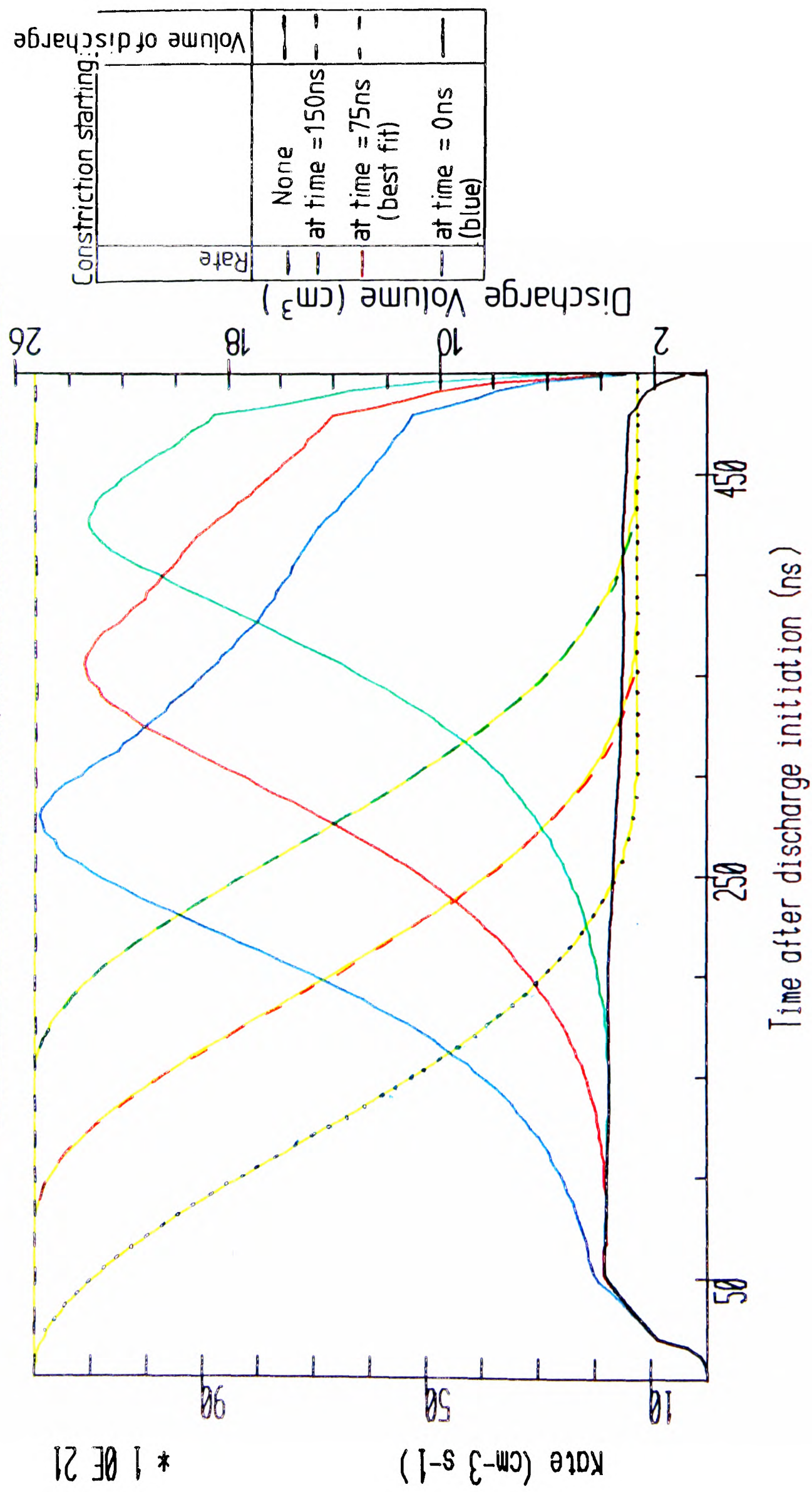
(b) The effect of varying the constriction function parameters:

The timing and degree of constriction introduced into the model in the manner shown in figure 6.2 was determined to a very large extent by the values used for the experimental transient loss. If one is to view the model at this final stage of its development, the effect of constriction on the predicted laser output cannot be separated from the effect of the transient loss.

Figure 6.13 contains curves of the predicted laser output in four constriction regimes: no constriction at all; constriction commencing at 150 and 75 ns after discharge initiation; constriction starting at discharge initiation. The corresponding curves of active volume and XeCl^* production in each of these cases are presented in figure 6.14. The curve for constriction starting at 75 ns is the same as that in figure 6.3 (the 'best fit' case) but on a different scale. Where constriction was included the active (discharge) volume was assumed to collapse to one tenth of its initial value by the end of the excitation pulse; the collapse in volume was introduced by reducing both the discharge (gain) length and the discharge thickness to a value ~ 0.32 of their initial values.

In the case where constriction is not present at all, the rate of production of XeCl^* via the ion channel is virtually constant for the duration of the excitation pulse. The corresponding power output curve, on the other hand, shows a dramatic rise late in the excitation pulse because the experimental transient loss is considerably smaller at this time. The introduction of constriction at 150 ns after discharge initiation results in a late rapid rise in the XeCl^* production rate - a rise which is also reflected by the laser power curve. In this case, however, the reduction in the gain length as a result of the constriction serves to limit the output to a value below that reached when constriction was not included. It is not until constriction is introduced at 75 ns that the predicted output begins to be similar to the experimental pulse shape; the predicted output was found to be very sensitive to t_{ac} and excursions of more than ± 3 ns from 75 ns resulted in an unacceptable prediction of laser output. Movement of the time of onset of constriction

FIGURE 6.14: XeCl* ion channel production rate vs. constriction



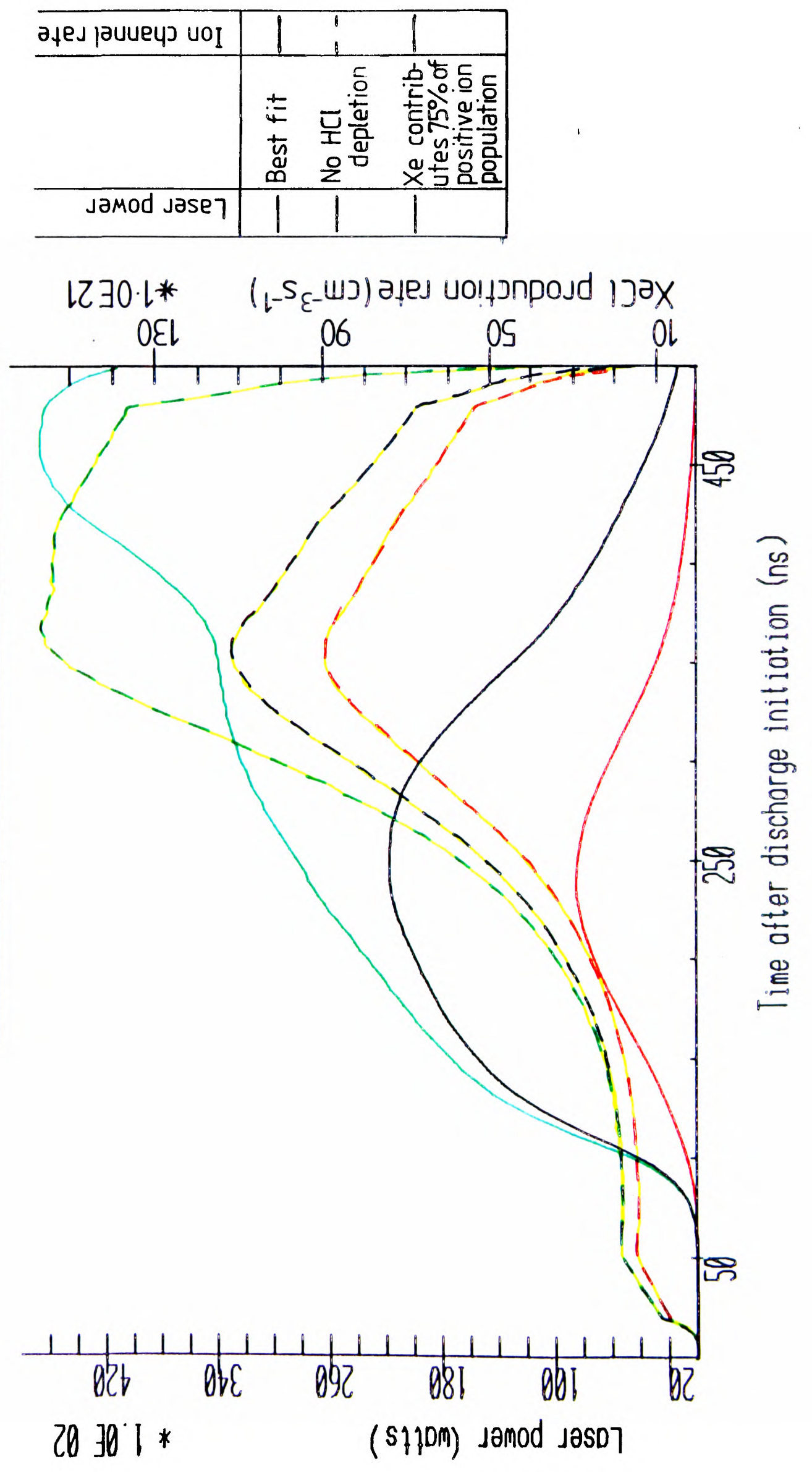
closer still to that of discharge initiation results in a prediction of a much shorter laser pulse duration. A combination of reduced gain length, high transient loss, and an earlier reduction of the rate of production of XeCl^* are responsible for this early termination. The earlier peaking of the XeCl^* production rate as the time of onset of constriction is moved closer to discharge initiation is due entirely to depletion of HCl within the active volume - either by destruction through Cl^- formation or by a reduction of the number density due to gas expansion. The rate of production of Cl^- , which governs the rate of production of XeCl^* , is reduced as the number density of HCl within the active volume is depleted.

(c) The effects of holding the HCl concentration constant or changing the proportion of positive ions due to xenon:

The effect of maintaining the HCl density constant against loss through Cl^- formation can be seen in figure 6.15. The rate of production of XeCl^* via the ion channel is greater for a maintained (gas expansion is still present here) HCl concentration than for that where the HCl number density is allowed to deplete (a 'best fit' curve is included for comparison). The predicted laser output bears no relation to the experimental result: the higher specific input power brought about by constriction is sufficient to overcome the one third reduction in gain length and a large peak in output is predicted at the tail end of the excitation pulse.

Also shown in figure 6.15 is the effect that a reduction of the proportion of the positive ion population made up by Xe^+ has on the laser output and the ion channel production rate. In this case Xe^+ was assumed to make up 75% of the total. The overall shape of both the laser output and the production rate are the same as for the 'best fit' case though there is a proportionate decrease in their magnitudes. It is emphasised, however, that the 25% of 'other' positive ions, thus assumed to be present, were allowed to react with Cl^- with the same speed as Xe^+ . This effectively reduced the rate of production of Cl^- - to which XeCl^* formation has been found to be so sensitive. When the 'other' ions were not allowed to react with Cl^- , a reduction in the proportion of Xe^+ had a

FIGURE 6.15: Effect of maintained HCl and reduced Xe+



negligible effect.

6.4 DISCUSSION:

The results presented for the 'best fit' case in the previous section only tell us that a plausible mechanism for premature termination of the laser emission from the LPD laser has been found. The values of many of the parameters used are probably incorrect: the rate coefficient for quenching of XeCl^* by electrons, in particular, was arbitrarily increased by a factor of two over the values generally quoted in the literature in the early stages of development of the model. At the time, this was done in an attempt to make the ratio of the predicted peaks of spontaneous emission with lasing to that without lasing more like the experimentally observed ratio (plate 4.3). Since these quenching rate coefficients are not known experimentally to any great degree of accuracy, once the rate coefficient was changed it was left as it was. If these rate coefficients had been reduced, the multiplying factor used for the rate coefficient for dissociative attachment of electrons to HCl would have had to be reduced to decrease the total output ('m' in section 6.2.2(d)). It is true that many other such rate coefficients could be changed but this would have had a snowballing effect in that other rate coefficients would have had to be altered to compensate. It was decided that considering the limited usefulness of the model such adjustments would have resulted in an unwarranted waste of time.

Initially, the model did not have constriction included nor did it use an experimental transient loss. The transient absorptions were included in the stimulated photon density equation (eq.6.32) using the photoabsorption processes which remain in the Cl^- and Xe^* equations in the final version. A further constant $1\% \text{cm}^{-1}$ loss for the duration of the pulse still had to be included to reduce the level of predicted emission. In some ways this approach was less artificial than the one used in the final version. Nevertheless, the laser emission predicted by the model when it was in this form did not correlate at all well to experimental observations: although the leading edge of the laser output pulse correlated well with experiment, the drop in emission brought about purely by depletion of the halogen donor, HCl , within the (constant) active

volume was not sufficiently fast to enable pulse termination to be explained by depletion alone.

Clearly, there are a number of serious shortcomings - the most important of which, as mentioned previously, is the way in which the rate coefficient for dissociative attachment of electrons to HCl was treated. From the results presented in the previous section it is seen that this rate dominates the rate of production of XeCl^* via the ion channel.

The useful conclusion which can be extracted from the model is that premature termination of the laser emission from the LPD laser may well be brought about by constriction of the discharge volume in time in combination with localised depletion of the halogen donor gas, HCl. It would seem that depletion of the actual halogen precursor, Cl^- , is not such a problem and that under certain circumstances it can actually increase in the later stages of the pulse without increasing the XeCl^* production rate. The experimental values of the transient loss were by no means sufficiently accurate. The predicted output power is very sensitive to the transient loss but the laser pulse is not terminated by it - a conclusion which is arrived at more as a result of the experimental evidence produced in chapter 4 and 5 than by the predictions of the code. The large experimental error in the transient loss meant that to a large extent the experimental values chosen as data in the code could be tailored to suit the needs for the predicted laser output. The predicted spontaneous emission curves provide a much better indication of the fact that this model does not provide a complete picture of the kinetic evolution within the LPD discharge.

It cannot be overemphasized that the significant point is not that well-matched predicted and experimental curves could be produced when constriction and HCl depletion were included, but rather that good agreement could not be obtained if they were not included.

The relative independence of XeCl^* production rates from their respective rate coefficients meant that the type of temperature dependence of the rate coefficient used in the model was unimportant. The place where this dependence

will make a difference is in the photon density equation since, as can be seen from fig. 6.12, the instantaneous Cl^- density increases as the ion channel rate coefficient becomes more dependent on the temperature. The increased absorption at the laser wavelength due to photodetachment of electrons from Cl^- might well be significant in a truly self-consistent kinetic model.

The degree to which the rate of production of XeCl^* via the ion channel is predicted by the model to be independent of the rate coefficient for the reaction is somewhat astounding. The magnitudes of the various Cl^- production and loss processes are such that the Cl^- rate equation (eq. 6.29) can be written to good approximation as:

$$d/dt(\text{Cl}^-) = K_6 \cdot [\text{HCl}] \cdot n_e - K_7 \cdot [\text{Cl}^-] \cdot [\text{Xe}^+] \quad (6.37),$$

and the rate of formation, R , of XeCl^* via the ion channel is given by

$$R = K_7 \cdot [\text{Cl}^-] \cdot [\text{Xe}^+] \quad (6.38),$$

where K_6 is the rate coefficient for dissociative electron attachment to HCl and K_7 is the ion channel rate coefficient. In the case where Xe^+ is the major positive ion species, $[\text{Xe}^+] \approx n_e$. When $d/dt(\text{Cl}^-) = 0$, it is clear that

$$K_7 [\text{Cl}^-] = K_6 \cdot [\text{HCl}] \quad (6.39)$$

which can be substituted directly into eq. 6.38. When this is done it is evident that ' R ' is independent of both the density of Cl^- or the value of the rate coefficient, K_7 . Indeed, the argument can be further extended to include an absence of any direct sensitivity of the rate of production of XeCl^* on the buffer gas number density. The major effect of an increase in the buffer gas density is to restrict the total volume occupied by the discharge. This indirectly increases the rate of production of XeCl^* by increasing the rate of dissociative electron attachment to HCl as a result of a higher electron density. The rate of change of the Cl^- density will only be negligible when $K_7 \gg K_6$. Since the major Cl^- loss process is via ion-ion recombination, any reduction in K_7 will be offset by an increase in $[\text{Cl}^-]$ - so long as the product $K_6 \cdot [\text{HCl}]$ does not change.

6.5 SUMMARY:

A 'semi-empirical' model of the kinetic evolution of the LPD laser

discharge has been developed. The model is semi-empirical because rather than determining the electron density via implementation of the steady-state Boltzmann equation, it is calculated using experimental values of the discharge voltage and current. Experimental values of the transient loss in the laser cavity are used in the photon density equation.

The major conclusion which arises from the predictions of the model is: it is possible to explain premature termination of the laser output in the LPD laser as being a consequence of constriction of the active discharge volume in time together with the depletion, within that volume, of the halogen donor species, HCl. It was found that when either of these effects was not taken account of the model could not provide a prediction of laser output which correlated with that which was observed experimentally.

Another interesting point which arose is that the rate of production of XeCl^* via the dominant formation channel - three body ion-ion recombination - is virtually independent of the rate coefficient for the reaction over a range of approximately three orders of magnitude. This is interpreted as a consequence of the fact that the rate coefficient for dissociative electron attachment to HCl is usually $\sim 10^4$ times smaller than that for the ion channel. It is concluded, in agreement with Kearsley[1980], that the rate of formation of Cl^- is the limiting step in XeCl^* formation. It is unlikely, however, that XeCl^* formation is independent of the density of Xe^+ as was stated by Kearsley[1980] since this population will be closely linked to the electron density.

The model is not generally applicable to other XeCl^* laser systems. It is nevertheless true that development of a self-evolving kinetic model for long pulse duration, discharge-excited systems will have to pay more attention to the effects of discharge instability, halogen donor depletion, and heating of the bulk gas.

Despite all of its shortcomings, however, the model presented here does attempt to take realistic account of the known facts, and almost certainly gives a closer approximation to the actual long pulse XeCl^* laser discharge than any of the descriptions published in the literature to date.

