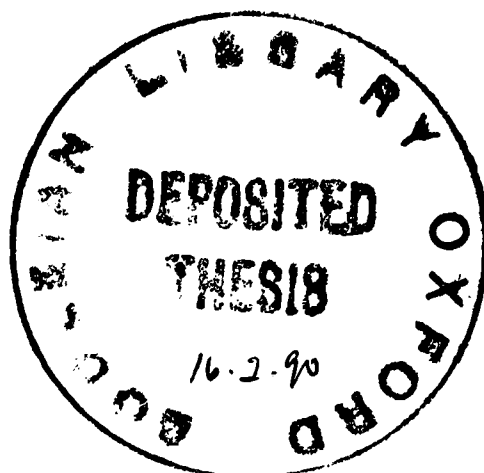


Spectroscopic Applications of Pulsed Metal Vapour Lasers

by

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New College
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A thesis submitted for the degree of
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The Clarendon Laboratory, Oxford.
Trinity Term, 1989.

"Spectroscopic applications of pulsed metal vapour lasers"
T G M Freearde, D Phil Thesis, 1989.

The author is aware of the following errata.

PAGE

40 The units of columns 2 and 3 of table IV-1 should be mm and μ rad's respectively.

51 The ellipsoid of wave normals is not a plot of refractive index against wavevector, although it defines this dependence. The surface is instead correctly defined by equation (V-9). Figure V-2 shows the refractive indices derived from the ellipsoid of wave normals.

52 The beginning of the final paragraph should read, "Phase matching is therefore achieved when the curves of figure V-2 for the fundamental and harmonic intersect."

56 In the format of the following calculations, equation (V-15) should read

$$E = E_0 \frac{\exp (-i k z)}{[(1+i \tau_x)(1+i \tau_y)]^{\frac{1}{2}}} \exp \left\{ -\left[\frac{x^2 / w_x^2}{1+i \tau_x} + \frac{y^2 / w_y^2}{1+i \tau_y} \right] \right\} \tag{V-15}$$

57 In S.I. units, the constant K should be given by

$$K = 2 \omega_1^2 d_{\text{eff}}^2 / \pi \epsilon_0 c^3 \eta_1^2 \eta_2$$

57 In equation (V-16), α_z should read α_1 .

58 In equation (V-17), α should read α_1 .

58 Equation (V-19) should read

$$E_2(x, y, z) = \frac{2 \pi i \omega_2 P_{0x} \exp (-\frac{1}{2} \alpha_2 l + 2 i k_1 z)}{4 \pi \epsilon_0 c \eta_2 (1+i \tau_x)^{\frac{1}{2}} (1+i \tau_y)^{\frac{1}{2}}} \int_0^l \frac{\exp (-\alpha z' + i \Delta k z')}{(1+i \tau_x')^{\frac{1}{2}} (1+i \tau_y')^{\frac{1}{2}}} \cdot \exp -2 \left[\frac{[x-\rho(l-z')]^2 / w_x^2}{1+i \tau_x} + \frac{y^2 / w_y^2}{1+i \tau_y} \right] dz' \tag{V-19}$$

M.S. D. Phil. c. 7828.

59 The final term in equation (V-23) should be [order τ_x^{-1}].
59 Equations (V-24) to (V-33) contain a few typographical errors and require conversion to the S.I. unit system. They should read

$$\begin{aligned} E_2(x,y,z) = & \frac{2\pi\omega_2\mathbf{P}_{0X}}{4\pi\epsilon_0c\eta_2}\cdot\frac{i}{(1+i\tau_x)^{\frac{1}{2}}(1+i\tau_y)^{\frac{1}{2}}}\exp\left[-\tfrac{1}{2}\alpha_2l+2ik_1z\right] \\ & \cdot\exp{-2\left[s^2(1-i\tau_x)+s'^2(1-i\tau_y)\right]}\cdot\int_0^l\exp{4i\beta\tau_x's}\frac{\exp(-\alpha z'+i\Delta kz')}{(1+i\tau_x')^{\frac{1}{2}}(1+i\tau_y')^{\frac{1}{2}}}dz' \end{aligned} \tag{V-24}$$

$$\begin{aligned} E_2(x,y,z) = & \frac{\omega_2\mathbf{P}_{0X}}{2\epsilon_0c\eta_2(\tau_x\tau_y)^{\frac{1}{2}}}\exp\left[2ik_1z-\tfrac{1}{2}\alpha_2l\right]\exp{-2\left[s^2(1-i\tau_x)+s'^2(1-i\tau_y)\right]} \\ & \cdot\int_0^l\exp{4i\beta\tau_x's}\frac{\exp(-\alpha z'+i\Delta kz')}{(1+i\tau_x')^{\frac{1}{2}}(1+i\epsilon^2\tau_x')^{\frac{1}{2}}}dz' \end{aligned} \tag{V-25}$$

$$H = \frac{1}{2\pi}\int_{-\epsilon_x(1-\mu)}^{\epsilon_x(1+\mu)}\frac{\exp{-\kappa\tau_x'}\exp{i\sigma'\tau_x'}}{(1+i\tau_x')^{\frac{1}{2}}(1+i\epsilon^2\tau_x')^{\frac{1}{2}}}d\tau_x' \tag{V-26}$$

Please note the missing subscripted x in the definition

$$\sigma' = \sigma_x + 4\beta s$$

$$H = \frac{1}{\pi b_x}\exp\left[\alpha f-i\Delta kf\right]\int_0^l\frac{\exp(-\alpha z'+i\Delta kz')\exp(4i\beta s\tau_x')}{(1+i\tau_x')^{\frac{1}{2}}(1+i\tau_y')^{\frac{1}{2}}}dz' \tag{V-27}$$

$$E_2(x,y,z) = \frac{2\omega_2\mathbf{P}_1b_x}{\epsilon_0\eta_1\eta_2c^2w_xw_y(\tau_x\tau_y)^{\frac{1}{2}}}\mathbf{d}_e\Gamma\Gamma\exp\left[-\frac{\alpha'}{2}l+\frac{\alpha l\mu}{2}-2(s^2+s'^2)\right]\cdot\mathbf{H} \tag{V-28}$$

where $\mathbf{P}_{0x} = \epsilon_0\mathbf{d}_{eff}\mathbf{E}_0^2 = 4\mathbf{P}_1\mathbf{d}_{eff}/[\pi\eta_1cw_xw_y]$. The second harmonic intensity is now defined by $\mathbf{S} = [\epsilon_0\eta_2c/2]\cdot|\mathbf{E}_2|^2$

$$\mathbf{S}(x,y,z) = \frac{2\omega_1^2}{\epsilon_0c^3\eta_1^2\eta_2}\mathbf{d}_{eff}^2\frac{4\pi\mathbf{P}_1^2k_1^2}{\tau_x\tau_y}\left[\frac{w_x}{w_y}\right]^2\exp\left[-\alpha'l+\alpha l\mu-4(s^2+s'^2)\right]|\mathbf{H}|^2 \tag{V-29}$$

$$\begin{aligned} P_2 = & \iint \mathbf{S}\,dx\,dy = w_xw_y\tau_x\tau_y\iint \mathbf{S}\,ds\,ds' \\ & \dots \text{ cont'd} \end{aligned}$$

$$= 4\pi K P_1^2 k_1^2 w_x w_y \left[\frac{w_x}{w_y} \right]^2 \exp \left[-\alpha' l + \alpha l \mu \right] \int_{-\infty}^{\infty} \exp -4s'^2 ds' \int_{-\infty}^{\infty} \exp -4s^2 |H|^2 ds \tag{V-30}$$

where $K=2\omega_1^2 d_{eff}^2 / [\epsilon_0 \pi c^3 \eta_1^2 \eta_2]$.

$$P_2 = K P_1^2 k_1 \exp(-\alpha' l) \left[\frac{\pi^2}{\epsilon_x} \exp \mu \alpha l \left[\frac{2}{\pi^{\frac{1}{2}}} e \int_{-\infty}^{\infty} \exp \left[-4s^2 \right] |H|^2 ds \right] \right] \tag{V-31}$$

where

$$H = \frac{1}{2\pi} \int_{-\epsilon_x(1-\mu)}^{\epsilon_x(1+\mu)} \frac{\exp -\kappa \tau_{x'} \exp i \sigma' \tau_{x'}}{(1+i\tau_{x'})^{\frac{1}{2}} (1+i e^2 \tau_{x'})^{\frac{1}{2}}} d\tau_{x'} \tag{V-32}$$

$$h_m = \left[\frac{\pi^2}{\epsilon_x} \exp(\mu \alpha l) \left[\frac{2}{\pi^{\frac{1}{2}}} e \int_{-\infty}^{\infty} \exp(-4s^2) |H|^2 ds \right] \right] \tag{V-33}$$

Equation (V-37) should read

$$\left| \left[\frac{1}{(1+i\tau)(1+ie^2\tau)} \right]^{\frac{1}{2}} \right|^2 = \frac{1}{(1+\tau^2)^{\frac{1}{2}} (1+e^4\tau^2)^{\frac{1}{2}}} \tag{V-37}$$

Table V-1. All angles are given in degrees. The acceptance angle Θ_{acc} is the full angle, defined for critical phase-matching by $\Theta_{acc}=\lambda/(\rho\eta_1 l)$ and equal to twice the value defined in equation (V-14) on p.55. Note that d_{11} for BBO mistakenly appears in the column for urea.

The final line of text should read, "amplitude radius w_0 is related to the radius, w_1 , at the focussing lens by" Kindly disregard the reference to "the figure below".

It is stated at the end of the first paragraph that practical laser dyes are limited to wavelengths above about 250nm. In practice, there are no dyes available for use below about 300nm.

The phrase, "as the dispersive elements," should be deleted from line 4.

The penultimate stage of the alignment procedure is to minimize, rather than maximize, the laser bandwidth.

Table VIII-2. The unit of the "corresponding peak absorption" is cm^{-1} .

The calculations of the acceptance angles of nonlinear crystals, in chapter V of this thesis, assume that the fundamental radiation is a plane wave. The effect of using focussed Gaussian beams is discussed by S Blit, E G Weaver and F K Tittel, "Wavelength temperature and angle bandwidths in SHG of focused beams in nonlinear crystals," Appl Opt 18 (5) 733-6 (1979)

**To my parents,
with many thanks.**

Abstract

Spectroscopic applications of pulsed metal vapour lasers.

T.G.M.Freearde, New College, Oxford.

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The early part of this thesis reports time-resolved measurements of the transverse coherence of radiation from a copper vapour laser (CVL), and shows the influence of an unstable resonator. Experiments to measure the visibility of fringes from pairs of pinholes with a range of spacings are described, and their results are shown to be consistent with a simple theory based upon geometrical optics. With a high magnification laser cavity, high transverse coherence can be achieved before the laser pulse has reached appreciable power. The average coherence over a number of pulses is found to be limited, however, because of distortions introduced by currents of warm air in the beam path. Substantial improvements have now been made.

The importance of the transverse coherence upon second harmonic generation is then investigated. By improving the transverse coherence of the laser beam, second harmonic generation has been demonstrated at efficiencies close to those theoretically predicted for stable Gaussian laser beams, and 200mW of ultraviolet radiation at 255nm has been produced with 10% conversion efficiency using the nonlinear material beta-barium borate (BBO). High birefringence in BBO causes cylindrical symmetry in the geometry for harmonic generation to be lost, and it has been suggested that the use of astigmatic focussing into the crystal might be advantageous. A theoretical analysis has therefore been made, and improvements are indeed predicted. Elliptical focussing is predicted to increase the typical harmonic power only by a factor of 1.3, however, and no experimental verification has been attempted.

The remainder of this thesis considers the longitudinal coherence of the copper vapour laser, and describes the design and initial performance of a narrow bandwidth dye laser for use with a CVL. Careful construction of a grazing-incidence grating arrangement similar to that of Littman has resulted in a laser which is particularly simple to align and which promises ease of tuning over a wide range. Operation confined to a single longitudinal mode has been demonstrated, corresponding to single shot bandwidths below 1GHz, but instabilities caused by vibration and uneven dye flow broaden the time-averaged width to around 3GHz. Suggested improvements have not yet been implemented.

I welcome the opportunity to thank

Professors Bill Mitchell, Bill Hayes, Pat Sandars and Roger Cowley for the use of the facilities of the Clarendon Laboratory,

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Tim Freegarde
Clarendon Laboratory
August 1989.

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Chapter I.

Introduction.

I-1. Spectroscopic applications of copper vapour lasers.

I-2. The layout of this thesis.

Introduction.

The copper vapour laser is the best known of the metal vapour lasers, and is the most powerful and efficient source of coherent visible radiation currently available. A single commercial device can now yield mean powers above 100W at efficiencies in excess of 1%, with a pulse energy above 25mJ and peak powers reaching 400kW. Devices are large but rugged, and a wide beam and high gain make alignment simple. The gain is sufficient for the laser to be used as a single-pass amplifier, for both power extraction and image intensification.

Laser oscillators, however, are also characterized by the longitudinal and transverse coherence of their radiation, and in this respect early copper lasers, and metal vapour lasers in general, have been particularly poor. Before the work described in this thesis was started, the copper vapour laser (CVL) was generally considered a source of high brightness, fixed wavelength radiation, and found application in dye laser pumping and image projection, where it replaced conventional high intensity lamps. The transverse coherence of the laser beam was poor, and although an unstable resonator configuration was known to offer an improvement, no such system had been characterized. Translation of the laser power to shorter wavelengths by nonlinear optical processes had been given only a cursory investigation yielding powers no greater than 20mW, and the development of high resolution CVL-pumped dye lasers was largely in its infancy. Little thought appeared to have been given to the longitudinal coherence of the laser beam and the potential for holographic applications, and virtually no importance had been attached to any structure within a single laser pulse; the copper laser was no more than a source of high intensity coloured flashes, and its applications were correspondingly limited.

In more recent years, the potential of the copper vapour laser as a high repetition rate source for laser spectroscopy has been recognized, and interest has been aroused in shifting the fixed wavelength laser emission to more useful regions of the spectrum. Pulsed laser spectroscopy is particularly useful for the investigation of nonlinear processes whose efficiency increases with the intensity of the probe beam, and although systems based around pulsed Nd:YAG lasers or, at shorter wavelengths, nitrogen and excimer lasers, are common, these generally run at pulse repetition rates below 10Hz and the time required to acquire a given data set can be considerable. Further applications of the copper vapour laser use its efficient combination of peak power and relatively high duty cycle, resulting from the high repetition frequency, and

indeed techniques such as isotope separation and resonance ionization mass spectrometry have motivated much of the development of CVL pumped dye lasers.

The work described in this thesis has been aimed at assessing the suitability of the copper vapour laser for use in a number of applications. The conclusions are of general relevance to spectroscopic applications of the range of metal vapour lasers, which share the characteristics of a long laser tube of large bore and a short laser pulse at high repetition rate.

I-1. Spectroscopic applications of copper vapour lasers.

A pulsed laser source for spectroscopic applications must fulfil several requirements. The system must produce pulses at an adjustable but otherwise stable wavelength; the pulse shape and timing must be reproducible; and the pulse energy should not vary. Wavelength tuning should be easy and its range should be large. The pulse energy and power should exceed any threshold levels in the experiment and yield a useful signal. Finally, the beam should have good focussing properties and longitudinal coherence.

The copper laser satisfies a number of these requirements and can form a good starting point for a spectroscopic system. Laser pulses of several millijoules can be produced under external control, at repetition rates in the typical range 3–30kHz and with timing instabilities of only 1ns. Pulse energies are generally stable to within a percent, and the pulse shape is reproducible and lasts around 30ns.

However, since the laser only operates at a pair of fixed wavelengths in the visible (511 and 578nm), conversion to tunable radiation is necessary. The most common route is via a dye laser, in which a solution of organic dye is pumped optically by the copper vapour laser and the wavelength of the emission is determined by suitable dispersive feedback components. If shorter wavelengths than those produced by the copper laser are required, nonlinear frequency conversion processes may be used: the dye laser output itself may be converted, or harmonic radiation derived directly from the copper laser may be used to pump a short wavelength dye. Alternatively, the frequencies of pump and dye lasers may be mixed to yield their sum. Dye laser systems using these principles are available commercially, but none is recommended for use with, or at the repetition rate of, the copper vapour laser.

As there are few sources of pulsed ultraviolet radiation with kilohertz repetition rates, there has been considerable interest in its own right in the efficient generation of harmonics of the copper laser frequencies. Spectroscopic applications in addition promoted an interest in copper laser pumped dye lasers of high resolution, and the combination of these two areas promised tunable radiation at wavelengths as low as 270nm. The original plan with the work reported in this thesis was therefore both to

develop a dye laser with tunable visible emission, and to investigate the nonlinear generation of ultraviolet radiation at the harmonics of the copper laser lines. Some combination of these arrangements was then to be used in a simple experiment to demonstrate the use of the copper vapour laser as a tool for spectroscopic research. Work commenced with the design of a high resolution CVL-pumped dye laser.

In the event, a painful delay in the manufacture of the dye laser allowed attention to be devoted to the problems of second harmonic generation, and little work has in fact been done to develop the dye laser to its full potential. Second harmonic generation was found to depend strongly upon the coherence characteristics of the copper laser, and effort was therefore put into determining and improving the CVL's transverse coherence. A complete study of second harmonic generation was then attempted, and only in the final weeks was limited attention given to the dye laser.

I-2. The layout of this thesis.

Chapters II to IV of this thesis are concerned with the copper laser and its coherence properties. Chapter II sets the scene by describing the construction and principles of operation of the CVL: a brief overview of the performance and characteristics of a typical device is given, and specific data are presented for the commercial model used in these experiments. The laser dynamics and atomic mechanisms are described in chapter III, and are used to predict the laser bandwidth and longitudinal coherence. Experimental determination of the actual longitudinal coherence is reported, and the implications for holography using copper laser radiation are considered.

Chapter IV then deals with the transverse coherence of the CVL. By developing a simple model of the laser resonator as a chain of alternately positive and negative lenses, the temporal development of the transverse coherence is predicted and its dependence upon the resonator magnification is derived. Time-resolved measurements of the visibility of Young's fringes from various pairs of pinholes then give sufficient information to allow the real transverse coherence to be determined. Laser beam instability is found to impose a serious limitation upon the shot-averaged transverse coherence of the laser beam, and some attempts to reduce this problem are reported.

Chapter V gives a theoretical account of second harmonic generation. Many crystals used for SHG can convert efficiently only radiation which enters within a limited angular range, which may be narrower in one dimension than in the other; it has therefore been suggested that matching the focussed laser beam to these angular ranges might offer a useful increase in conversion efficiency. An established theoretical description of second harmonic generation using circular Gaussian beams has therefore

been extended to permit a degree of ellipticity in the focussing, and the results of calculations based upon this extended theory are presented. A small but useful increase in the conversion efficiency is indeed predicted, although no attempt at experimental verification has been made.

A recently developed nonlinear material, beta-barium borate, has a number of qualities which commend it for use with the copper vapour laser; chapter VI describes experimental investigation of second harmonic generation using this crystal with conventional focussing of various strengths and a range of laser resonators. Predictions based on the theory of chapter V, with reference to the transverse coherence measurements of chapter IV, are compared with the experiment. As a result, a number of improvements to the apparatus, which should be included in any practical implementation of the arrangement, are suggested, and an extension of the results to predict the performance at higher laser powers is attempted. A preliminary investigation has also been made using a crystal of the material ammonium dihydrogen phosphate, which is used in a rather different configuration. Some special apparatus was constructed for this study, and is described in this chapter. Experiments, in which severe thermal limitations were found, are also reported.

Finally, in chapters VII and VIII, the design of a copper laser pumped dye laser is addressed. Chapter VII reviews the history and present state of pulsed dye lasers and considers a number of different arrangements in current use. The suitability of various designs for use with the CVL is then discussed at the beginning of chapter VIII, and finally the design and initial operation of a CVL-pumped narrowband dye laser are described. Intended as a low power oscillator, with a bandwidth below 1GHz and with simplicity of tuning, this dye laser is simple, compact, and easily aligned. Single longitudinal mode operation has been demonstrated over a wide spectral range, and output powers are sufficient to allow the future use of a single stage dye amplifier. However, insufficient time has been available to explore the prevention, by the synchronous scanning of cavity length and grating angle, of mode-hopping during tuning, nor to remove some wavelength instability due to vibration and turbulent dye flow.

Chapter II

The copper vapour laser.

II-1. Introduction

II-2. History of the copper vapour laser

II-3. Physical properties of the copper vapour laser

- 3.1 Dependence of laser power upon buffer gas pressure
- 3.2 Dependence of power upon copper vapour pressure
- 3.3 Dependence of pulse energy upon pulse repetition rate
- 3.4 Spatial dependence of the laser emission and gain

II-4. Oxford Lasers model CU10

Chapter II.

The copper vapour laser.

II-1 Introduction

The copper vapour laser is one example of a class of pulsed metal vapour lasers, and is noted for its combination of high efficiency and high average power. The laser is intrinsically pulsed, for the lower laser levels are metastable, and emission is shared between green (510.6nm) and yellow (578.2nm) lines, usually favouring the green. The copper is maintained in the vapour phase by heat generated in the pulsed electrical discharge and it is this discharge which provides the energy to excite the atomic copper thus liberated. The laser operates with high gain at high pulse rates, with a typical efficiency of 1%.

The laser head, illustrated in figure II-1, is based around a ceramic tube, typically a centimetre in radius and perhaps a metre long, which contains a few tens of grammes of copper wire distributed in half a dozen pieces along its length. A coaxial Pyrex tube acts as a vacuum jacket – the intervening space is filled with insulating alumina fibre – and flanges at each end complete the vacuum system. These flanges contain foil electrodes and form the connection to the laser power supply. Gas connections are also made to the end flanges, and a buffer gas of helium or neon flows slowly through the tube with a pressure of a few tens of millibars.

The laser is heated from cold by a pulsed discharge in the buffer gas at an average power of a few kilowatts. As the operating temperature of around 1500 C is approached, the vapour pressure of atomic copper increases and the discharge begins to run in the copper vapour. Electron impact excites the copper atoms to the upper laser levels, and from here the majority of the population is further excited towards ionization, participating in the discharge. A small fraction, however, may be stimulated into radiative decay to the lower laser levels: here population accumulates, and the population inversion is eventually cancelled after a pulse duration of perhaps 30ns. Relaxation of the metastable lower laser levels takes several tens of microseconds, and puts an upper limit on the pulse repetition rate. Typical devices may be limited to pulse rates of 20kHz; nonetheless, such frequencies are only barely within reach of the previous families of pulsed gas discharge lasers using nitrogen or rare gas-halide excimers.

The high repetition rate is one of the main advantages of the copper vapour laser and may be matched only by quasi-cw lasers involving Q-switching or

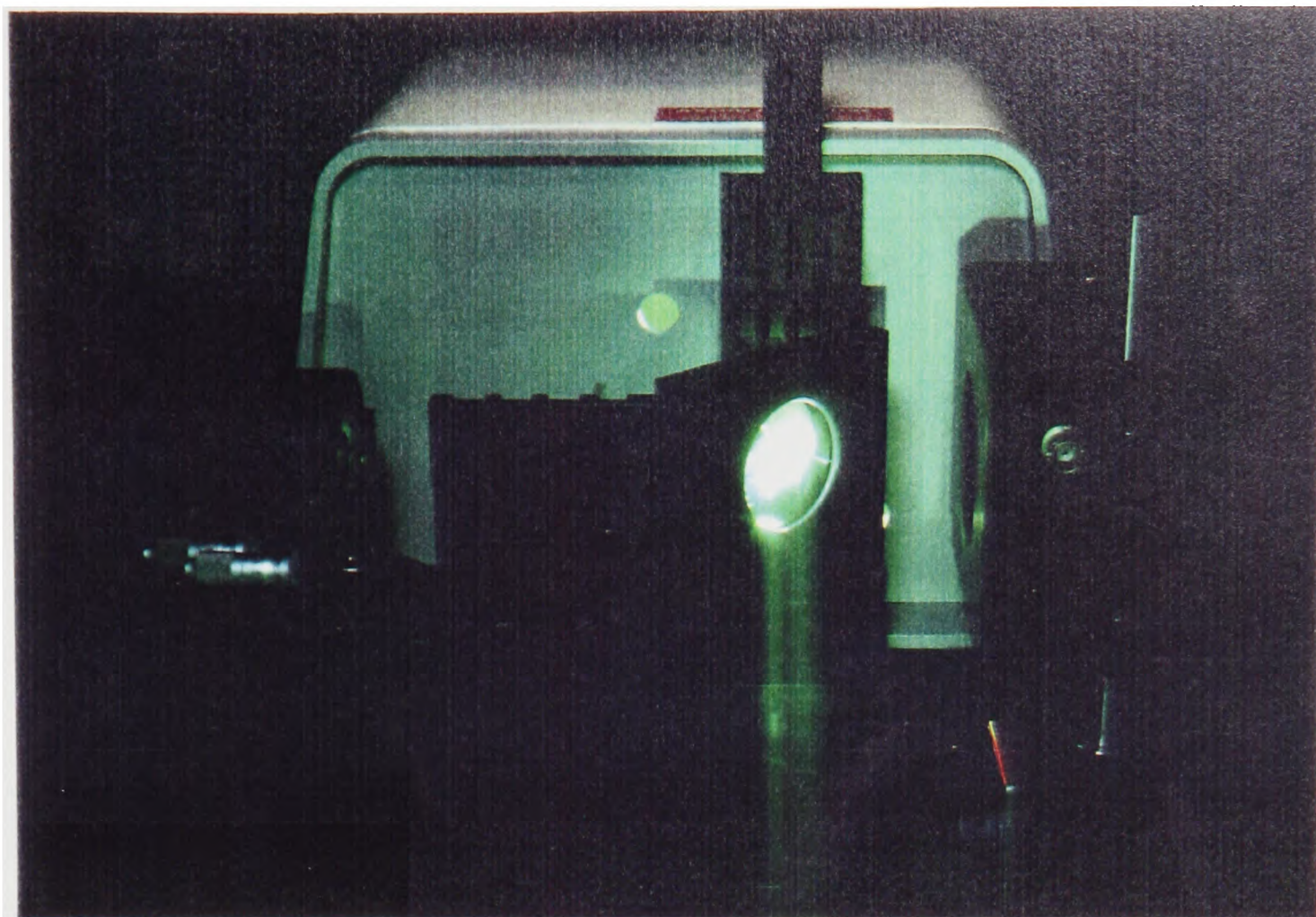


Plate II-1. The copper vapour laser with the unstable cavity side-arm.

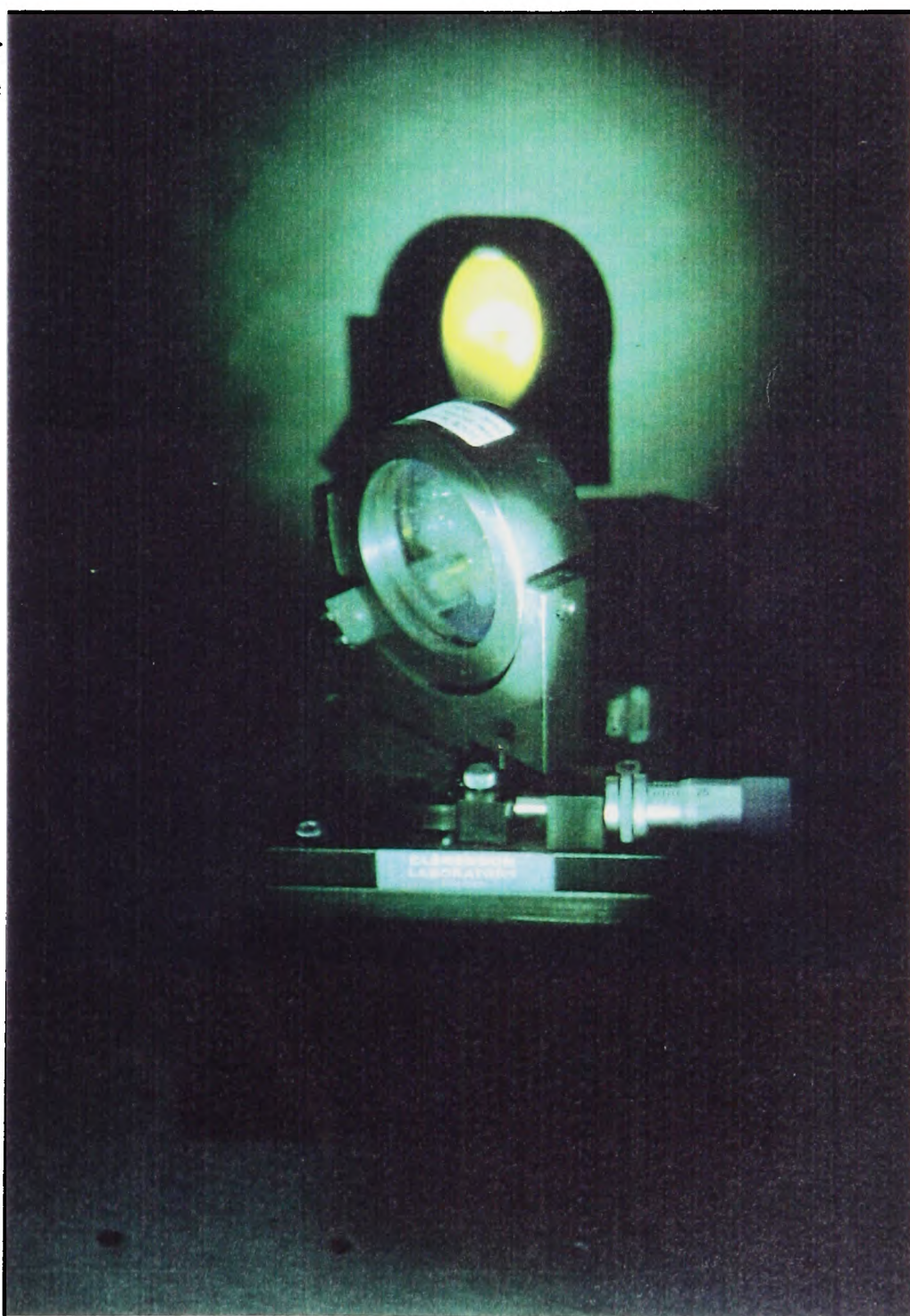


Plate II-2. A dichroic beamsplitter transmits the yellow, 578nm, radiation and reflects the green, 511nm line. The green halo is caused by scattering from a dirty laser window.