C H A P T E R S E V E N

Discussion and Conclusions

7.1 REVIEW OF MAJOR FINDINGS:

The object of this study was to extend the duration of laser emission obtainable from discharge excited XeCl^* lasers. This object was achieved with the construction of a device which employed a resistively ballasted pin array as one of the electrodes of the transverse discharge. A FWHM laser pulse duration of 270 ns was obtained from the 'LPD' laser.

Although in comparison to other devices the laser pulse duration was long, the major problem which arose in the study of the properties of this device was the identification of the cause of the termination of the laser emission well before the end of the electrical excitation pulse. In the early stages of the investigation, this premature termination of the laser pulse presented something of a paradox because:

1. an increase in the density of transient species which absorb at the laser wavelength of 308 nm could, on the basis of experimental observations, be ruled out immediately as the cause of termination;
2. recordings of voltage and current waveforms appeared to indicate that the specific power input to the discharge was constant for the full 500 ns duration of the pulse forming network driver;
3. observation of the discharge uniformity for a normal laser gas mixture by eye did not reveal the occurrence of gross arcs - on the contrary the entire volume between the electrodes appeared to be uniformly illuminated;
4. depletion of the halogen donor species, HCl, within the discharge volume was identified as the probable cause of the termination since ~10% of the

HCl initially present could be accounted for on the basis of observed laser emission - yet a computer simulation of the discharge kinetics indicated that, although some drop in output could be expected on this basis, it could not explain the sharpness of the termination.

Time-resolved spectroscopic analysis of the discharge was carried out in order to obtain a more comprehensive view of the properties of the discharge. This analysis provided the crucial evidence that the discharge uniformity was not being maintained as had been previously supposed. The occurrence of intense emission from high lying xenon ion energy levels in the later stages of the electrical excitation pulse provided the conclusive proof that the volume occupied by the discharge was constricting in time - an interpretation which was confirmed by time-resolved photography of the discharge uniformity.

The cause of this temporal constriction of the volume occupied by the discharge (TVC) was linked unquestionably to the presence of HCl in the laser gas mixture. A parametric study of the effect of gas mixture composition on the discharge uniformity and its characteristic emission revealed that increasing the HCl partial pressure had a further effect - that of reducing the total volume occupied by the discharge from the moment of discharge initiation (static parametric contraction or SPC). Although one cannot be adamant that SPC is the major reason that discharge-excited XeCl^* lasers in general are so sensitive to the partial pressure of HCl, it certainly provides a more attractive explanation than those which appeal to quenching processes, for example, because of its comparative simplicity.

A 'semi-empirical' model of the kinetics of the LPD laser discharge supported the conclusion that the combined effects of TVC and depletion of HCl within the active volume are primarily responsible for the premature termination of lasing from the LPD laser.

A number of points remain to be discussed in this chapter. Firstly, and most importantly, proposals as to possible cause of SPC and TVC will be put forward. Following this there will be a discussion of why a difference in laser performance is observed when different buffer gases are used. Finally, the

prospects for obtaining even longer laser pulse durations from discharge-excited XeCl^* systems will be discussed and suggestions given as to the direction of further study of the subject.

7.2 DISCHARGE CONSTRICTION:

One of the problems which has not been addressed so far is the the reason why SPC and TVC should arise when HCl is present in the laser gas mixture. We can only provide unconfirmed conjectures as to why these effects should occur: the development of a complete theory will clearly require further detailed work. In chapter 2, various theories of the growth of instabilities in the high pressure discharge were examined. It is instructive at this stage to look at them again briefly.

7.2.1 Previous work:

A crucial difference between an RGH laser discharge and other discharges containing electronegative gases is the *greater* speed of the three-body ion-ion recombination reaction in comparison to that of negative ion formation (via dissociative attachment). The greater speed of the ion-ion recombination reaction prevents the build up of a significant negative ion population within the active volume of the RGH discharge. The attachment instability described by Douglas-Hamilton and Mani[1974] for CO_2 discharges is a result of the discharge becoming dominated by negative ion current flow. As was seen in the previous chapter, even for the case where the 'ion' channel rate coefficient was given a $T_{\text{gas}}^{-5/2}$ dependence, the maximum Cl^- density was ~ 40 times less than the electron density.

It is unlikely that either the thermal instability mechanism described by Jacob and Mani[1975]

is applicable to the present situation.

In the studies presented here, it was observed that for discharges containing a rare gas alone, or a rare gas with a small amount of a non-attaching molecular gas such as nitrogen, discharge instabilities did not

occur. The instability mechanism proposed by Daugherty, Mangano and Jacob[1978], was discussed in chapter 2. Although their model strictly applies to an e-beam sustained discharge not a self-sustained discharge, its predictions are couched in terms of assumptions concerning RGH laser discharges which are not valid. In particular:

1. the XeI metastable population was found here to be saturated with respect to the electron density. It is highly improbable that under similar conditions in a KrF^* laser that the KrI metastable population would not also be similarly saturated.
2. the Xe^* population was observed to increase as the halogen concentration was increased. An increase in the rare-gas metastable population as the halogen within the volume is depleted is the key feature of Daugherty et al.'s instability mechanism.
3. increases in halogen donor concentration destroy discharge uniformity. This observation is not limited to the XeCl^* -LPD laser: it has been found in the gas laser development group at the Clarendon laboratory that discharge uniformity in 'conventional' self-sustained discharge KrF^* lasers is also adversely affected by increased halogen concentration. This is totally at odds with the stability enhancement properties attributed to electron attaching gases by Brown and Nighan[1979].

7.2.2. Suggestions as to the cause of constriction:

The problem which seems to have been studiously avoided to date is the intrinsically spatial nature of the onset of instabilities within the RGH discharge.

In view of what has been observed experimentally, spatially non-uniform depletion of the electron-attaching gas, HCl, within the discharge volume would appear to be the major cause of discharge constriction(TVC). It is unlikely, of course, that other effects, such as expansion of the bulk gas due to localised heating, will not play some part, but the observed sensitivity of the discharge to the presence of HCl makes any process in which the presence of HCl plays a

major part very attractive.

(a) Temporal constriction of the discharge volume (TVC):

From the observation that the LPD laser pulse duration was observed to increase when attention was paid to the surface quality of the unballasted anode (see appendix 2), it is postulated that the uniformity of the electric field between the electrodes is a crucial element in the maintenance of discharge uniformity.

In comparison to the rate coefficients for excitation of XeI to XeI^* and ionisation of XeI^* to Xe^+ , the rate coefficient for dissociative electron attachment to HCl is approximately independent of E/N .

Let us consider now the case where there are a number (it does not really matter how many) of small variations in the average electric field along the length of a pair of transverse electrodes - as might well be caused by the metal-insulator interface at each the pins of the ballasted array in the LPD laser - and that the electron density is approximately uniform.

The rate of loss of electrons due to attachment to HCl across a cross section of discharge parallel to the two electrodes will be almost constant. The rate of production of electrons will not be so constant, however, since the mean electron energy will be higher in the regions of higher average field. Since the net rate of change of electron density in the high field regions will be greater than that in the low field regions, the uniformity of electron density initially present will not be maintained. The rate of attachment of electrons to HCl will be slightly higher in the regions where the density of electrons is slightly higher and, as time progresses, a significant difference will arise between the HCl number density in the high field regions compared to the low field regions. Although increased attachment as a result of a higher electron density would initially tend to inhibit any large increase in the proportion of the (driver circuit limited) total current carried by the higher average field regions, a point will be reached where the reduction in density of HCl in the high field regions will tend to reduce the loss rate of electrons on a per electron basis. A positive feedback loop will ensue whereby the greater

net production of electrons in the high field regions will further increase the density of electrons which will deplete the HCl at an even faster rate in comparison to the lower average field regions. All the while, the average field required to sustain the total current flow will be becoming smaller, the rate of production of electrons, being more sensitive to the mean electron energy, will start to fall below the rate of loss in the low field regions and the discharge will be observed to constrict.

Let us now review the last sentence without leaving out any of the logical steps: the electric field everywhere will start to fall below its initial value - the non-uniformities in the electric field remain however. The rate of production of electrons - by collisional ionization of XeI^* which, as stated before, is particularly sensitive to E/N - will everywhere decrease. The rate of loss of electrons due to attachment on a per electron basis, however, will maintain (in comparison to the rate of production of electrons) its original value in the 'low field' regions while its value in the 'high field' regions, where depletion of HCl has occurred, will be smaller. The original equilibrium between production and loss of electrons which sustained the discharge in the 'low field' regions will now be destroyed with the result that $n_e(t)$ will die away there. The effect of a net increase in conduction in the 'high field' regions and net electron loss in the low field regions will therefore lead to constriction of the current into a number of filamentary channels. Given enough time there is no doubt that the discharge would eventually reach a true arc phase.

An explanation along these lines is consistent with all of the observations of the behaviour of the LPD laser. For example, the only reason that the discharge is observed to constrict uniformly into columnar discharges associated with each individual pin is that the resistive ballasting encourages the total current to be shared equally amongst all the pins - in a manner superficially the same as that described in chapter 2. The earlier onset of XeII emission, and therefore, constriction as the impedance of the driver circuit is decreased, can also be explained via this mechanism: the local rate

of depletion of HCl is higher as a result of a higher total current (and therefore electron density). The point in time at which a critical level of depletion is reached is earlier as a consequence.

It should be noted that this proposal does not require any increase in the rate of production of electrons as a function of time - and in fact it requires, to some extent, that the rate of production should decrease. What is required is that there is a spatially non-uniform rate of production from the time of discharge initiation. Constriction is more a consequence of a spatially non-uniform reduction in the rate of loss of electrons from the active volume.

The mechanism proposed here is equally applicable to the case of a discharge between a pair of continuous smoothly profiled electrodes, except that the cause of the original field non-uniformity could be the result of microscopic imperfections in the electrode surface for example.

Taylor[1983] has recently performed a framing camera study of the temporal evolution of discharge uniformity in an X-ray preionized XeCl^* laser with a reasonably long (~ 150 ns) discharge excitation pulse. He has found that termination of laser output from this device is closely associated with the appearance of very many filamentary channels in the discharge. Taylor has also found that an increase in the initial HCl partial pressure in that device results in the earlier appearance of these filaments and a corresponding earlier termination of laser output. A time integrated view of that discharge does not reveal the presence of the filamentary structure (ie. it always appears totally uniform) - even when large HCl concentrations are used. This fact probably explains the reluctance of Levatter[1983], who has done much work on X-ray preionized XeCl^* systems, to accept that discharge stability could be a problem in X-ray preionized devices.

(b) Static parametric contraction of the discharge volume (SPC):

The discussion of the preceeding paragraph goes some way to explaining the cause of TVC, the reason for the occurrence of SPC is somewhat different however. The earlier collapse of the discharge into constricted columns for a higher given initial HCl number density is explained in much the same way that decreasing the Impedance of the driver circuit hastens the onset of TVC: given that SPC occurs, the specific input power to the active volume is higher for higher initial HCl densities.

The cause of SPC must be related to the effect that HCl has on the initial density of electrons resulting from preionization. Following the same lines of argument put forward by Kearsley[1980] (p.80) to explain the short (in comparison to CO₂ lasers) delay required between preionisation and the firing of the main discharge, it must be true that the lifetime of free electrons within the volume will be shortened as the density of HCl is increased - purely on the basis that an electron-HCl encounter will be more likely. Thus, as the initial HCl density is increased, the initial density of 'seed' electrons which aid uniform avalanching will be reduced.

The avalanching process in the high pressure discharge is not yet well understood. Undoubtedly, however, small variations in the electric field applied at discharge initiation are amplified exponentially. The greater the initial density of seed electrons, the smaller will be the electron multiplication required to achieve the total current dictated by the driver circuit. Regions of high field will avalanche more rapidly than those of low field - if the total 'running' current is achieved before the low field regions have fully avalanched the discharge will not exist there.

7.2.3 Helium vs. Neon as the buffer gas:

The better performance of the LPD laser when neon was used as the buffer gas rather than helium is the result of a combination of interrelated factors:

1. Electron transport properties in the two gases are different. In the region of interest, neon has a much lower momentum transfer cross section for electrons than either Helium or Argon, the electron drift velocity at a given E/N is therefore higher in neon. Put another way, the average field required to sustain current flow in neon is lower than either helium or argon. This probably has a significant bearing on the difference in discharge stability achieved with the three gases.
2. Transient absorption in Helium buffered mixtures was found to be greater than in Neon buffered mixtures (see chapter 5). The computer model presented in chapter 6 indicated that laser output is extremely sensitive to the transient loss.
3. According to Flannery[1978] the three-body ion-ion recombination rate coefficient is smaller and has a faster dependence on the bulk gas temperature in Helium than in Neon in the pressure region of interest. For the reasons discussed in chapter 6 this will not affect the rate of production of XeCl^* via the ion channel. It is possible, however, that the instantaneous Cl^- population is higher in Helium than in Neon and this might explain the greater transient loss.

It is not possible to single out any one of these factors in particular as that responsible for the observed superiority of neon as a buffer gas since they are all so closely related to one another.

7.3 THE PROSPECTS FOR LONGER LASER PULSE DURATION XeCl^* LASERS:

It could be said that the question of whether or not significantly longer laser pulse durations may be obtained from XeCl^* systems excited by self-sustained discharges is still open. No intrinsically kinetic limitation was discovered in the course of this work, rather the problem would still seem to be mainly one of maintaining the volume occupied by the discharge as a function of time. Depletion of the halogen donor gas does play a major role in termination of laser emission in the system described here however, and its effects would be substantially reduced if the discharge volume could be

maintained. If all other obstacles can be overcome depletion of HCl will prove to be an inescapable limitation.

Another factor which may become important (before that of halogen depletion), if total discharge stability is achieved, is the build-up of the total transient absorption in the laser cavity. If the three-body ion-ion recombination coefficient is strongly dependent on the bulk gas temperature absorption due to increases in the instantaneous Cl^- population may well extinguish laser oscillation.

The solution to the problem of maintaining the discharge volume as a function of time will not be easy to find. Termination in the LPD laser cannot be blamed totally upon the poor electrode profile of the ballasted array: the ballasting has a more beneficial effect than that which can be achieved by having a smooth electrode profile. This is evidenced by the fact that X-ray preionised devices, which used well profiled electrodes, have only achieved ~75% of the pulse duration (FWHM) of the LPD laser (Levatter et al.[1981]).

Excitation via microwave or RF discharges has not resulted in any improvement in discharge stability despite the claims of Young et al.[1982] that such devices are intrinsically more stable than transverse discharges. Their results show a similar sharp, unexplained, reduction in the intensity of spontaneous emission to that observed in the LPD laser. They comment that the device suffers from a 'skin depth' effect - this may well be true but the terminology clouds the basic issue - that they find increased conduction in some areas of their discharge volume with respect to others. It would be tempting to offer an alternative explanation on the basis of the findings of this work: namely, a spatially non-uniform rate of electron loss as a result of non-uniform depletion of HCl. Both Waynant[1983] and Wisoff[1983] have found, however, that the discharge collapse they have observed in each of their devices occurs whether or not HCl is present. Waynant[1983] has proposed that a plasma wave process - as a result of interactions of the RF radiation with the walls of the gas containment vessel - is the probable cause of discharge collapse in those devices. The mechanism is not well understood.

Laser pulse durations could possibly be extended by the use of smooth profile distributed resistance electrodes - such as can be manufactured from amorphous germanium (Gibson[1977b]) and have seen use in the stabilisation of HF discharges. Although such a device would be interesting to construct from the academic viewpoint, it is unlikely that it would ever have widespread application since the electrodes suffer from severe etching by the corrosive halogen donor gases. Continual refurbishment of the electrodes in order to maintain the benefits of a smooth electrode profile is not a practical option for applications use.

7.4 FURTHER WORK:

One of the major objects for wanting to extend the pulse duration of RGH lasers - to improve the beam quality - has been achieved by other means since this work was started. The use of unstable resonator optics for RGH lasers has virtually eliminated the requirement for a large number of cavity roundtrips to achieve a diffraction limited beam (the reader is referred to Smith[1982] for a full description).

The need for narrow linewidth laser output still remains. The question which must be asked, however, is whether it might not be better to consider using a frequency-doubled CW dye laser to injection lock short pulse RGH laser amplifiers for most applications. At this stage, it would appear that the law of diminishing returns militates against further attempts to extend the laser pulse duration of self-sustained discharge XeCl^* lasers.

In this study many problem areas have been identified and it is likely that other RGH lasers (KrF^* , ArF^* etc.) will suffer from very similar limitations. At the time of writing, however, no work has been reported on attempts to extend the pulse duration of discharge-excited KrF^* lasers, for example. A study of long pulse excitation of KrF^* has just been commenced and initial indications are that discharge stability is even more difficult to maintain in KrF^* than in XeCl^* . This work is continuing.

It is obvious from what has been discussed above, however, that the most

urgent need for further work is in the area of the theory of discharge stability in high-pressure, strongly electron-attaching gas mixtures.

7.5 SUMMARY:

Investigations of the response of the XeCl^* (308nm) laser system to relatively long (500 ns) duration self-sustained transverse discharge excitation revealed that, although significant improvements in the duration of laser emission over conventional devices could be obtained, temporal constriction of the discharge volume in conjunction with localised depletion of the halogen donor gas, HCl, caused premature termination of the laser pulse.

The efficacy of resistive stabilisation of the RGH discharge for the prevention of arcs was proven. The laser, which employed a discrete pin electrode resistively ballasted with an electrolytic solution, produced laser emission of maximum duration 270 ns FWHM with a peak power of $\sim 20\text{kW}$ ($\sim 5\text{--}10\text{ mJ/pulse}$). At the time of writing this is the longest pulse duration yet reported for a self-sustained discharge excited RGH laser.

A comprehensive parametric investigation of the laser was undertaken which, in conjunction with detailed spectroscopic analysis of the discharge, revealed that a transition of the discharge from a uniform glow filling the entire volume into a number of discrete columnar discharges was occurring. Termination of the laser output well before the end of the discharge excitation pulse was found to be intimately associated with this transition in the spatial characteristics of the discharge.

A semi-empirical numerical model of the kinetics of this laser was developed as an aid to the identification of the cause of the premature termination of laser emission. The findings of this model supported the conclusion - arrived at on the basis of the experimental results - that laser pulse termination was caused jointly by depletion of the halogen donor species, HCl, within the active volume and by constriction of the total discharge volume.

Experimental evidence was found which contradicted the only theory yet reported which deals specifically with the onset of discharge instabilities in

RGH laser discharges. Rather than showing that the strongly electron attaching nature of the halogen donor aids discharge stability - as was predicted by that theory - it was found that the presence of the halogen bearing species was primarily responsible for the onset of discharge instabilities within the device.