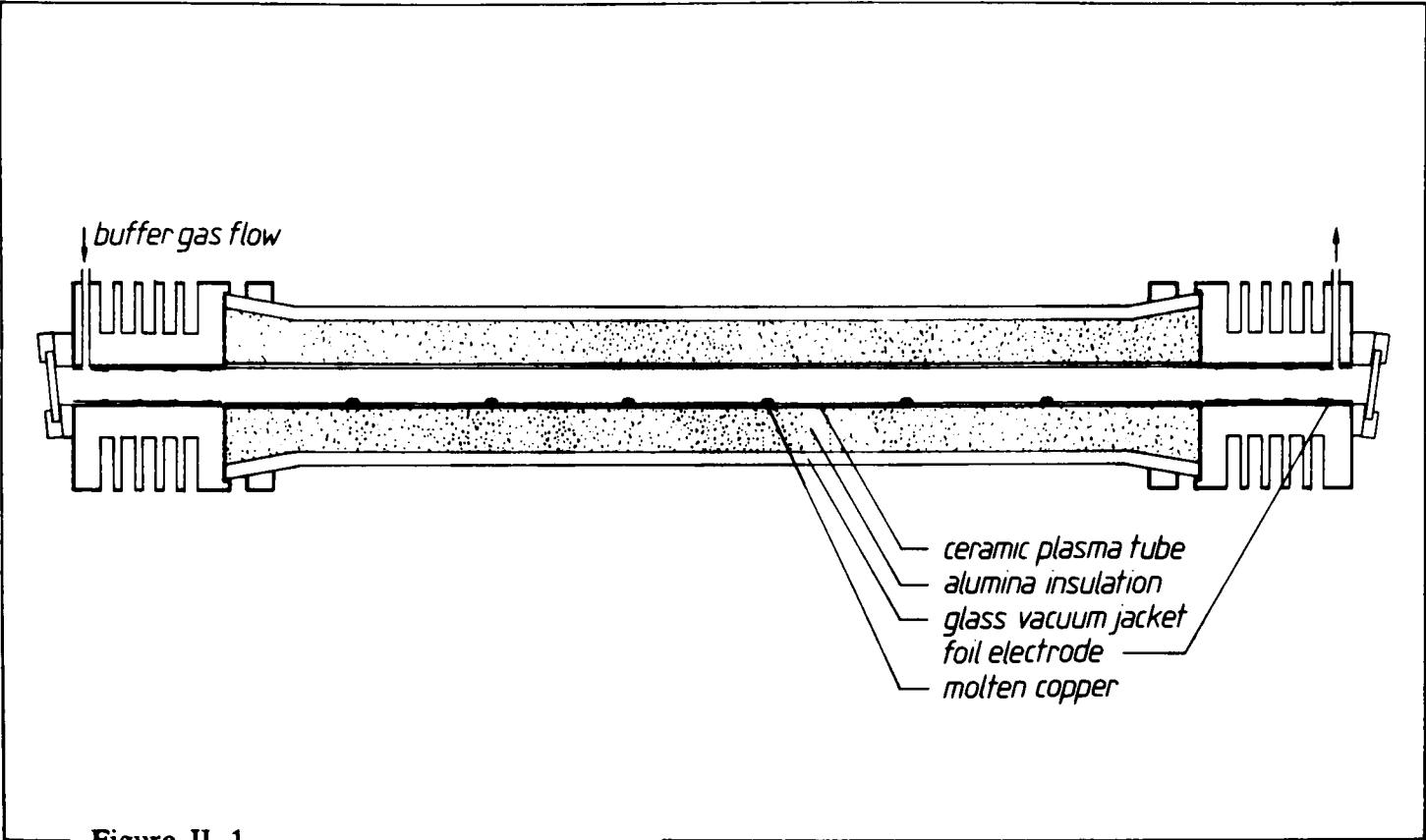


Figure II-1
The copper vapour laser head. The CU10 used for all the experiments described here used a 70cm long plasma tube with a bore of 25mm, giving an active volume of around 300cm³.

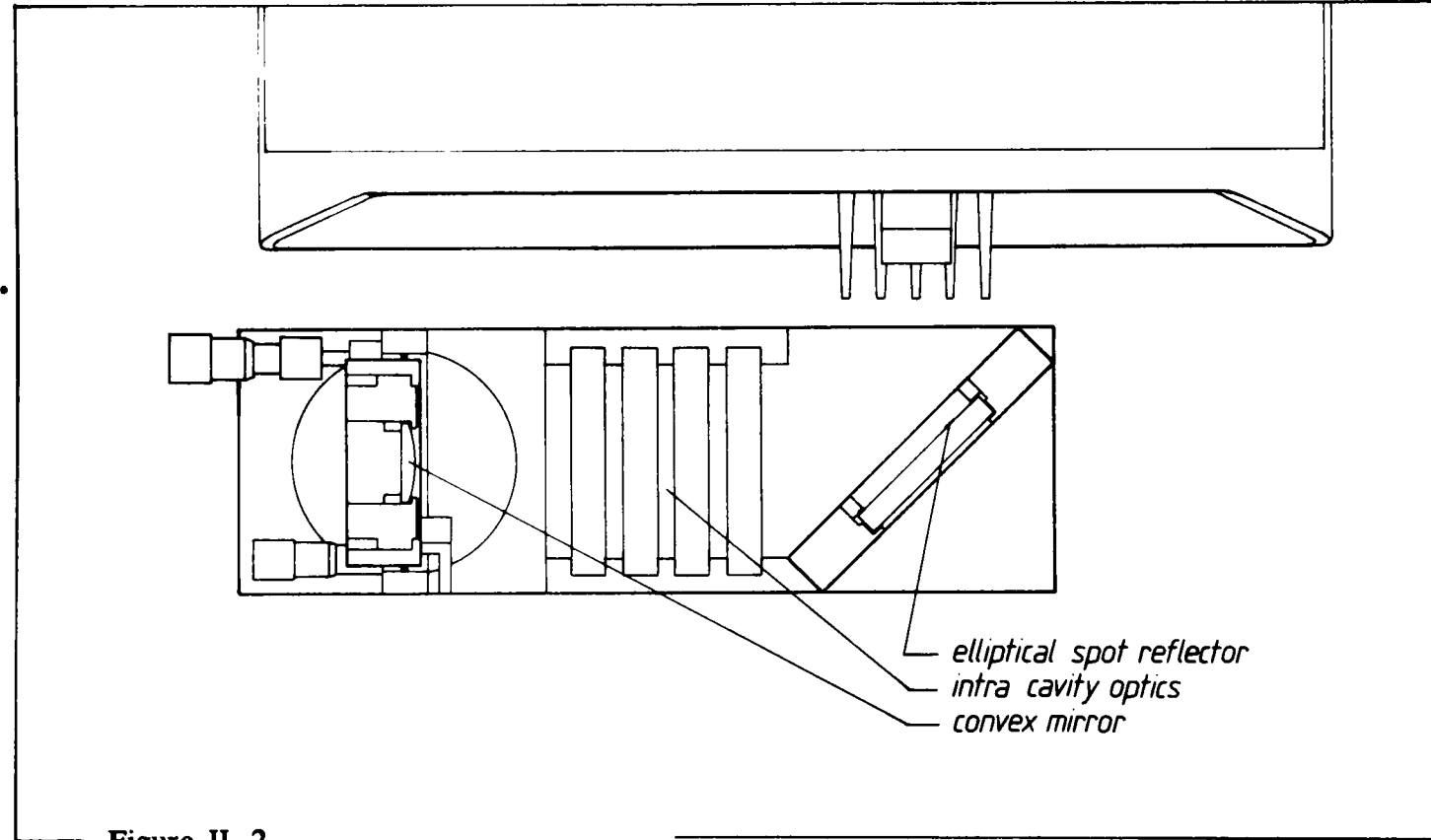


Figure II-2
The unstable cavity side-arm.

mode-locking. The pulse energy and peak power which may be extracted from a copper laser are relatively low in comparison, being typically a few millijoules (at maximum perhaps 25mJ) and 100kW, but at these high repetition rates, metal vapour lasers are capable of high mean powers, from 10 to 100W, and are the most powerful laser sources of visible radiation available. The average power reflects the high overall efficiency of the metal vapour lasers – copper lasers typically approach 1% conversion of electrical power into light.

It is the high efficiency, average power, and repetition rate which have driven the search for spectroscopic applications of this unique laser system.

This chapter is intended to review the basic properties of copper vapour lasers, and in particular the dependence of mean and instantaneous power, pulse energy and length, upon the laser's operating parameters. We shall later find that the optical characteristics of the copper vapour laser, such as the transverse coherence, depend upon these properties, and hence the best performance of systems incorporating copper lasers for purposes such as dye pumping, harmonic generation and holography, will depend upon the optimization of these parameters.

II-2 History of the copper vapour laser

The first report of laser action in copper was made by Walter et al in 1966, following similar work in the analogous systems of lead and manganese. All these devices had used discharge excitation of the metal vapour, but it was not until 1972 that the discharge assumed the role of primary – and indeed sole – source of heating for the laser, with a device made by Isaev which gave an output power of 15W at an efficiency of 1%.

Repeated problems with fracturing of the discharge tubes had prompted an interest in devices which would operate at lower temperatures by using compounds of copper, which could be dissociated by a discharge to generate the required pressure of the atomic vapour, and work concentrated around copper halide based devices which have since become available commercially in eastern Europe as reliable, if relatively low power, units. It was at this stage in the early '70s that work with metal lasers was started at the Clarendon Laboratory, with the first transverse discharge copper halide laser being reported in 1974 and the achievement of room temperature operation for the first time, using copper acetylacetonate $[\text{Cu}(\text{C}_5\text{H}_7\text{O}_2)_2]$ in 1977. However, interest revived in elemental copper lasers in the mid '70s, and by 1978 lasers of 15W average power were being sold by General Electric in the United States. The achievable average power was on the increase: in 1977, Isaev had reported transient operation in excess of 40W at 1% efficiency. Three years later, the

record had been pushed to 55W at the Lawrence Livermore National Laboratory in the U.S. as part of the Laser Isotope Separation programme.

In 1980, work on longitudinal-discharge elemental copper lasers was started in the Clarendon Laboratory following initial investigations of the dynamics of metal vapour lasers by Richard Hollins. A successful design, from which today's reliable high power commercial units have stemmed, was developed by Rhys Lewis and Tony Andrews in the early 1980's. A 100W laser to this basic design is soon to join the family of lower power commercial devices, and the Oxford Lasers CU10 used in the experiments described in this thesis is its smallest relative.

Lewis made extensive studies of the physical characteristics of copper vapour lasers, and his D.Phil. thesis stands as the most complete accumulation of such knowledge available. Many conclusions regarding the internal dynamics of the copper vapour laser were reached as a result of this work, and a number of improvements to commercial devices have resulted from its practical implementation. Yet Lewis never had the opportunity to consider the structure within the laser pulse. The temporal development of laser properties is both governed by and reflected in the gross laser characteristics he studied; the exact form, of crucial importance to processes such as frequency doubling, has been the subject of more recent work in this laboratory, of which some is described in this thesis.

II-3 Physical properties of the copper vapour laser

A schematic diagram illustrating the dynamics of the copper vapour laser is given in figure II-3. The atomic copper ground state is excited, by electronic collision in the discharge, predominantly to the upper laser levels. The discharge is sustained by ions created by further collisional excitation into ionization and accounts for the major losses out of the upper laser level, but perhaps a tenth of this population may be stimulated into radiative decay to the lower laser levels.

The laser transitions appear to correspond to a two electron jump, from the $3d^{10} 4p$ upper levels to the $3d^9 4s^2$ lower configuration. However, there is significant configuration mixing of the upper levels with the $3d^9 4s 4p$ states, and as a result the transition probabilities approach those of the resonance transitions. At first sight, the resonance transition appears to present a serious problem, for it would appear that the most significant route for decay of the upper laser levels would be directly back to the ground state. However, the large resonance transition probability means that this ultraviolet radiation is trapped by re-absorption in the copper vapour. Radiation trapping dramatically lengthens the upper laser level lifetime, and under normal lasing conditions the primary mechanism for direct decay of the upper laser levels is by non-radiative quenching by hot electrons ($\Delta E = 3.8\text{eV}$).

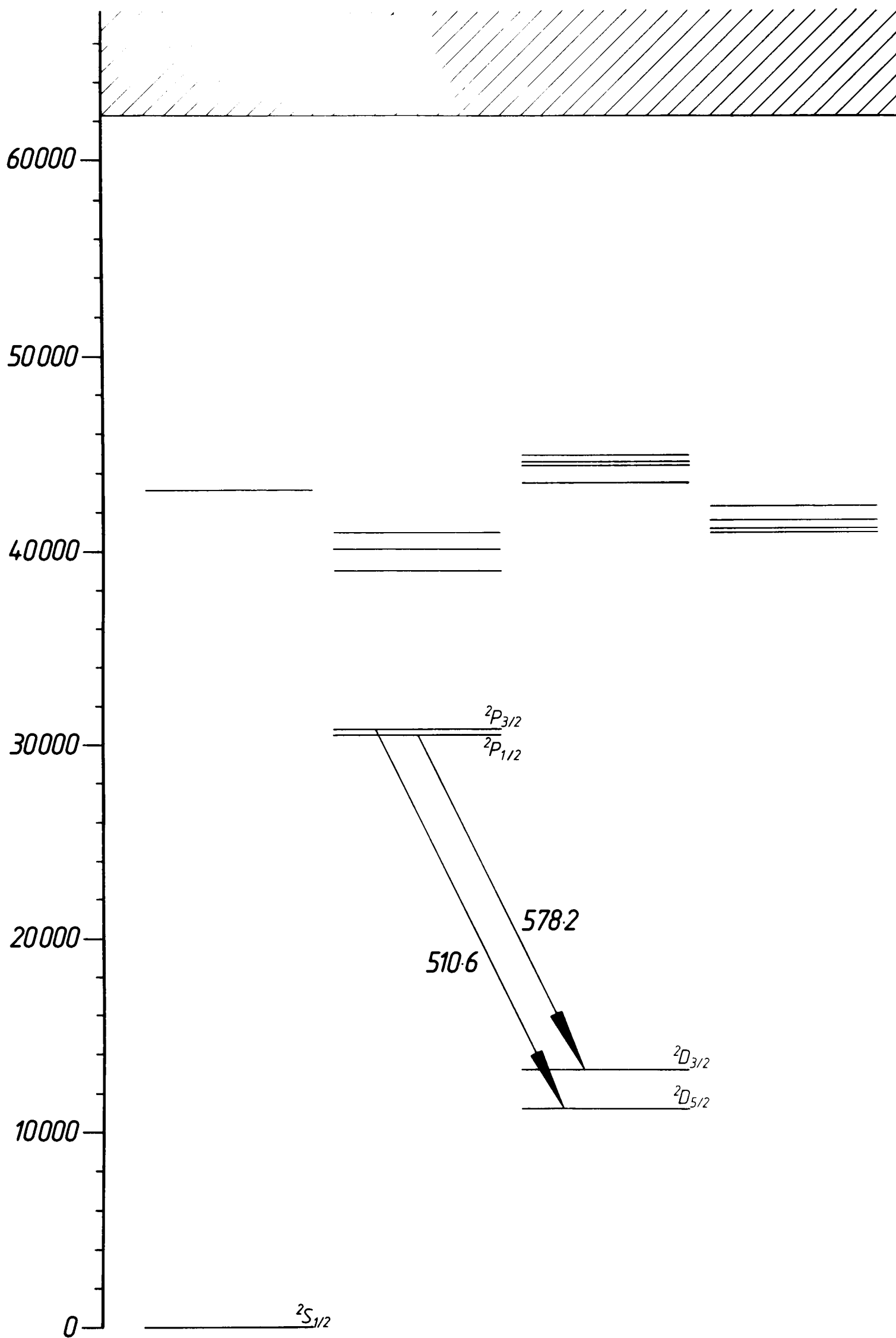


Figure II-3
Energy levels of atomic copper showing the low-lying energy levels and the relative position of the ionization level, Cu^+ .

The lower laser levels are metastable. They are the lowest excited levels, and radiative transitions to the ground state are forbidden by conservation of both parity and angular momentum. The relaxation of these lower laser levels, normally taking in excess of 1msec, is quenched by superelastic collisions with cool electrons ($\Delta E \sim 1.8\text{eV}$), but the lifetime nonetheless far exceeds that of the laser transition and the laser is thus intrinsically self-terminating. Unfortunately, the reverse of this process affords a mechanism for direct pumping of the lower laser level by the discharge, and at low electron energies this route is preferred to the excitation of the upper laser levels. High buffer gas densities can limit the energy reached by a discharge electron, and we shall see that low buffer gas pressures are thus preferred.

II-3.1 Dependence of laser power upon buffer gas pressure

The buffer gas is necessary initially in order that a room temperature discharge may be sustained, and this discharge fulfills the important role of heating the copper. As the copper melts and evaporates, the discharge will begin to switch to the copper vapour with its lower ionization potential, and under normal lasing conditions the partial pressure of the copper vapour is around 0.2mbar. This corresponds to an atomic density of 5×10^{14} to 10^{15} cm^{-3} . The pulse energy from each cubic centimetre of gain medium is around $4 \mu\text{J cm}^{-3}$, or around $10^{13} \text{ photons.cm}^{-3}$.

Under running conditions, one of the prime roles of the buffer gas is in stabilizing the discharge and anchoring it to the electrodes in preference to the gas ports. Without the buffer gas a stable discharge is impossible. However, the buffer gas also plays an important part in the dynamics of the copper laser, for the free electrons are cooled between pulses principally by contact with the buffer gas, and a low electron temperature is necessary for quenched relaxation of the lower laser level. Finally, the buffer gas plays a practical role by reducing the copper diffusion rate, thus reducing the rate of loss from the laser and discouraging the copper from depositing on the windows.

An excess buffer gas pressure will steal the discharge from the copper vapour and will also reduce the mean electron energy by reducing the electron mean free path. At high enough pressures the discharge will be constricted into an arc. The lower electron temperature has a further effect in reducing the pumping of the upper laser levels in preference to the lower levels, and thus the dynamics of the laser are again affected. In general, then, the buffer gas pressure is determined loosely by the desire for a uniform glow discharge, and more tightly by the optimization of differential pumping of the upper laser level against effective quenching of the lower one. In practice there remains some degree of freedom, and one uses the highest buffer gas pressure within this range to limit the loss of copper from the laser.

Both neon and helium are effective as buffer gases. The optimum pressure of neon is around 100mbar; with helium it is significantly lower at some 7mbar. How the buffer gas affects the laser output is not well understood, but it is perhaps surprising that the green line operates with the same efficiency in both gases whereas the yellow line is somewhat weaker in helium than in neon. The green and yellow lines operate independently, and no collision-less transfer of population from one to the other is possible.

All the experiments described in this theses used neon as a buffer gas, at pressures around 75mbar. In practice, there is only a small variation in the laser pulse energy or pulse shape below 120mbar.

II-3.2 Dependence of power upon copper vapour pressure (laser temperature)

The gain of a laser depends upon the population inversion density. At low copper vapour pressures, the inversion density will be too low for significant gain to occur, and the discharge characteristics will be determined by the buffer gas alone. At the highest temperatures, the high copper vapour density will restrict the mean electron energy or temperature, and the selective excitation of the upper laser levels will be reduced. There is thus an optimum copper vapour pressure, some 0.2mbar, which corresponds to a laser temperature of around 1500 C.

The efficiency of resonance trapping, and thus of upper laser level pumping, will fall sharply below a certain copper density. This copper density, however, lies well below the usual threshold of gain for the copper vapour laser.

II-3.3 Dependence of pulse energy upon pulse repetition rate

For a given excitation voltage, the pulse energy from the copper laser is essentially determined by the lower laser level population remaining after the previous pulse. The pulse energy therefore drops when the inter-pulse period is comparable with or shorter than the relaxation time of the lower laser level. At lower pulse repetition rates, the pulse energy is constant. The lower limit on the repetition rate of the copper vapour laser will eventually be determined by the decay of residual ionization which preionizes the next pulse, allowing the launch of a uniform discharge; this lower limit is around one pulse a second.

In practice, the discharge power must remain constant for the equilibrium temperature to be maintained, and thus for a given device the excitation voltage and energy will be linked to the pulse repetition frequency. For the CU10 used in these experiments, figures II-4 and II-5 show the dependence of the output power and

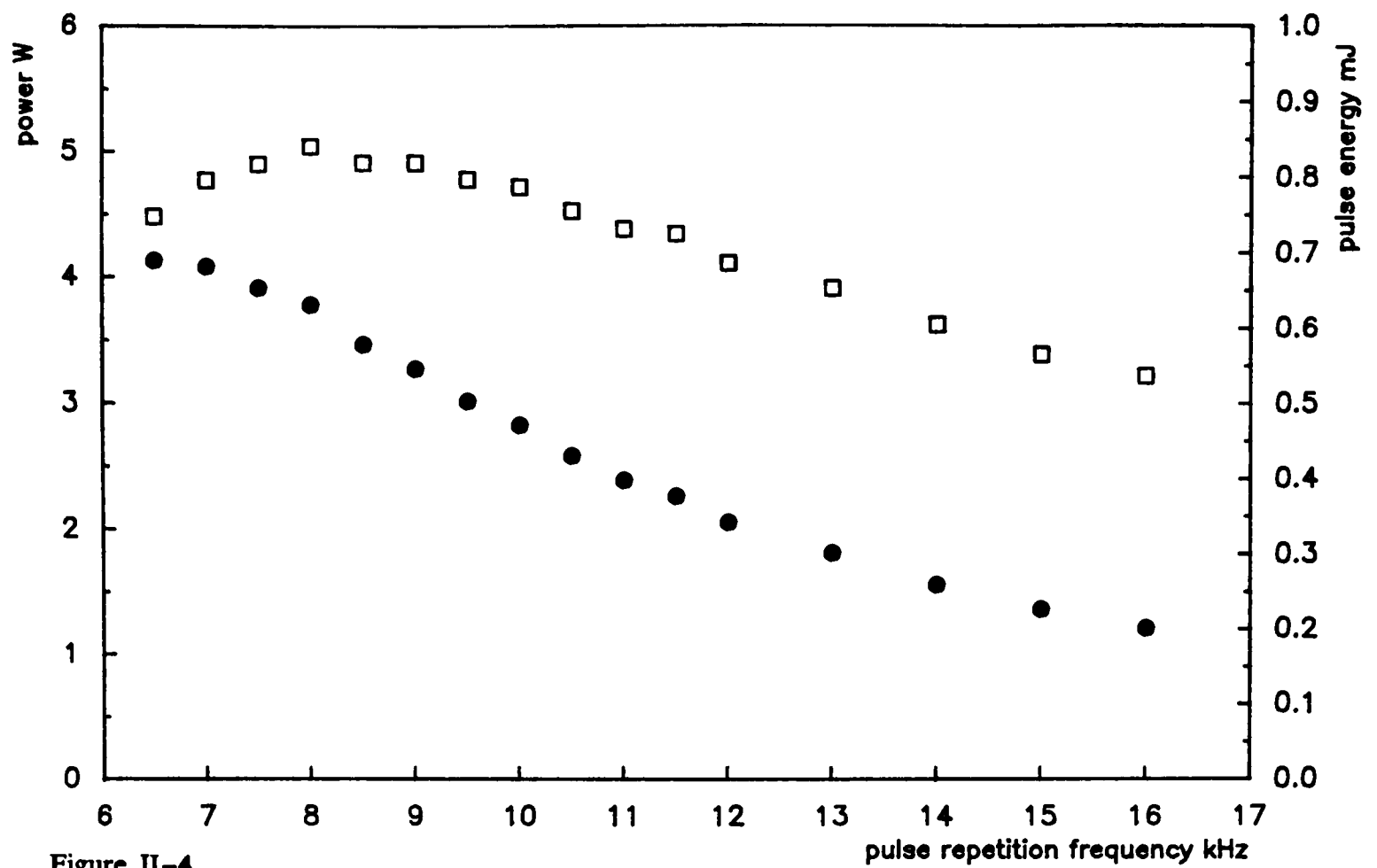


Figure II-4
Variation of green laser power and pulse energy with prf. Open squares show laser power: full circles show pulse energy.

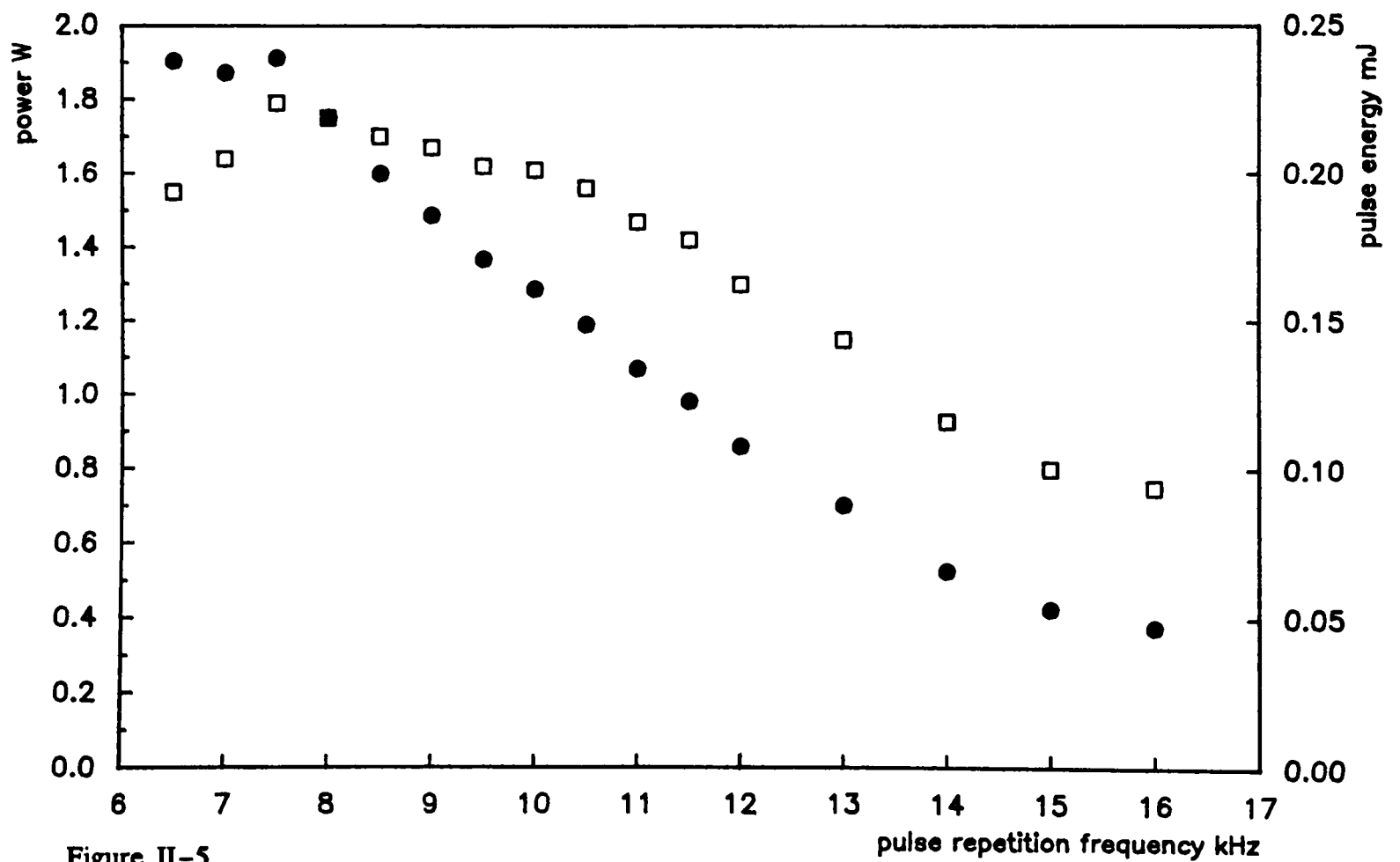


Figure II-5
Variation of yellow laser power and pulse energy with prf.

pulse energy of the two laser lines upon pulse repetition frequency at a constant input power of 1.65kW, which has been found experimentally to correspond to the optimum tube temperature. The highest mean laser power, and hence efficiency, occurs at around 8 kHz, and the poorer performance at lower repetition rates reflects the fact that with a given power supply circuit the pulse voltage may not be independently optimized. The power supply was not capable of delivering the required 1.65kW at frequencies below about 6.5 kHz, but above this value the pulse energy clearly falls with inter-pulse period.

An investigation of the pulse shapes over this range shows that the peak pulse power varies only slightly, and that the change in pulse energy comes from a variation in pulse length. The dependence of pulse length upon repetition rate is shown in figure II-6.

II-3.4 Spatial dependence of the laser emission and gain

Under steady running conditions, there will be a radial temperature variation, and the average gas temperature at the tube centre may be 200 C higher than that at the edge. The radial temperature distribution and uniform copper vapour pressure lead to a variation in copper density. This will be highest at the tube walls, and for this reason, a cool copper laser will show gain only around the edges of the laser tube, where the threshold population inversion is exceeded. Above normal operating temperatures, the higher copper density at the wall will lead to more significant reduction in the discharge electron energies, and thus the gain is localized at the tube centre.

II-4 Oxford Lasers model CU10

The copper vapour laser used for these experiments was an Oxford Lasers model CU10, and was built by the author at the Oxford Lasers factory in 1986. Although the laser is nominally completely air cooled, some water cooling has been added to the power supply since its manufacture. The laser is specified to give a mean power of 10W with a plane-plane cavity; in practice, powers up to 15W may be obtained.

The power supply is of traditional design, mains voltage being transformed up to some 5 kV before being bridge rectified and smoothed by a large capacitor bank. This supply is used to recharge resonantly a smaller capacitor bank, which is then switched into the laser by a hydrogen filled thyatron. The CU10 power supply is separate from the laser head, and the two are connected by a single 'umbilical' tube, through which all power and gas supplies and control signals are passed.

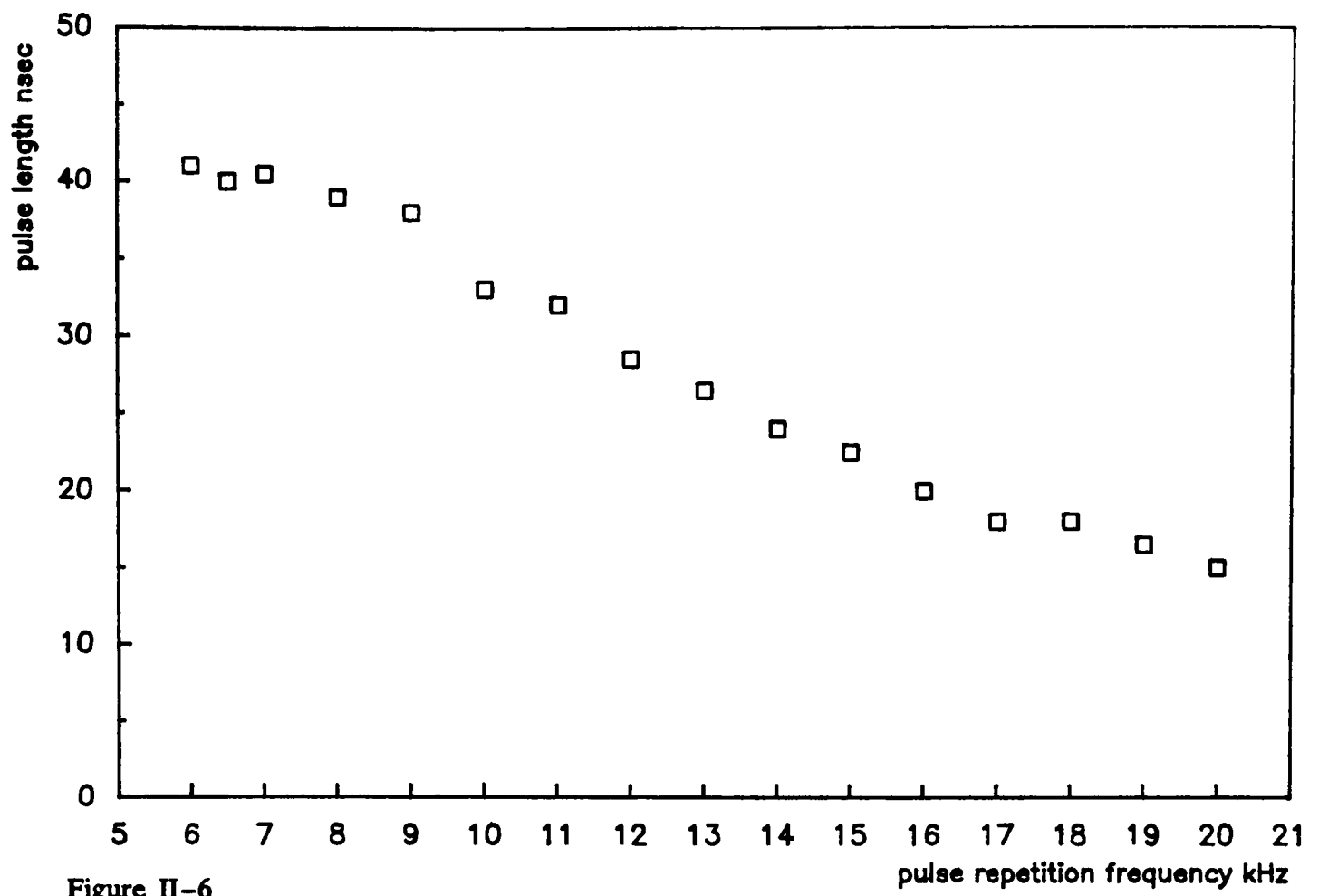


Figure II-6
Variation of laser pulse length with prf.

There is no active regulation of the voltage applied to the laser head, and some drift in laser power with mains voltage therefore occurs. Further, there is 100Hz modulation of the laser power; the rms value of this variation is less than 1% of the mean laser power.

It is common practice with wide bore, high gain, short pulse lasers to use an unstable optical resonator, and this has indeed been the case for most of the experiments described in this thesis. The unstable cavity is considered in some detail in chapter IV (transverse coherence); it is of the side-arm arrangement as shown in figure II-2, which has the advantage that additional optical components such as polarizers and filters may be included in the cavity without interfering with, or being subject to, the full laser output. This is particularly important if the additional element may be damaged by the high laser output power for the flux, if not the intensity, is lower in the side arm.

It is not difficult to align an unstable resonator around a wide bore laser. It is, however, more difficult to ensure that the laser tube lies along the axis of the resonator, and thus the unstable cavity used in these experiments has been aligned using a small helium-neon laser to define the direction of the output beam and the path of the beam within the cavity. A pair of goggles intended for use with the copper vapour laser attenuates the viewed copper laser beam to a similar intensity to that of the He-Ne. In this way, the He-Ne beam can still be seen as it passes through the working copper laser, and the copper laser beam can be readily aligned relative to it. A further advantage of the He-Ne in the alignment method is that the reflectivity and transmission of multilayer dielectric coatings used with the copper laser are similar, and thus the He-Ne will follow both paths.

The copper laser head was mounted on a Photon Control optical bench, which formed a breadboard for these experiments. The unstable cavity is outside the laser head box and is mounted directly onto the optical table. All light beams lie in the same horizontal plane, and all optical components are at the same height. Black anodised aluminium panels then surround the system and prevent stray beams from passing into the room. This has proved a very safe and easy system to use.

References

R R Lewis, D Phil thesis, Oxford University, 1985.

Chapter III

Longitudinal Coherence and Lineshape

III-1 Structure of the copper spontaneous emission lineshape

III-2 Theory of longitudinal coherence

III-3 Measurement of the longitudinal coherence of the 510.6nm laser line

III-4 Measured longitudinal coherence of the copper vapour laser

III-5 Implications for holography using a copper laser source

III-6 Conclusion

Longitudinal Coherence and Lineshape

III-1. Structure of the copper spontaneous emission lineshape.

The copper vapour laser operates at two wavelengths: 510.6nm, corresponding to transitions between the $3d^{10} 4p \ ^2P_{3/2}$ and $3d^9 4s^2 \ ^2D_{5/2}$ levels; and 578.2nm, which results from the corresponding $\ ^2P_{1/2}$ to $\ ^2D_{3/2}$ transition. These two wavelengths, in the green and yellow respectively, are further split by hyperfine structure and isotope shifts.

A schematic energy level diagram for a single copper isotope is shown in figure III-1. The $\ ^2P_{1/2}$ state has two hyperfine components with $F=1$ and 2 ; the $\ ^2P_{3/2}$ and $\ ^2D_{3/2}$ levels are each split into four components corresponding to $F=0,1,2$ and 3 ; and the $\ ^2D_{5/2}$ state has four components with $F=1,2,3$ and 4 . Assuming the copper atom to be L-S coupled, the frequency shifts of the hyperfine components relative to the centres of gravity of the levels (γJ) are given by

$$\Delta\nu = A \frac{C}{2} + B \frac{\frac{3}{2} C(C+1) - 2I(I+1)J(J+1)}{4 I J (2I-1)(2J-1)} \quad (\text{III-1})$$

where $C=F(F+1)-J(J+1)-I(I+1)$. A and B are the magnetic dipole and electric quadrupole splitting factors, measurements of which have been collated by Tenenbaum et al [III-1] together with the values of the isotope shifts between the ^{63}Cu and ^{65}Cu isotopes. These are the only naturally abundant isotopes of copper, and are found in the ratio 69:31. Both isotopes have nuclear spin $3/2$.

Tenenbaum et al [II-1] have used these data to calculate the spontaneous emission lineshapes of the copper laser lines. However, there are inconsistencies in the printed tables which indicate errors in their calculation; the correct values for the hyperfine splittings, and resulting shifts in wavelength for the laser emission lines, are given in table III-1 at the end of this chapter. This table also lists the relative intensities of the hyperfine spectral components, which in the LS coupling approximation are given by Sobelman [III-2] to be

$$I \propto (2F_1+1)(2F_2+1) \left\{ \begin{matrix} J_1 & F_1 & I \\ F_2 & J_2 & 1 \end{matrix} \right\}^2 \left| \langle \gamma_1 J_1 | D | \gamma_2 J_2 \rangle \right|^2 \quad (\text{III-2})$$

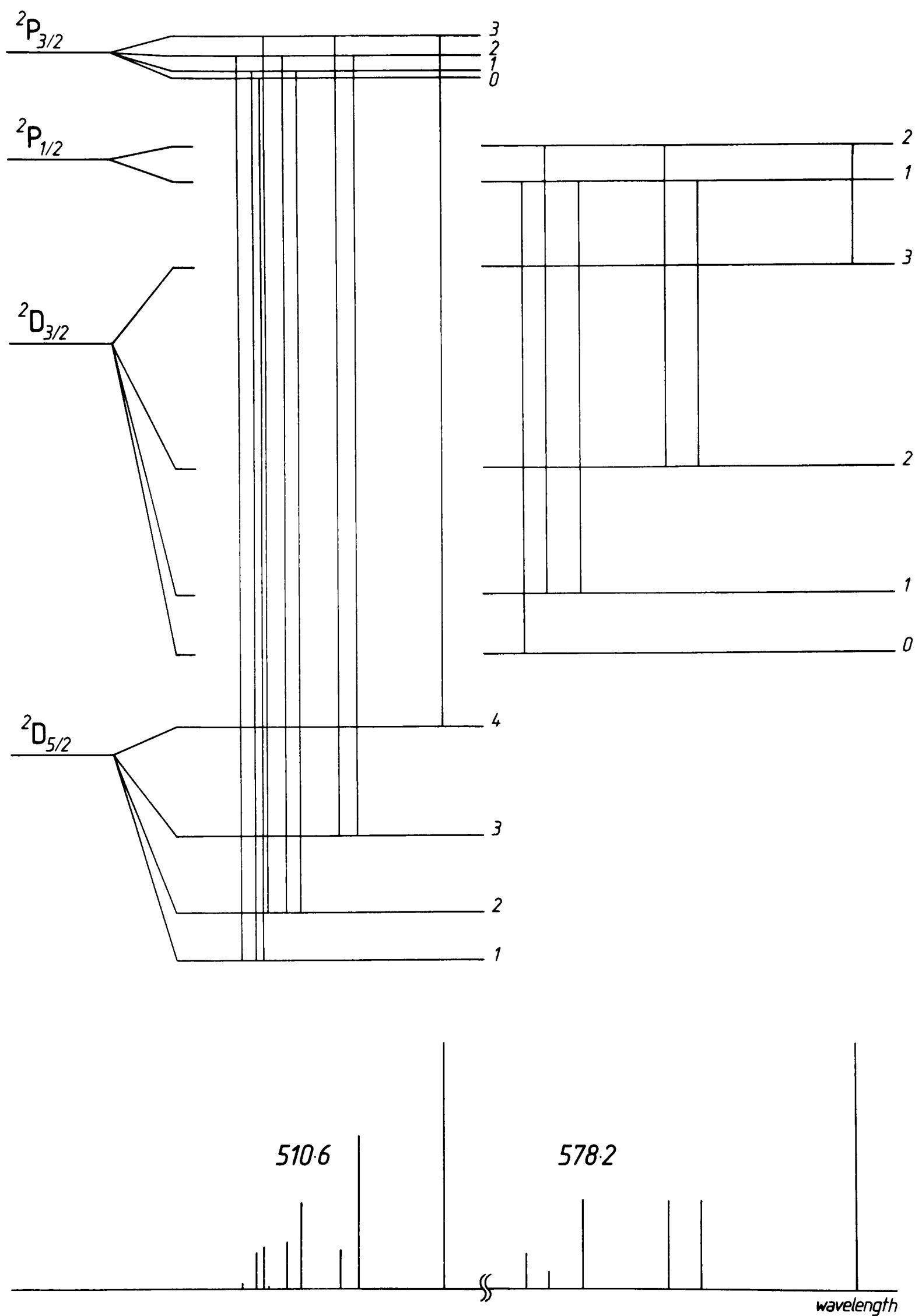


Figure III-1
Hyperfine structure in copper showing the laser transitions between components split according to F. The green laser line has nine components for each of the two copper isotopes, and the yellow line similarly has six for each. Both isotopes have nuclear spin $3/2$.

where the subscripts 1 and 2 refer to the initial and final states. For a given laser band $(\gamma_1 J_1, \gamma_2 J_2)$, the term $|\langle \gamma_1 J_1 | D | \gamma_2 J_2 \rangle|^2$ is a common factor, so we may write

$$I \propto (2F_1+1)(2F_2+1) \begin{Bmatrix} J_1 & F_1 & I \\ F_2 & J_2 & 1 \end{Bmatrix}^2 \quad (\text{III-3})$$

and neglect the fact that $|\langle \gamma_1 J_1 | D | \gamma_2 J_2 \rangle|^2$ will in fact be zero for the laser transitions which are forbidden in the LS coupling approximation. The relative positions and intensities of the hyperfine components are shown at the bottom of figure III-1.

The spontaneous lifetimes of the upper laser levels are around 10nsec, and the dominant broadening mechanism is the Doppler effect. The spontaneous emission lineshape is thus a convolution of the spectrum of figure III-1 with a Gaussian distribution whose width (FWHM), at temperatures around 1500 C, is 2.2 GHz. The overall observed spontaneous emission spectrum may thus be expected to be the combination of these three effects: hyperfine structure, isotope shifts, and Doppler broadening. The laser lineshape will be further complicated by the spectral gain variation, which will tend to exaggerate peaks in the spontaneous emission spectrum.

Tenenbaum et al [III-1] have measured the laser lineshape as a function of tube temperature, and find that gain narrowing is particularly significant. At low temperatures, and hence low copper densities, only the strongest transitions show gain and thus these dominate the laser lineshape; higher copper densities at higher tube temperatures are required for the whole of the spontaneous structure to be apparent. Unfortunately, a meaningful definition of tube temperature is difficult, since significant thermal gradients occur along the laser tube and the line profile is the accrued result of gain at all points in the tube, and thus all investigations of these effects in real lasers can only be qualitative. The Doppler width, it may be noted, is only a weak function of temperature ($T^{1/2}$) and is thus essentially constant over the narrow temperature range within which normal laser operation occurs.

III-2. Theory of longitudinal coherence.

The extent to which the phase of radiation at one point is related to that at another longitudinally displaced from it is given by the normalized longitudinal coherence $C(\tau)$, defined by

$$C(\tau) = \frac{\langle \xi(t+\tau) \xi^*(t) \rangle}{\left[\langle \xi(t) \xi^*(t) \rangle \right]^{1/2} \left[\langle \xi(t+\tau) \xi^*(t+\tau) \rangle \right]^{1/2}} \quad (\text{III-4})$$

This is just the autocorrelation of the scalar radiation field $\xi(t)$ and is related to the spectral intensity distribution $S(\omega)$ by the Wiener-Khintchine theorem,

$$C(\tau) = \frac{1}{2} \int S(\omega) \exp(i\omega\tau) d\omega. \quad (\text{III-5})$$

The spectral intensity distribution and the longitudinal coherence function thus form a Fourier transform pair. Essentially, the light can be coherent only over a distance in which the number of wavelengths is known to within one cycle. The scale of this coherence is relevant to all processes which involve interference of light following different optical path lengths. Holography is perhaps the most obvious example, although a particularly short coherence length will have an adverse effect upon second harmonic generation as well.

The spontaneous emission lineshape can be represented as the convolution of the components of the structure: the hyperfine and isotope structure, and the Doppler broadening Gaussian. If necessary, we could split this into three functions by considering the isotope shift and hyperfine structure separately. The lineshape may therefore be written as

$$S(\omega) = (\text{hyperfine and isotope structure}) \otimes (\text{Doppler Gaussian})$$

or

$$S(\omega) = \sum_{i=1}^N a_i \delta(\omega - \omega_i) \otimes \exp\left[-\omega^2/\Delta\omega^2\right] \quad (\text{III-6})$$

where the i summation runs over the range of individual components of figure III-1. It now follows from the convolution theorem that the longitudinal coherence function may be written as a product of the Fourier transforms of the individual distributions giving

$$C(\tau) = F.T. \left[\sum_{i=1}^N a_i \delta(\omega - \omega_i) \right] \cdot F.T. \left[\exp(-\omega^2/\Delta\omega^2) \right]$$

or

$$C(\tau) = \exp(-\pi^2 \Delta\nu^2 \tau^2) \cdot \sum_{i=1}^N a_i \cos(2\pi\nu_i \tau) \quad (\text{III-7})$$

where $\Delta\nu$ is the $1/e$ half-width in frequency of the Doppler Gaussian and the components of figure III-1 are defined by frequencies ν_i and amplitudes a_i . This expression, then, describes the theoretical longitudinal coherence of spontaneous emission from a copper vapour.

It may be assumed that as the laser excitation is increased, components will appear above threshold in order of spontaneous emission strength, and a range of coherence functions for varying numbers of such components has been calculated for the 510.6nm line and are shown in figure III-2. The curve corresponding to a single component shows the pure Doppler limited coherence with a half width at half

maximum of 0.20ns, while the predicted spontaneous emission coherence corresponds to the curve with all eighteen components correctly weighted. Intermediate curves are shown for the strongest 2, 5 and 10 components oscillating with relative strengths given by their spontaneous emission probabilities. From these curves we would expect a coherence length (HWHM) of between 21 and 60mm.

The following section describes direct measurement of the longitudinal coherence of the 510.6nm laser radiation.

III-3. Measurement of the longitudinal coherence of the 510.6nm laser line.

It is readily shown that the longitudinal coherence of a beam of light may be measured directly using a Michelson interferometer, in which case the coherence function $|C(\tau)|$ is equal to the visibility of the fringes formed,

$$v(\tau) = \frac{I_{\max} - I_{\min}}{I_{\max} + I_{\min}} \quad (\text{III-8})$$

The following experiment therefore used a Michelson interferometer, with fringes imaged onto a point detector, to measure the fringe visibility at a number of mirror displacements over a range of $\pm 3\text{cm}$.

The range of required mirror displacements places severe demands upon a standard Michelson interferometer, in which the mirrors must remain aligned as the displacement is varied. This problem may be eased by using corner-cube retro-reflectors instead of mirrors, but no such instrument was available at the time of these experiments. It was therefore decided to construct a Michelson interferometer which would permit only a limited adjustment of the mirror displacement between discrete positions but which could be built using existing laboratory optical mounts and fishtail rails. The lack of a continuous measurement of intensity with displacement made such an instrument unsuitable for use as a Fourier transform spectrometer, but the measurement precision approaching 0.1mm was quite adequate for investigating the longitudinal coherence of laser sources. It was initially intended that the instrument would be realigned for each new mirror position, and straight line fringes, produced by slightly misaligning one mirror, would then be scanned by hand using a standard translation stage. In this way, the fringe visibility could be measured with simple apparatus at a number of points up to very large mirror displacements. The ultimate limit upon mirror displacement was expected to be determined by the rapidly falling fringe size and increasing sensitivity to misalignment.

In practice, a change was made to this procedure, for the stability of the instrument was found insufficient to produce stationary fringes; instead, the fringe pattern was observed to drift slowly with time, and to respond to air currents and any table distortions caused by the experimenter. The idea of moving the detector

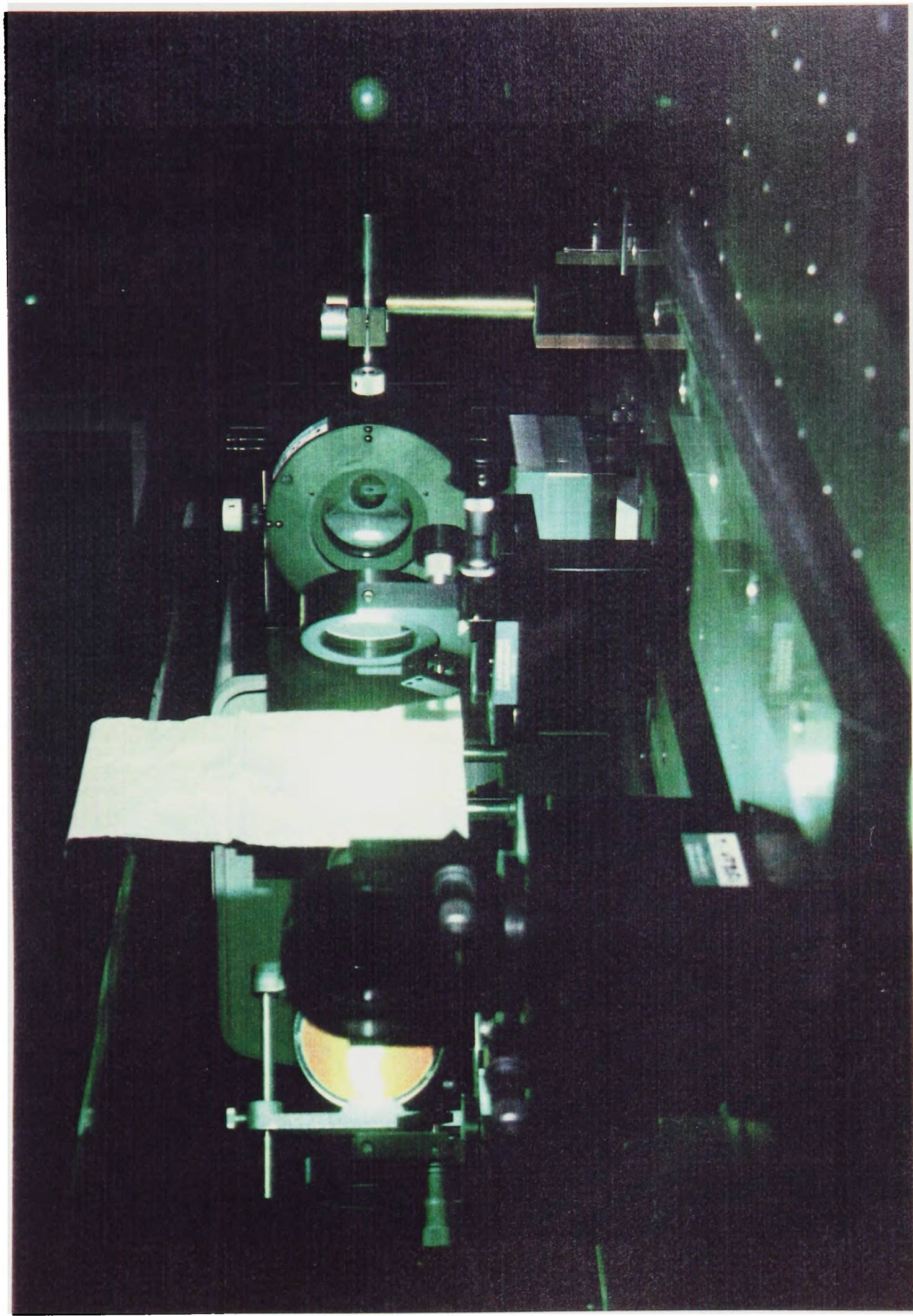


Plate III-1 Apparatus for measuring the longitudinal coherence of the CVL.

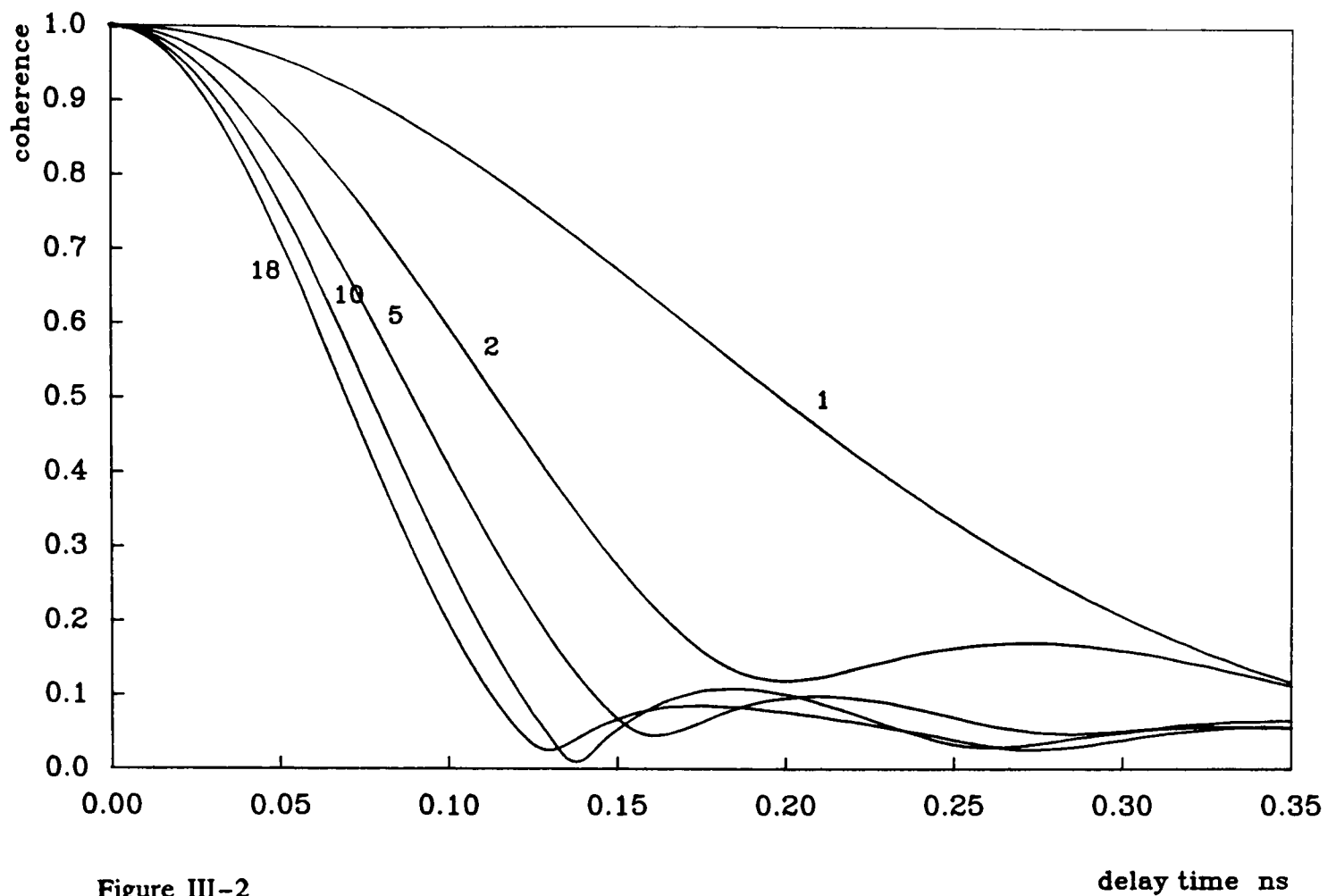


Figure III-2
Coherence functions for various numbers of lasing components showing the calculated longitudinal coherence for the strongest 1, 2, 5 and 10, and all 18, components of the green laser transition, assuming the positions and strengths listed in table III-1 and a temperature of 1500 C.