Further extensions of the laser pulse duration of discharge excited XeCl^* (and probably all the RGH lasers) will have to await a solution to the problem of the onset of spatial non-uniformities in the discharge. Since it would seem that the cause of discharge constriction is, at its most basic level, associated with the kinetic problem of reforming the halogen bearing species once it has been dissociated, it is unlikely that pulse durations will be extended significantly beyond the 270ns already achieved. Further development of the theory of discharge stability in high pressure, electron-attaching gas mixtures is the most urgent need. The major findings of this study have already been published in outline (Hogan et. al.[1983]).

A P P E N D I X O N E

Mirror coatings in the wavelength range 200-350 nm

The author wishes to thank Mr.C.W.Goodwin for writing this appendix and for preparing all of the mirrors used in the study.

All the reflective coatings described here have been produced in the Clarendon Laboratory using either the 12" or the 19" coating plant made by Edwards High Vacuum (U.K.).

The standard method used to produce thin film optical coatings^{1,2,3} is to place chemically cleaned substrates in a high vacuum chamber evacuated to 10^{-5} torr or better. An AC glow discharge, in either air or argon, with a current of 250mA at a voltage of 3kV is run at 10^{-1} torr for 10 minutes. The discharge is then turned off and the vacuum reestablished. The material (or materials) to be deposited on the substrate is heated to evaporation temperature before a shutter is opened to allow the material into the vacuum chamber. The shutter is left open until the required thickness of material is deposited on the substrate: this is judged by observing the transmission of the required wavelength of light through a monitor blank. By choosing the correct source to substrate geometry it is possible to produce very uniform thickness films.²

Several types of coating, all of which required different coating techniques, were produced for the gas laser development group at the Clarendon Laboratory for use with rare-gas halide lasers. They can be classified into three types:

1. Metallic coating.
2. Metallic coating with dielectric overcoat.
3. Multilayer dielectric coating.

Metallic coatings:

Metallic coatings were mainly used for mirrors required to have good reflectivity at 193 nm (ArF^*). They were produced in a 12" belljar coating plant (Edwards type 12E7). The silica substrates (normally 2"x1/4" Spectrosil B) were placed in a rotating jig after a standard cleaning procedure. A maximum of four substrates could be accommodated in the jig at one time. The chamber was evacuated to 2×10^{-5} torr before a running an air discharge for 10 minutes. (It was found that running this cleaning discharge was a crucial factor in increasing the damage threshold of the coating; the coating was destroyed almost immediately by the laser when the cleaning discharge was not used in its manufacture.) As soon as the 2×10^{-5} vacuum was reestablished the evaporation was carried out.

A 3 cm length of 1 mm diameter, 99.999% purity aluminium was electrically heated in a tungsten boat (manufactured by Balzers). The rate of evaporation was required to be as high as possible without increasing the pressure in the chamber beyond 2×10^{-5} torr. A beam of light was passed through a monitor glass situated at the centre of rotation of the coating jig and onto a photodiode. The output of the photodiode showed the transmission of the film as it was deposited onto the monitor blank. Approximately 10 seconds was required for the film to become opaque but the total evaporation time was allowed to be twice this. The final thickness of the film was ~ 100 nm.

Coatings of this type were nearly always used as back surface reflectors on a Spectrosil B substrate and thus doubled as windows for the laser gas envelope. Recent measurements⁴ have shown that a back face mirror of this type which, incidentally, has very broad band reflectivity, has a damage threshold some three times higher than one used as a front face reflector and overcoated with magnesium fluoride. Unfortunately, they are susceptible to mechanical damage and cannot be cleaned. Another problem is inherent absorption in the aluminium which, when freshly deposited, is ~10% and this value increases with time. The thickness of the aluminium oxide layer which grows on the surface of the film plays a significant part in the absorption of UV radiation. One may overcoat the metallic film with dielectric materials in order to stop this Al_2O_3 growth problem.

Metallic coatings with a dielectric overcoat:

Much has been written on the subject of metallic thin films overcoated with a dielectric⁵⁻¹⁰. Several of this type of coating were manufactured using a 19" coating plant (Edwards type E500). This coating plant has the capability of optically monitoring the film thickness, changing the monitoring disc during the run, utilising up to four separate shuttered coating material sources, and evacuating the coating chamber at much higher speeds than the plant described above.

As before, the substrates were scrupulously cleaned. Aluminium was placed in a tungsten boat and magnesium fluoride (granular form supplied by Balzers) was placed in a covered molybdenum boat. The chamber was evacuated to $\sim 10^{-5}$ torr and a 10 minute cleaning discharge was run. Within 3 minutes of completing the cleaning discharge, the aluminium was evaporated onto the substrate (the aluminium had already been preheated by passing a current of 300 A through it with the shutter closed to prevent it being released into the chamber). The transmission of the monitor blank was observed to fall to zero within a few seconds once the shutter was opened. Since the plant was equipped with two high current power supplies the magnesium fluoride could also be preheated. A clean monitor disc, inserted by remote control, replaced that used for monitoring the deposition of aluminium. A half wavelength thickness (for the specific laser wavelength of interest) was deposited on top the aluminium film when the MgF_2 shutter was opened. It was found to be easier to optically monitor the deposition of a quarter wave thickness at twice the wavelength (i.e. in the visible). After an upgrade of the capabilities of the coating plant it was decided to use silicon dioxide, evaporated from an electron beam source, rather than MgF_2 as the overcoating material.

Although overcoating can minimise extra absorption by preventing the growth of aluminium oxide it does not overcome the problem of inherent absorption by the aluminium in the UV. Low-loss dielectric coatings are required for further improvements - especially for high-power pulsed laser applications where damage thresholds are all important.

Multilayer dielectric coatings:

To produce high reflectivity low loss mirrors it is necessary to deposit many quarter-wave layers of alternately high and low refractive index materials onto a substrate.^{2,5} In the visible region of the spectrum there are many materials available which have either high or low refractive indices and low absorption.

Ideally, the materials chosen for the mirror will have as large a difference in refractive index as possible. This allows not only a wide bandwidth reflectivity to be achieved but also reduces the number of layers required to achieve a high total reflectivity. For example, at 550 nm, a mirror consisting of 13 layers of zinc sulphide ($n=2.35$) and cryolite ($n=1.35$) has a

bandwidth of 100 nm and a reflectivity of over 99%. In the ultraviolet, however, the choice of suitable, low absorption, materials - especially those with a high refractive index - is considerably reduced. The literature on the subject¹¹⁻¹⁶ does not help here since there is often disagreement about the wavelength at which a particular material starts to absorb. Other problems which arise are the chemical and mechanical durability of a particular material and the compatibility of paired dielectrics.

Soft dielectric coatings:

Initially only thermal evaporation boats (i.e. electrically heated) were available for use in the coating plant and this further limited the choice to 'soft' (as opposed to 'hard') dielectric materials. At this stage then, the available technology permitted only the manufacture of mirrors for 308 nm (XeCl^{*}): a combination of antimony trioxide ($n=2.3$) and cryolite ($n=1.35$) was used. These are low evaporation temperature materials, compatible for mechanical stress, have a reasonable ratio of refractive index, and are easy to work with. On the other hand they are soft and hygroscopic so they are liable to chemical and mechanical damage.

As before the substrates were cleaned and placed in the chamber. A monitor disc (also Spectrosil B) was placed on the axis of rotation of the coating jig in the same plane as the substrates. After pumping out and a 10 minute cleansing discharge, a quarter-wave layer of antimony trioxide (powdered) was evaporated from a covered boat (Balzers). Coating was terminated by closing the shutter and then cryolite (Na_3AlF_6) was evaporated on top of this layer from a similar boat powered by a separate low voltage power supply. The same monitor disc was used for all layers - amplification of the signal was required as the transmission fell. To summarize the results for antimony trioxide/cryolite coatings: 1, 3, 5, 7, 9, and 13 layers give 25%, 67%, 87%, 95%, 98%, 99%, and 99.6% reflectivity respectively at 308 nm: this assumes no absorption since transmission, not reflectivity, was measured. (Optical monitoring was used on all occasions).

Antimony trioxide starts to absorb at $\sim 300\text{nm}$ and is therefore not suitable for 249 nm (KrF^*) or 193 nm (ArF^*) mirrors. Lead fluoride, evaporated from a platinum boat,¹⁷ was tested in combination with cryolite. It was found to be difficult to evaporate this material in a controlled manner but, more importantly, it was found to absorb at 249 nm which was totally unsatisfactory.

Hard dielectric coatings:

At this time a 4-hearth, 10 kW electron gun (Electrotech type ET15/4), a 2 kW quartz substrate heating unit, and oxygen flow equipment for reactive evaporation, were installed in the 19" chamber to increase the range of usable dielectrics. A large area of the evaporant was heated at once using a large electron beam spot in combination with full longitudinal and lateral scanning. A relatively low accelerating voltage ($\sim 6\text{kV}$) was applied to the gun at all times. These factors are important since it is very easy to dissociate these dielectric materials when the e-beam spot is too intense. A maximum beam current of 1 amp was possible. Very high temperatures could be attained without running into problems of boat or crucible reactions since the dielectric formed its own crucible within the water cooled copper hearth. This apparatus allowed refractory oxides and other high melting point materials to be evaporated easily and in a controlled manner. Such materials produce very hard and durable films which can be cleaned repeatedly.

Al_2O_3 , HfO_2 , MgF_2 , MgO , SiO_2 , Y_2O_3 , and ZrO_2 were all tested to ascertain the best material² for 249 nm mirrors. Some were² tried in combination with Na_3AlF_6 . All of the high evaporation temperature materials were found to prefer substrates heated to $\sim 200^\circ\text{C}$.

MgO ($n=1.8$, obtained from Balzers in tablet form) in combination with MgF_2 was the first material tested for mirror suitability at 308 nm. With the substrates heated to 180°C and no cleaning discharge, 31 layers of MgO/MgF_2 were put down with a half wave layer of MgF_2 on top. Oxygen was not used. Although the mirrors were satisfactory upon removal from the chamber (99% reflectivity, 30 nm bandwidth centred at 308 nm), a bloom had appeared a few days later making them unusable - this has also been observed by other workers¹⁸.

Al_2O_3 ($n=1.7$, granules) in combination with MgF_2 onto a cold substrate (33 layers) peeled off as soon as air was readmitted to the chamber. This combination was later successfully employed as a two layer anti-reflection coating at 249 nm when substrate heating was used. The resultant coating (Al_2O_3 next to substrate) proved to be very durable.

HfO_2 ($n=2.1$, tablets from BDH(Merck)) in combination with SiO_2 was tested as had been done by others.¹⁹ In order to increase the refractive index of the HfO_2 , it had to be evaporated onto a hot substrate (250°C) in the presence of oxygen. The optimum O_2 pressure was found by experiment. A base vacuum of 4×10^{-5} torr was first obtained with the substrate heating on and then oxygen was admitted until the pressure rose to 2×10^{-4} torr. The HfO_2 was evaporated using 220 mA at 6 kV: it took about two minutes to achieve a quarter-wave thickness. Deposition was terminated simply by switching off the power. SiO_2 was deposited in the absence of oxygen using 60 mA at 6 kV to power the e-beam gun.

It was found that many materials showed a wavelength shift (and change in transmission) upon readmission of air in the cases of both thermal and e-beam evaporation. The magnitude of the shift varied with both the material and substrate temperature at the time of deposition. This was caused by variations in the packing density when the materials condensed on the substrate.^{12,16} $\text{HfO}_2/\text{SiO}_2$ showed a consistent 5% shift to longer wavelengths when used as described above. The $\text{HfO}_2/\text{SiO}_2$ combination gave good results: 21 layers consistently gave ~99% reflectivity, 30 nm bandwidth at 249 nm. Similar results were achieved at 308 nm. All of the coatings were measured with either a Perkin Elmer 552 or Pye Unicam spectrophotometer.

$\text{HfO}_2/\text{SiO}_2$ gave hard durable coatings which could be cleaned many times and which had damage thresholds in excess of $1 \text{ J}/\text{cm}^2$ at 249 nm: a threshold which compares well with commercially produced coatings. This became the standard pairing for coatings down to 249 nm. Mirrors of lower reflectivity were also manufactured: 1, 3, 5, 7, and 9 layers gave reflectivities of 25%, 50%, 70%, 80%, and 90% respectively. The refractive index ratio of this pair is about the best that can be achieved. Materials such as Y_2O_3 ($n=1.87$), NdF_3 ($n=1.61$) and LaF_3 ($n=1.6$) require many more layers to achieve the same reflectivity and result in a narrower bandwidth reflector when combined with either MgF_2 or SiO_2 : they were not investigated fully. These materials may be useful at 193 nm²⁰, however, and work is just about to start on producing mirrors in this region of the spectrum. Stelmark and Flint(1980) give some indication as to materials which may be useful around 193 nm and below - they include mainly the fluorides and oxides of the rare-earth and alkali-earth elements. Unfortunately, all these materials have low refractive indices and approximately 40 layers will be required for high reflectivity mirrors.

Recently published results²² indicate that the use Sc_2O_3 in conjunction with MgF_2 (using similar techniques as described above for $\text{HfO}_2/\text{SiO}_2$) enables the production of mirrors with damage thresholds as high as $6 \text{ J}/\text{cm}^2$ at 249 nm. Trial runs on these are also about to be conducted with powdered materials obtained from Cerac. Damage thresholds may be increased by preconditioning with irradiation at a level below that resulting in damage²³ and also by overcoating with half wave layers of MgF_2 .²² Coating designs have been simulated using programs run on a 380Z computer and they agree well with the results of actual manufactured mirrors.

In conclusion, work will continue on these coatings to improve damage thresholds and to expand the range of mirrors to include 193 nm multilayer reflectors. Better control of the dielectric evaporation rate is hoped to be achieved shortly by the use of quartz crystal monitoring equipment linked to a microprocessor. This will also be linked to the optical monitor to provide more accurate layer termination as suggested by Gibson et al.²⁴

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A P P E N D I X T W O

Specific details of the construction of the LPD laser head

This appendix provides a more detailed account of the construction of the LPD laser head than was given in chapter 4.

A2.1 The gas envelope:

The laser gas envelope was required to have good vacuum integrity (to permit a base pressure of $\sim 10^{-3}$ mbar with a small vacuum pump) and be able to withstand internal pressures of at least 3.5 atmospheres. Over 450 metal screws would penetrate the walls of the gas envelope and effective vacuum and pressure seals had to be made at all of the metal-insulator interfaces.

Since reasonable success had been gained with the use of 'stripped' PCB in the prototype laser, it was decided to construct the entire gas envelope from commercially available, 1.3 cm thick, epoxy glass-fibre sheet. Four slabs of this material, ~ 10 cm x ~ 40 cm, made up the side walls of the envelope. Before being finally bonded together the appropriate 'facilities' were provided in each of the slabs. The top face had a hole drilled in it at each end to suit 'Edwards SC10' vacuum fittings and a 50 mm aperture for the 'sidelight' window was provided at its mid-point. In the bottom face 2BA holes were drilled and tapped at 1 cm spacings along its length; brass screws (bonded with epoxy cement into these holes provided electrical 'side-feed' connections to the external circuit (fig.4.1(b)). The two side faces each contained a ballasted-pin array: one for the ballasted electrode, the other for the preionization. A channel (6.5 mm deep x 1 cm wide) was milled along most (38 cm) of the length of each side to form the main part of the electrolyte containment reservoirs. The ballasted electrode was of similar construction to the prototype electrode: about 105 10BA screws were bonded into a single row of tapped holes which were

spaced at 3.6 mm intervals. These screws were milled flush with the glass-fibre board (the screw heads were removed in this process) on both sides to improve the electrode uniformity. The pin array for the preionization consisted of three rows of 10BA screws, approximately 330 screws in all. The extremities of each screw hole were at least 1.8 mm from those of other screws. However, since the whole array was left ~ 1 cm proud of the fibre-glass support plate with screw heads intact (in order to minimize the distance between the preionization sparks and the discharge region), the actual screw spacing at the tips was ~ 1 mm.

Merely bonding these four slabs of fibre-glass board together at the edges with epoxy resin (Araldite) would not have resulted in a structure capable of safely withstanding an internal pressure of 3.5 atmospheres. This internal pressure corresponds to a total outward force on each side of the gas envelope equivalent to ~ 1000 kg. Technical data from the manufacturers of 'Araldite' epoxy adhesive indicated that the strength of a bond was considerably greater when subjected to shear forces than when under tension. In order to take advantage of the 'shear' strength of the bond a further eight (smaller) slabs of glass-fibre were cut, four for each of the top and bottom faces of the envelope. 'Tongue and groove' slots were milled in all sections and finally the structure was glued together with 'Araldite'. As can be seen in figure 4.1, this meant that the sides containing the ballasted-pin arrays were held in place by the 'tongue and groove'. The top and bottom of the gas envelope were held in place by large surface area glue joints which would have to be 'sheared' to separate.

The two detachable end flanges were also made of glass-fibre board. Each flange had a 50 mm aperture - sufficiently large and so positioned as not to clip the laser beam - and each had a neoprene O-ring of ~ 10 cm diameter seated in grooves in them. It was found that where an O-ring passed over the end joints between the different component slabs of glass-fibre of the main body a good vacuum seal could not be obtained. The solution to this problem was to provide annuli of PCB (stripped of their copper cladding), with radii similar to that of the O-rings, glued to each end of the main body to cover the end

joints. Each end flange was secured by four strong screws tapped into each end face.

A2.2 Preionization and the 'see-through' anode:

The possibility of obtaining more uniform and intense preionization by using a 'behind the grid' preionization array was the reason that method was chosen for the LPD laser. In the design stage, meshes made from 0.1 mm thick copper sheet and having a transparency of ~80% were readily available. The photographic services section of the Clarendon Laboratory was able to produce such meshes with lengths up to 40 cm using the photographic reduction and etching process more commonly employed in making printed circuits. The maximum available mesh length imposed an effective upper limit on the length of the laser discharge (since it was clearly undesirable to have a sharp edged join in one discharge electrode). However, the relatively short limit imposed on the discharge length helped in another respect: it eased the problems of driver circuit design. The smaller volume resulting from a shorter discharge length would reduce the power input requirement which, at a typical $\sim 10 \text{ MWcm}^{-3}$ for state-of-the-art discharge-excited RGH lasers, was a major consideration.

Preionization was to be provided by the three rows of ballasted pins arcing directly to the mesh electrode. Unfortunately all attempts to construct a suitable mesh electrode failed. It was almost impossible to constrain the thin copper mesh to conform to a flat plane when attached to a support structure. Consequently, some parts of the mesh electrode were always closer to the ballasted electrode than others. The discharge was observed to concentrate at these points rather than spreading uniformly along the length of the discharge gap. It had been foreseen in the design stage that modification of the anode in the light of experimentation might well be required: the laser head was designed for simple removal of the anode structure for this reason.

The anode shown schematically in figure 4.3 was the result of an 'educated guess' of a good compromise between the advantages of a uniform electrode profile and those of 'back preionization'. It proved to be so successful that further development of the anode structure was not necessary. The principal

features of the anode are:

1. a brass support plate with a wide (3 cm) hole milled along most of its length to allow the UV preionizing radiation to illuminate the discharge volume;
2. a slim (3.2 mm thick), profiled and polished copper blade electrode set edgewise onto the support plate and attached only at its ends;
3. a series of wires, at 1 cm intervals, connecting the free standing blade of the electrode to the support plate. The purpose of these wires was to reduce the length of the path for current returning via the middle portion of the blade electrode compared to that of paths near its ends. It is not known whether or not this precaution was necessary since the wires were included in the electrode structure at the outset - not as an afterthought.
4. two strips of thin copper sheet attached to each side of the slot in the support plate and overhanging the entire length of the 38 cm slot. The sharp edges of these sheets promoted arc formation between them and the row of ballasted pins closest to each (during operation no arcs were observed to emanate from any of the central row of ballasted preionization pins).

The major disadvantages of this electrode structure, as compared to the mesh electrode, are firstly the fact that parts of the main discharge (or active) volume close to the anode are shielded from the preionizing radiation, and secondly that the source of preionization is slightly further away from the active volume. However this must be balanced against the benefit of having a far more uniform electrode profile. It was found that discharge uniformity was quite sensitive to the smoothness of the profile of this anode. An increase in laser pulse duration of ~20% was achieved by removing small irregularities from the electrode profile and by giving the surface a highly polished finish. This observation can, with the benefit of hindsight, be seen as providing an inkling into some of the conclusions which will be drawn about the ballasted electrode profile in the final chapter.

A2.3 The Electrolyte Reservoirs:

The electrolyte reservoirs were completed by attaching brass plates to the outside faces of the gas envelope with screws. A gasket cut from 1 mm thick rubber sheet prevented leakage of the solution at the metal-fibreglass interface. The depth of electrolyte sandwiched between the backing plates and the pin-arrays was, taking compression of the gasket into account, about 7 mm. This was approximately double the depth of the electrolyte layer in the prototype laser. The reasons for doubling the depth of the electrolyte bath were twofold. Firstly, the depth of the bath in the prototype represented a compromise between minimizing the ballast resistance and avoiding dielectric breakdown of the solution. It was known from the results presented in figure 3.6 that the optimum ballast resistance was quite high, ~ 1.5 ohm, and because the electrolyte bath of the prototype was a sealed unit there was no way of checking for dielectric breakdown (though it is unlikely that it was occurring). Secondly, the electrolyte was known to become hot under repetitive operation. It was hoped that increasing the depth of the bath would make dielectric breakdown even less likely, and prevent excessive loss of water by evaporation, which, over the course of a long experiment, could alter the resistivity of the solution.

In order to facilitate draining of the electrolyte (or to provide the facility to be able to flow it: if it proved necessary), the backing plates had small pipes soldered into holes situated slightly below the level of the electrolyte reservoir. A 5 mm slot along the greater part of the length of each backing plate, and slightly above the level of the pin-array, allowed the observation of the electrolyte whilst it was being pulsed. Initially, glass bulbs covering these slits prevented overflow of electrolyte. These bulbs proved unsatisfactory because of their depressing tendency to crack and they were subsequently replaced by milled strips of epoxy-glass-fibre (as shown in figures 4.1 and 4.2). Before the glass bulbs were replaced, careful observation of the electrolyte was made whilst the circuit was pulsed. There was no indication that dielectric failure of the electrolytic solution was occurring.

A P P E N D I X T H R E E

Manufacturer's quoted purity for the various gases used in the study

HYDROGEN CHLORIDE:

Supplier: BDH Chemicals, Poole, Dorset

Typical analysis:	Hydrogen Chloride	99.8%
	Moisture	0.01%
	Hydrogen Fluoride	8 ppm
	Dichlorodifluoromethane	< 0.1%
	Inerts (gas phase)	1.2% v/v

XENON: (Research Grade)

Supplier: BOC Special Gases, Deer Park Road, London

Typical Analysis:	Xenon	99.995%
	Krypton	40 vpm
	Argon	< 1 vpm
	Nitrogen	5 vpm
	Oxygen	< 1 vpm
	Carbon Dioxide	< 1 vpm
	Hydrocarbons	< 1 vpm
	Hydrogen	< 1 vpm
	Moisture	< 1 vpm

NEON: (CP grade)

Supplier: BOC Special Gases, Deer Park Road, London

Typical Analysis:	Neon	99.998%
	Helium	5 vpm
	Nitrogen	5 vpm
	Oxygen	2 vpm
	Hydrogen	< 1 vpm
	Hydrocarbons	< 1 vpm
	Moisture	2 vpm

HELIUM: (Grade A)

Supplier: BOC Special Gases, Deer Park Road, London

Typical Analysis:	Helium	99.995%
	Neon	5 vpm
	Nitrogen	10 vpm
	Oxygen	2 vpm
	Argon	< 1 vpm
	Hydrogen	< 1 vpm
	Carbon Dioxide	< 1 vpm
	Hydrocarbons	< 1 vpm
	Moisture	3 vpm

A P P E N D I X F O U R

Listing of the semi-empirical discharge model

A listing of the computer program used to model the LPD laser discharge follows. It was written in RML Algol for use on a RML 380Z microcomputer. The 380Z is an 8-bit machine and is therefore badly suited to this type of numerical analysis from the point of view of accuracy and speed. Many of the routines had to be altered to reduce truncation and rounding errors. The input/output support procedures are not included in the listing. The functions of the various program variables are defined at the end of the listing.

```

BEGIN
  REAL balres,vdrift,delta t,old delta t,final time,loss,old time, ch time,
    en,time,chek1,chek2,xenon,hcl,pretime,lgain,tdisch,ch temp,temp,
    old temp,cpionch,current,old current,volts, old volts,gasfrac,
    total,newtotal,buffer;
  INTEGER finish,dat one,dat two,dat three,dat four,
    dat five,res one,res two,res thr,step1,step2,
    step3,i,raspberry,kk,rr,tata,select,p,sourgrapes,
    printer,samps,numcoeffs;
  INTEGER ARRAY ct[0:7];

  COMMENT insert input/output support routines;
  LIBRARY "A:INPUTOUT.LIB"
  LIBRARY "A:SETDISP.LIB"
  LIBRARY "A:MESSAGE.LIB"
  numcoeffs:=24;
  samps:=11+numcoeffs-4+7;
  setdisp;
  raspberry:=0;
  ioc(22);

  COMMENT open data input files;
  dat two:=findinput("C:DATTWO.DAT");dat three:=findinput("C:DATTHR.DAT");
  dat four:=findinput("C:DATFOR.DAT");dat five:=findinput("C:DATFIV.DAT");

  COMMENT read in counters;
  ct[2]:=read(dat two);ct[6]:=read(dat two);ct[3]:=read(dat three);
  ct[4]:=read(dat four);ct[5]:=read(dat five);close(dat five);
  datone:=findinput("C:DATONE.DAT");

```

```

ct[1]:=read(dat one);

COMMENT set up dynamic arrays;
BEGIN
  REAL ARRAY init[0:ct[1]],rate[0:ct[2],0:ct[6]],confrac[1:3],popfrac[2:10],
             conrate[0:ct[3]+ct[6]-1],vi[0:ct[4],0:3],
             driftv[0:ct[5],1:2],pops[0:11],coeff[0:numcoeffs],
             del[0:10],popres[0:126,0:samps],maxmin[0:samps],minmax[0:samps];
  BOOLEAN b1,b2,b3,b4,b5,b6,b7,b8,b20;
  LIBRARY "A:FORMAT.LIB"
  error(LABEL out);

  COMMENT read in initial values;
  FOR step1:=1 STEP 1 UNTIL ct[1] DO
    init[step1]:=read(dat one);
  close(dat one);
  {init[21]=0 for self-evolving Xe+ population;
   =1 for charge neutrality constraint};
  IF init[21] > 0.5 THEN b4:=FALSE ELSE b4:=TRUE;
  {init[23]=0 for constant HCL;=1 for varying HCL};
  IF init[23] > 0.5 THEN b5:=FALSE ELSE b5:=TRUE;
  dat five:=findinput("C:DATFIV.DAT");
  ct[5]:=read(dat five);

  COMMENT read in rate processes versus E/N;
  FOR step1:=1 STEP 1 UNTIL ct[2] DO
    FOR step2:=1 STEP 1 UNTIL ct[6] DO
      rate[step1,step2]:=read(dat two);
    close(dat two);

  COMMENT read in constant rate processes;
  FOR step1:=1 STEP 1 UNTIL ct[3] DO
    conrate[step1]:=read(dat three);
  close(dat three);

  COMMENT read in ballast resistance and V, I versus time;
  balres:=read(dat four);
  FOR step1:=1 STEP 1 UNTIL ct[4] DO
    FOR step2:=0,1,2,3 DO
      vi[step1,step2]:=read(dat four);
    close(dat four);

  COMMENT read in values of V drift;
  IF ct[5]=1 THEN b1:=TRUE ELSE b1:=FALSE;
  IF b1
  THEN vdrift:=read(dat five)
  ELSE
  FOR step1:=1 STEP 1 UNTIL ct[5] DO
    FOR step2:=1,2 DO driftv[step1,step2]:=read(dat five);close(dat five);

  COMMENT calculate real voltage versus time;
  FOR step1:=1 STEP 1 UNTIL ct[4] DO
    vi[step1,2]:=vi[step1,2]-(balres*vi[step1,3]);

  COMMENT calculate initial number densities and set initial values;
  FOR step1:=1 STEP 1 UNTIL 11 DO pops[step1]:=0.0000000;
  pops[1]:=init[9]*((init[10]+15.0)/15.0);buffer:=pops[1];
  pops[2]:=init[9]*init[12]*1e-3;xenon:=pops[2];
  pops[3]:=init[9]*init[11]*1e-3;hel:=pops[3];
  total:=buffer+hel+xenon;newtotal:=total;
  confrac[1]:=buffer/total;confrac[2]:=xenon/total;confrac[3]:=hel/total;
  pops[4]:=init[16];ch temp:=0.0000000;gasfrac:=1.0000000;temp:=init[30];

```

```

cpionch:=conrate[2]*(init[30]^(abs(init[35])));
oldtime:=0.000000;delta t:=1e-12;final time:=vi[ct[4],1];
select:=entier((final time/125)+0.5);p:=10;kk:=1;rr:=3;ct[7]:=1;

COMMENT calculation loop starts here;
FOR time:=0.000000 STEP (delta t*1e9) UNTIL final time DO IF ct[7] <=126 THEN
BEGIN

COMMENT finish up early facility;
finish:=emt(29);
IF finish=&^S OR finish=&^s
THEN GOTO out
ELSE
BEGIN

COMMENT choose plot variable;
IF finish=&^P OR finish=&^p
THEN
BEGIN
LIBRARY "A:CHNGEPLT.LIB"
END
ELSE
BEGIN

COMMENT print out variables facility;
IF finish=&^D OR finish=&d
THEN LIBRARY "A:PRINT.LIB";

COMMENT finding gain length and discharge thickness at time (constriction);
IF time <= init[26]
THEN BEGIN tdisch:=init[18];lgain:=init[3] END
ELSE
BEGIN
IF time >= init[26]+init[27]
THEN
BEGIN
tdisch:=init[18]*init[29];lgain:=init[3]*init[28]
END
ELSE
BEGIN
tdisch:=init[18]*(1-((1-init[29))*((sin(1.5708*(time-init[26]))
/ (init[27])^2)))));
lgain:=init[3]*(1-((1-init[28))*((sin(1.5708*(time-init[26]))
/ (init[27])^2)))));

END;
END;

COMMENT find voltage,current and transient loss at this time;
FOR step2:=1 STEP 1 UNTIL (ct[4]-1) DO
BEGIN
IF (time >= vi[step2,1] AND time < vi[step2+1,1])
THEN
BEGIN
IF abs(time-vi[step2,1]) < 0.01
THEN
BEGIN
loss:=vi[step2,0];volts:=vi[step2,2];current:=vi[step2,3];
END
ELSE
BEGIN
loss:=vi[step2,0]+(((vi[step2+1,0]-vi[step2,0])*
(time-vi[step2,1]))/(vi[step2+1,1]-vi[step2,1]));

```

```

    volts:=vi[step2,2]+(((vi[step2+1,2]-vi[step2,2])*
        (time-vi[step2,1]))/(vi[step2+1,1]-vi[step2,1]));
    current:=vi[step2,3]+(((vi[step2+1,3]-vi[step2,3])*
        (time-vi[step2,1]))/(vi[step2+1,1]-vi[step2,1]));
    END;step2:=ct[4];
    END;
    END;

    COMMENT check for temperature dependence;
    ch time:=time - old time;
    IF time < 0.000001
    THEN
    BEGIN
        old volts:=volts;old current:=current
    END;

    COMMENT inserting temperature dependence: only allow access every ns;
    IF (ch time >= 1 AND init[31] > 1)
    THEN
    BEGIN
        ch time:=ch time*1e-9;old temp:=temp;
        FOR step2:=2,4,5 DO popfrac[step2]:=pops[step2]/xenon;
        FOR step2:=3,7,8,9 DO popfrac[step2]:=pops[step2]/hcl;
        popfrac[10]:=pops[9]/xenon; popfrac[6]:=popfrac[7]+popfrac[8]+popfrac[9];
        ch temp:=init[32]*(((init[31]/newtotal)*(volts+old volts)*
            (current+old current)*ch time*0.25)/(lgain*tdisch*init[8]));
        temp:=temp + ch temp;
        gasfrac:=(init[30]/temp)+((1-(init[30]/temp))*(exp(-(time*1e-9)
            /init[33]))));

        IF gasfrac < 0.999995 THEN
        BEGIN
            newtotal:=total*gasfrac; buffer:=confrac[1]*newtotal;
            xenon:=confrac[2]*newtotal; hcl:=confrac[3]*newtotal; pops[1]:=buffer;
            FOR step2:=2,4,5 DO pops[step2]:=popfrac[step2]*xenon;
            FOR step2:=3,6,7,8 DO pops[step2]:=popfrac[step2]*hcl;
            pops[9]:=((popfrac[9]*hcl)+(popfrac[10]*xenon))/2;
        END;
        conrate[2]:=cpionch*(temp^(init[35]));old volts:=volts;
        old current:=current; old time:=time;
    END;

    COMMENT calculate E/N at this time;
    en:=volts/(init[8]*pops[1]);

    COMMENT find vdrift at E/N of this time;
    IF NOT b1
    THEN
    BEGIN
        IF en > driftv[ct[5],1]
        THEN vdrift:=driftv[ct[5],2];
        ELSE
        FOR step2:=1 STEP 1 UNTIL ct[5]-1 DO
        IF (en >= driftv[step2,1] AND en <= driftv[step2+1,1])
        THEN
        BEGIN
            IF en = driftv[step2,1]
            THEN vdrift:=driftv[step2,1]
            ELSE vdrift:=driftv[step2,2]+(((driftv[step2+1,2]-driftv[step2,2])*
                (en-driftv[step2,1]))/(driftv[step2+1,1]-driftv[step2,1]));
            step2:=ct[5];
        END;
    END;
    END;

```

```

COMMENT find electron density at this time;
pops[11]:=current/(init[17]*vdraft*lgain*tdisch);

COMMENT charge neutrality and particle conservation restraints,
      init[22] gives Xe+ as fraction of total positive ion population,
      b4 chooses self-evolving Xe+ or charge neutrality constrained,
      b5 chooses constant[TRUE] or varying[FALSE] HCL concentration;
IF NOT b4
THEN pops[4]:=init[22]*(pops[11]+pops[7]);
pops[2]:=xenon -(pops[4]+pops[5]+pops[9]);
IF b5
THEN pops[3]:=hcl
ELSE pops[3]:=hcl-(pops[9]+pops[7]+pops[8]);

COMMENT find values of rate processes at this E/N;
FOR step2:=1 STEP 1 UNTIL (ct[2]-1) DO
BEGIN
  IF (en >= rate[step2,1] AND en < rate[step2+1,1])
  THEN
  BEGIN
    IF en=rate[step2,1]
    THEN
    BEGIN
      FOR step3:=1 STEP 1 UNTIL (ct[6]-1) DO
        conrate[ct[3]+step3]:=rate[step2,step3+1]
      END
    ELSE
    BEGIN
      FOR step3:=1 STEP 1 UNTIL (ct[6]-1) DO
        conrate[ct[3]+step3]:=rate[step2,step3+1]+(((rate[step2+1,step3+1]
          -rate[step2,step3+1])*(en-rate[step2,1]))/(
            rate[step2+1,1]-rate[step2,1]))
      END;
    END;
  END;
  step2:=ct[2]
END
END;

COMMENT increase electron att. to HCL by factor of init[20] at t=init[19]:
      this is to account for increased att. due to vibrational excitation
      of HCL;
IF time < init[19] AND init[19] > 0
THEN conrate[ct[3]+5]:=conrate[ct[3]+5]+conrate[ct[3]+5]*time*init[20]
      /init[19]
ELSE conrate[ct[3]+5]:=conrate[ct[3]+5]+conrate[ct[3]+5]*init[20];

COMMENT define coefficients of rate equations;
coeff[1]:=delta t*conrate[ct[3]+1]*pops[11]*pops[2];
coeff[2]:=delta t*conrate[ct[3]+5]*pops[11]*pops[3];
coeff[3]:=delta t*conrate[ct[3]+6]*pops[11]*pops[4];
coeff[4]:=delta t*init[1]*lgain*init[14]*pops[5]*pops[10]/init[2];
coeff[5]:=delta t*init[1]*lgain*init[13]*pops[7]*pops[10]/init[2];
coeff[6]:=delta t*conrate[ct[3]+4]*pops[11]*pops[5];
coeff[7]:=delta t*conrate[1]*pops[7]*pops[6];
coeff[8]:=delta t*conrate[2]*pops[7]*pops[4];
coeff[9]:=delta t*conrate[3]*pops[6]*pops[8];
coeff[10]:=delta t*conrate[4]*pops[5]*pops[3];
coeff[11]:=delta t*init[6]*pops[9];
coeff[12]:=delta t*init[1]*init[7]*lgain*pops[9]*pops[10]/init[2];
coeff[13]:=delta t*conrate[ct[3]+2]*pops[11]*pops[2];
coeff[14]:=delta t*conrate[ct[3]+3]*pops[11]*pops[5];
coeff[15]:=delta t*conrate[5]*pops[9]*pops[11];
coeff[16]:=delta t*conrate[6]*pops[9]*pops[2];