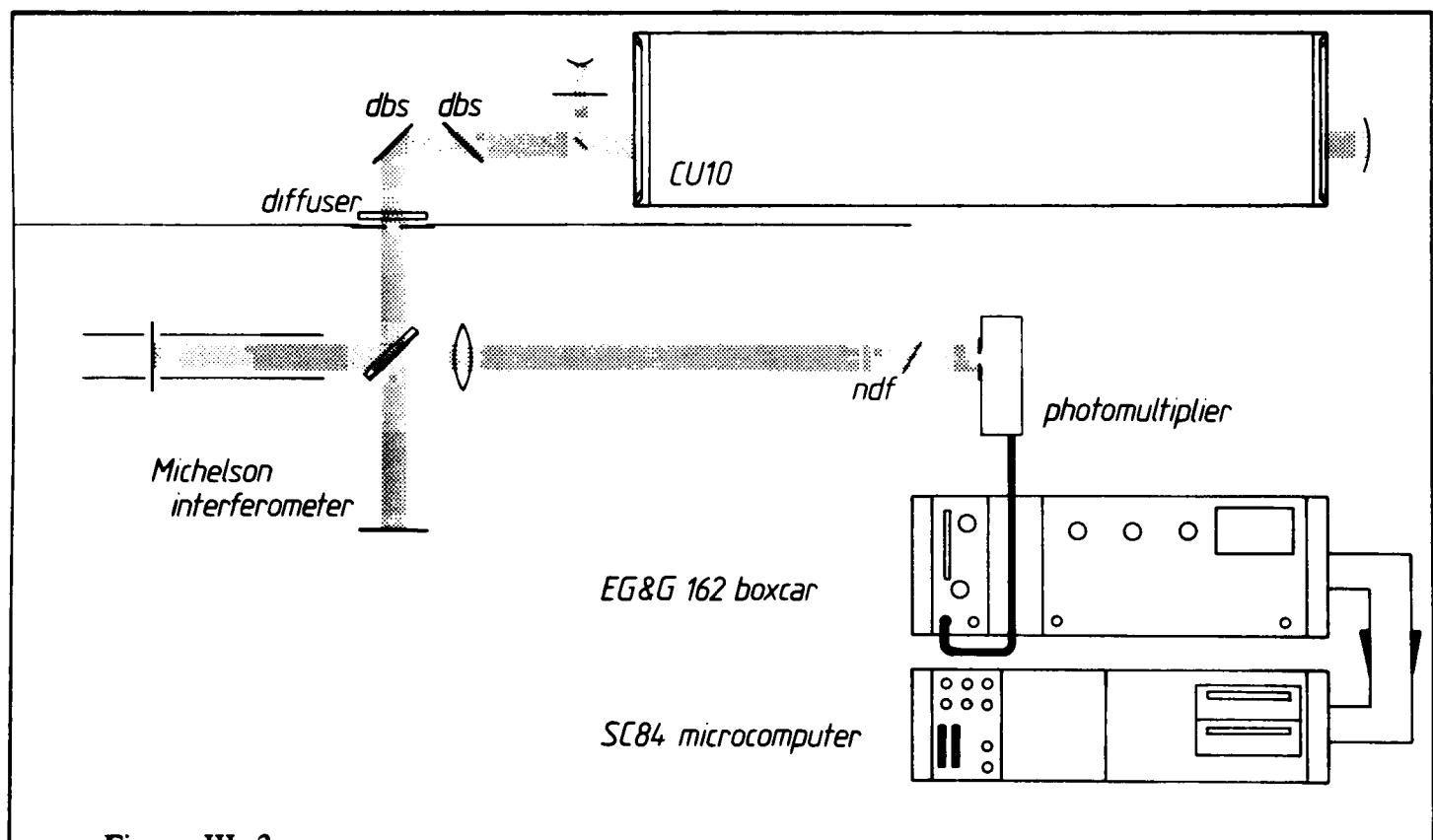


Figure III-3
Experimental arrangement for measurement of longitudinal coherence using a Michelson interferometer made from standard laboratory optical mounts. The green copper vapour laser emission is filtered by a pair of dichroic beam splitters (dbs), and the fringe pattern may be attenuated before it strikes the photomultiplier by a neutral density filter (ndf).

laterally to scan the fringe pattern was therefore abandoned in favour of a stationary detector and continuous measurement of the central fringe intensity as the fringe pattern wandered – either randomly or under the influence of the experimenter's body weight. A computer was then used to note the extremes of the intensity variation by building up a distribution of instantaneous intensities from a series of measurements. By losing information about the exact path length, this method had become sensitive to fluctuations in the intensity of the ingoing laser beam, but the recorded distributions had sharp edges and the data were felt to be good.

The experimental layout is illustrated in figure III-3. The 510.6nm line from the copper laser is selected by a pair of dichroic beamsplitters (dbs), the first of which rejects the yellow by reflection and the second by transmission. These filters together reduce the proportion of yellow light in the beam by an order of magnitude. The filtered green light eventually falls upon a diffusing screen made by bead-blasting a piece of Perspex™. This forms the diffuse illumination source for the Michelson interferometer.

The interferometer itself is based around a flat ($\lambda/10$) silica plate which is coated on one surface with a dielectric anti-reflection film, and on the other with a partially transmitting layer of aluminium designed to have approximately equal coefficients of transmission and reflection. Further aluminium coated mirrors of similar optical quality reflect the two beams thus produced, and the recombined beam is focussed using a 1m focal length lens onto the detector, a photomultiplier masked by a brass foil containing a $230\mu\text{m}$ pinhole. In order to reduce the intensity of the beam to a reasonable level, a reflective neutral density filter (ndf) may be inserted before the detector. Photographs of an early configuration of this experiment, before the addition of screens to reduce scattered light and draughts and using a piece of tissue paper as a rudimentary diffuser, are reproduced as plate III-1.

It will be noticed that the interferometer used in this experiment lacks the "compensating plate" usually included to correct for dispersion in the beamsplitter. Dispersion is not a problem with the narrow bandwidth sources used here; but there is an interesting consequence of the absence of this plate in that clear fringes are generated at zero path difference.

There are two effects by which the measured visibility may differ from the longitudinal coherence of the laser emission. The more important is that of fluctuations in the intensity of the laser beam, and is significant with this particular experiment only, for they could be removed by normalization of the Michelson fringe signal against the average incident laser power. The second is inherent in the use of the Michelson interferometer and concerns the finite size of the sampling pinhole. Both these effects are more pronounced at large path differences, although they have opposite consequences for the apparent coherence which is increased by laser fluctuations and reduced by an excessively large sampling pinhole. Neither effect was

calculated to be particularly significant in the cases reported here, but some correction for the effect of intensity fluctuation has nonetheless been included in the following results.

It is likely, nonetheless, that the coherence measurements made in this experiment underestimate the true longitudinal coherence. The need to realign of the apparatus at each new mirror displacement, and the increasing difficulty in achieving this as the displacement was increased, makes clear fringes less likely at the larger spacings. The reason for this difficulty may be attributed both to the reduced size of the fringes and to the reduced contrast itself.

III-4. Measured longitudinal coherence of the copper vapour laser.

The measurements of longitudinal coherence $C(\tau)$ obtained in these experiments are illustrated in figure III-4. Standard laser running conditions of 6.5kV, 245mA, 6.1kHz and around 50mbar of neon were used.

The experiment yields a coherence length (half width at half maximum) of 40 ± 2 mm, which is around two-thirds of that predicted for a single Doppler broadened component (60mm) but slightly higher than is calculated for laser action on the two strongest lines (34mm). Tenenbaum et al [III-1] found similar results at their temperature T_2 . In the experiment described here, the tube temperature was unknown. No structure is obvious in the distribution of figure III-4.

The data plotted have been adjusted to take into account fluctuations in the mean intensity as described above. The correction factor was found empirically by observing the fluctuations when one arm of the interferometer was blocked, and assuming the same fractional fluctuation for each visibility measurement. The exact magnitude of this correction is important only for the extreme points on the graph, and should not affect the overall estimate of coherence length.

The laser lineshape cannot be expected to match the calculated lineshape for spontaneous emission with any accuracy. Only if laser action is saturated for all the hyperfine components will the structure of figure III-1 be appropriate, and even then some gain narrowing of the Doppler broadened profile will occur. The calculated lineshapes for different numbers of lasing components, then, serve to define the range within which practical shapes are likely to occur.

III-5. Implications for holography using a copper laser source

The longitudinal coherence of a source laser used for recording holograms determines the maximum path difference allowed in the optical layout. This path

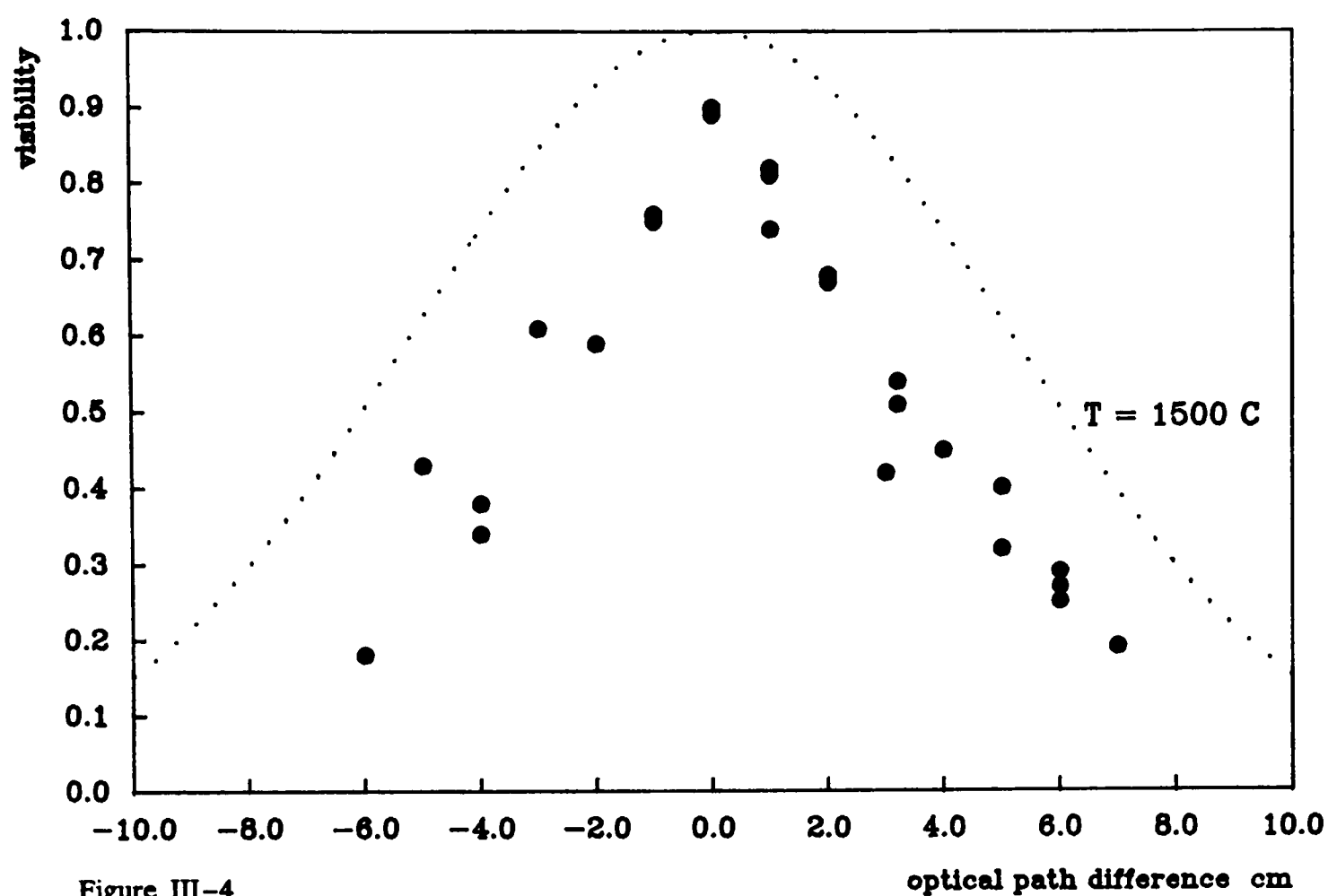


Figure III-4
Measured longitudinal coherence as a function of mirror position. The curve shows the calculated longitudinal coherence for a single hyperfine component corresponding to the Doppler limit at a temperature of 1500 C.

difference arises from the difference in the mean distances travelled by the object and reference beams, and from the illumination geometry and size of the object itself. The intensity of the reconstructed image is proportional to the visibility of the recorded interference pattern. A short longitudinal coherence length therefore limits the size of the object.

However, in a typical holographic recording layout, the reference beam will be from 3 to 10 times more intense than the scattered beam. The large reference intensity has two important effects: firstly, it ensures that the exposure of the hologram defines the amplitude of the interference pattern uniquely, by forcing the amplitude never to change sign; secondly, it uses the linear region of the film response by providing a background intensity throughout. Since the second effect involves the artificial reduction in contrast over the maximum theoretically possible, there is some trade-off possible here which would allow the effects of a limited coherence to be overcome.

The copper laser green line, then, should be suitable for recording holograms involving path differences as large as 100mm, in which case the extremes of the image will be some 40% as intense as the central portion.

III-6. Conclusion

This direct measurement of the longitudinal coherence of the copper vapour laser gives results which are thoroughly consistent with predictions based upon the lineshape data of Tenenbaum et al. Under the conditions prevailing at the time of the experiment, the measured coherence length (hwhm) was 40 mm, which is 0.67 of the coherence length calculated for a single hyperfine component having a Doppler-broadened profile. This coherence length may be expected to fall with increasing laser temperature as further components reach threshold, with a lower limit of 22mm.

It may be assumed, therefore, that the limited longitudinal coherence will be of no consequence for second harmonic generation, where typical crystal lengths are around a centimetre. Holographic applications, however, will be subject to a maximum path difference variation of around 100mm, depending upon the acceptable minimum visibility.

III-References

1. J Tenenbaum, I Smilanski, S Gabay, L A Levin, G Erez, S Lavi, "Structure of 510.6 and 578.2nm copper laser lines," Opt Comm 32 (3) 473-477 (1980)
2. I I Sobelman, "Atomic spectra and radiative transitions," Springer-Verlag, 1979.

Table 1. Corrected version of Table 2, Tenenbaum et al.

Frequency shifts of hyperfine components of the 510.6 and 578.2nm transitions relative to the centres of gravity of the ^{63}Cu transitions. Intensities relative to the strongest hyperfine transition (taken as 100). Corrections, based on the references quoted in [III-1], are shown in bold type.

λ (nm)	F	F'	^{63}Cu			^{65}Cu		
			$\Delta\nu$ (MHz)	I	Des.	$\Delta\nu$ (MHz)	I	Des.
510.6	3	4	-2423.6	100	a	-328.3	45	a'
	2	3	164.8	62	b	2424.9	28	b'
	3	3	721.6	16	c	3025.6	7	c'
	1	2	1909.2	35	d	4312.4	16	d'
	2	2	2328.4	19	e	4757.2	9	e'
	3	2	2885.2	1	f	5357.9	0.5	f'
	0	1	3034.6	17	g	5544.5	7	g'
	1	1	3258.6	15	h	5780.2	7	h'
	2	1	3677.8	2	i	6225.0	1	i'
578.2	2	3	-3818.9	100	a	-1868.1	45	a'
	1	2	857.7	36	b	3124.3	16	b'
	2	2	1871.5	36	c	4210.9	16	c'
	1	1	4422.3	36	d	6964.8	16	d'
	2	1	5436.1	7	e	8051.4	3	e'
	1	0	6135.9	14	f	8821.4	6	f'

Transverse coherence of the copper vapour laser.

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Transverse coherence of the copper vapour laser.

"If we see light at the end of the tunnel, it's the light of the oncoming train"
– Robert Lowell, "Day by Day".

IV-1. Introduction

The transverse coherence of a laser determines how tightly the beam may be focussed, and what intensities may be reached. It is a property which is of importance for dye pumping applications and for optical projection systems, when it may indeed be the only way in which the laser is distinguishable from a classical light source; in such applications, the transverse coherence properties of a laser are commonly but loosely described by the qualitative term "beam quality", or by the more meaningful "beam divergence".

The transverse coherence is of fundamental importance if the laser is to be used as the light source for holography: the process will mix light from all points across the beam profile, and poor coherence will lead to a weakening of the holographic interference pattern. However, it is when the laser is used for nonlinear processes, of which second harmonic generation is a good example, that the transverse coherence assumes vital importance. The signal strength from a coherent nonlinear process depends upon quadratic and higher powers of the fraction of the beam which is coherent, and will thus fall sharply as the transverse coherence is reduced. It is the transverse coherence of the laser which most determines the efficiency of second harmonic generation, and the relevance of transverse coherence to second harmonic generation has been the prime motivation of the work described in this chapter.

The origin of the transverse coherence of pulsed lasers is found to be quite different from those for c.w. devices. It can be shown that there is not only a fundamental divergence caused by diffraction from the laser output aperture, but that for pulsed lasers there is an equally fundamental limit to the transverse coherence which depends upon the laser geometry and duration of the light pulse, and which can prevent the divergence from falling to its diffraction limit. Transverse optical cavity modes are no longer an appropriate description of the optical field and the output of a copper laser behaves like that of a flashlamp enclosed within a delay line with associated gain. The copper laser is truly a light amplifier, and cavity oscillation

at optical frequencies is not an appropriate description of the operation of the device.

The coherence properties of a pulsed laser are commonly improved by the use of an unstable resonator configuration. The CU10 laser has been used throughout with an unstable cavity, and in the course of these experiments, cavities with a variety of magnifications have been investigated. By considering the path equivalent to an "unfolded" laser cavity, it is possible to derive a simple theory which describes the temporal development of the transverse coherence of a pulsed laser. This does not take account of non-uniform gain profiles or the complex nature of the initial laser emission: however, such a theory does help us to understand some of the aspects of the actual temporal evolution of the transverse coherence of the copper vapour laser. In practice, the predictions of this theory describe the measured properties of the copper vapour laser surprisingly well.

This chapter, then, will describe and account for the transverse coherence of the radiation from a copper vapour laser. Time-averaged measurements have been made of the transverse coherence for both the green and yellow lines, and the yellow line appears to have slightly better coherence than the green. Time-resolved studies have also been made to investigate the development of the coherence as the 510nm, green, laser pulse progresses, and are believed to be the first time-resolved studies of the transverse coherence of any pulsed laser. It will be concluded that there are two important effects governing the transverse coherence of a copper vapour laser. Firstly, the single shot coherence is determined by the magnification of the unstable cavity used, and the beam may be essentially diffraction limited for the majority of the pulse. Secondly, there is a significant variation in the direction of the beam over a number of shots, resulting from hot air currents around the laser head and discharge flicker within it, and this limits the shot-averaged coherence to an order of magnitude below the single pulse value. The disturbances due to thermal air currents outside the laser head have been all but removed during the course of this work, and the improvement in beam quality is significant.

IV-2. Theory of transverse coherence of light from a diffuse source.

Figure IV-1 shows a Young's fringes experiment in which an extended incoherent source is used. Fringes are formed as the result of interference between the light from the two pinholes. However, the point in the source from which the light originated is unknown, and thus the position of the fringes (their phase) is a little uncertain. The observed Fraunhofer diffraction pattern is therefore the average of these individual fringe patterns. It is clear that the visibility, or depth of modulation, of this pattern will depend upon the spacing of the pinholes, and that if the angular width of the source is much larger than the angular fringe spacing (ie. if the phase

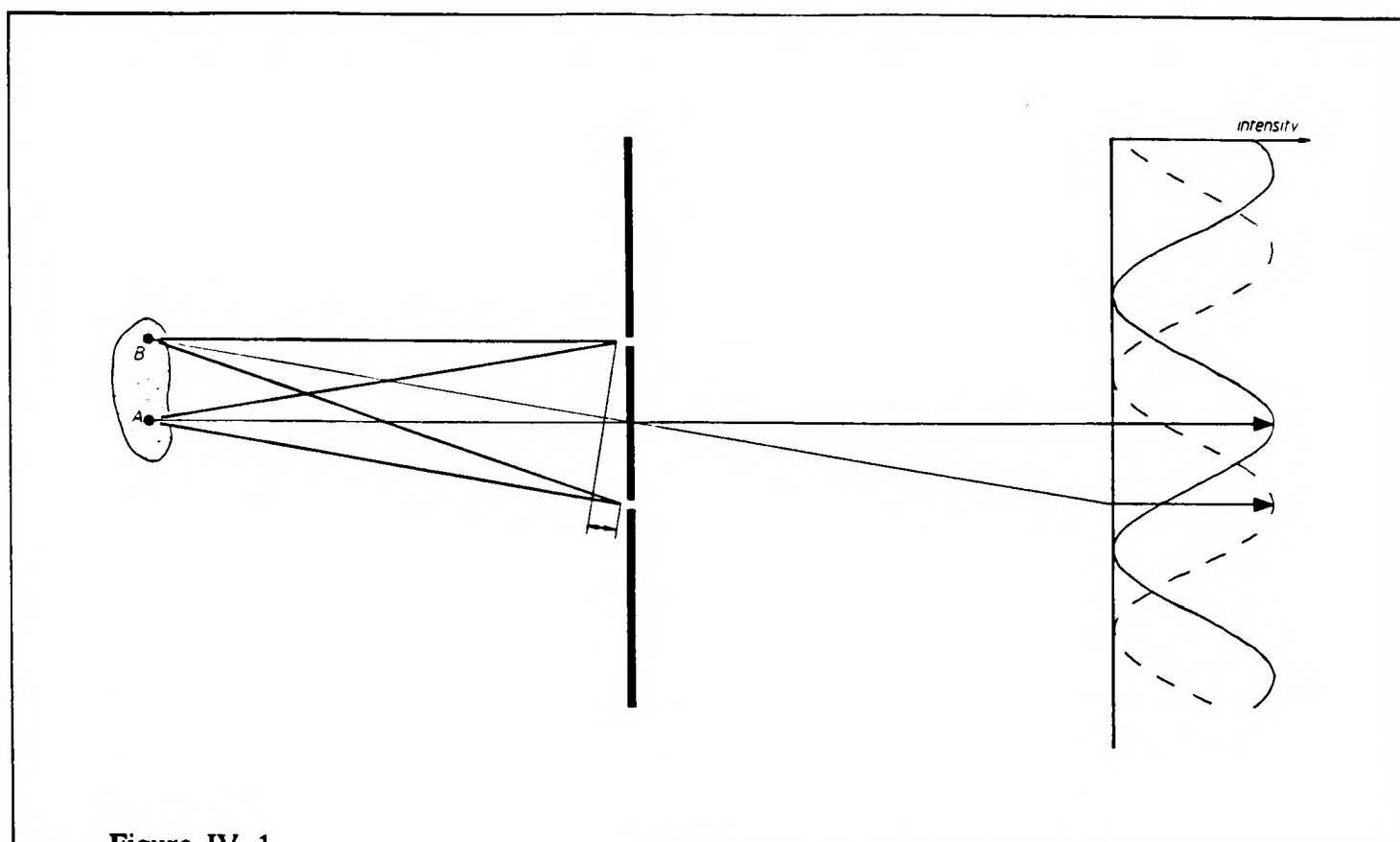


Figure IV-1

Transverse coherence. Young's fringes formed by light from a range of points in the source are displaced, so that source A produces the fringe pattern shown by the full line and the dotted line represents the interference of light from B. All such patterns, which are uncorrelated, then add causing a reduction in the overall visibility of the fringe pattern. The relative position of the fringe pattern is determined by the phase difference between the light reaching the two pinholes, and thus to the path difference indicated. The uncertainty in fringe position thus follows the uncertainty in relative phase between the pinholes.

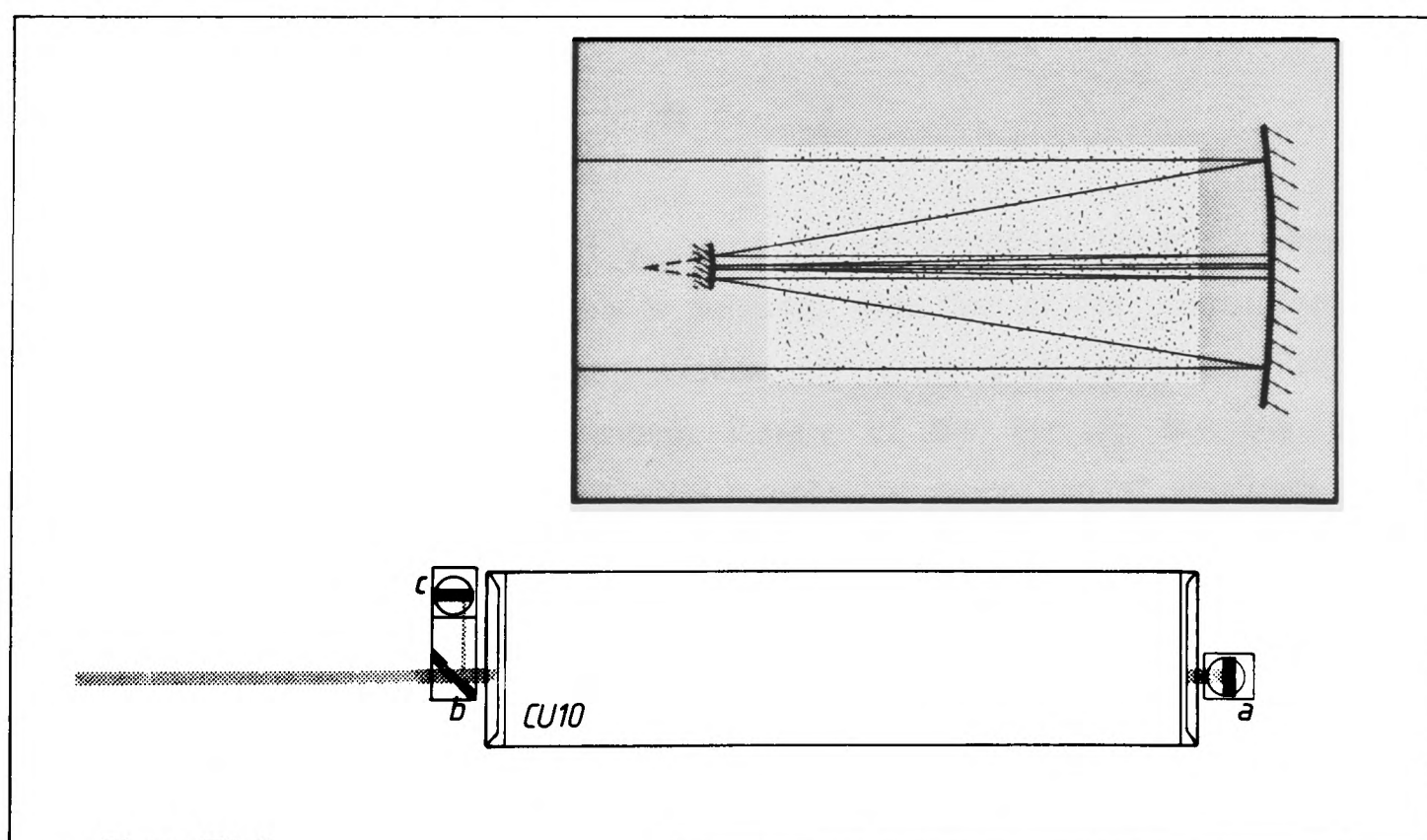


Figure IV-2

Unstable cavity geometry. The copper vapour laser has been used with an external unstable cavity. Mirror a is a full aperture concave mirror with a 1600mm focal length; various short focal length convex mirrors may be inserted at c, and are included in the cavity via a small reflecting area deposited onto an otherwise antireflection coated beamsplitter b. The reflecting area is to the side of the laser beam to make use of the earlier emission at the tube walls. The output beam will show a shadow of this small reflector. Inset: general confocal unstable cavity. The two mirrors share a common focal point, resulting in a parallel output beam. The majority of the gain medium is only seen in the final pass before emerging.

difference between the two pinholes can vary over more than a cycle) then the fringe pattern will be completely blurred out.

The transverse coherence of any given illumination is the extent to which the phase of the light field at one point defines the phase at another point transversely separated from it. It is defined strictly as the normalized correlation of the two radiation fields. It may also be shown that it is the proportion of the field amplitude at one point which is coherent with that at the other, and is equal to the visibility of fringes resulting from the interference of the two fields provided they have the same intensity. The coherence between two points as a function of their relative position is determined by the Van Cittert-Zernike theorem to be the Fourier transform of the angular intensity distribution of the light source. This theorem is the spatial analogue of the Wiener-Khintchine theorem which described the longitudinal coherence in chapter III.

The normalized coherence function $\gamma_{1,2}$, defined as

$$\gamma_{1,2} = \frac{\langle E_1 E_2^* \rangle}{[I_1 I_2]^{\frac{1}{2}}} \quad (\text{IV-1})$$

is related to the visibility of fringes (see equation (III-8)) by $V=|\gamma|$ if the pinholes are of equal size and illumination. E_1 and E_2 are the scalar electric fields whose coherence is being studied and I_1 and I_2 are the associated light intensities. We may also write the general relation in the form [IV-1]

$$I = I_1 + I_2 + 2I_1^{\frac{1}{2}}I_2^{\frac{1}{2}}\gamma_{1,2} \quad (\text{IV-2})$$

and specifically

$$I_{\text{max,min}} = I_1 + I_2 \pm 2I_1^{\frac{1}{2}}I_2^{\frac{1}{2}}|\gamma_{1,2}| \quad (\text{IV-3})$$

from which it is apparent that $\gamma_{1,2}$ may be interpreted as being the proportion of the amplitude at one pinhole which is coherent with that at the other. The remainder of the amplitude, which from conservation of energy must be $(1-\gamma_{1,2}^2)^{\frac{1}{2}}$, will not be coherent, and will thus contribute merely to the average intensity of the fringe pattern. In summary, $\gamma_{1,2} = \text{visibility} = \text{coherent proportion of amplitude}$.