```

```

coeff[17]:=delta t*pops[1]*conrate[7]*pops[9]*pops[2];
coeff[18]:=delta t*init[1]*ln(1/(init[4]-init[34]))*pops[10]/2*init[2]);
coeff[19]:=delta t*init[5]*init[6]*pops[9];
coeff[20]:=delta t*init[1]*init[3]*loss*pops[10]/init[2];
coeff[21]:=delta t*conrate[8]*pops[9]*pops[3];
coeff[22]:=delta t*conrate[9]*pops[9]*pops[1];
IF NOT b4
THEN coeff[23]:=delta t*conrate[2]*pops[7]*pops[4]*(1-init[22])
ELSE coeff[23]:=0;
coeff[24]:=delta t*conrate[10]*pops[11]*pops[7];

COMMENT find delta(population);
del[2]:=(coeff[1]+coeff[4]+coeff[6])-(coeff[3]+coeff[8]);
del[5]:=(coeff[2]-(coeff[8]+coeff[5]+coeff[7]+coeff[23]+coeff[24]));
del[1]:=(coeff[7]+coeff[9])-(coeff[2]+coeff[10]);
del[4]:=(coeff[2]+coeff[10])-(coeff[7]+coeff[9]);
del[6]:=(coeff[11]+coeff[12]+coeff[5]+coeff[15]+coeff[16]+coeff[17]
+coeff[21]+coeff[22]+coeff[23]+coeff[24])-(coeff[9]);
del[3]:=(coeff[13]-(coeff[10]+coeff[4]+coeff[6]+coeff[14]));
del[7]:=(init[24]*coeff[8]+init[25]*coeff[10])-(coeff[11]+coeff[12]
+coeff[15]+coeff[16]+coeff[17]+coeff[21]+coeff[22]);
del[8]:=(coeff[19]+coeff[12]-(coeff[18]+coeff[20]));

COMMENT check that rate of change of every population is
      small and change time step if necessary;
old delta t:= delta t;chek2:=0.0000000;
FOR step2:=1 STEP 1 UNTIL 8 DO
BEGIN IF pops[step2+2]=0 THEN chek1:=0.0 ELSE
  chek1:=abs(del[step2]/pops[step2+2])*1e2;
  IF chek2 < chek1
  THEN chek2:=chek1
END;
step3:=0;
FOR step3:=step3+1 WHILE chek2 >10 DO
IF delta t >= 1e-12
THEN
BEGIN IF step3 > 10 THEN text(1,"stuck in chek2 > 10 loop*N");
  delta t:=delta t*1e-1;
  chek2:=chek2*1e-1
END
ELSE chek2:=0.5;
step3:=0;
FOR step3:=step3+1 WHILE chek2 < 1 DO
IF delta t < 1e-9
THEN
BEGIN IF step3 > 10 THEN text(1,"stuck in chek2 < 1 loop*N");
  delta t:=delta t *1e1;
  chek2:=chek2*1e1
END
ELSE chek2:=5;
IF time<1e-2 AND delta t>1e-11 THEN delta t:=1e-11;
IF delta t>1e-10 THEN delta t:=1e-10;
FOR step2:=1 STEP 1 UNTIL 8 DO del[step2]:=del[step2]*(delta t/old deltat);
FOR step2:=1 STEP 1 UNTIL numcoeffs DO
coeff[step2]:=coeff[step2]/old delta t;

COMMENT increment populations - inhibit negative pops.;
FOR step2:=3 STEP 1 UNTIL 10 DO
BEGIN pops[step2]:=pops[step2]+del[step2-2];
  IF sign(pops[step2])=-1
  THEN
  BEGIN text(1,"*NThe following population has become negative:");

```

```

        write(1,step2);pops[step2]:=0.0000000
    END
END;

COMMENT get values of populations and coefficients every
        (final time/125) nanoseconds;
b6:= ct[7]=1;
b7:=+(time+(delta t*1e9)-entier(popres[ct[7]-1,0]+0.5)) > select;
IF b6 OR b7
THEN
BEGIN
    popres[ct[7],0]:=time; {time};
    FOR step3:=1 STEP 1 UNTIL 11 DO popres[ct[7],step3]:=pops[step3];
    stepl:=11;
    FOR step2:=1 STEP 1 UNTIL numcoeffs DO
    BEGIN
        IF step2#3 AND step2#7 AND step2#9 AND step2#24
        THEN
        BEGIN
            stepl:=stepl+1; popres[ct[7],stepl]:=coeff[step2]; {set coefficients};
        END
    END;

COMMENT set special quantities to some popres;
popres[ct[7],11+numcoeffs+1-4]:=tdisch*lgain*init[8]; {volume(t)};
popres[ct[7],11+numcoeffs+2-4]:=en; {E/N}
popres[ct[7],11+numcoeffs+3-4]:=volts; {volts at this time}
popres[ct[7],11+numcoeffs+4-4]:=current; {current at this time}
popres[ct[7],11+numcoeffs+5-4]:=temp; {temperature of the gas}
popres[ct[7],11+numcoeffs+6-4]:=gasfrac; {gas multiplying factor}
popres[ct[7],11+numcoeffs+7-4]:=conrate[2]; {rate coeff. for ion chan}
popres[ct[7],10]:=1.08e-29*init[1]*init[1]*pops[10]
        *(1-init[4])*init[8]*tdisch; {power out at this time};
popres[ct[7],6]:=(coeff[11]*tdisch*lgain)/(init[3]*init[18]); {spontaneous
        emission coefficient multiplied by normalised volume};
popres[ct[7],27]:=loss; {time-dependent cm-1 loss};
    FOR step3:=0 STEP 1 UNTIL samps DO
    BEGIN
        IF ct[7]=1
        THEN
        BEGIN
            maxmin[step3]:=popres[1,step3]; minmax[step3]:=popres[1,step3];
        END
        ELSE
        BEGIN
            IF maxmin[step3] < popres[ct[7],step3]
            THEN maxmin[step3]:=popres[ct[7],step3];
            IF minmax[step3] > popres[ct[7],step3]
            THEN minmax[step3]:=popres[ct[7],step3];
        END;
    END;
END;
IF b6
THEN vtint(2,16,time)
ELSE
BEGIN
    INTEGER d2,d3,d4; d3:=15;d4:=0;
    FOR d4:=d4+1 WHILE d3 < entier((popres[ct[7]-1,0]/(select*5))+6.5) DO
        d3:=d3+10;
        d2:=d3-entier((popres[ct[7]-1,0]/(select*5))+6.5); d4:=d4*5-3;
        vtint(d3,d4,time);

COMMENT plotting routine;

```

```

BEGIN
  PROCEDURE grid(px1,pxh,pyl,pyh);
  VALUE px1,pxh,pyl,pyh;
  INTEGER px1,pxh,pyl,pyh;
  BEGIN
    INTEGER step,i;i:=(pxh-px1)/5;
    FOR step:=i STEP i UNTIL i*4 DO
      BEGIN
        penup;plposn(px1+step,pyl);pendown;plposn(px1+step,pyh);penup
      END;
      i:=(pyh-pyl)/4;
      FOR step:=i STEP i UNTIL i*3 DO
        BEGIN
          penup;plposn(px1,pyl+step);pendown;plposn(pxh,pyl+step);penup
        END
      END of grid;
      resolution(0,2);plot(-1,-1,2);pspace(120,220,30,110);border;
      grid(120,220,30,110);map(0,entier(final time+0.5),0,maxmin[p]);
      plot(-1,-1,3);penup;plgoto(popres[1,0],popres[1,p]);pendown;
      FOR step3:=1 STEP 1 UNTIL ct[7] DO
        plgoto(popres[step3,0],popres[step3,p]);
        penup;vtline(40,8,mes[p],6);vtreal(44,6,maxmin[p],0,5);
      END of the live plotting library;
    END;
    ct[7]:=ct[7]+1;
  END;
END
END of the calculation loop;

COMMENT output initial values and results;
IF ct[7] > 126 THEN ct[7]:=126;
out:sourgrapes:=chin(13);priter(3,0);
IF sourgrapes >= 0 THEN
  outvalue(3,"*NAN ERROR HAS OCCURRED =",sourgrapes,6,0,"*N");
  raspberry:=raspberry+1;
IF raspberry # 1 THEN BEGIN b2:=TRUE;GOTO fin END;

COMMENT output graphical data;
can:ioc(14);
res one:=findoutput("C:TEMPDATS.SER"); res two:=findoutput("C:TEMPDATG.GRP");
res thr:=findoutput("C:TEMPDATI.INT");write(res thr,ct[7]);
write(res thr,samps);rwrite(res thr,time,0,6);close(res thr);
FOR step2:=1 STEP 1 UNTIL samps DO
  BEGIN
    caption(res two,step2);rwrite(res two,maxmin[step2],0,6);
    rwrite(res two,minmax[step2],0,6);
    FOR step3:=1 STEP 1 UNTIL ct[7] DO
      BEGIN i:=popres[step3,step2]*255/maxmin[step2];chout(res two,i) END;
      FOR step3:=ct[7]+1 STEP 1 UNTIL 256 DO BEGIN i:=0;chout(res two,i) END;
    END; close(res two);

COMMENT reduce size of popres and output serial data;
res two:=findoutput("C:TEMPDATA.ALL");
FOR stepl:=res one,res two DO
  BEGIN
    initial out(stepl);outarr(1,ct[1],"init",init,stepl);
    outarr(ct[2],ct[6],"rates vs. E/N",rate,stepl);
    outarr(ct[4],3,"losses,time,voltage,current input data",v1,stepl);
    IF NOT b1
      THEN outarr(ct[5],2,"Vdrift vs. E/N",driftv,stepl)
      ELSE text(stepl,"*N*N*NVdrift is constant*N");

```



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      IF stepl=res two
      THEN outarr(ct[7]-1,samps,"Calculated populations",popres,stepl);
      outarr(1,samps,"maximums",maxmin,stepl);
      outarr(1,samps,"minimums",minmax,stepl);
    END;
    close(res two);close(res one);
    b2:=FALSE;fin:
    IF b2 THEN BEGIN i:=newext(res one,"ERR");i:=newext(res two,"ERR") END;
    resolution(0,2);vtclr(0,0,79,23);
  END
END
FINISH

```

END of MODEL LISTING

IDENTIFICATION OF PROGRAM VARIABLES

THE DATA FILES:

There are five input data files required:

"DATONE.DAT" "DAT TWO.DAT" "DAT THR.DAT" "DAT FOR.DAT" "DAT FIV.DAT"

"DATONE" REQUIRES THE FOLLOWING:

ctl - an integer which indicates the number of initial values or constraints on the operation of the model.

Then follows a list of numbers which are read into the real array init[0:ctl].

	UNITS
init[1]= c, the speed of light	cm/sec
init[2]= l(cavity), the length of the cavity	cm
init[3]= l(gain), the gain length(pre-constriction)	cm
init[4]= R, the reflectivity of the output coupler	percent
init[5]= f, the fraction of the spontaneous emission which passes along the optical axis	-unitless
init[6]= A, the Einstein coefficient(spontaneous)	1/sec
init[7]= sigma, the optical cross-section(stimulated)	cm*cm
init[8]= d, the discharge gap	cm
init[9]= Loschmidt's number =2.7e19	1/cu.cm
init[10]= Total pressure of gas	p.s.i.g.
init[11]= quantity of halogen	mbar
init[12]= quantity of rare gas	mbar
init[13]= sigma(phd), the photodetachment cross-section	cm*cm
init[14]= sigma(phi), the photoionization cross-section	cm*cm
init[15]= gamma, the total losses incurred due to all things apart from particles and output coupling	1/cm
init[16]= the initial electron density (time=0)	1/cu.cm
init[17]= the electronic charge (postive)	coulomb
init[18]= w, the width of the discharge(pre-constriction)	cm
init[19]= the time at which attachment peaks	ns
init[20]= the attachment multiplying factor	unitless
init[21]= choose self-evolving(0) or charge neutrality constrained(1) Xe+ population.	'1' or '0'
init[22]= the fraction of positive ions due to Xe+ in the charge-neutrality constrained regime.	unitless
init[23]= choose constant(0) or time-evolving(1) HCl conc.	'1' or '0'
init[24]= choose ion channel active(0) or inactive(1)	'1' or '0'
init[25]= choose excited channel active(0) or inactive(1)	'1' or '0'
init[26]= duration of steady-state disch(pre-constriction)	ns

init[27]= duration of constriction phase	ns
init[28]= fraction of discharge length after constriction	unitless
init[29]= fraction of discharge thickness after constriction	unitless
init[30]= The initial gas temperature	K
init[31]= Avagadro's number/molar specific heat	K/Joule
init[32]= Fraction Fraction of energy into gas heating	
init[33]= Relaxation time of the gas	seconds
init[34]= Fraction Transmission(etc.) of far mirror	
init[35]= Exp dependence on T of ion channel rate coefficient	

"DAT TWO" requires the following:

this file gives a list of rate versus E/N for those rate constants which have been chosen for this type of dependence:

ct2=the number of ordered pairs which will be entered

ct6=the number of rates which are E/N dependent plus one

Then follows a list in the following form which is put into the real array
rate[0:ct2,0:ct6]

	UNITS
rate[0:ct2,1]= E/N	volts*sq.cm
rate[0:ct2,2]= K(ion), the ionization rate of the rare gas at E/N	cu.cm/sec
rate[0:ct2,3]= K(ex), the excitation of the rare gas by electrons at E/N	cu.cm/sec
rate[0:ct2,4]= K(dex), the de-excitation rate of the rare gas at E/N	cu.cm/sec
rate[0:ct2,5]= K(ionex), the ion exchange rate of the rare gas at E/N	cu.cm/sec
rate[0:ct2,6]= K(att), the attachment rate of electrons onto halogen donor at E/N	cu.cm/sec
rate[0:ct2,7]= K(rec), the recombination rate of electrons and positive ions (rare gas) at E/N	cu.cm/sec

"DAT THR" requires the following:

this file gives a list of rates which are taken as being constant

ct3=the number of rates

then follows a list in the following form which is put into the real array
conrate[0:ct3]

	UNITS
conrate[1]= K(HCl-), the H + Cl(-) recomb. rate coeff.	cu.cm/sec
conrate[2]= K(ion channel), the ion channel rate coeff. (an effective two-body rate is used)	cu.cm/sec
conrate[3]= K(HCl), the H + Cl recomb. rate coeff.	cu.cm/sec
conrate[4]= K(ex.ch.), the excited channel rate coeff.	cu.cm/sec
conrate[5]= K(quench), the rate coeff. of quenching of XeCl* by electrons	cu.cm/sec
conrate[6]= K(2)(quench), the rate coeff. of quenching of XeCl* by Xe	cu.cm/sec
conrate[7]= K(3)(quench), the rate coeff. of loss of XeCl* by Xe2Cl formation	cu.cm/sec
conrate[8]= K(HCl-quench), the rate coeff. of quenching XeCl* by HCl	cu.cm/sec
conrate[9]= K(Buffer-quench), the rate coeff. of quenching of XeCl* by the buffer gas	cu.cm/sec
conrate[10]= K(Electron impact detachment from Cl-), the rate coeff. for detachment of electrons from Cl- by electron impact.	cu.cm/sec

"DATFOR" requires the following:

this file is a list of voltage (including the ballast resistance voltage) and current versus time (obtained experimentally).

ct4= number of ordered pairs

balres= the ballast resistance (Ohms)

Then follows an array vi[ct4,x]

	UNITS
vi[ct4,0]= distributed transient loss	cm-l
vi[ct4,1]= time	seconds
vi[ct4,2]= voltage	volts
vi[ct4,3]= current	amperes

"DATFIV" requires the following:

this file contains values of the electron drift velocity versus E/N.

ct5= number of ordered pairs

if ct5=1 then only one number-constant Vdrift -is entered
otherwise, the following array is entered:

	UNITS
driftv[0:ct5,1]= E/N	V-cm2
driftv[0:ct5,1]= Vdrift at E/N	cm/sec

THE PROGRAM VARIABLES:Non-subscripted variables:

samps-	the upper bound on the array popres
numcoeffs-	the upper bound on the array coeff
balres -	the ballast resistance as read in from "DATFOR.DAT" in ohms
vdrift-	the electron drift velocity, if electron drift velocity is constant "vdrift" is assigned the value in "DATFIV.DAT" and maintains that value. If drift velocity is non-constant then "vdrift" is assigned a different value as n(electrons) vs. time is determined.
delta t-	initially set at 1ns but can be changed if the % change in the population with the largest % change is not within the limits (0.1- 1)%
old delta t-	the delta t used before % change in populations was checked
final time-	this value is taken from the last time listed in the array vi[ct4,1]
en-	this is the determined value of E/N at a particular stage in the calculation loop
lgain-	this is the determined value of the the gain length at a particular stage in the calculation loop after taking into account constriction
tdisch-	this is the determined value of the discharge thickness at a particular stage in the calculation loop after taking constriction into account.
time-	this is the incremental variable in the calculation loop. when the difference between "time" and the previous time in the result file -popres[ct7-1,0] - plus the present "delta t" is greater than 5ns results are put into popres[x]
chek1-	shows last calculated % population change
chek2-	shows maximum % population change found in this iteration

loop

```

dat one-
dat two-
dat three-    the stream numbers associated with each input data file
dat four-
dat five-

res one-
res two-      the stream numbers associated with the hard copy output files
res thr

step1-
step2-        "for" statement counters
step3-

d2-
d3-           associated with 'live' output display
d4-

loss-         the distributed transient absorption at this time(experimental)
old time-     time at which temperature change was last executed
ch time-      allows access to temperature changing routine only once/ns
gasfrac-      the fraction the gas density at this time is of the initial
               density (gas expansion due to local increase in T(gas))
total-        the total gas number density at time=0;
newtotal-     the total gas number density * gas frac
buffer-       the total initial buffer gas density * gasfrac
xenon-        the total initial Xenon number density * gas frac
hcl-          the total initial HCl number density* gas frac
ch temp-      the calculated change in the bulk gas temeprature
temp-         the bulk gas temperature
old temp-     the bulk gas temperature on previous exit from this routine
cpionch-      constant of proportionality for 3-body recomb. rate coefficient
current-      current at time (experimental value)
old current-  current at last access of this routine
volts-        volts at time (experimental value)
old volts-    volts at last acces of this routine
finish-       'live' option flag
raspberry-    error flag
sourgrapes-   error flag
select-       time interval at which data is selected
p-            the default 'live' plot variable

```

BOOLEANS:

```

b1-    TRUE if vdrift is constant
b4-    FALSE if Xe+ is charge-neutrality constrained
b5-    TRUE if HCl population is kept constant (unlimited source)
b6-    if TRUE allows access to data collection routine when t=0
b7-    if TRUE allows access to data collection routine at 'select' interval

```

Subscripted variables:

```

ct[1]-    number of initial values
ct[2]-    number of ordered pairs in rates vs. E/N
ct[3]-    number of constant rates
ct[4]-    number of loss(t), volt(t) and current(t) points
ct[5]-    number of Vdrift(E/N) points
ct[6]-    number of rates which are not constant
ct[7]-    a counter for iterating popres[0:ct7,0:38]

```

```

init[0:ct1]-    described in "DATONE"

```

```
del[1]= delta[HCl]           del[2]= delta[Xe+]           del[3]= delta[Xe*]
del[4]= delta[Il]            del[5]= delta[Cl-]           del[6]= delta[Cl]
del[7]= delta[XeCl*]          del[8]= delta[photons]
```

VARIOUS populations and parameters sampled every 5ns

```

popres[ct7,0]= time ( the calculation loop counter)
popres[ct7,1]= [Buffer]
popres[ct7,2]= [Xe]
popres[ct7,3]= [HCl]
popres[ct7,4]= [Xe+]
popres[ct7,5]= [Xe*]
popres[ct7,6]= (Rate of spontaneous emission/cm-3)*fraction of initial volume)
popres[ct7,7]= [Cl-]
popres[ct7,8]= [Cl]
popres[ct7,9]= [XeCl*]
popres[ct7,10]= laser power out
popres[ct7,11]= [electrons]
popres[ct7,12]= Rate of electron impact ionisation of Xe (ground state)
popres[ct7,13]= Rate of electron dissociative attachment to HCl
popres[ct7,14]= Rate of photoionisation of Xe*
popres[ct7,15]= Rate of photodetachment of electrons from Cl-
popres[ct7,16]= Rate of electron impact ionisation of Xe*
popres[ct7,17]= Rate of production of XeCl* via the ion channel
popres[ct7,18]= Rate of production of XeCl* via the excited channel
popres[ct7,19]= Rate of spontaneous emission
popres[ct7,20]= Rate of stimulated emission
popres[ct7,21]= Rate of electron impact excitation of Xe
popres[ct7,22]= Rate of electron impact de-excitation of Xe*
popres[ct7,23]= Rate of quenching of XeCl* by electrons
popres[ct7,24]= Rate of quenching of XeCl* by Xenon
popres[ct7,25]= Rate of loss of XeCl* by Xe2Cl formation
popres[ct7,26]= Rate of loss of photon density through output coupler
popres[ct7,27]= Transient loss (experimetal value)
popres[ct7,28]= Rate of loss of photon density (transient loss)
popres[ct7,29]= Rate of quenching of XeCl* by HCl
popres[ct7,30]= Rate of quenching of XeCl* by the buffer gas
popres[ct7,31]= Rate of loss of Cl- by 3-body processes not involving Xe+
                  (Depends on guess of % of positive ions which are Xe+)
popres[ct7,32]= Time dependent volume
popres[ct7,33]= E/N
popres[ct7,34]= Volts (experimental value - IxR(ballast))
popres[ct7,35]= Current (experimental value)
popres[ct7,36]= Bulk gas temperature
popres[ct7,37]= Gas fraction (expansion as a result of T increase)
popres[ct7,38]= Rate coefficient for ion channel (T(gas) dependence)

```

MAXIMUM values of each value of popres[0:ct7,0:11]