From the Van Cittert-Zernike theorem, we may calculate the transverse coherence for light from a uniform circular source as a function of the distance r between the sampling points. The result is an Airy function, $\gamma_{1,2} = 2J_1(kr\theta)/(kr\theta)$, whose width is given by a coherence radius $r_{\text{coh}} = 0.411\lambda/\theta$ (half width at $1/e$ of maximum) where θ is the angle subtended by the radius of the source at the plane of observation, and $k=2\pi/\lambda$. If the source is a disc of radius ρ at a distance R from the plane of observation, then we obtain

$$r_{\text{coh}}=0.411\lambda R/\rho. \quad (\text{IV-4})$$

We note that these results are based upon Fraunhofer diffraction theory, and are thus accurate only when the plane source is in the far field of the coherence area. The copper laser always satisfies this criterion.

The copper laser is quite unlike a cw or long pulse laser – there is time for only three or four round trips of the cavity during the laser pulse, and thus the CVL tends to behave more like an incoherent source than like a cw laser. The transverse coherence of the copper laser may indeed tend towards the diffraction limit within the duration of the laser pulse, but the period during which the coherence develops towards this limit is always going to be of significance.

IV-3. The role of the unstable cavity.

It is common with high gain pulsed lasers of large cross-section to use an unstable optical cavity, which yields rather better transverse coherence than stable configurations. The typical unstable cavity geometry is illustrated in figure IV-2, and strongly resembles a Newtonian reflecting telescope; this similarity will become apparent in the treatment presented overleaf.

Early attempts [IV-2] to treat the unstable resonator theoretically retained from work with stable geometries the idea of transverse modes, and the unstable cavity was considered to offer a higher discrimination against those of higher orders. But the concept of transverse modes is hardly relevant to the unstable cavity, for it originates in the very stability which is lacking in the unstable arrangement.

A more useful approach was adopted by Zemskov et al [IV-3], who considered instead the confinement and transformation by the cavity of diverging and converging beams of spontaneous radiation. In a later paper [IV-4], the authors acknowledge that this method is inappropriate for a detailed treatment of the properties of the beam of radiation during the build up time of the pulse, but the theory nonetheless yields results consistent with the approach used in the following section, in which the improved coherence of the unstable cavity will be considered the result of a rapidly diminishing source cross-section. This latter explanation is the more appropriate for short pulse, long cavity lasers such as the copper vapour laser in which there is barely time for three or four round trips during the gain period, and a recent account of this model has been published by Eggleston [IV-5].

The following model will be considered: the copper vapour laser consists of a plasma tube of uniform bore, which partially fills the cavity formed by two mirrors, one large and concave and one small and convex. The laser pulse is assumed to start with a flash of spontaneous emission of short duration throughout the laser medium, which is initially considered to fill the whole cavity. This acts as the source for all subsequent emission, and thereafter the laser acts purely as a light amplifier working

upon this initial seeding. This assumption is equivalent to assuming a fast build up of the intra-cavity flux towards saturation. Cavity modes are regarded as irrelevant, and the resonator is unfolded to give a chain of lenses and gain media as shown in figure IV-3. The model thus assumes that the laser emission is seeded by an incoherent disc source with the diameter of the laser plasma tube, which recedes down the unfolded cavity at the speed of light away from the unfortunate observer. The transverse coherence of the laser emission is thus determined by the apparent size and position of this incoherent source.

The evolution of the transverse coherence will be considered in four stages. The first stage corresponds to the start of the pulse when the source is making its first pass down the laser tube and is not yet influenced by the cavity. The second and third stages correspond to the second and third passes along the tube, when first the concave mirror and then the convex reflector are involved. Finally, the development of the coherence after an arbitrary number of round trips of the cavity will be considered. The combination of these individual stages allows the form of the coherence to be calculated for any time, and this has been used to compute the development of the transverse coherence for each cavity magnification. Throughout, the laser will be characterized by a cavity length L_c , magnification M , and tube radius a .

Ultimately, the laser beam will be essentially coherent over its entire width. Further reductions in the effective source size will no longer have a significant effect, and the output will behave as if plane parallel light were incident upon an aperture having the diameter of the laser plasma tube. This beam is now said to be "diffraction limited", since its divergence and focusability are determined entirely by diffraction from the finite width of the laser beam. Except for the complication of some transverse mode structure, the laser now has the coherence properties of a stable c.w. laser. With a high magnification unstable cavity, this limit is reached by a copper vapour laser before the peak of the laser pulse.

IV-4. Modelling the transverse coherence.

IV-4.1 The first pass, and the plane-plane cavity

The coherence radius for illumination of a surface by an incoherent circular disc of radius ρ , when the coherence radius is defined as the $1/e$ point of the function $2J_1(kr\theta)/(kr\theta)$, is

$$r_{\text{coh}} = \alpha \lambda R / \rho \quad (\text{IV-5})$$

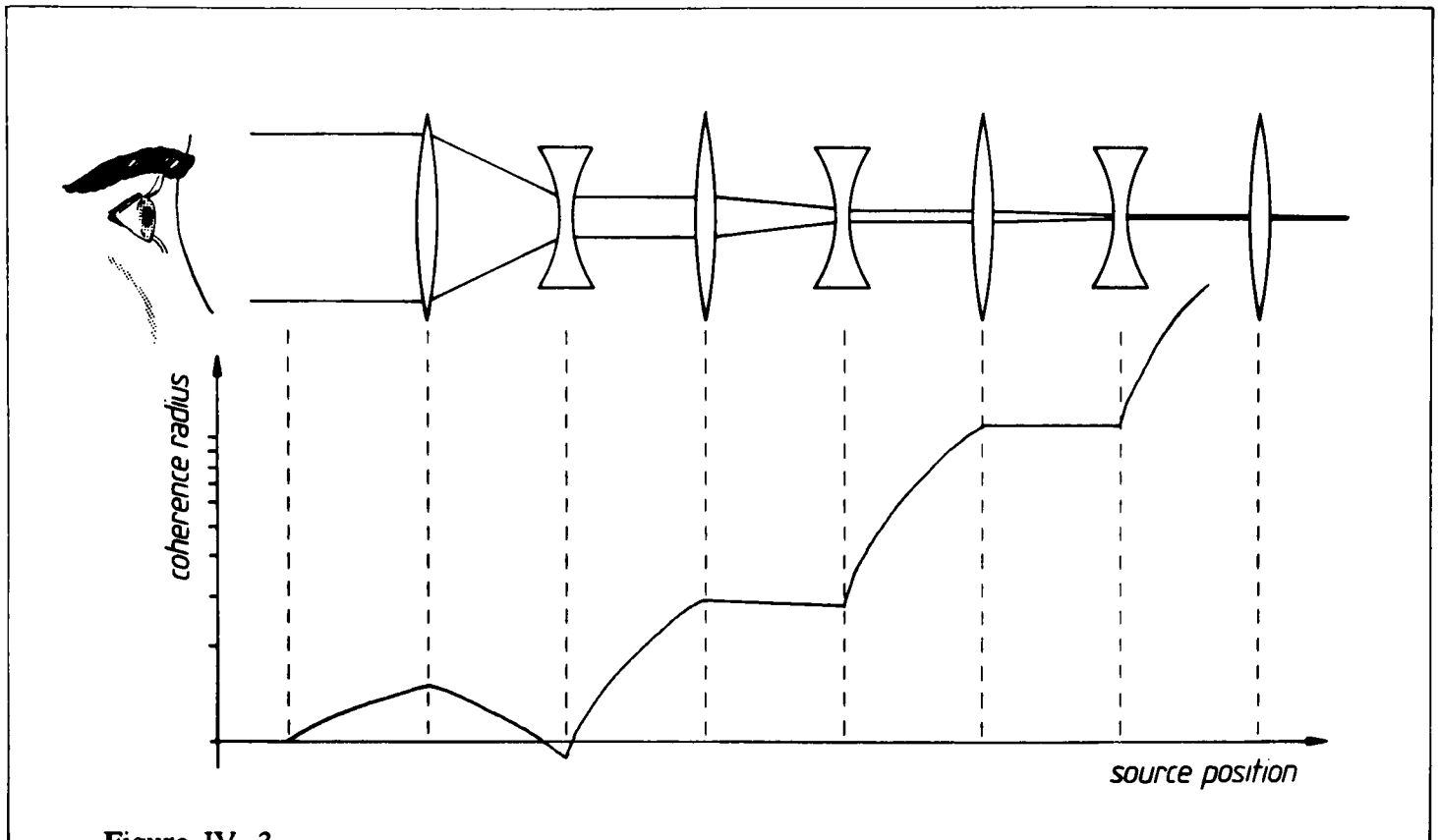


Figure IV-3
Model of the unstable cavity. Transverse mode structure is unimportant, and the cavity is therefore unfolded to be represented by a chain of lenses. The initial laser emission recedes down this chain and its apparent size to the observer tends to reduce. The coherence radius thus develops as indicated by the computed example plotted. The vertical axis is logarithmic.

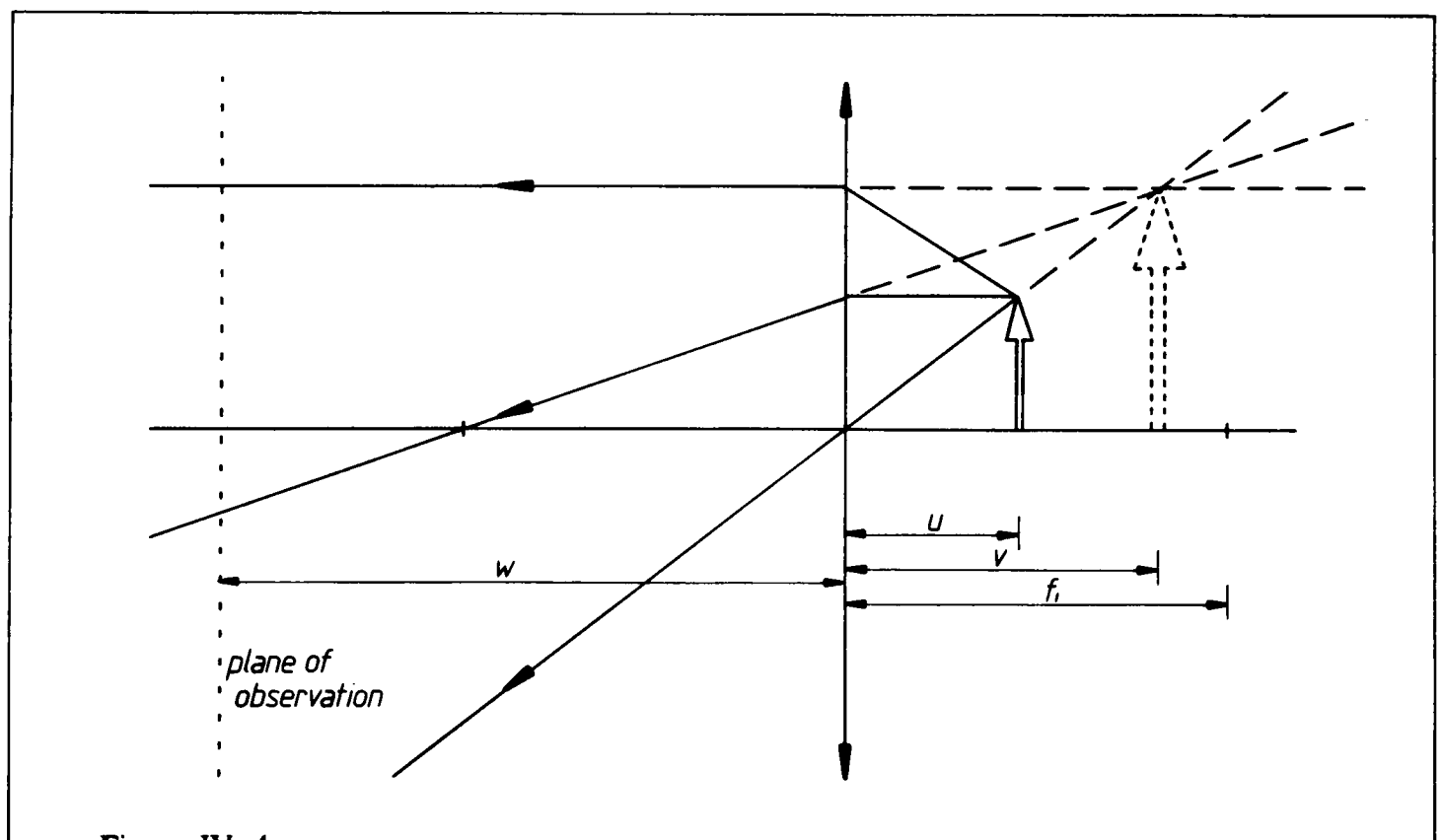


Figure IV-4
Calculation of apparent source size. The true source, represented by the solid arrow, is viewed in this case through a single converging lens, which is in practice the concave laser cavity mirror. The apparent source has a size and position indicated by the broken arrow.

where $\alpha = 0.411$. A similar form holds for other source shapes with different values of α . For the first pass down the laser tube, we simply put $R=ct$, ρ =tube radius, and obtain

$$r_{\text{coh}} = \alpha \lambda ct / \rho. \quad (\text{IV-6})$$

This describes the evolution of the coherence radius during the first pass down the laser tube, before any magnifying optical components have been involved, and corresponds to the first, linear, section of figure IV-4. It also fully describes the development of the transverse coherence for a plane-plane cavity. In this case, it will be perhaps $3\mu\text{s}$ before the coherence function has grown to fill a copper laser output aperture; typically, a copper laser pulse lasts only 25ns.

IV-4.2 The second pass along the tube (the first return trip)

From the geometry illustrated in figure IV-3 it may be shown that the first converging mirror gives the source an apparent position and size defined by $v=uf_1/(f_1-u)$ and $\rho'/\rho=f_1/(f_1-u)$, where u and v are the object and image distances from the lens f_1 . Putting $ct=w+u$, where w is the distance between the lens and the observer, we obtain

$$r_{\text{coh}} = \alpha \lambda \left[(1-w/f_1)ct + w^2/f_1 \right] / \rho \quad (\text{IV-7})$$

Note that if the plane of observation is more than f_1 in front of the first lens, then the coherence radius actually decreases with time during this first return trip, for the lens or mirror is acting as a magnifying glass. This result applies to the cavity alone, and during the second pass the laser plasma tube will obscure the edges of the image, so that the coherence radius will in fact be a little larger than this estimate. Incomplete filling of the laser cavity by the plasma tube makes rather more careful calculation of this effect pointless, and has the consequence that the peaks and troughs of figure IV-4 will be clipped. The seeding of the laser pulse must have occurred within the gain region, and thus it is the earliest available seeding which is significant. When the gain region does not fill the entire cavity, there will be times when this seeding cannot have originated at the very start of the pulse.

IV-4.3 The third pass along the tube

A similar calculation may be made for a full round trip of the cavity. A page of algebra quickly yields the result

$$r_{\text{coh}} = (\alpha\lambda/\rho) \cdot [(f_1/f_2 - f_2/f_1)w - f_1/f_2 \cdot ct + (f_1 + f_2)^2/f_2] \quad (\text{IV-8})$$

This section of the curve of r_{coh} against t thus has a gradient equal to $(-f_1/f_2)=M$, the cavity magnification, as illustrated in figure IV-4. Once again, some aperturing of the apparent source may occur for the initial part of this stage, but by the end the whole source should again be visible.

IV-4.4 After an arbitrary number of round trips

Eggleston [IV-5] has shown that the apparent radius and position of the source after n round trips of the cavity are given by

$$R_s = M^n R_a$$

and

$$L_s = L_c [M^{2n} - 1]/[M^2 - 1], \quad (\text{IV-9})$$

where R_a and L_c are respectively the true source radius and the cavity length respectively. Eggleston assumes a cavity in which the angular magnification occurs entirely at the beginning of the cavity transit. To generate results consistent with the previous treatment, we consider a cavity round trip in which the light passes initially through a diverging lens, after which free propagation over one cavity length occurs before the light passes through the second, converging, lens. This is illustrated by figure IV-5. The second pass down the cavity is then made. The resulting round-trip ray transfer matrix is now

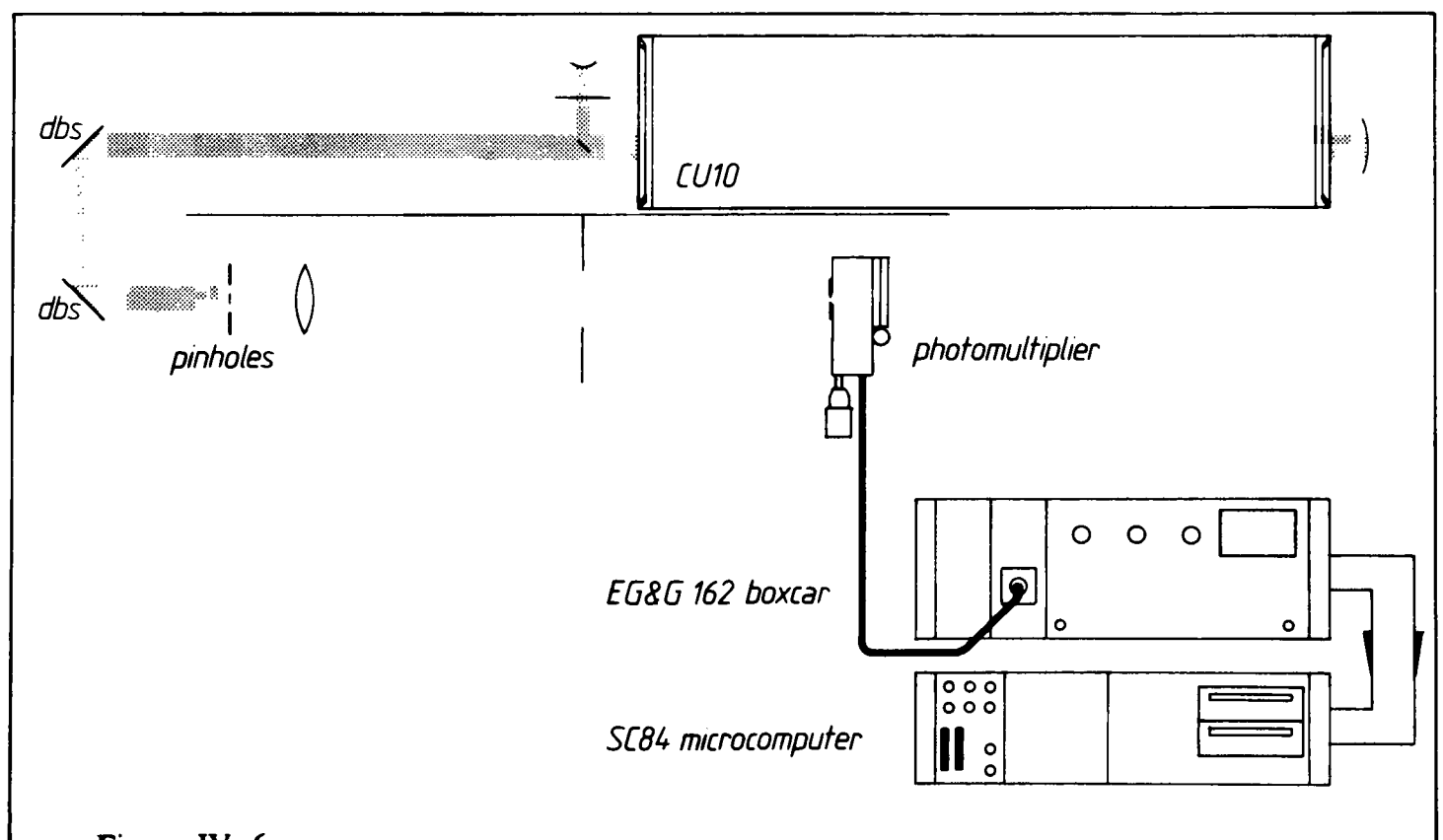
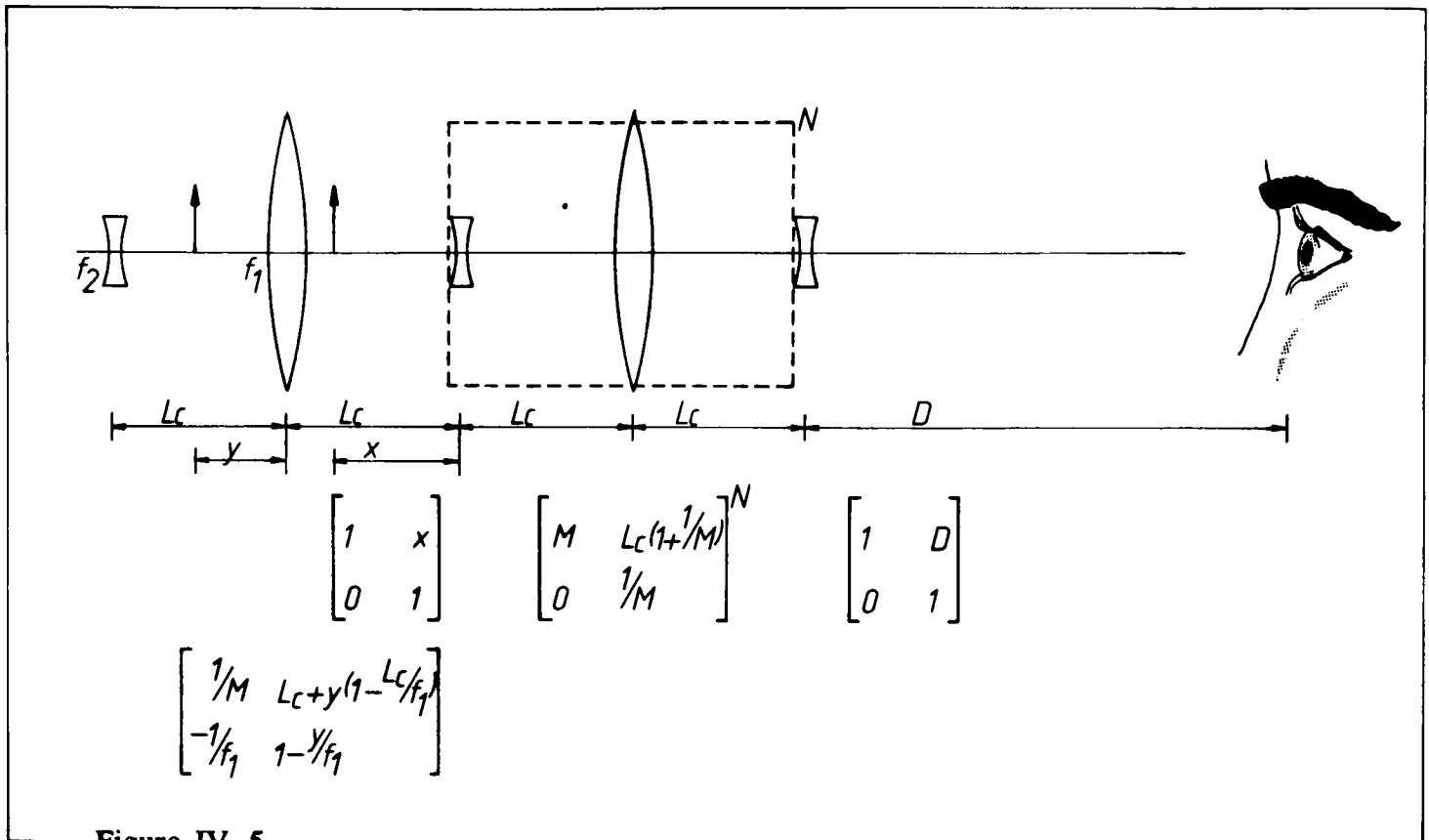
$$\begin{bmatrix} M & L_c(1+1/M) \\ 0 & 1/M \end{bmatrix} \quad (\text{IV-10})$$

Applying Eggleston's result for the n^{th} power of such a matrix we find that the effective size and position of the source are given by

$$R_s = M^n R_a$$

and

$$L_s = L_c [M^{2n}-1]/[M-1] \quad (\text{IV-11})$$



We thus arrive at a general result for the coherence radius after n round trips:

$$r_{\text{coh}} = (\alpha\lambda/\rho) L_c [M^{2n}-1]/[M-1][M^n] \quad (\text{IV-12})$$

This result agrees with those of parts IV-4.2 and IV-4.3 for the case of one round trip.

Since $L_c = f_1[1-1/M]$, we have,

$$r_{\text{coh}} = (\alpha\lambda/\rho) f_1 [M^{2n}-1] / [M^{n+1}] \quad (\text{IV-13})$$

Then, making the approximation $M^{2n} \gg 1$, we obtain

$$r_{\text{coh}} = (\alpha\lambda f_1/R_a) M^{n-1}. \quad (\text{IV-14})$$

This allows us to calculate the number of round trips required before a diffraction limited beam is achieved, by putting $r_{\text{coh}} = R_a$, whence

$$\ln\{R_a^2/\alpha\lambda f_1\} = (n-1)\ln M \quad (\text{IV-15})$$

or

$$n = 1 + \ln M_0 / \ln M \quad \text{where } M_0 = R_a^2 / \alpha\lambda f_1 \quad (\text{IV-16})$$

This result is consistent, to within small arbitrary constants in defining M_0 , with the results obtained by Zemskov et al [IV-3, IV-4].

For practical purposes, the precise value of α is of little importance. Nonetheless, the value 0.411 will be used henceforth, consistent with equation (IV-4). Using a cavity length of 1.5m and tube bore of 25mm, we obtain

$$n = 1 + 6.21/\ln M \quad (\text{IV-17})$$

Thus for

$M = 15,$	$n = 3.3$
$M = 115$	$n = 2.3$
$M = 148$	$n = 2.2$

and the time difference between the $M=15$ and $M=150$ cavities reaching the diffraction limit should thus be some 10ns.

It is worth considering the significance of the plasma tube bore upon the transverse coherence of the copper vapour laser. The tube bore determines both the size of the initial source and the final coherence area which must be filled; a large tube diameter will thus take longer to reach the diffraction limit on both counts. Equation (IV-16) shows that the dependence of n upon the tube bore is given by

$$n = 1 + \ln(R_a^2 / \alpha \lambda f_1) / \ln M$$

$$= 1 + 2 \ln R_a / \ln M - \ln(\alpha \lambda f_1) / \ln M \quad (\text{IV-18})$$

by which doubling the tube bore will increase n by 0.6 at a magnification of 10, or by 0.3 when the cavity magnification is 100.

IV-4.5 Divergence of the copper laser beam

The propagation of a beam outside the laser cavity is determined by the form of the wavefronts emerging from the laser tube, and the optical field distribution is given by the diffraction pattern of this source. The far-field amplitude distribution of the laser beam is therefore

$$a(\theta) = \int a(\underline{r}) \exp [ik\underline{r} \cdot \underline{\theta}] d\underline{r} \quad (\text{IV-19})$$

where $\underline{r}$ and $\underline{\theta}$ are vectors defining the position within the laser output aperture and the angular coordinates in the far field. The far-field intensity distribution is therefore given by

$$\begin{aligned} I(\theta) &= \left| \int a(\underline{r}) \exp [ik\underline{r} \cdot \underline{\theta}] d\underline{r} \right|^2 \\ &= \iint a(\underline{r}_1) a(\underline{r}_2)^* \exp [ik\underline{r}_{12} \cdot \underline{\theta}] d\underline{r}_1 d\underline{r}_2 \\ &\quad \text{where } \underline{r}_{12} = \underline{r}_2 - \underline{r}_1 \\ &= \iint \langle a(\underline{r}_1) a(\underline{r}_2)^* \rangle \exp [ik\underline{r}_{12} \cdot \underline{\theta}] d\underline{r}_1 d\underline{r}_2 \\ &\quad \text{when the time-average is taken} \\ &= \iint J_{12}(\underline{r}_{12}) \exp [ik\underline{r}_{12} \cdot \underline{\theta}] d\underline{r}_{12} \\ &\quad \text{from the definition of the mutual intensity} \\ &\quad J_{12} \text{ between points 1 and 2 [IV-1]} \\ &= \int d\underline{r}_2 \cdot \int J_{12}(\underline{r}_{12}) \exp [ik\underline{r}_{12} \cdot \underline{\theta}] d\underline{r}_{12} \\ &\quad \text{if } J_{12} \text{ is independent of } \underline{r}_2 \\ &\propto \int \gamma_{12} \exp [ik\underline{r}_{12} \cdot \underline{\theta}] d\underline{r}_{12} \quad (\text{IV-20}) \\ &\quad \text{if } J_{11} \text{ and } J_{12} \text{ are independent of } \underline{r}_2 \end{aligned}$$

γ_{12} is the normalized complex degree of coherence. Thus if the optical field over the output aperture has uniform amplitude and constant coherence, the far field intensity distribution will be the Fourier transform of the coherence function.

This makes the effect of the laser output aperture particularly clear, for the

coherence function will be bounded by the laser output aperture, and thus it may be included in the above expression which, upon application of the convolution theorem, becomes

$$I(\underline{\Theta}) = \int \gamma_{1,2} \exp \left[i k \underline{r}_{1,2} \cdot \underline{\Theta} \right] d\underline{r}_{1,2} \otimes F(\underline{\Theta}) \quad (\text{IV-21})$$

where $F(\underline{\Theta})$ is a function equal to the intensity of the Fraunhofer diffraction pattern which would be generated by the laser aperture. The divergence of the laser beam cannot therefore be less than that determined by the diffraction pattern of the aperture. This is the fundamental diffraction limit for all lasers, and if the divergence of the laser is defined by the 1/e intensity full-angle, then a circular output aperture of radius a will have a divergence Θ given by

$$\Theta = 0.610 \lambda/a \quad (\text{IV-22})$$

If the coherence radius is rather smaller than the radius of the output aperture, however, the laser beam is not diffraction limited, and the divergence is determined by the coherence radius. For a Gaussian coherence function, the full divergence angle is now given by

$$\Theta = 2\lambda/\pi r_{\text{coh}} \quad (\text{IV-23})$$

The typical time-averaged r_{coh} of 2mm which will be met in section IV-5 thus corresponds to a divergence of $160\mu\text{rad}$, or $140\mu\text{rad}$ fwhm. It will be useful to bear in mind the coherence radius due to spontaneous emission after the 3m journey along the optical bench: this is around $50\mu\text{m}$, and corresponds to a divergence of 5mrad fwhm.