```

maxmin[0:38]- maximums of popres[0:ct7,0:38]
minmax[0:38]- minimums of popres[0:ct7,0:38]

```

A P P E N D I X F I V E

Optical Multichannel Analyser (OMA): detector head principles of operation and specifications

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3.1 BASIC VIDICON SCANNING

A typical vidicon detector target consists of a disk of silicon 16 mm in diameter, with a microscopic array of several million photodiodes on it (Figure III-1). The diodes have a common cathode and isolated anodes, which are selectively addressed by the scanning electron beam.

The beam serves both to read out signal from the target, and to recharge it. When light falls upon the target, electron-hole pairs are generated, which recombine to deplete the surface charge. As the beam scans the depleted area, a recharging current flows which is proportional to the depleted charge. This recharging current is sensed by the preamplifier, and integrated for one complete channel in the preamplifier to convert current back to charge.

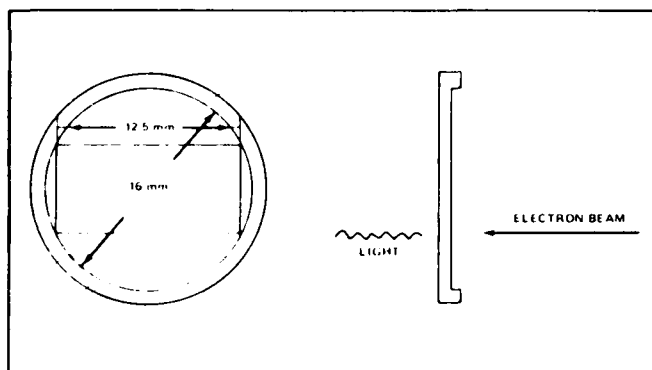


Figure III-1. SILICON TARGET

The OMA 2 Silicon Vidicon detectors are scanned over a 12.5 mm x 12.5 mm square area by an electron beam approximately 25μ in diameter. Figure III-2 shows the relationship between the electron beam and the Si Vidicon target. Scanning of the target progresses from bottom to top, left to right, making the target the equivalent of the first quadrant in a cartesian display. Parameters are 512 x 512 picture elements. A picture element is defined for our

purposes as the instantaneous area under the scanning electron beam. In practice, this is a circle approximately 25μ in diameter, encompassing a number of photodiodes.

A channel is a vertical line of picture elements, one picture element wide. It may have any starting point, from 0 to 511, and any height from 1 to 511, provided that the sum of the channel height ΔY and the starting position Y_0 do not exceed 511.

A track in the OMA 2 is a number of consecutively scanned adjacent channels. The channels per track may be as large as 512, or as small as 1. As with the channel height, the sum of the starting channel (X_0) and the track width (number of channels) may not exceed 512.

3.2 SIDE CHANNELS

Signal produced by the detector will be affected by the charge state of those picture elements immediately adjacent to the scanned track. For that reason, the OMA 2 system scans additional channels on either side of the programmed area. Data from these channels is not transmitted to the 1215 console. These extra or "side" channels account for the fact that the OMA 2 console deals with 500 point curves while the detector can be scanned over a 512 channel area. The twelve channels are used to remove the effect of charge bleed into the programmed scan area and hysteresis effects in the scanning beam. 9 Channels precede the desired scan, 3 follow it.

The side channels are programmed by the console regardless of the size of the track. Thus, if one channel were requested to be scanned, in fact 13 would be, but only one of them would yield data.

In producing complex multi-track or "2D" scan patterns, the operator must take these side channels into consideration. It is possible to overlap scanning of nearby tracks, without realizing that it's being done. The result of multiple scanning of a given area of the detector target is to reduce signal levels in the overlapped area.

3.3 DARK CURRENT

Dark current in the vidicon target is charge leakage from target due to thermal effects. Thus an unilluminated detector will exhibit a signal profile. This profile, plus whatever stray light is present in the experimental system, will form the background signal.

The level of dark current is a function of target temperature, among other things. In normal OMA 2 operation, presuming the detector is kept at a reasonably stable temperature, a background curve will remain valid for a substantial period of time.

Where the work is of a general exploratory nature, and numerous samples are to be taken, it is possible to acquire a background, take several samples in one format, then to take a second background curve at the conclusion of that run. Comparison of such background curves over as much as an hour will show very little difference. They are not likely to be as uniform, however, if the OMA 2 detector is subjected to radical change in the temperature of its operating environment. Where very precise numbers are sought, of course, such techniques are not recommended.

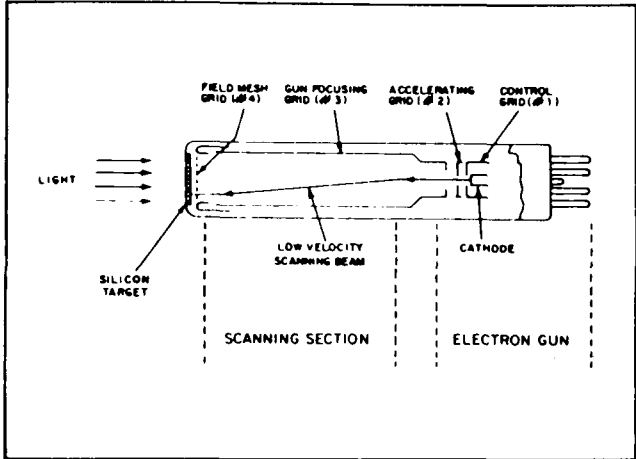


Figure III-3. SILICON VIDICON

3.4 INTENSIFIED DETECTORS

The Silicon Intensified Target (SIT) vidicon (Model 1254) and double intensified (ISIT) vidicon (Model 1256) differ from the basic detectors in that they employ one or two electrostatically focused image intensifiers prior to the vidicon target. An intensified target vidicon is shown in Figure III-4.

The incident light is imaged on a fiber optic facial plane and is carried to a photocathode, which emits photoelectrons. These photoelectrons are accelerated by the anode potential (up to 9 kV in our application). The high energy electrons bombard the Si target material, producing sub-

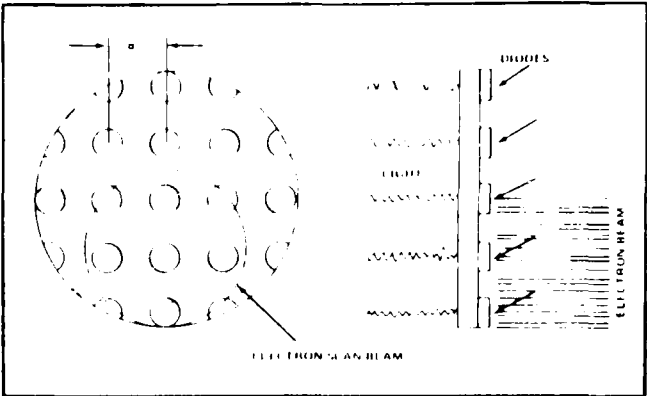


Figure III-2. SILICON TARGET EXPANDED VIEW

stantially more electron-hole pairs than would the incident light alone. This results in in-target amplifications of the signal. The amplification factor is approximately 1500. However, it is offset by the loss in the fiber optics, and quantum efficiency of the photocathode, such that net gain is on the order of 200. Figure I-1 displays the relative sensitivities of the intensified and non-intensified tubes, along with their spectral responses. The intensified SIT, or ISIT, couples an additional intensifier on the front, providing an optical gain of 10. This permits a preamplifier gain reduction of 5 which allows detection of a single photoelectron.