For a uniform disc source, then, equation (IV-14) may be used to give the divergence for a pulsed laser after n round trips

$$\Theta_{\text{lim}} = 1.549 \frac{R_a}{f_1} M^{-(n-1)} \quad (\text{IV-24})$$

In the derivation of equation (IV-20), the coherence function $\gamma_{1,2}$ was assumed to be independent of $\underline{r}_2$, the position in the beam cross-section. If the source is circular and the coherence is not affected by either beam occlusion or spatial gain variations, this should indeed be true. The results plotted as figure IV-11 offer initial confirmation that this is the case.

IV-4.6 Computer modelling of the transverse coherence

The ray transfer matrix treatment of the development of the transverse coherence of the copper vapour laser may be extended to describe the coherence at any time in the laser pulse. The incomplete round trip of the laser from the time of initial seeding is represented by one of two matrices depending upon whether or not the converging lens is to be included. This is then multiplied by the N^{th} power of the round trip matrix which accounts for the N complete passes up and down the tube, and finally a translation matrix describes the propagation of the laser beam from the laser output aperture to the pinhole pair. These matrices are illustrated against their respective sections of the cavity transit in figure IV-5, and this model has been used as the basis of a computer program CALCOH.BAS. This is listed in Appendix IV-3 and calculates the evolution of the predicted coherence radius for a given laser configuration. The results of this computer model are shown in figure IV-3, and indeed follow the explicit results derived in the preceding sections.

IV-5. Experimental determination of transverse coherence of the copper laser

The theory presented above gives a reasonably rigorous prediction of the transverse coherence properties of a pulsed laser, provided that the assumptions which have been made are valid. In practice, it is not known how long it takes the copper vapour laser to reach saturation, and thus how long the seeding by spontaneous emission lasts, nor whether the spatial dependence of the gain will affect the transverse coherence. Most importantly, there is considerable uncertainty regarding the timing of the start of the laser pulse relative to the peak power. This last effect determines how long the coherence has had to develop before significant power is extracted from the laser. This is particularly difficult to measure directly, for the initial spontaneous emission should be many orders of magnitude weaker than the a.s.e. of the laser pulse.

It is therefore essential to measure the transverse coherence of the laser. It would be particularly interesting to perform time resolved measurements to examine the development of the coherence within the laser pulse. To achieve this, we use the fact that the coherence between two points is equal to the visibility of Young's fringes from evenly illuminated identical pinholes at those points. By measuring the visibility of fringes produced by pinhole pairs with a variety of pinhole spacings, the profile of the transverse coherence may be determined.

IV-5.1 Experimental details

The basic apparatus used in this experiment is illustrated in figure IV-6. The experiment was based around an array of pairs of pinholes used previously in a study of the transverse coherence of a KrF excimer laser [IV-6]. This, in turn, was inspired by the original work by Thompson and Wolf [IV-7].

The pairs of pinholes, with spacings from $35\mu\text{m}$ to 10mm , were made by focussing the beam from a Nd:YAG laser onto $5\mu\text{m}$ thick nickel foil. Each pair was separated from its neighbours by 1mm , and the nominal pinhole diameter was $10\mu\text{m}$. The foil was mounted on a vertical translation stage behind a slit aperture which allowed the copper laser radiation to strike only one pinhole pair at a time.

The resulting fringe pattern was focussed using a 1m focal length lens onto a small pinhole ($115\mu\text{m}$ radius), mounted in front of a photomultiplier on a translation stage. This allowed the detector to be scanned across the fringe pattern. Black cardboard was extensively used to mask the fringe pattern from the scattered copper laser beam, for the intensity of the fringe pattern from the pair of $10\mu\text{m}$ pinholes was easily matched by laser light scattered off the ceiling, for example.

The photomultiplier signal was then taken to a boxcar averager, triggered directly from the copper vapour laser control box. For the first experiments an EG&G 162 boxcar system was used, with the model 163 sampled integrator giving a gate width of 350ps . An analogue connection to the microcomputer allowed data to be stored on floppy disc for later analysis. Later, the EG&G unit was replaced by a Stanford Research Systems boxcar, with the model 250 gated integrator operated with a gate width of around 2ns . Connection to the microcomputer in this case was made digitally by an RS232 serial interface. In both cases, time resolved measurements of the fringe intensity were possible, allowing the development of the transverse coherence during the laser pulse to be studied.

Using the basic apparatus just described, a series of experiments was performed. The usual technique was to find the fringe maximum or minimum by maximizing or minimizing the measured intensity, and thus recording three measurements: the maximum intensity, the minimum intensity, and a 'background' intensity with the pinhole pair masked. In each case, the recorded signal could be either a single time-averaged value using a gate width greater than the laser pulse length or a time-resolved signal recorded by using a narrow gate width and scanned delay of the measurement with respect to the laser trigger. It may be noted that the 'background' trace provides more than just a 'zero' reference. There is significant electrical noise on the signals, generated by the laser discharge, and this is removed by subtracting a recorded background signal.

The results were processed by subtracting the background from the maximum and minimum traces, and then calculating the visibility ($= [I_{\text{max}} - I_{\text{min}}] / [I_{\text{max}} + I_{\text{min}}]$).

The time resolved experiments yielded a data set of visibility as a function of both pinhole separation and time. By fitting each visibility-pinhole separation set to a Gaussian, and defining a "coherence radius" by the 1/e point of this curve, the results have been reduced to the temporal evolution of this single parameter.

It was found in practice that there was some variation in the size of the pinholes, and indeed it is not even clear that all the pinholes were truly circular. This was of particular concern to Graeme Hirst who spent much effort in measuring the ratio of the areas of each pair of pinholes. Hirst's techniques are described in chapter 7 of his thesis [IV-6], and yielded ratios which were trusted to within 0.5% of which the worst was 0.63. The effect of non-equal pinholes is that the fringe pattern cannot fall to zero intensity even for complete coherence. It is shown in appendix IV-1 that the measured visibility differs from the coherence function in this case by a factor $(A^{\frac{1}{2}} + A^{-\frac{1}{2}})/2$, where A is the ratio of the pinhole areas. If we put $A=1+a$ and expand the resulting expression, we obtain a factor of $1+a^2/8 + \dots$, and it is thus obvious that this correction is not a strong function of A. For most of the pinholepairs used in this experiment, the correction was less than 1%.

A further allowance had to be made for the diameter of the sampling pinhole, which in some cases was comparable with the fringe spacing. It is shown in appendix IV-2 that the visibility in this case differs from the coherence function by a factor of

$$\frac{\text{measured visibility}}{\text{true visibility}} = 1 - \frac{4}{\pi} \int_0^1 [1-\beta^2]^{\frac{1}{2}} \sin^2 \gamma \beta \, d\beta \quad (\text{IV-25})$$

where $\gamma = \pi r / \text{fringe spacing}$

In general, this factor was less than 20%, although some corrections up to 50% were used.

It is difficult to detect the initial weak spontaneous emission which seeds the laser pulse, for any detection system will have to cope with the full laser pulse power which follows it. No reliable measurements have been possible of the point at which emission first occurs.

The laser had a plasma tube bore of 25mm, and the laser beam thus was somewhat narrower than this due to edge effects or masking from the accumulation of unmolten copper. The eventual beam width was around 20mm. Figure IV-11 shows that the time-averaged coherence has a radius of around 0.5mm, and thus the cross-section of the coherent radiation is around 20 times smaller than the full diameter of the copper laser beam. The initial build-up of the transverse coherence is seen clearly in the traces of figure IV-7, and the slower rise of the lower magnification cavity is also clearly apparent. Figures IV-7 are at first puzzling in the way that the coherence radius appears to reach a limiting value which is independent of cavity magnification and remains constant for the remainder of the laser pulse; limits due to a.s.e. or non-uniform gain would both depend strongly upon the varying

laser conditions as the pulse progresses, and the sensitivity to these effects would be expected to depend upon the cavity magnification.

This phenomenon is consistent with the presence of a random beam-steering effect which causes the fringe pattern move, and which will therefore blur out the time-averaged fringe pattern for small fringe spacings. Light beams sampled by the pinholes are considered coherent if there is a well defined phase difference between them; beam 'wander' means that this phase difference varies slowly with time, so that the coherence is good over short timescales but poor over periods of seconds.

This interpretation has been convincingly demonstrated by direct observation of the fringe pattern through a microscope, when clear but unstable fringes could be seen for even large pinhole separations. It was suggested that this effect could be due to hot air currents outside the laser head, to steering of the beam by the flickering discharge within it, or to mechanical vibration from the cooling fans and vacuum pump. In practice, the first two causes have been investigated, and the former is the more significant.

The effects of thermal air currents were the easiest to investigate. Initially, crude beam guards made of cardboard and sticky tape were fitted between the laser head and the ends of the laser head box. The fringes, viewed through the microscope, were observed to be more stable, but the jitter was seen to be further reduced if the cooling fans were covered with pieces of card, and to be still more improved if the fans were temporarily turned off.

Thus motivated, a pair of foam beam guards was fitted. These beam guards have been fitted to the most recent commercial CVLs merely to prevent scattered light from emerging through the fan holes in the head box, but they were also thought to provide some degree of air seal. With these guards installed, the fringe jitter was reduced to a level equal to that achieved earlier by switching off the fans. Some underlying movement remained, and was of a more random and sudden nature than before: the fringe pattern would be stationary for periods of perhaps a second or two, and would then jitter for a short period before steadying again. There is some evidence that this disturbance was caused by distortion of the optical path by the discharge switching anchor points at the electrodes. Separate investigations in this laboratory of the effect of the discharge in a copper vapour laser have confirmed that this explanation is plausible, although the possibility of perturbation by residual air currents cannot be eliminated. Beam steering could also be caused by vibration from cooling fans and pumps. All these forms of variation would be too slow to affect the coherence of a single pulse from the copper laser.

With the foam beam guards fitted, the apparatus was reassembled to repeat the determination of the time resolved transverse coherence, giving the results shown in figures IV-8 to IV-10. With a 1600mm focal length concave mirror, convex mirror radii of curvature of 180, 28 and 22 gave magnifications of 18, 115 and 148

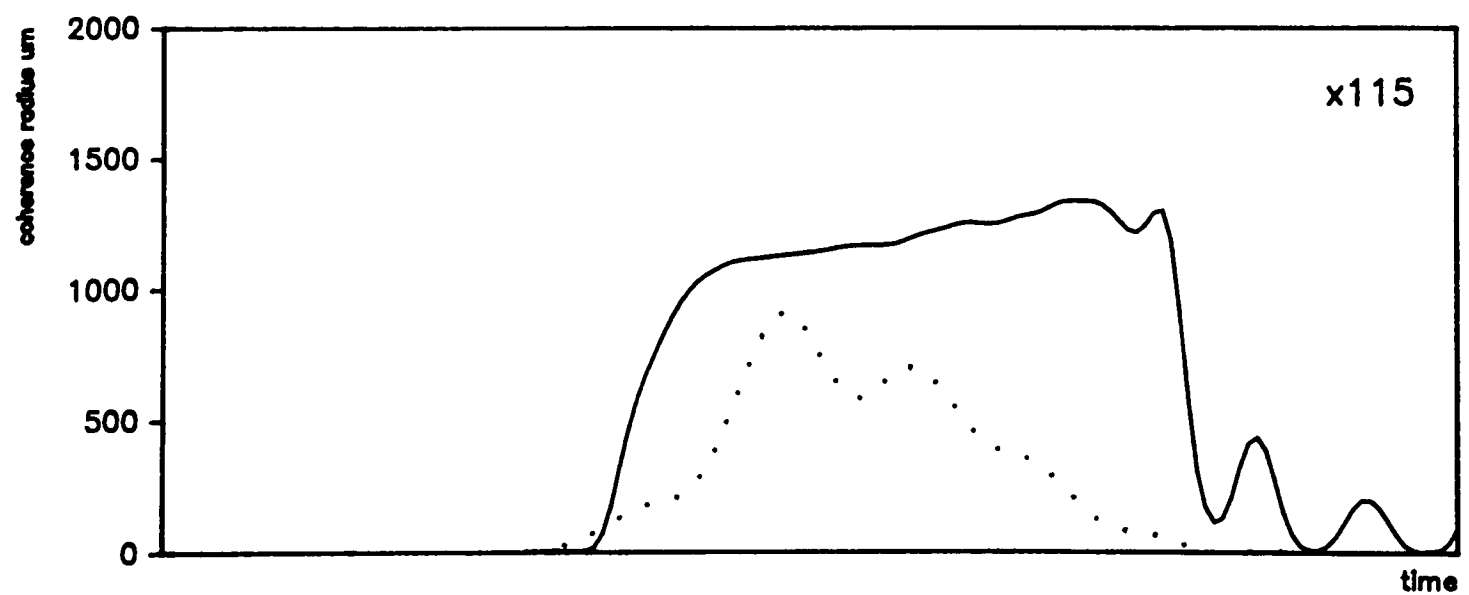
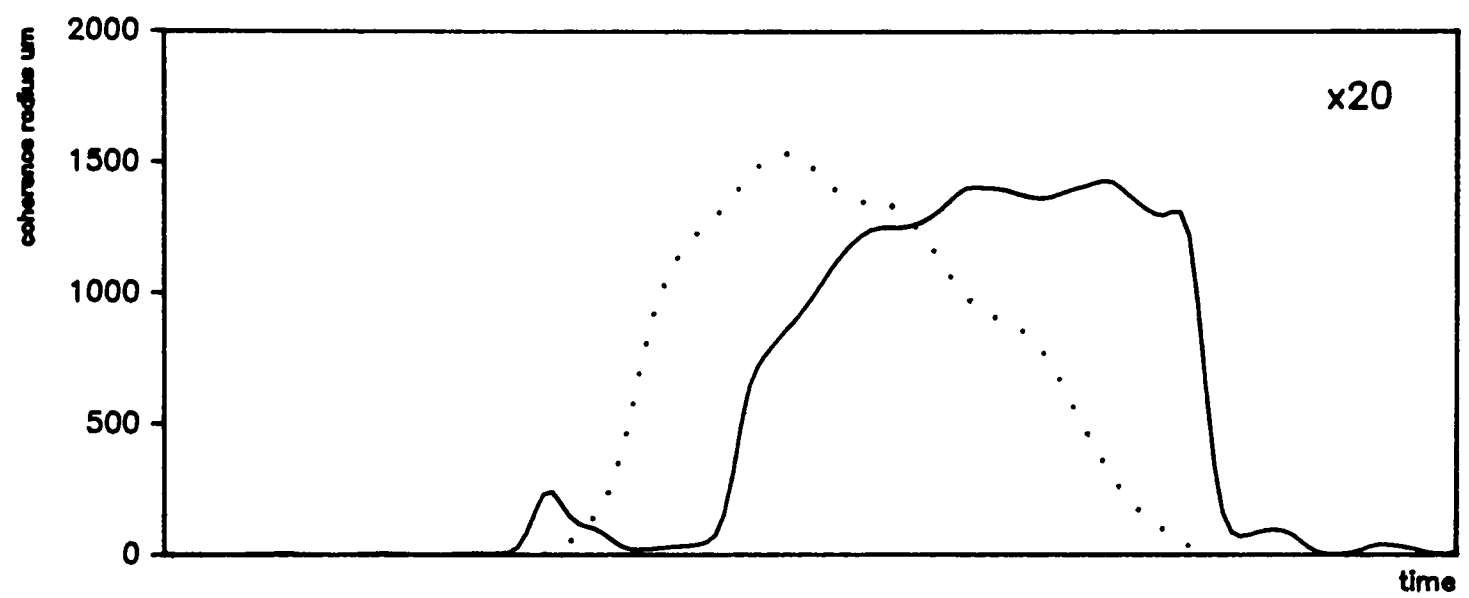
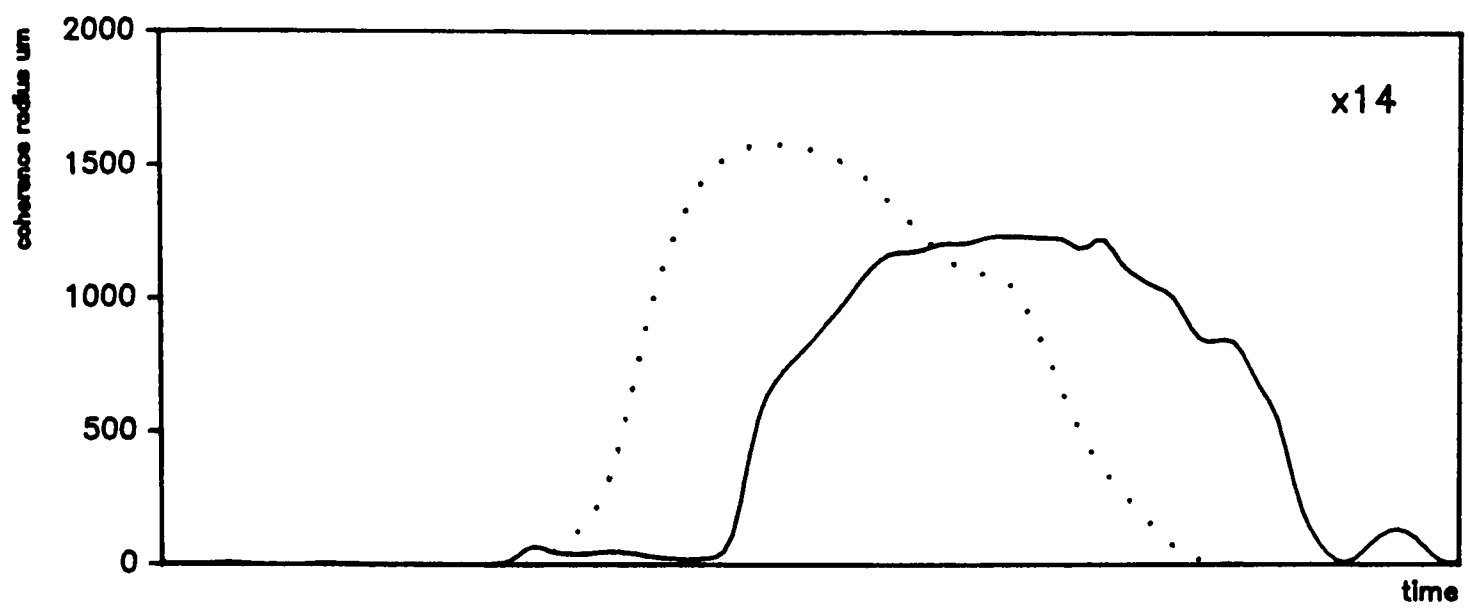


Figure IV-7
 Evolution of coherence radii without beam path covers. The coherence radius, defined by the pinhole separation for $1/e$ visibility, is shown here for three magnifications of unstable cavity. The coherence radius in each case is the full line, and the dotted curve shows a typical laser pulse for comparison. The horizontal axis spans about 100ns.

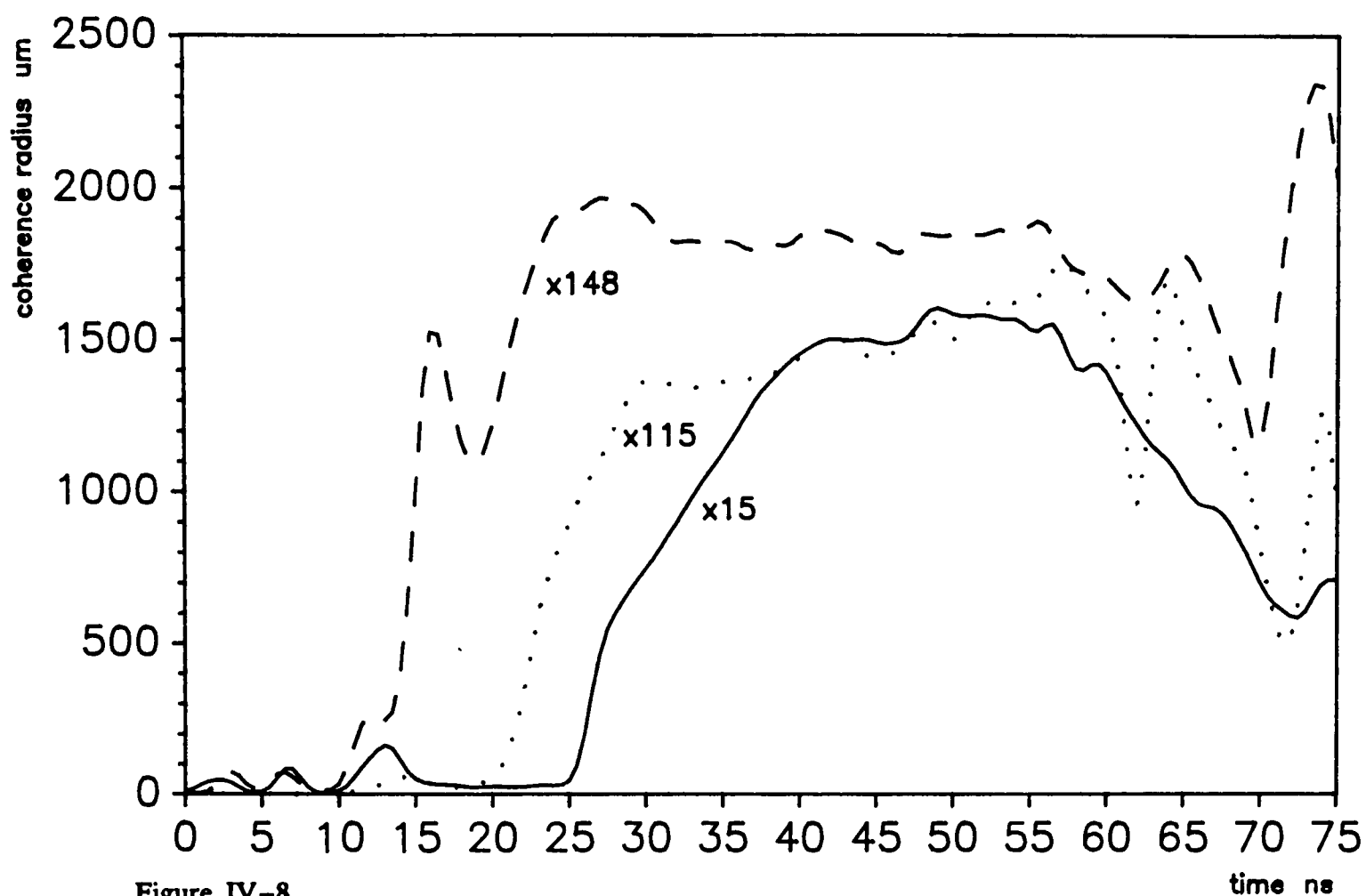


Figure IV-8
Evolution of coherence radii with beam path covers. The coherence radius is clearly seen to develop earlier when higher magnification cavities are used, but reaches a limit which is believed due to beam wander from shot to shot. The higher limiting value with the $\times 148$ cavity is not believed to be cavity related.

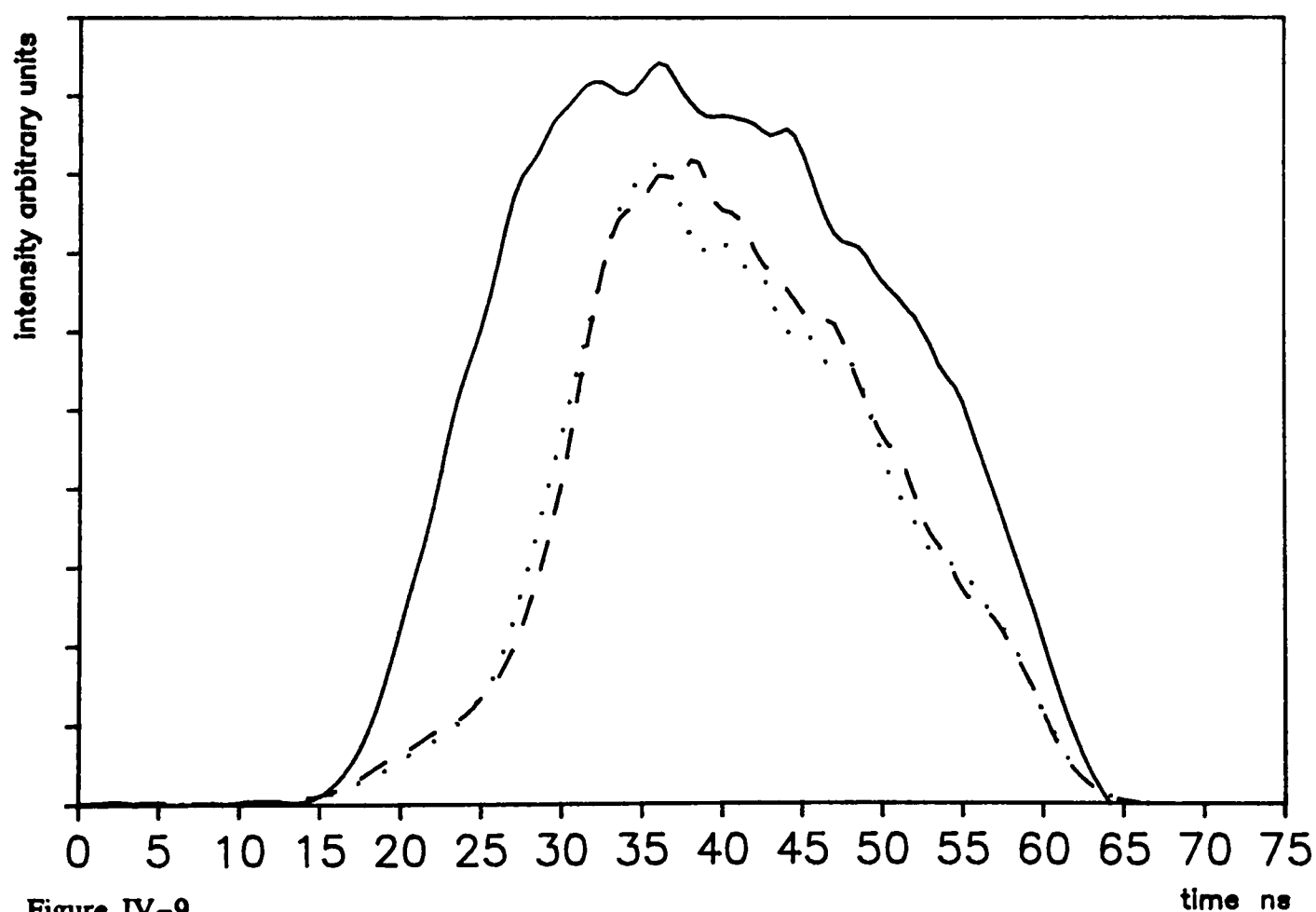


Figure IV-9
Example pulses overlaid showing the relative timing of the output from the three unstable cavity magnifications, the linetype matching those of figure IV-8 above. The higher losses of the $\times 115$ and $\times 148$ cavities result in a delayed start of the pulse, although the pulses finish together. There is no appreciable difference between the traces obtained from a $\times 15$ cavity and a plane-plane stable resonator, which has the same loss as would be incurred from an unstable cavity with a magnification of 5. Vertical scales are different for the three cavities: the powers from the $\times 115$ and $\times 148$ cavities were 0.78 and 0.71 respectively of that from the $\times 15$ cavity.

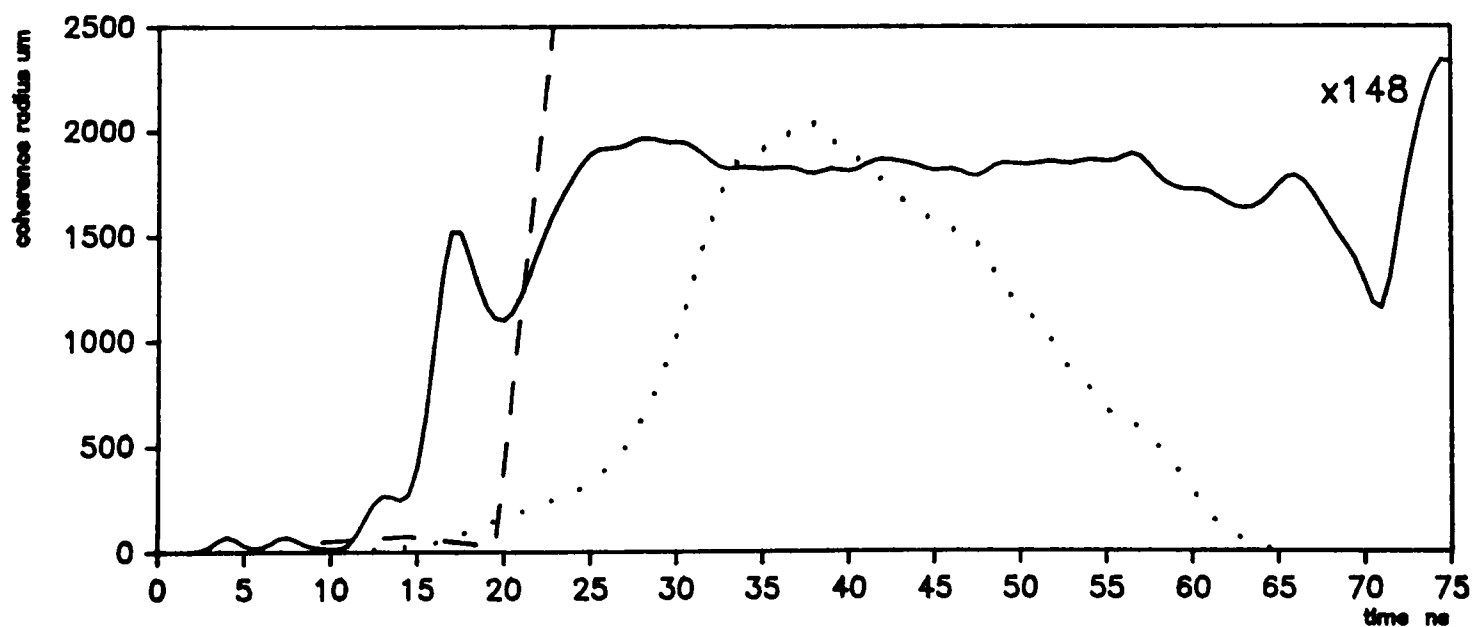
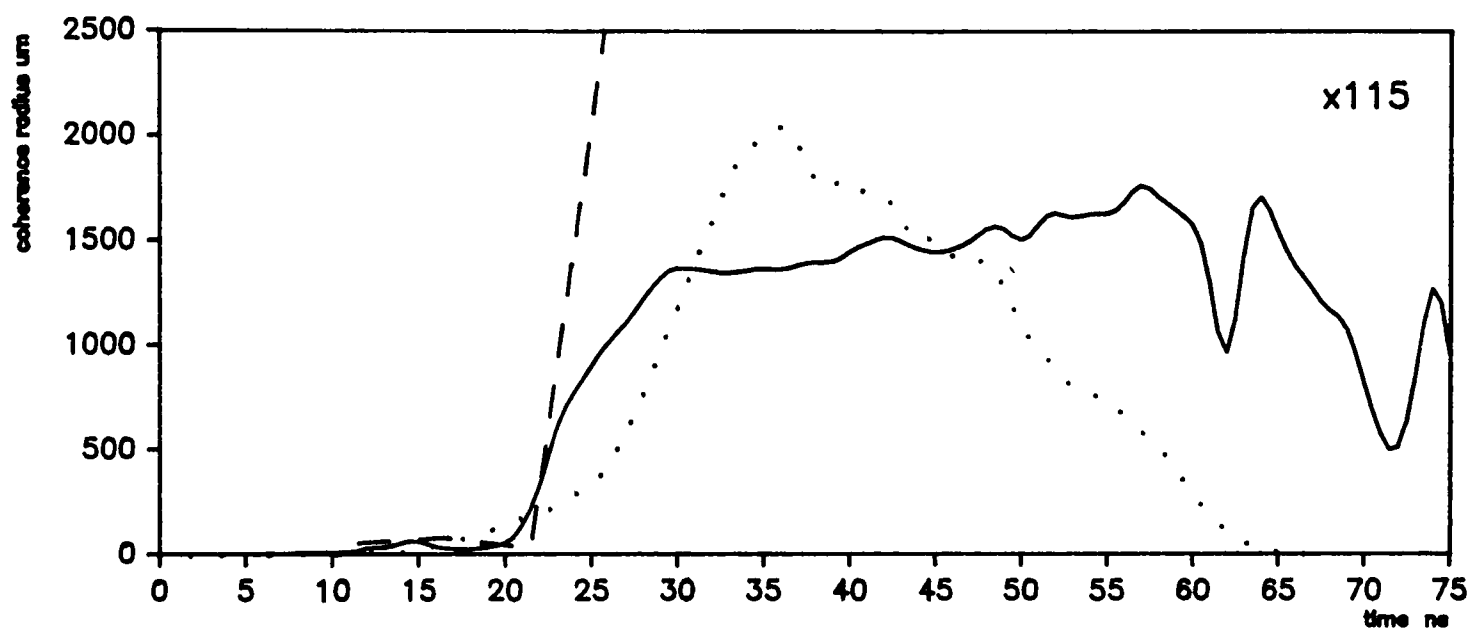
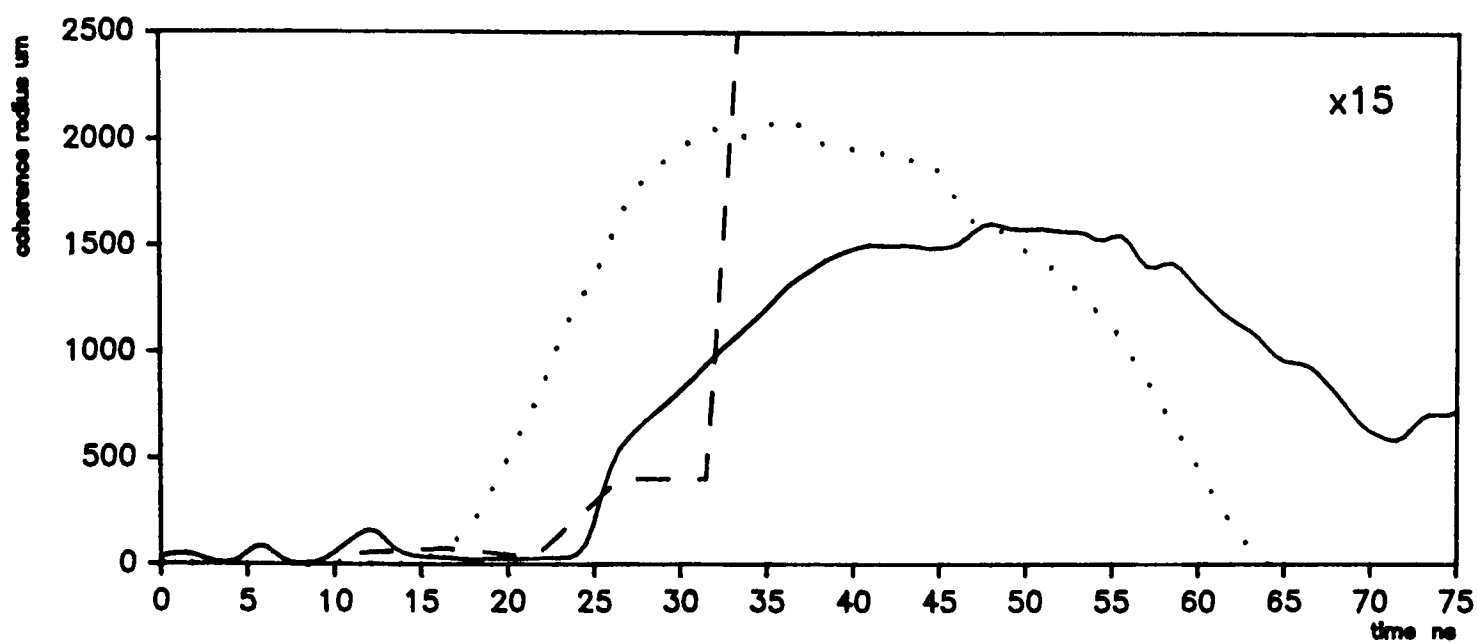


Figure IV-10
Evolution of coherence radius with different cavity magnifications. Each graph shows the measured coherence radius by a full curve and the computed development is drawn dashed after fitting by eye to a suitable timescale. A typical laser pulse is in each case indicated by a dotted line. The initial peak in coherence radius with the $\times 148$ cavity corresponds to laser intensities comparable with noise in the measurements and has therefore been ignored in the fitting process.

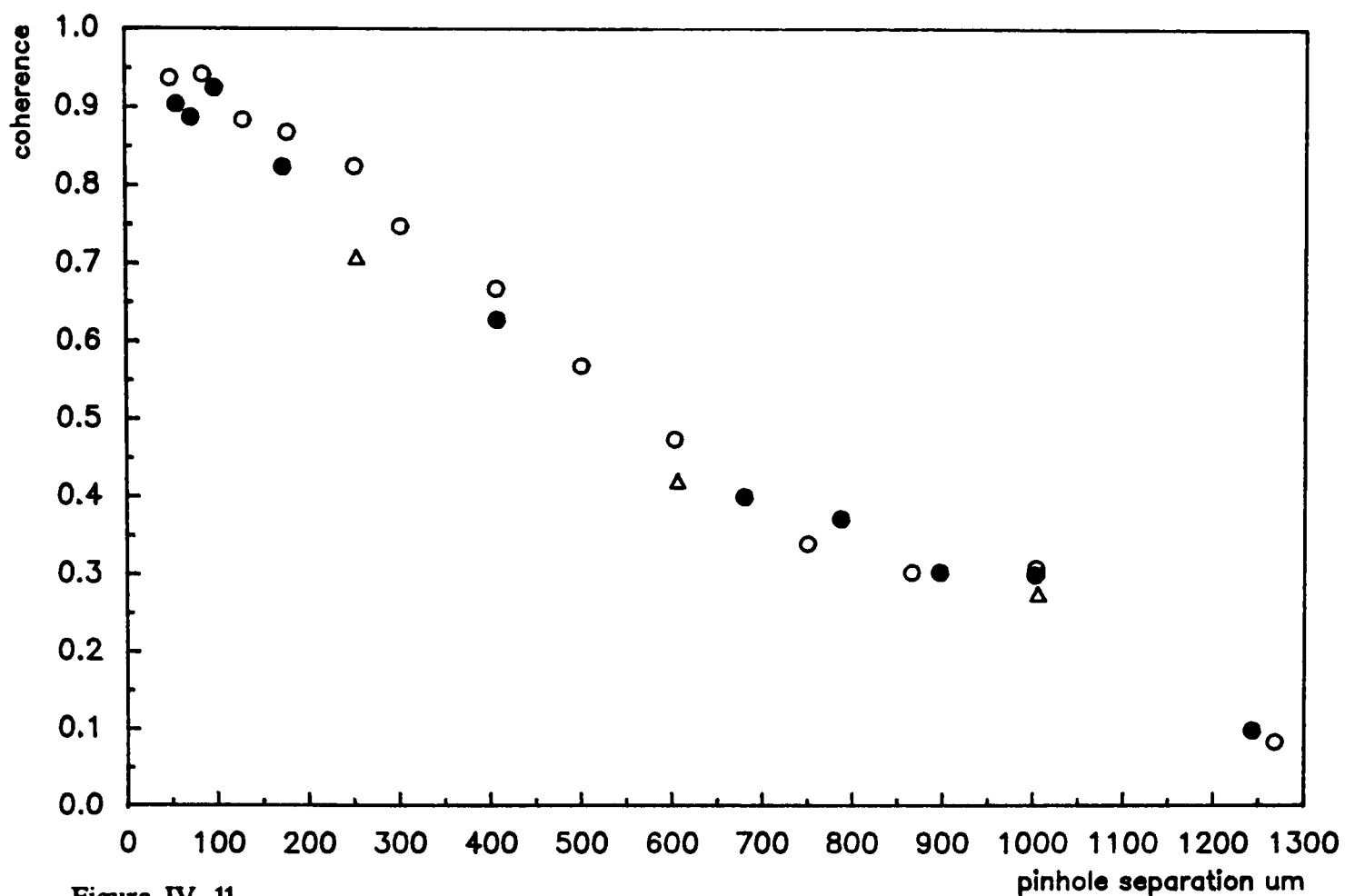


Figure IV-11

Time averaged transverse coherence of the green line with a central spot unstable cavity of magnification 10 without draughtproofing. Coherence radius = 670um. The symbols indicate different sets of data, the circles being from a pair of experimental runs with the pinholes separated horizontally and the triangles corresponding to vertical separation. The coincidence of the sets of data suggests that around the beam centre, at least, the transverse coherence is independent of orientation and position of the pinhole pair.

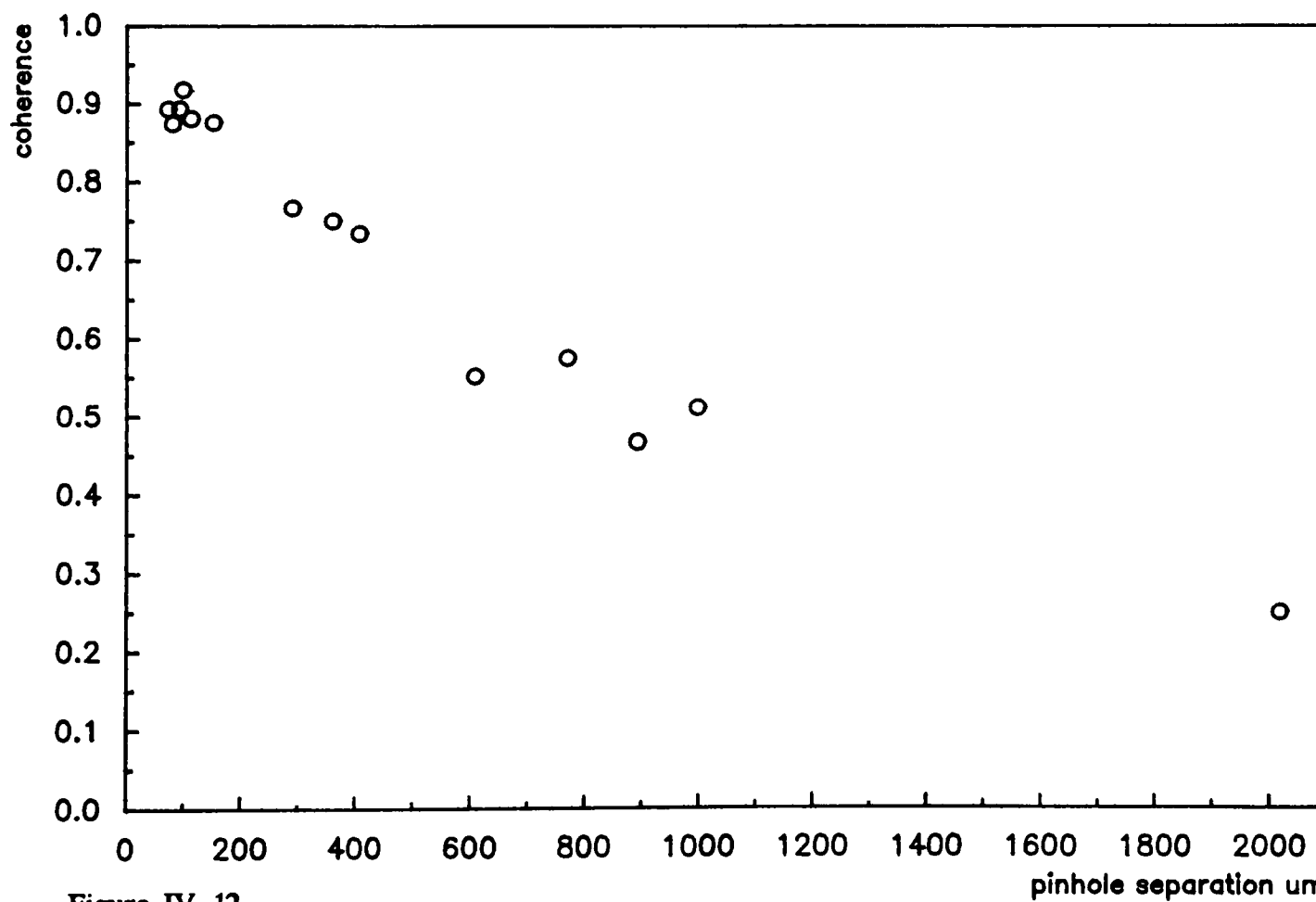


Figure IV-12

Time averaged transverse coherence of the yellow line with a central spot unstable cavity of magnification 10 without draughtproofing. Coherence radius = 1100um.

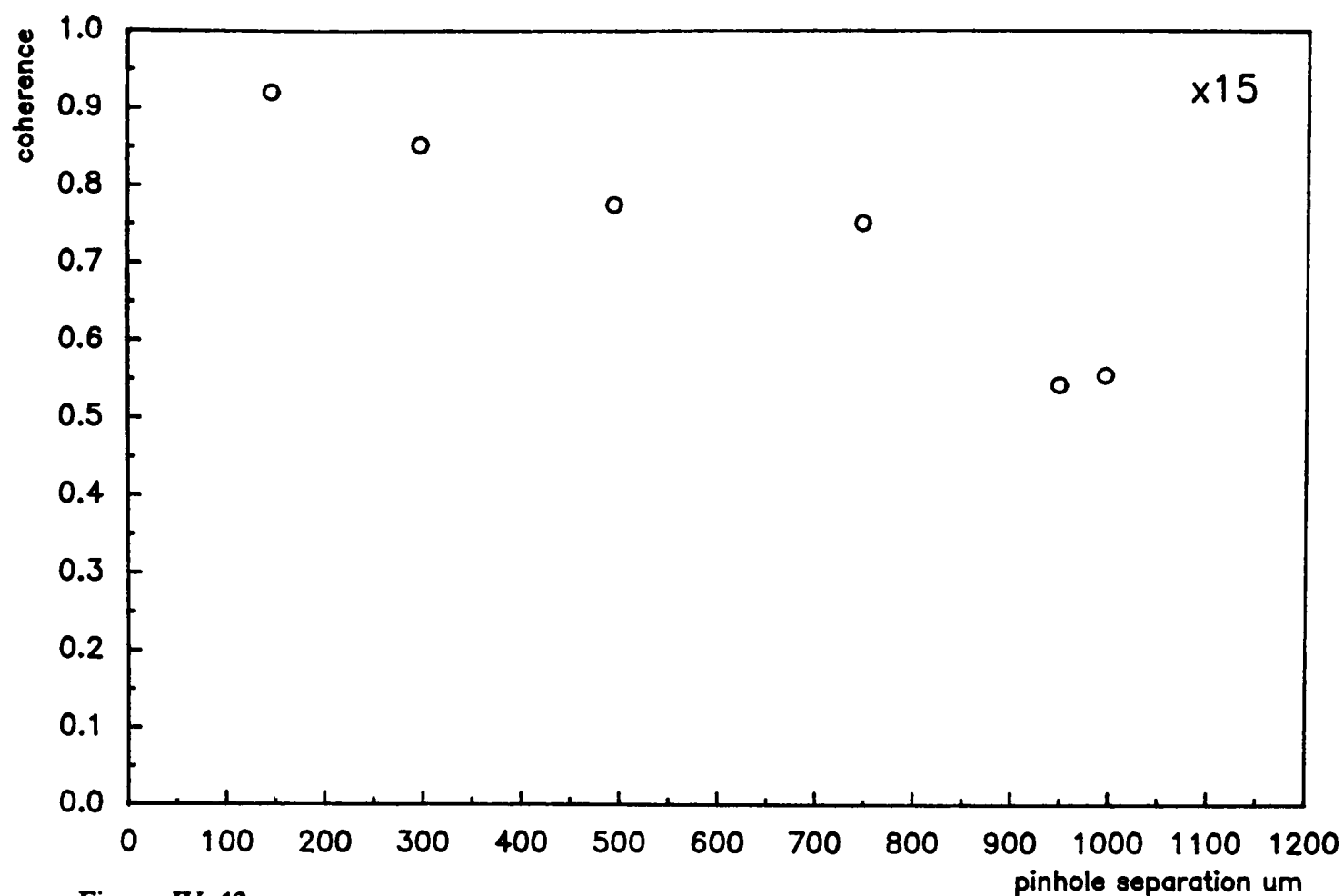


Figure IV-13
Time averaged transverse coherence of the green line with a cavity magnification 15 and with draughtproofing.

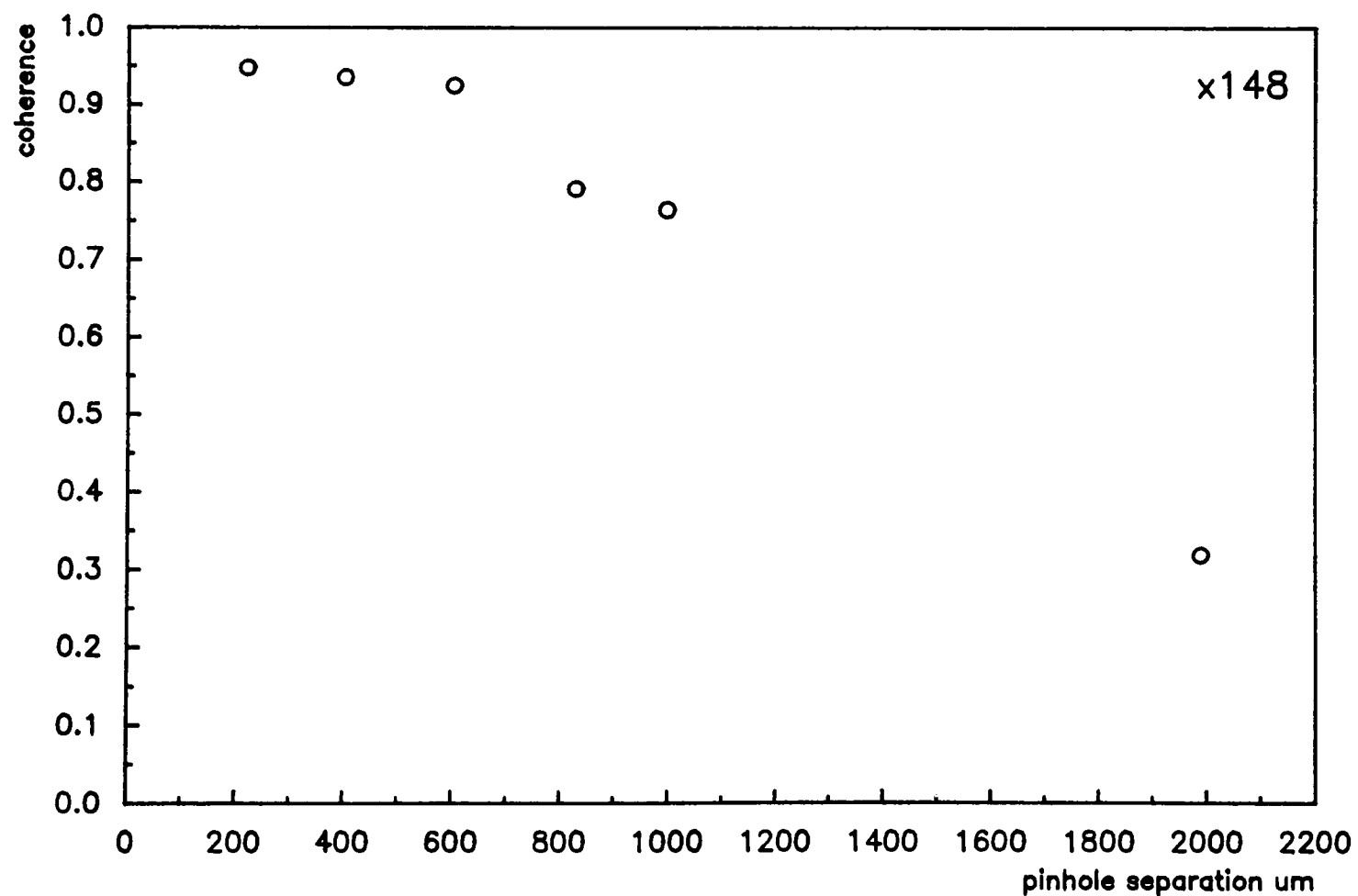


Figure IV-14
Time averaged transverse coherence of the green line with a cavity magnification 148 and with draughtproofing.

respectively. These experiments were performed using the SRS boxcar, which is a superior instrument all round and especially is considerably more stable than the EG&G system. Once again, the development of the single shot coherence dominates the early part of each trace, and accounts for the initial rising edge whose slope and position are dependent upon the cavity magnification. After a certain point, however, the improvement in coherence comes to a sudden halt and the coherence radius remains essentially constant for the remainder of the pulse. This is the effect of the beam wander or jitter. The higher value for this time-averaged coherence with the $\times 150$ cavity is not related to cavity magnification, but rather corresponds to a noted steadying of the projected fringes during the course of the experiment.

IV-5.2 Dependence of laser power upon cavity magnification

During the course of these experiments an investigation was made of the dependence of laser power upon cavity magnification. There is little effect upon the peak laser power achieved during the pulse, and the cavity dependence is apparent as a change in the pulse length. The green power available from the $\times 115$ magnification cavity is thus only 70-80% of that emerging from the lower magnification $\times 15$ resonator, whilst the $\times 150$ magnification arrangement yielded only 50-70% of the $\times 15$ configuration. There is, however, no noticable fall in the power of the yellow line over this range of cavity magnification. It is worth noting that the usual plane-plane resonator has a reflectivity of only 4% at the output coupler, and therefore the loss is the same as from an unstable cavity with a magnification of 5. There is no difference in pulse shape between the plane-plane arrangement and the $\times 15$ magnification unstable cavity.

Similar results have been reported by Zemskov et al [IV-3], who compared cavities with magnifications of 1 (a plane-plane cavity), 6, and 250. The generation of diffraction limited beams was convincingly demonstrated, and the drop in output power with a high magnification cavity was indeed found to be small, the $\times 250$ cavity giving around half the power available from the plane-plane configuration. A rather larger dependence has been recorded by Amit et al [IV-9], who found that at a magnification of 50 the power was reduced to 40% of its maximum. The much weaker dependence of the power in the yellow beam, however, was confirmed.

IV-6. Interpretation of coherence measurements

Figure IV-8 shows the measured development of the coherence radii for the three cavity magnifications after correction to a common timebase (with a correction

precision of 2ns). The rising edge of the coherence radius clearly advances with increasing cavity magnification, so that the leading edge of the $\times 15$ cavity is delayed by nearly 10ns relative to that of the $\times 150$ resonator. Typical laser pulses for each cavity magnification are also shown, in figure IV-9.

The curves of figure IV-8 are reproduced, together with the appropriate calculated development of the coherence radius, in figure IV-10. Since the time of seeding of the laser pulses has not been measured directly, each computed coherence trace has been fitted by eye to the timing of the measured curves with an overall precision of some 2ns. Within this tolerance the three computed traces thus defined all start at the same time. The timing of the rise of transverse coherence of the copper vapour laser is thus well predicted by a simple approach using geometrical optics.

It is clear that associated with an increase in the cavity magnification is a sharpening of the rate of rise of coherence radius. Further, higher magnification resonators have higher cavity losses, and thus a slower rise in laser power at the beginning of the pulse, so that the start of the pulse appears to be delayed. These effects combine when the cavity magnification is high to give the rise in coherence a large advance relative to the start of the laser pulse. The rising edge of the pulse from a low magnification cavity occurs around one round trip after the apparent start of the pulse, and extrapolation of the leading edges of the curves of figure IV-10 therefore suggests that diffraction limited performance is not approached until half way through the laser pulse. At the higher magnifications, however, this delay has increased to 1.5-2 round trips, and the coherence radius has reached its limit well before the pulse power has become appreciable.

There is no reason why, within a single pulse, the divergence of the beam from a copper laser with a high magnification unstable cavity should not become diffraction limited. The rate of rise of coherence radius should increase as the pulse develops, and the beam from the copper vapour laser is thus believed to assume diffraction limited coherence shortly after the multiple shot averaged coherence has reached its maximum. This hypothesis is supported by the direct observation of clear but moving fringes at large pinhole spacings, and by the results of attempted second harmonic generation which are reported in chapter VI. A large coherence area does not necessarily mean, however, that the beam is of good quality, as a plane wave passed through any optical component will maintain its coherence properties irrespective of the distortions introduced to the wavefront. Nonetheless, experiments performed by Graham Naylor of Oxford Lasers Limited using long focal length lenses have shown that the majority of the power from a copper vapour laser may be focussed into a spot of twice the diffraction limited radius, which moves within a circle of perhaps three times this radius. This not only suggests that the copper laser has good beam quality over its coherence area, but also reinforces the belief that the beam

approaches diffraction limited coherence on a single shot basis.

From the curves of time-averaged transverse coherence presented as figures IV-13 to IV-16, the beam divergence may be estimated using equation (IV-23). Table IV-1, below, summarizes these results.

Table IV-1.

figure	r_{coh} (mm ⁻¹)	divergence (fwm/e μrad ⁻¹)	
IV-13	0.6	500	510nm, without draughtproofing
IV-14	1.2	250	578nm, without draughtproofing
IV-15	1.5	200	510nm, with draughtproofing, ×15
IV-16	2.0	160	510nm, with draughtproofing, ×148
	5.0	60	diffraction limit

The divergences given above may be considered to be the angular ranges of the beam wander in each case. It will be shown that this is a useful interpretation when the implications of these results upon second harmonic generation are considered, in section IV-8 and in chapter VI.

Throughout, arbitrary curves have been fitted to Gaussian profiles in order that they be reduced to a single parameter, the coherence radius. There is no good reason why these distributions should be Gaussian in shape: the coherence function itself, for example, is the Fourier transform of the imaged spontaneous emission, subject to the accrued non-uniform gain of the laser. Similarly, the pointing angle distribution might well be dominated by a handful of preferred discharge anchoring points. Nonetheless, the Gaussian is a simple form which is easy to manipulate, and which is likely to be as good an approximation as any other arbitrary shape. The widths derived from it - the coherence radius and divergence - have far more physical significance than the full experimental curves from which they are derived, and in practice lose little in precision.