3.5 DETECTOR RESPONSE

Most of the vidicon detectors show a loss of efficiency below about 1100 nm and above 300 nm (Figure I 1). The 1252B, the least expensive of the available detectors, is useful over the entire visible spectrum, and into the near Ultraviolet and Infrared. The 1252B UV response falls off principally due to its glass window. The 1252G offers a quartz window which extends ultraviolet sensitivity to about 300 nm, although its response is identical to that of the 1252B in the region from 400 nm and longer.

The 1252E is designed for use at wavelengths from 600 nm to 1100 nm. Because of a thicker target, it is more sensitive at the longer wavelengths. The silicon is semi-transparent, however, and generates a Moire interference pattern, which shows up as spurious signals. This is suppressed by means of a special anti-reflective coating.

For applications where extended ultraviolet response is required, a scintillator is added to the SIT. This 1254/01 scintillator is a coating on the vidicon face which is virtually transparent to visible light, but which converts UV radiation to visible wavelengths such that they can be detected.

3.6 COOLED OPERATION

In applications where extremely small signals are to be detected, noise levels can be reduced by cooling the detector. Dry ice cooling using 1212 cooled housing allows

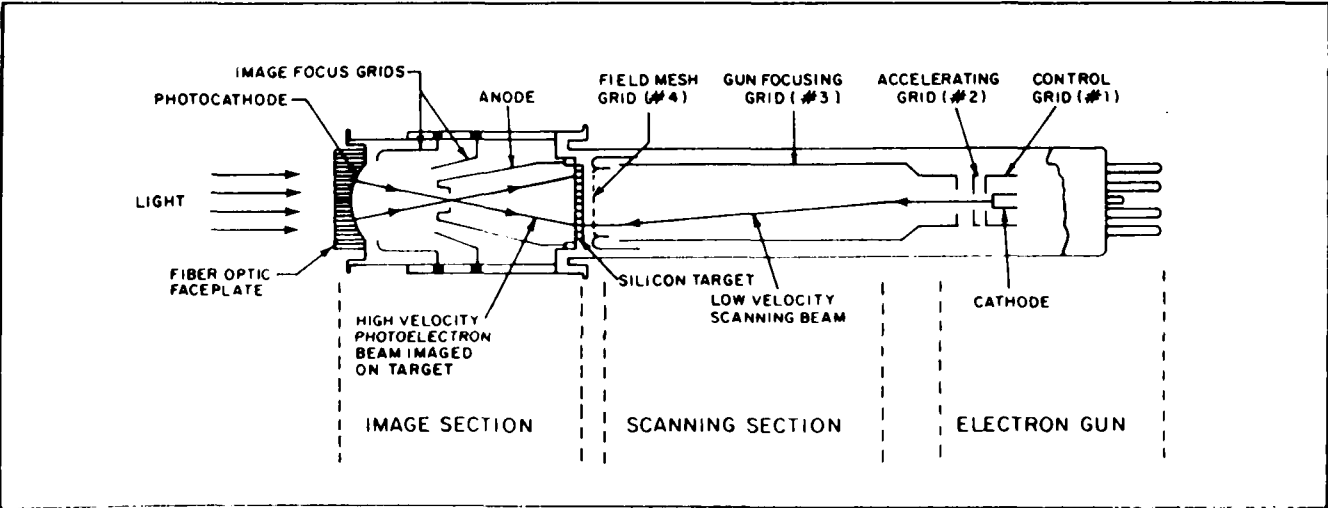


Figure III 4 SILICON INTENSIFIED TARGET VIDICON

1.2D 1254 SIT DETECTOR

SENSITIVITY: Approximately 2 detected photons per count.

BACKGROUND NOISE⁶: 1.5 counts rms max (1 count rms typical).

FULL SCALE⁷: 16,383 counts/channel, each frame.

DYNAMIC RANGE⁸: 1×10^4 typical for line spectrum

LINEARITY AS A FUNCTION OF INTENSITY: $\pm 2\%$.

GATING RATIO: 10^4 minimum.

UNIFORMITY OF SENSITIVITY⁹: $\pm 10\%$ typical (over center 10 mm dia. area of target with ΔY minimum of 250 channels).

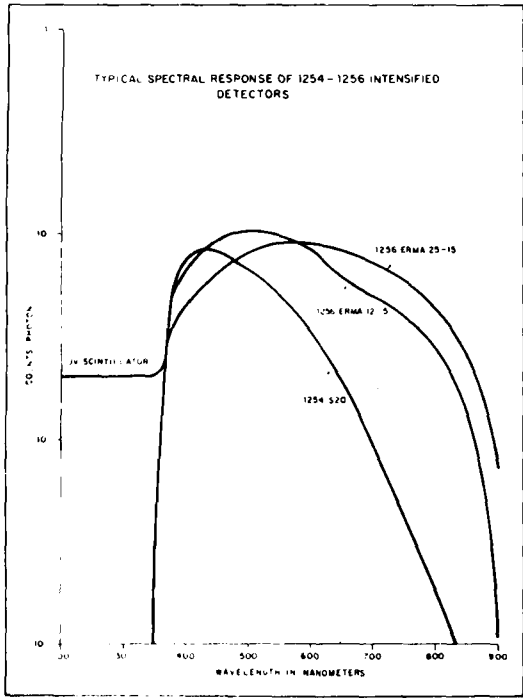
TARGET DIAMETER: 16 mm. Internal mask limits scan width and height to 12.5 mm.

GEOMETRIC DISTORTION: ± 2 channels (± 50 micron) typical between channels 100 and 400 for 2.5 mm scan centered pattern.

CHANNEL CROSS TALK⁶: Side channels $\leq 60\%$ of peak channel.

USEFUL SPECTRAL RANGE: See detector sensitivity curves

- 6. Measured using a scanned area of 3.50 x 12.5 mm and a frame time of 30 ms.
- 7. Minimum for an image height of 3.50 mm and a frame time of 30 ms
- 8. Dynamic range = full scale signal/background rms noise
- 9. Minute blemishes can cause a non uniformity in excess of 5% for channel data taken with very small scanned areas. However, the computational capability of the Model 1215 can eliminate these effects.



A P P E N D I X S I X

Compilation of experimental absorption spectra around 467 nm and 484 nm

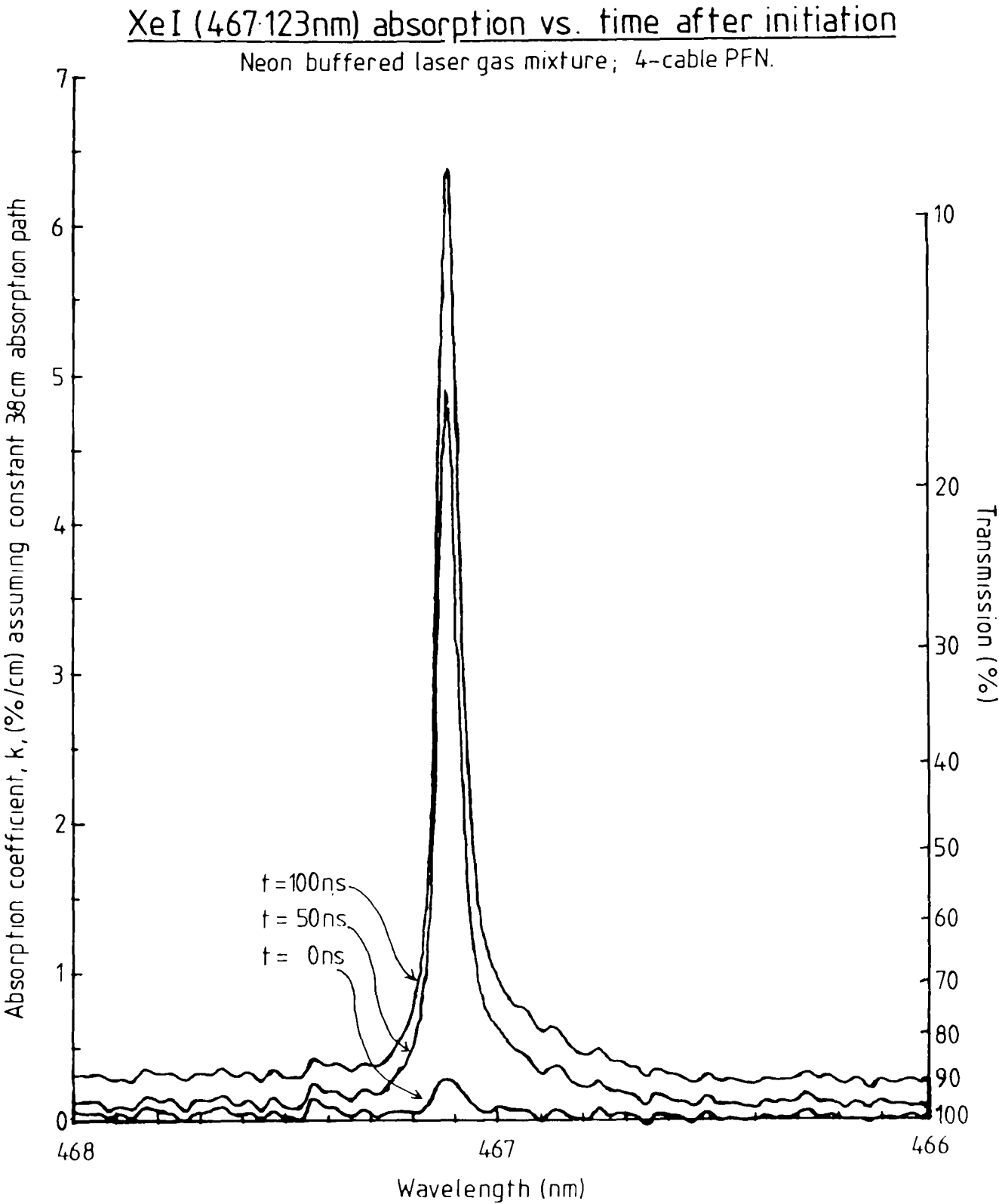


Figure A6.1

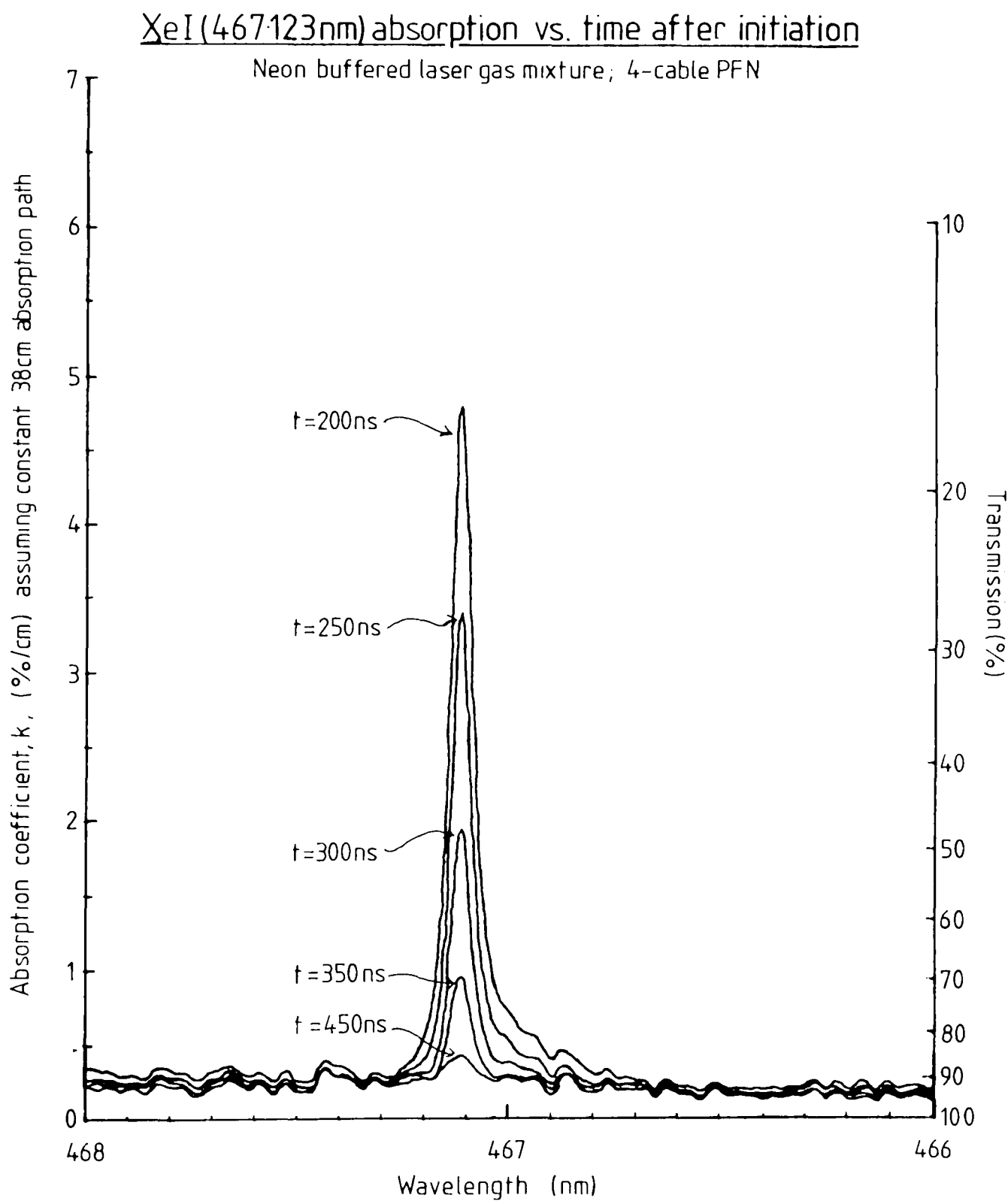


Figure A6.2

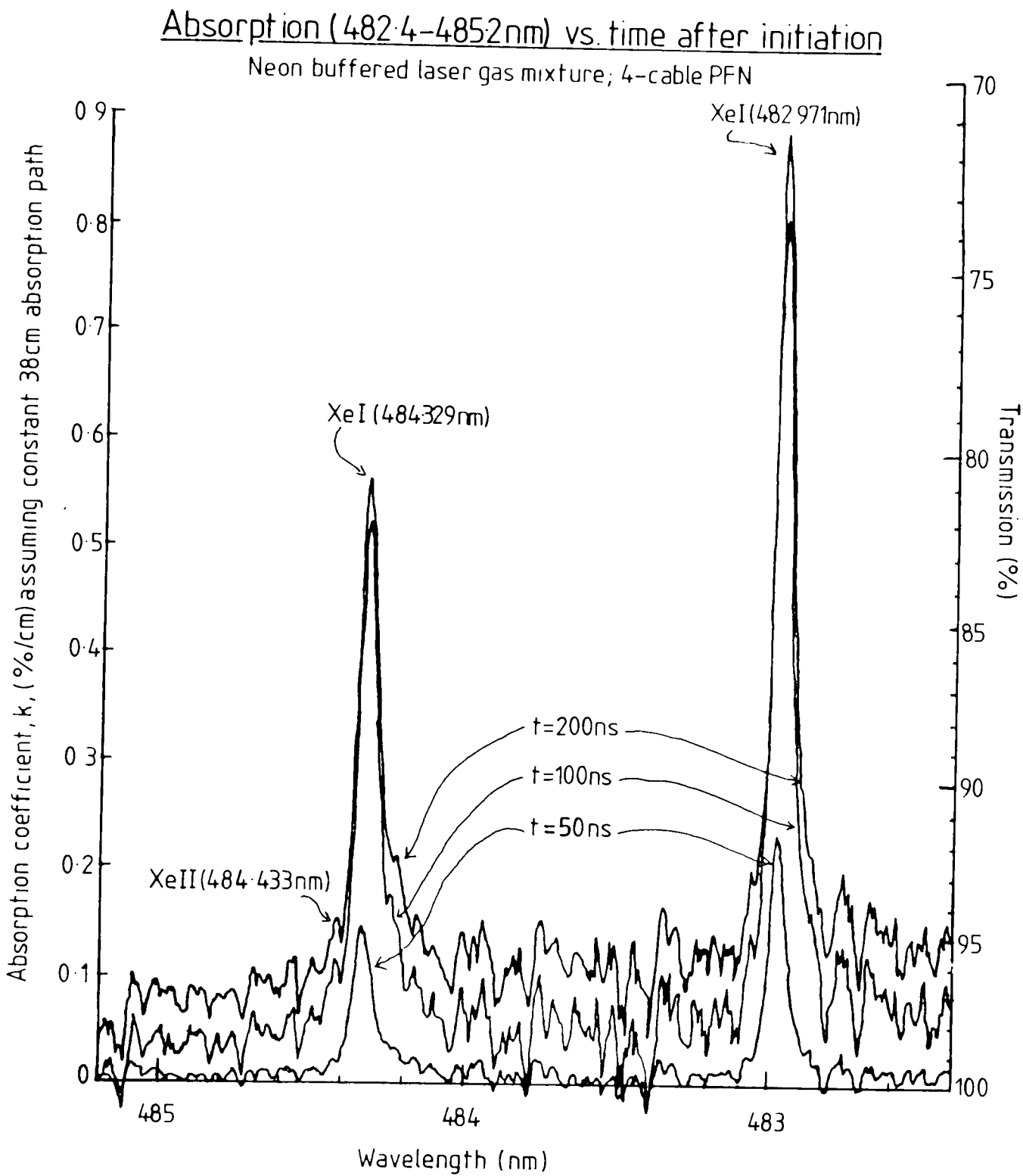


Figure A6.3

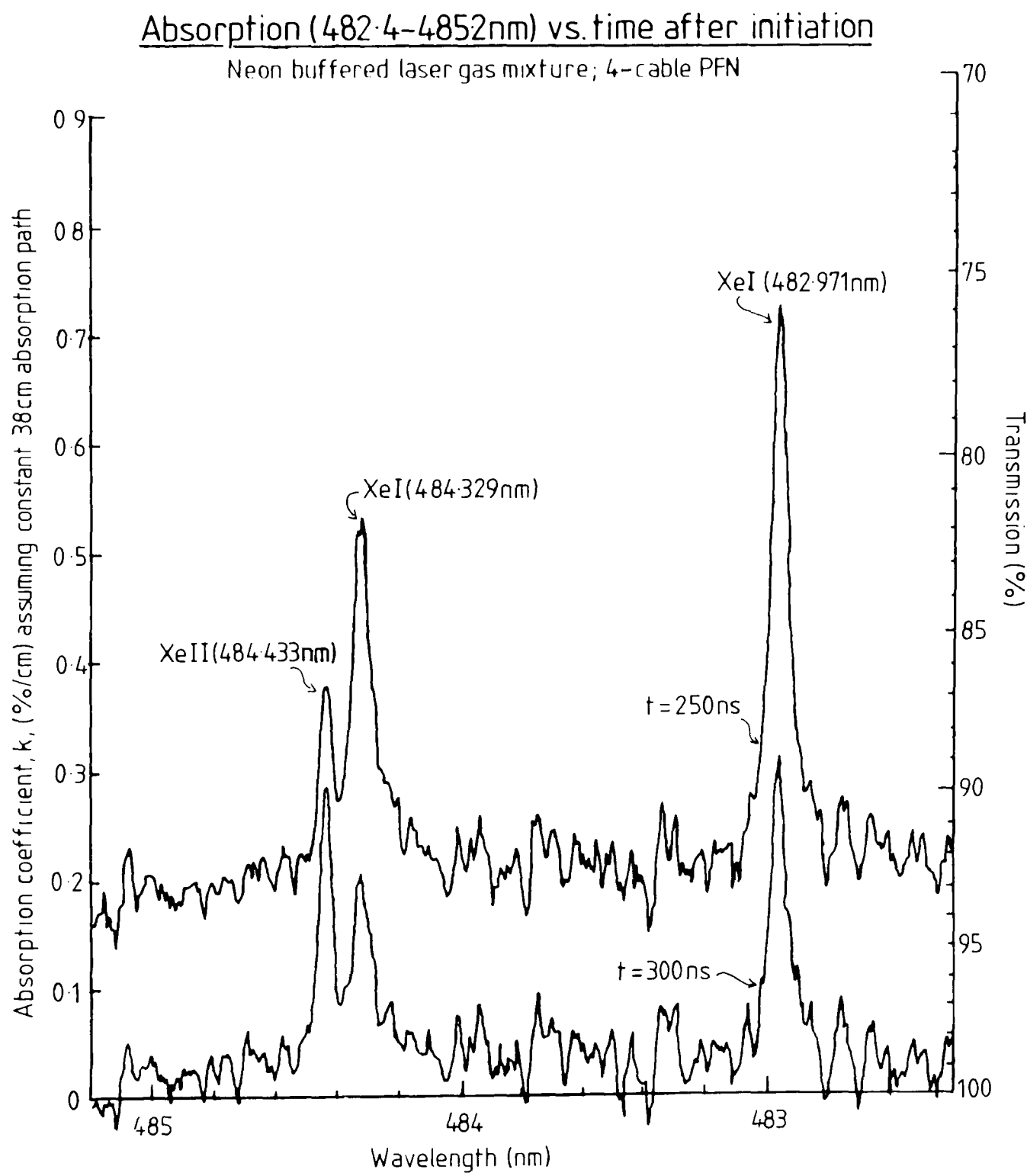


Figure A6.4

Absorption (482.4-485.2nm) vs. time after initiation

Neon buffered laser mixture; 4-cable PFN

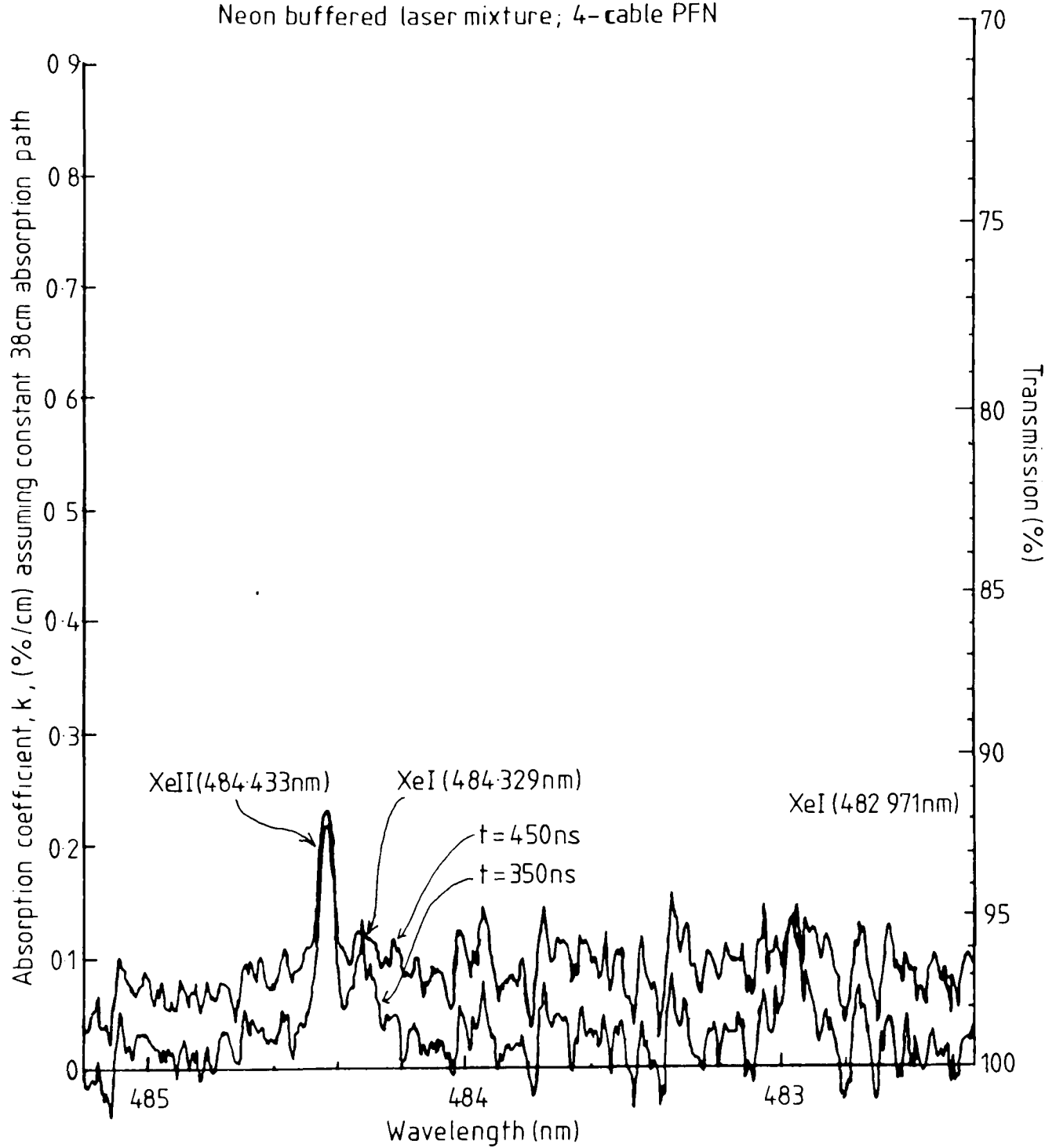


Figure A6.5

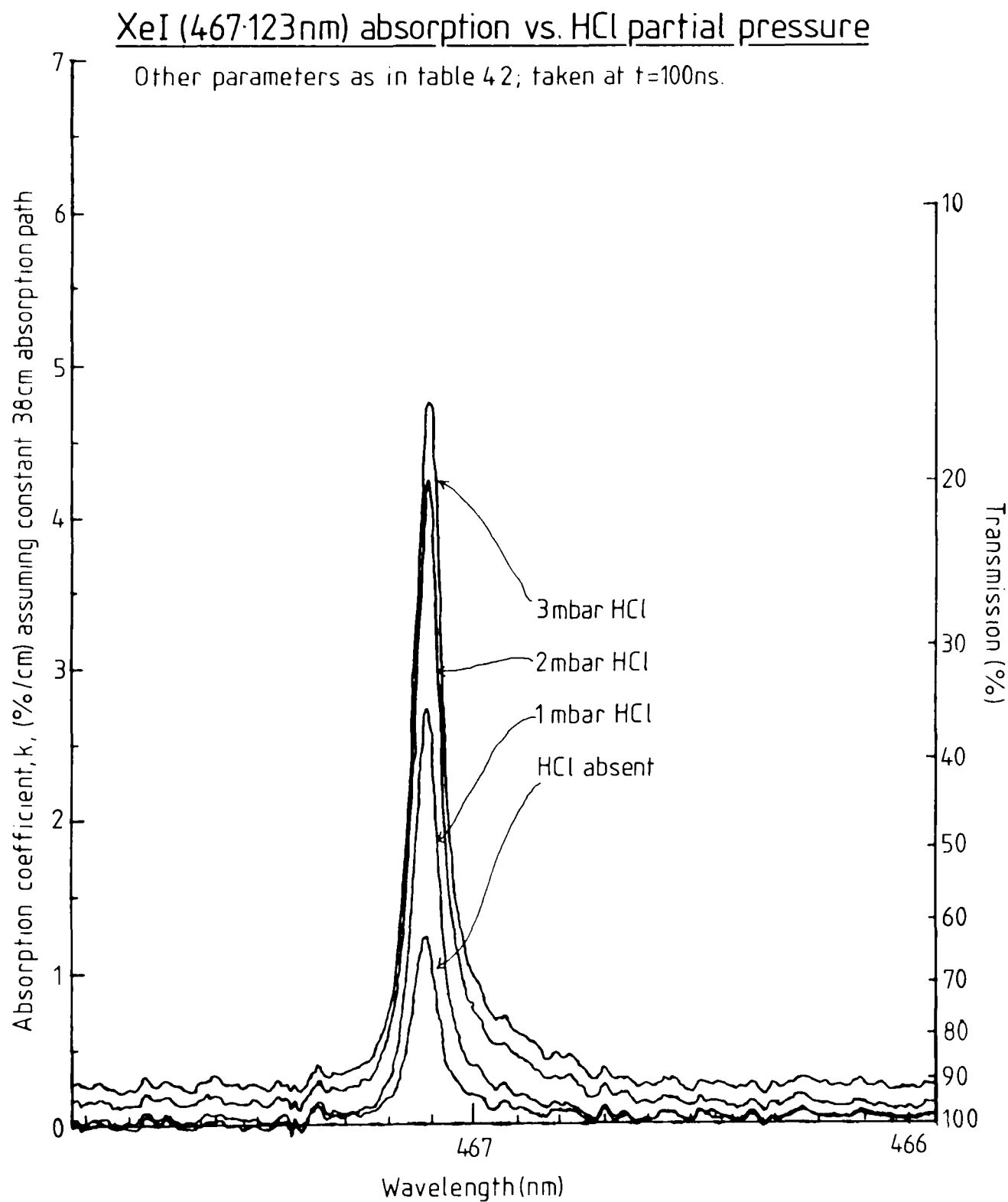


Figure A6.6

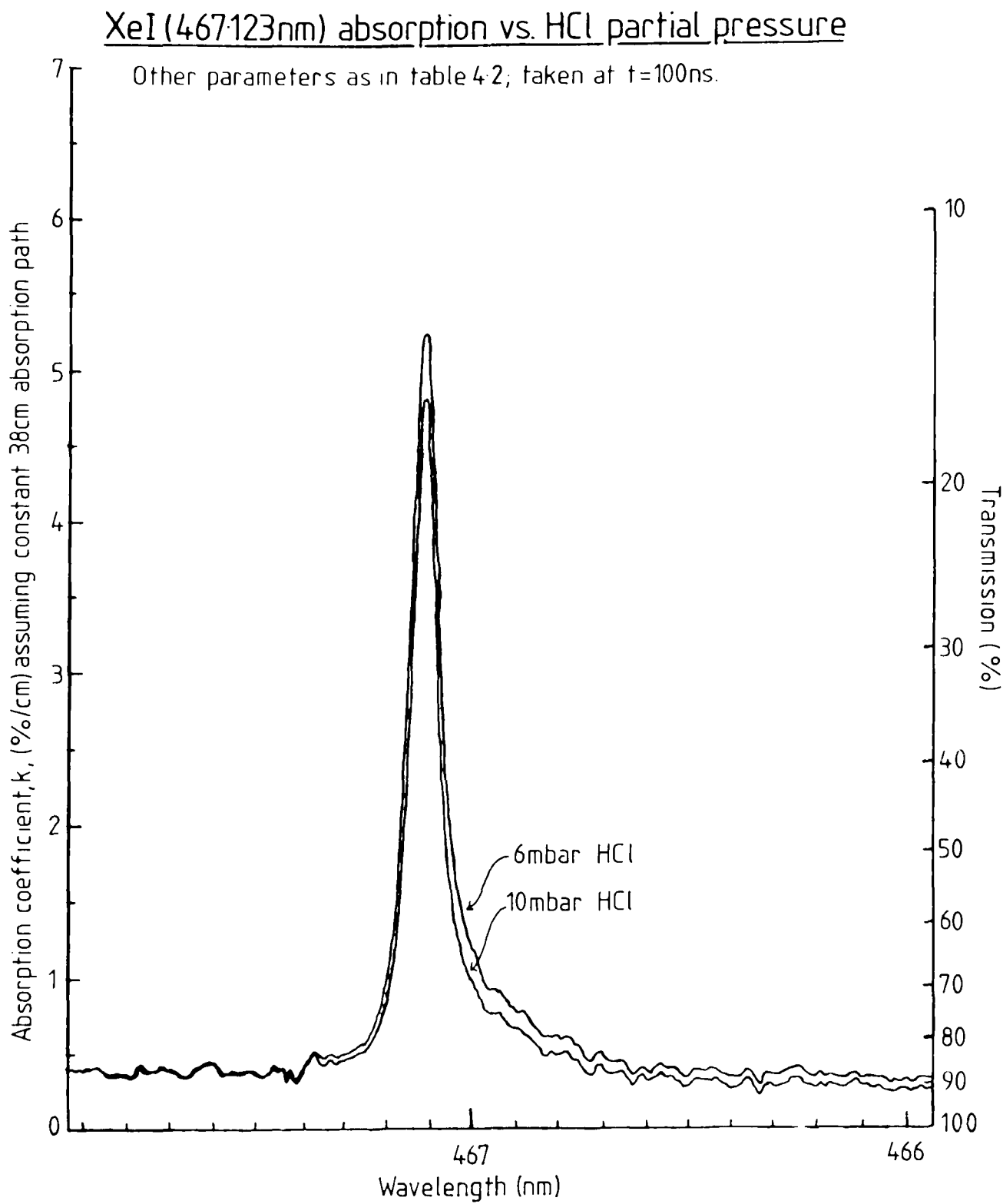


Figure A6.7

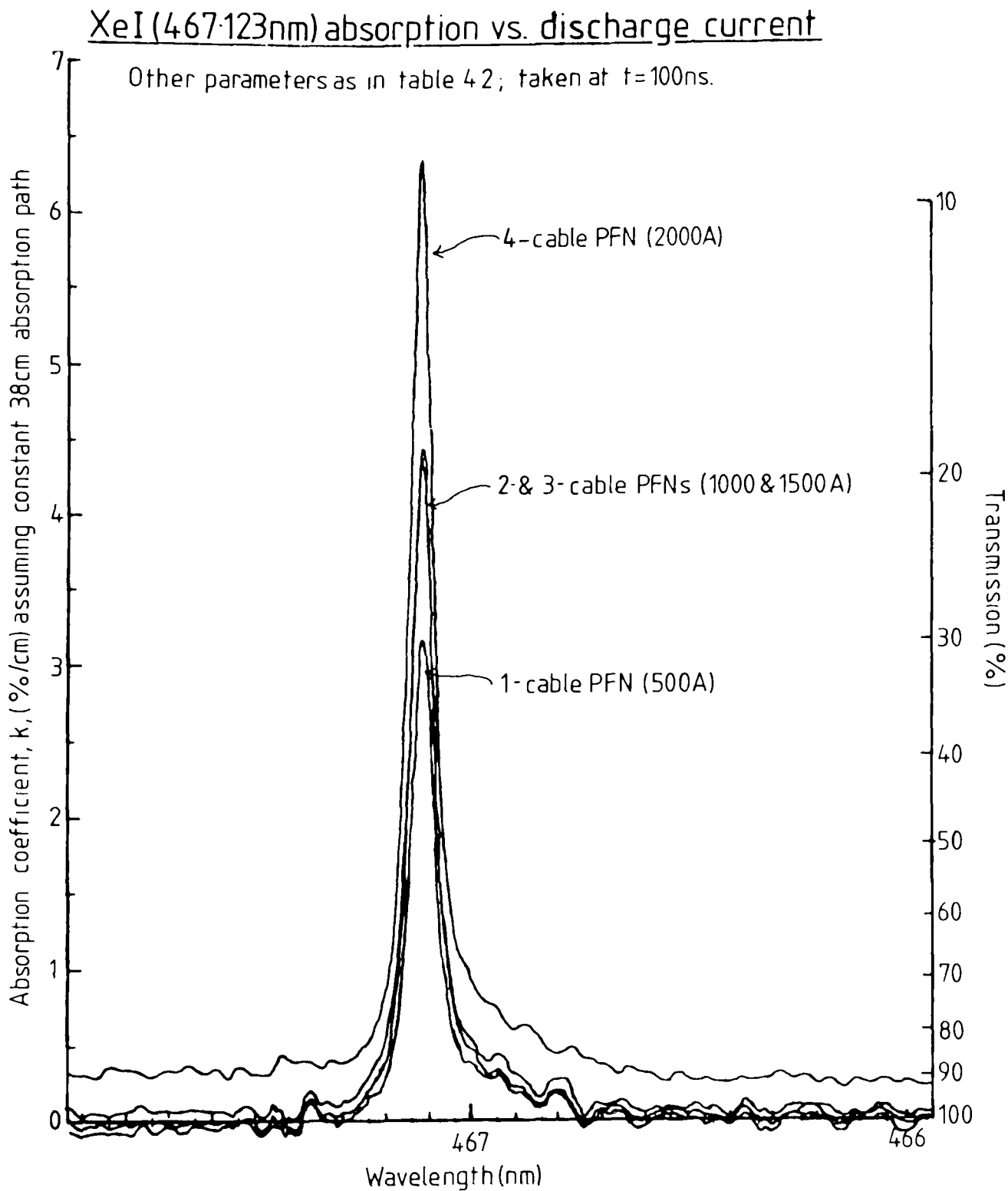


Figure A6.8

A P P E N D I X S E V E N

Completion of the derivation of the stability criterion of Daugherty et al.[1976]

In this appendix, the derivation of the stability criterion described by eq.2.1, which was started in section 2.2.2, will be completed. It was claimed by Daugherty et al.[1976] that this criterion described the onset on instabilities in discharge-excited RGH lasers. The criterion has been shown to be unrealistic in this thesis and this derivation is only included for the sake of completeness. Continuing now from where the discussion left off in section 2.2.2 (this derivation is due to Webb[1976a]):

We now make the additional assumption that:

- (5) during the early stages of the instability, when the electron density is only just starting to 'run away', the two terms of eq.2.3 are much larger than dn_e/dt , i.e.

$$\frac{d}{dt} [Kr^*] \approx 0$$

and,

$$[Kr^*] = \tau_d \langle \sigma v \rangle_* [Kr] n_e = \alpha' n_e \quad (A7.1),$$

where

$$\alpha' = \langle \sigma v \rangle_* \tau_d [Kr] \quad (A7.2),$$

and substituting from eq.A7.4 into eq.2.2

$$v_1 = \alpha' \langle \sigma v \rangle_1 n_e = \alpha n_e \quad (A7.3),$$

where

$$\alpha = \alpha' \langle \sigma v \rangle_1 \quad (A7.4).$$

The master equation becomes

$$\frac{dn_e}{dt} = S_{eb} + (\alpha n_e - \beta) n_e \quad (A7.5)$$

where S_{eb} is the contribution to the rate of change of electron density from the e -beam sustainer of Daugherty et al.'s device. The solution of eq.A7.5 will lead to some equilibrium value of electron density, $n_e(0)$. If a small harmonic perturbation occurs in n_e , then

$$n_e = n_e(0) + \delta n_e$$

where $\delta n_e = n_e(0) \cdot e^{-i\omega t}$.

So,
$$\frac{d}{dt} (n_e(0) + n_e) = S_{eb} - \beta(n_e(0) + \delta n_e) + \alpha(n_e(0) + \delta n_e)^2$$

but,
$$\frac{d}{dt} (n_e(0)) = 0, \text{ and } S_{eb} - \beta n_e(0) + \alpha n_e(0)^2 = 0,$$

thus,
$$\frac{d}{dt} (n_e) = -\beta \delta n_e + 2\alpha n_e(0) \delta n_e,$$

where terms in δn_e^2 have been neglected. Thus,

$$-i\omega = -\beta + 2\alpha n_e(0).$$

But the condition for the perturbation to damp out as a function of time is

$$\text{Im}(\omega) < 0$$

which gives

$$\beta \geq 2\alpha n_e(0) \text{ or equivalently } \beta \geq 2\nu_i,$$

which is equation 2.1, as the condition for stability.

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XeI Term Diagram - Racah Notation - j-j coupling

Core configuration: $5p^5(^2P^o_{3/2})$ - undashed; $5p^5(^2P^o_{1/2})$ - dashed. (x) denotes 'x' levels too close to resolve here.

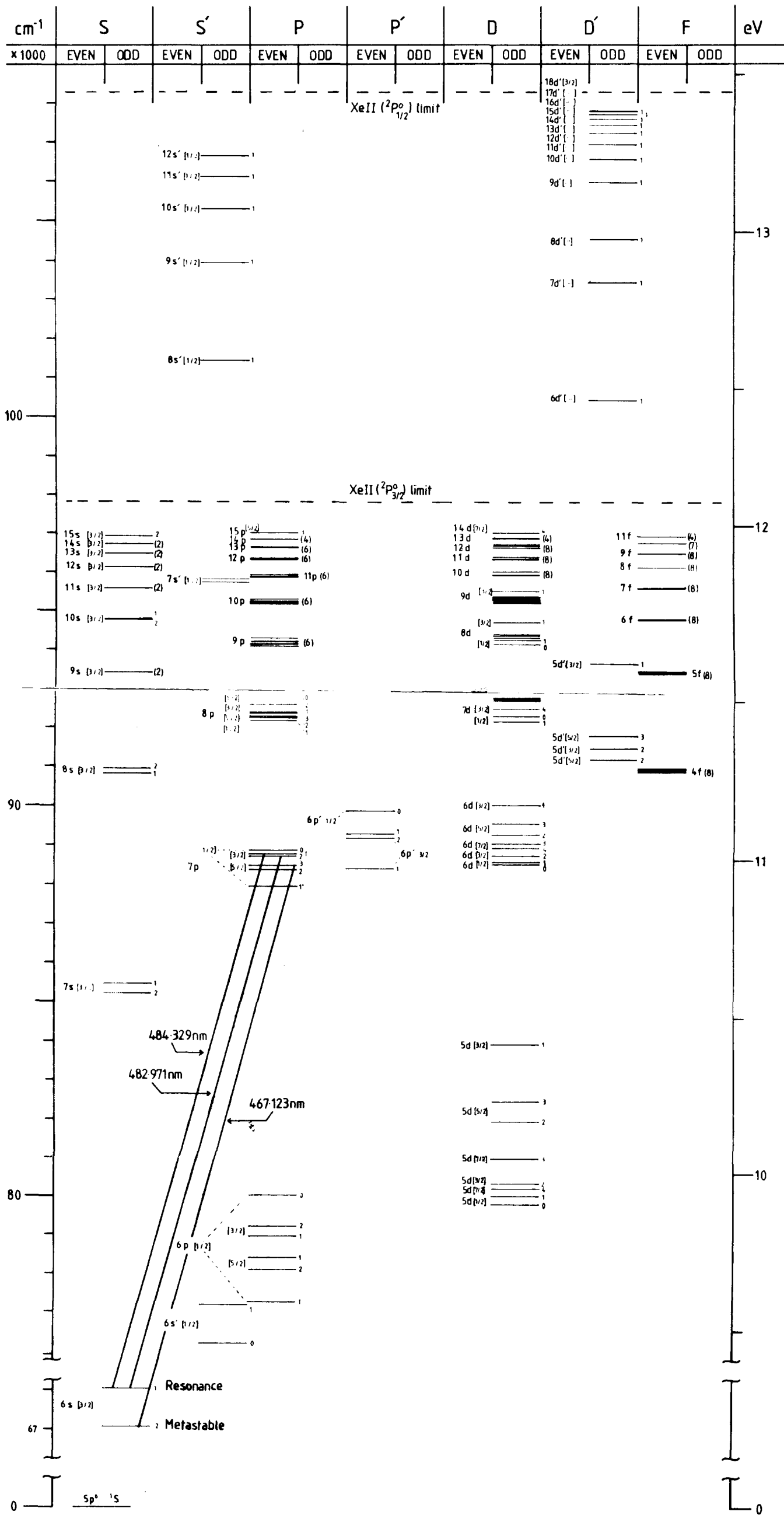


FIGURE 5.20