IV-7. Conclusion

The transverse coherence of the radiation from a copper vapour laser has been measured and is in accordance with the predictions of simple geometrical optics. The single shot coherence appears to approach the diffraction limit when a reasonably high cavity magnification is used, so that with magnifications of around 100 the majority of the pulse energy occurs in a diffraction limited beam. When averaged over periods of the order of seconds, however, the time-averaged coherence may thus be an order of magnitude worse than the diffraction limited case, and no averaged coherence radii above 2mm have been measured. The overall pulse energy is not drastically reduced even with cavity magnifications as high as 150.

These experiments have shown that the transverse coherence of the copper vapour laser is the product of two effects. Firstly, the development of the transverse coherence at the beginning of the pulse is determined by the unstable cavity which is being used. Spontaneous emission seeds the resonator some 10ns before powerful laser emission can occur, and the high cavity losses associated with high magnification cavities can delay the laser pulse further. A high magnification cavity thus allows the coherence to have developed completely to an essentially diffraction limited level by the time the pulse has properly begun. However, there is some residual beam wander, due to thermal and discharge-induced fluctuations in the optical beam path. Assuming gentle disturbances, this may be approximated to a linear variation in phase shift across the beam. It is thus possible to produce highly coherent laser pulses, but the direction in which they propagate will wander with a timescale around a second.

Significant variations in the beam quality are going to result from changes in the pulse length and timing. A delay in the pulse relative to the initial seeding gives the coherence longer to develop, and a longer pulse will similarly take advantage of the improving beam quality with time. It may therefore be advantageous, say, to run at higher repetition rates which might correspond to a delayed pulse start. It is likely to be the difference in timing which gives the yellow line the better transverse coherence at low magnifications.

IV-8. Implications for second harmonic generation

Second harmonic generation is a nonlinear process. The generated harmonic power is therefore dependent upon the area over which the beam maintains plane, or at least well defined and smooth, wavefronts. A high conversion efficiency approaching that appropriate to a single transverse mode laser will therefore indicate a large single shot coherence area and good beam quality.

A varying direction of propagation of the laser beam, however, means that the angle of incidence upon the crystal will vary, and thus the extent to which phase matching occurs will change. In comparison with the beam divergences of section IV-6, which vary from $500\mu\text{rad}$ down to $160\mu\text{rad}$, the acceptance angle for beta-barium borate is around 0.03° (full width), or $500\mu\text{rad}$; it therefore appears that for a significant fraction of the time phase matching will be missed. If the overlap of the phase matching sinc^2 function with a Gaussian representing the beam wander is calculated, it is found that the harmonic power may lie between 20% (without beam path covers) and 50% (with covers) of that theoretically available from a stable laser beam.

We note in passing that if the lens system used to focus the copper laser beam for second harmonic generation is arranged so as to produce at infinity an image of

the perturbing area, then any beam wander due to a linear spatial variation in refractive index will only move the focal spot around the crystal face and will not affect the angle of incidence itself. This arrangement has not, however, been tested experimentally; nor has the nature of the spatial variation in optical path been further investigated.

IV-References

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IV-Appendix 1 Correction factor for non-equal pinhole areas

Consider two pinholes, whose dimensions are much less than the pinhole separation, of areas 1 and $A=a^2$. Let the pinhole of unit area be the radiation field against which the coherence of light from the second pinhole is measured, and let the proportion of the amplitude at this second pinhole which is coherent be c . The total coherent amplitude from the second pinhole is thus ac , giving an intensity $(ac)^2$, whilst the remaining intensity $a^2(1-c^2)$ corresponds to an amplitude $a(1-c^2)^{\frac{1}{2}}$ which is not coherent with the first pinhole.

The minimum fringe intensity will be that when the coherent portions oppose each other, and will thus be

$$I_{\min} = a^2(1-c^2) + (1-ac)^2$$

the sum of the incoherent intensity and the intensity of the net coherent portion, and the maximum fringe intensity will similarly be

$$I_{\max} = a^2(1-c^2) + (1+ac)^2$$

This correctly gives a mean fringe intensity of $1+a^2$.

The fringe visibility may now be calculated to be

$$\frac{I_{\max}-I_{\min}}{I_{\max}+I_{\min}} = \frac{4ac}{2(1+a^2)}$$

and the coherence c is thus

$$c = \frac{1+a^2}{2a} \frac{I_{\max}-I_{\min}}{I_{\max}+I_{\min}}$$

Thus the coherence is related to the measured visibility by

$$\frac{\text{coherence}}{\text{measured visibility}} = \frac{A^{\frac{1}{2}} + A^{-\frac{1}{2}}}{2}$$

IV-Appendix 2 Correction factor for finite measuring pinhole size

Consider a pinhole of radius r sampling a fringe pattern $a(1+\cos\theta)$, where θ is a linear function of position x . The light transmitted by the pinhole (which lies centred on the fringe maximum) will be given by the overlap integral

$$\int_{-r}^r 2y(x) a[1+\cos\theta(x)] dx$$

where $y(x)$ is the pinhole cross-section at x , given by $x^2+y^2=r^2$. Now define $\theta=\alpha x$, so that the fringe separation is $2\pi/\alpha$, and the transmitted light amplitude becomes

$$4a \int_0^r y(x) [1+\cos\alpha x] dx$$

which on substitution for $y(x)$ gives

$$4ar^2 \int_0^1 [1-\beta^2]^{\frac{1}{2}} [1+\cos\alpha r\beta] d\beta$$

where $\beta=x/r$. This integral may be rewritten as

$$4ar^2 \left[\frac{\pi}{2} + \int_0^1 [1-\beta^2]^{\frac{1}{2}} [\cos\alpha r\beta - 1] d\beta \right]$$

or finally as

$$2\pi r^2 a \left[1 - \frac{4}{\pi} \int_0^1 [1-\beta^2]^{\frac{1}{2}} \sin^2 \gamma \beta d\beta \right]$$

$\gamma = \alpha r/2 = 2\pi r/\text{fringe separation}$

The effect upon fringe measurements is to multiply the depth of modulation by the bracketted term above, so that

$$\frac{\text{measured visibility}}{\text{true visibility}} = 1 - \frac{4}{\pi} \int_0^1 [1-\beta^2]^{\frac{1}{2}} \sin^2 \gamma \beta d\beta$$

IV-Appendix 3 Computer program CALCOH.BAS

```

1000 REM *****
1010 REM *** Routine to calculate coherence radius as function ***
1020 REM *** of time for unstable cavity. ***
1030 REM *** Tim Freearde 9/11/88 ***
1040 REM *****
1050 REM
1100 PRINT "Enter magnification:";TAB(45);
1105 INPUT M
1110 LC = 1.5 :REM cavity length in metres
1120 F1 = LC/(1-1/M) :REM focal length in metres
1130 F2 = LC/(1-M) :REM focal length in metres
1140 R1 = 0.0125 :REM tube radius in metres
1150 C = 3.0E+08 :REM speed of light in metres/sec
1160 AL = 2.096E-07 :REM alpha.lambda
1170 DT = 0.5E-09 :REM step in seconds
1180 DX = C*DT :REM step in metres
1190 D = 3 :REM laser-pinhole distance in metres
2000 AD = 1
2010 BD = D
2020 CD = 0
2030 DD = 1
5000 E$ = "D:FAULT.DAT"
5010 PRINT "Enter output filename [default ";E$;"]:";TAB(45);
5020 INPUT D$
5030 IF D$="" THEN D$=E$
5040 PRINT "Saving data in";TAB(47);D$
5050 CREATE #10,D$
5060 QUOTE #10
5070 OPEN #10,D$
5080 PRINT #10,D$
5090 C$="Calculated coherence: M =" +STR$(M)+" R1 =" +STR$(R1)+" D
    =" +STR$(D)
5100 PRINT "Comment will be";TAB(45);C$
5110 PRINT #10,C$
10000 REM
10010 NM = 5 :REM number of round trips to
    calculate
10100 N = 0: X = 0
10110 AM= M↑N
10120 BM= M*LC*(1+1/M)/M↑N*(M↑(2*N)-1)/(M↑2-1)
10130 CM= 0
10140 DM= 1/M↑N
10200 Y = X - (2*N*LC) :REM START OF X LOOP
10210 A1 = 1
10220 B1 = Y
10230 C1 = 0
10240 D1 = 1
10250 A2 = AM*A1 + BM*C1
10260 B2 = AM*B1 + BM*D1
10270 C2 = CM*A1 + DM*C1
10280 D2 = CM*B1 + DM*D1
10290 A1 = AD*A2 + BD*C2
10300 B1 = AD*B2 + BD*D2
10310 C1 = CD*A2 + DD*C2
10320 D1 = CD*B2 + DD*D2

```

```

10400      Z = B1/((A1*D1-C1*B1)*R1)
10410      RC = Z*AL*1.0E6           :REM Coherence radius in microns
10420      PRINT #10,RC,X*1.0E9/C    :REM RC, time in nanoseconds
10500      X = X+DX
10510      IF (X-2*N*LC)<LC THEN 10200
11100          Y = X-(2*N+1)*LC      :REM START OF Y LOOP
11110          A1 = 1/M
11120          B1 = LC + Y*(1-LC/F1)
11130          C1 = (-1)/F1
11140          D1 = 1 - Y/F1
11150          A2 = AM*A1 + BM*C1
11160          B2 = AM*B1 + BM*D1
11170          C2 = CM*A1 + DM*C1
11180          D2 = CM*B1 + DM*D1
11190          A1 = AD*A2 + BD*C2
11200          B1 = AD*B2 + BD*D2
11210          C1 = CD*A2 + DD*C2
11220          D1 = CD*B2 + DD*D2
11300          Z = B1/((A1*D1-C1*B1)*R1)
11310          RC = Z*AL*1.0E6       :REM Coherence radius in microns
11320          PRINT #10,RC,X*1.0E9/C :REM RC, time in nanoseconds
11400          X = X+DX
11410          IF (X-(2*N+1)*LC)<LC THEN 11100
11500          N = N+1
11510          IF N<(NM+1) THEN 10110
12000      CLOSE #10
19000      END

```

Theory of second harmonic generation.

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Chapter V

Theory of second harmonic generation.

V-1. Introduction

It is possible, in a number of crystals and other media, to induce an electric polarization whose strength is not directly proportional to that of the inducing field, but rather is correctly expressed as an expansion in ascending powers of this field. The first nonlinear term, the quadratic, results in the generation from a fundamental oscillating field of a second harmonic with twice its frequency. With the high field strengths available from laser sources, this can be an efficient method of converting visible light to the ultraviolet.

This chapter describes the theory of second harmonic generation. It accounts for the variation in conversion efficiency with crystal parameters and orientation, and therefore explains how to maximize the second harmonic power under practical constraints. It also permits a comparison of the predicted performances of various crystal types for a given laser system. The treatment is written in terms of ideal, Gaussian laser beams; that the beam from a copper vapour laser is less ideal will be considered in later chapters.

Reviews of the basic theory of second harmonic generation have been made by Yariv [V-1], Franken and Ward [V-2], Byer [V-3], and Yariv and Pearson [V-4], and the treatment of the groundwork for the theory presented here follows these references. Similarly, the nonlinear properties of crystals are covered more than adequately by Nye [V-5]. The full theory of second harmonic generation with elliptical Gaussian beams, however, is a development of the classic treatment by Boyd and Kleinman [V-6] and is presented here in full for the first time: a summary was given at the Optical Society of America's *Topical Meeting on Nonlinear Optical Properties of Materials* in Troy, N.Y., in August 1988 [V-7].

V-2. Crystal susceptibility and nonlinearity.

The electric polarization of a crystal in response to an electric field may be expressed as a tensor expansion in powers of the applied field:

$$\underline{P} = \epsilon_0 \left[\chi^{(1)} \underline{E} + \chi^{(2)} \underline{E}^2 + \chi^{(3)} \underline{E}^3 + \dots \right] \quad (V-1)$$

where $\chi^{(1)}$ is the linear susceptibility responsible for the refractive index of the crystal and dominant for low field strengths. For work at optical frequencies, it is customary to define the susceptibility tensors in terms of Fourier components of the fields, so that in the relation above, $\underline{P}$ and $\underline{E}$ are replaced by the amplitudes of the oscillating field and polarization. In the presence of several fields at different frequencies, the full expression looks like

$$\underline{P}(\omega_1) = \epsilon_0 \left[\sum_p \chi^{(1)}(\omega_1) \underline{E}_p(-\omega_1, \omega_1) + \frac{1}{2} \sum_{pq} \chi^{(2)}(-\omega_1, \omega_p, \omega_q) \underline{E}_p(\omega_p) \underline{E}_q(\omega_q - \omega_1 - \omega_p) + \dots \right] \quad (V-2)$$

where the factor $\frac{1}{2}$ appears in the second term to account for the conversion from field to amplitude. (Yariv [V-1] and Byer [V-3] show this by writing out $\underline{E}$ as the sum of a complex term and its conjugate; it's much more obvious to write the square of $\underline{E} = \underline{E}_0 \cos \omega t$ as $\underline{E}^2 = \frac{1}{2} \underline{E}_0^2 [\cos 2\omega t + 1]$.) $\chi^{(2)}$ accounts for second harmonic generation, frequency mixing and parametric oscillation, the Pockels (linear electro-optic) effect, and dc rectification. $\chi^{(3)}$ is responsible for two-photon absorption, third harmonic generation, the Kerr (quadratic electro-optic) effect, and Brillouin, Raman and Rayleigh scattering.

Second harmonic generation, then, involves the $\chi^{(2)}$ susceptibility tensor, and the polarization $\underline{P}(2\omega)$ at the second harmonic results from a fundamental electric field $\underline{E}(\omega)$ according to

$$\underline{P}(2\omega) = \frac{1}{2} \epsilon_0 \chi^{(2)}(-2\omega, \omega, \omega) \underline{E}(\omega) \cdot \underline{E}(\omega) \quad (V-3)$$

The second harmonic generation tensor $\underline{d}$ is then defined as $\frac{1}{2} \chi^{(2)}(-2\omega, \omega, \omega)$, so that

$$\underline{P}(2\omega) = \epsilon_0 \underline{d} \underline{E}(\omega) \cdot \underline{E}(\omega) \quad (V-4)$$

For fixed polarizations of $\underline{E}$ and $\underline{P}$, this expression may be written in a scalar form with effective scalar d_{eff} , so that

$$P(2\omega) = \epsilon_0 d_{\text{eff}} E(\omega)^2 \quad (V-4a)$$

V-3. A plane wave treatment of second harmonic generation.

We may derive from Maxwell's equations a set of equations for the electromagnetic fields generated by the induced nonlinear polarization (see for example Byer [V-3]). For steady-state plane-wave second harmonic generation, and making the approximation of slowly-varying amplitudes, these become

$$\frac{dE(\omega)}{dz} + \alpha_{\omega}E(\omega) = iKE(2\omega)E^*(\omega) \exp[-i\Delta kz] \quad (V-5)$$

and

$$\frac{dE(2\omega)}{dz} + \alpha_{2\omega}E(2\omega) = iKE(\omega)E(\omega) \exp[i\Delta kz] \quad (V-6)$$

with $\Delta k = 2k_{\omega} - k_{2\omega}$ and $K = \omega d_{\text{eff}}/\eta c$, η being the refractive index of the medium. α is the electric field loss coefficient, $\mu_0 \sigma c/2$. In the case of low conversion efficiency, then there is no significant depletion of the fundamental, and when $\alpha_{\omega} = 0$, the total second harmonic field strength from a given length of crystal l becomes

$$E(2\omega) = KE^2(\omega) l \frac{\sin(\Delta k l/2)}{\Delta k l/2}. \quad (V-7)$$

The oscillating modulation when $\Delta k \neq 0$ is the result of coherent addition from a line source along the line of propagation. If $\Delta k = 0$, so that all the fields from the line source add with the same phase, then *phase matching* is said to occur. This corresponds to the condition that the fundamental and second harmonic should propagate at the same speed within the crystal. They should thus share the same refractive index; dispersion usually decrees that they do not.

Phase matching is therefore achieved by modifying the refractive indices in some way, and for this the intrinsic birefringence of the crystal is used. By appropriate polarization of the fundamental and harmonic, their refractive indices may be made equal either by rotating the crystal to a special orientation or by exploiting the temperature dependence of the refractive indices.

V-4. Linear susceptibility and phase matching.

The propagation of the fundamental through the crystal is determined by the linear component of the susceptibility, $\chi^{(1)}(\omega)$, defined below:

$$[P_x, P_y, P_z] = \begin{bmatrix} \chi_{11} & \chi_{12} & \chi_{13} \\ \chi_{21} & \chi_{22} & \chi_{23} \\ \chi_{31} & \chi_{32} & \chi_{33} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix} \quad (V-8)$$

This may be compared with the relation $P = \epsilon_0(\eta^2 - 1)E$ for isotropic media. $\chi^{(1)}$ can always be diagonalized by judicious choice of the crystal axes x, y, z , and such are known as the *principal crystal axes*.

In an isotropic medium, the polarization will always be in constant proportion to the applied field, and thus the components of the diagonalized linear susceptibility will be given by $\chi_{11}=\chi_{22}=\chi_{33}$. To all intents and purposes $\chi^{(1)}$ will look like a scalar: the refractive index will be constant. More generally, however, the refractive index will be a function of the direction of polarization (eg. for propagation along the x axis, $\chi_{22}\neq\chi_{33}$). This phenomenon is known as birefringence, and it may be shown that for any given direction of propagation there will be two possible refractive indices, corresponding to the two eigenmodes of polarization which satisfy Maxwell's equations. Intermediate polarizations do not by themselves satisfy Maxwell's equations for propagation, but rather must be resolved into components of the only two polarization eigenstates which do and which propagate independently according to their different refractive indices. There is nonetheless always at least one orientation for which the refractive index does not vary with polarization, and this direction of propagation is known as the *optic axis*.

It is possible to represent the linear susceptibility tensor by a multitude of physically significant three-dimensional surfaces, each of which is known by a number of different names. It is possible to represent refractive index, susceptibility, phase velocity and "ray velocity", as functions of wavevector and Poynting vector. Only two of these surfaces are of interest here: the *ellipsoid of wave normals*, which is a plot of refractive index as a function of wavevector; and the *ray surface*, which describes the ray velocity as a function of Poynting vector and therefore illustrates the development of a wavefront. These should not be confused with the *ray* (or *Fresnel*) *ellipsoid* $\epsilon_x x^2 + \epsilon_y y^2 + \epsilon_z z^2 = 1$, or the (wave-) *normal surface* which describes the variation of phase velocity with wavevector.

The ellipsoid of wave normals – otherwise described as the *optical indicatrix*, *index ellipsoid*, or *reciprocal ellipsoid* – is given by the equation

$$\frac{x^2}{\epsilon_{11}/\epsilon_0} + \frac{y^2}{\epsilon_{22}/\epsilon_0} + \frac{z^2}{\epsilon_{33}/\epsilon_0} = 1 \quad (\text{V-9})$$

and is illustrated in figure V-1. This surface defines the components of the electric displacement $\underline{D}$ for a constant energy density $\underline{D} \cdot \underline{E}$. Its practical significance is a little complicated and is described in some length by Yariv [V-1], but it allows a procedure for finding the refractive index appropriate for propagation of light in any direction. The intersection of the surface with a central plane is an ellipse, shown in figure V-1 by the solid line, whose axes by their orientation and magnitude define the directions and refractive indices of two polarizations. These polarizations are those allowed by Maxwell's equations to propagate with wavevector $\underline{k}$ normal to the plane, and intermediate polarizations will propagate as if resolved into components along these axes. The polarization is defined by the electric displacement $\underline{D}$, which is in general different from the electric field $\underline{E}$ but which is necessarily perpendicular to $\underline{k}$.

Figure V-1
Ellipsoid of wave normals. Intersections with the planes of symmetry are shown dotted. The solid line is the intersection of the ellipsoid with a plane through the origin and normal to the wavevector $\underline{k}$. The axes of this ellipse define the polarizations which are allowed to propagate with this wavevector.

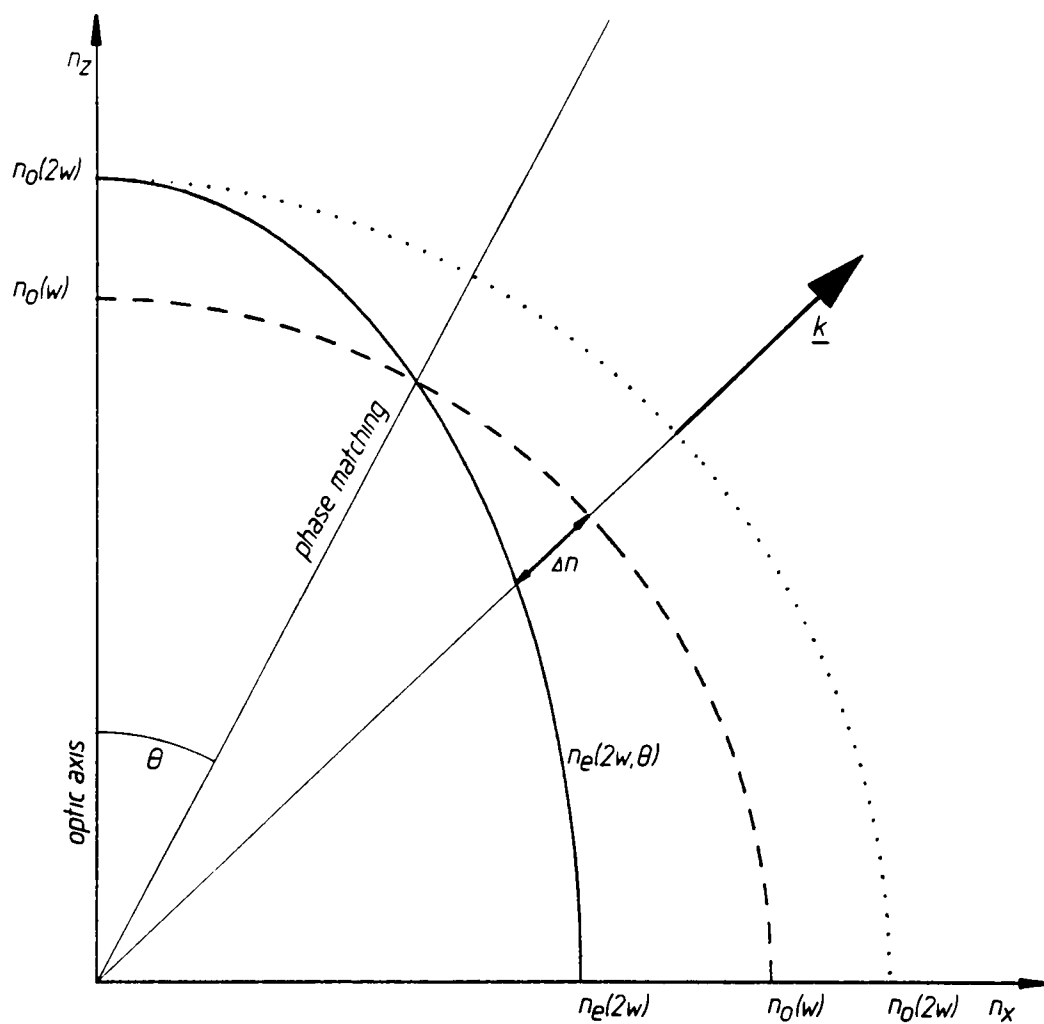
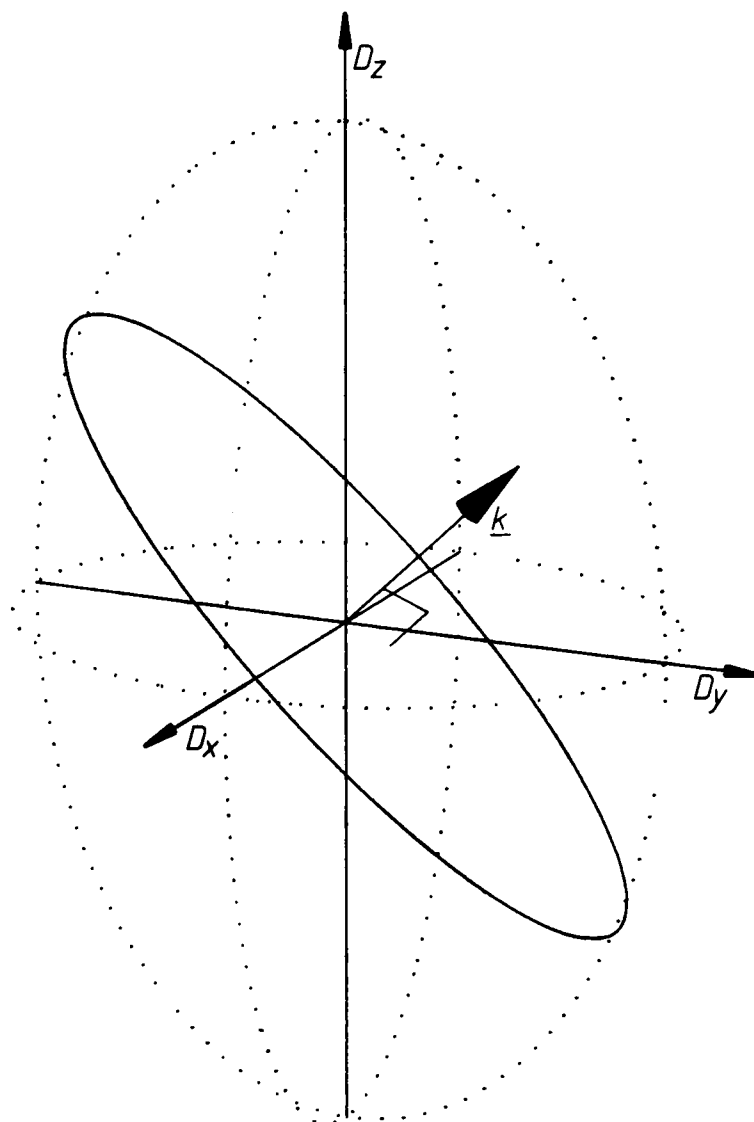


Figure V-2
Type I phase matching is shown here for a negatively birefringent crystal ($\eta_e < \eta_o$) with normal dispersion ($\eta^{2\omega} > \eta^\omega$). For a general direction of wavevector $\underline{k}$ there will be a difference $\Delta\eta$ between the refractive indices of the fundamental and harmonic beams. This results in a phase mismatch $\Delta k = 2k_1 - k_2 = 2\omega_1 \Delta\eta / c$. Only at the phase matching angle θ does $\Delta\eta = 0$.

The other surface of interest is the ray surface (also termed *ray velocity surface* or *wave surface*), and is the locus of the *ray velocity* – the speed of propagation of a wavefront along the Poynting vector. The phase velocity, the speed of propagation along wavevector, is the component of this ray velocity normal to the wavefront. The ray surface has the same shape as the wavefront emanating from a point source, and it is for this property that it is useful, for it highlights the distinction between wave-vector and Poynting vector. The electric displacement $\underline{D}$ is tangential to the ray surface (again since $\underline{D} \cdot \underline{k} = 0$; see Appendix V-2).

As already stated, the optic axis of a crystal is defined as being the direction of propagation of a beam whose refractive index is independent of polarization: the two polarizations are thus indistinct and indeterminate, and a section through the index ellipsoid perpendicular to the optic axis is therefore circular. Uniaxial crystals, not suprisingly, are those which have but one optic axis. This is a principal crystal axis, and about it the index ellipsoid has rotational symmetry. Biaxial crystals [V-8], in contrast, have two optic axes, neither of which coincides with a principal axis of the crystal. The rest of this chapter is concerned only with crystals which are uniaxial.

V-5. Primary effect of birefringence: phase matching in uniaxial crystals.

It is seen that in general the refractive index for propagation in a given direction can take two values according to the polarization of the beam. One polarization is inevitably perpendicular to the optic axis and is termed the *ordinary* polarization: the other will be orthogonal to both the ordinary polarization and the wavevector, and this is the *extraordinary* polarization. As the crystal orientation is changed, the ordinary refractive index remains constant, whilst the extraordinary refractive index varies. Only with the wavevector along the optic axis the two are equal. Confusingly, the extraordinary ray propagating perpendicular to the optic axis, and therefore polarized along a principal crystal axis, is said to have *the* extraordinary refractive index. This specific value is usually distinguishable from the general one only by the absence of an implied angular dependence (as in $\eta_e(\theta)$).

This is an appropriate point at which to define the terms *positive uniaxial* and *negative uniaxial*, which refer to the relative magnitudes of the ordinary and extraordinary refractive indices. For a positive uniaxial crystal, $\eta_e > \eta_o$.

Phase matching is therefore achieved when the index ellipsoids of the fundamental and harmonic intersect. Dispersion usually requires that the two have different polarizations, and indeed the orientation of the crystal can be used to achieve this – a process known as *angle tuning*. The most common arrangement is that known as Type I phasematching, in which the fundamental propagates as the ordinary ray and the second harmonic is extraordinary; it is this arrangement which is

appropriate for negative uniaxial crystals with normal dispersion ($d\eta/d\omega < 0$) and which will be considered in the following calculations. The alternative arrangement called type II phase matching, which allows the tuning range of crystals to be slightly extended, will not be further considered. In this method, the fundamental propagates as a mixture of ordinary and extraordinary waves whilst the harmonic may be either pure ordinary or pure extraordinary, and an equivalent set of equations may be derived. Indeed, many of the expressions given for type I phase matching are also appropriate for type II.

The geometry of type I phase matching is illustrated in figure V-2. The phase matching angle θ is given by

$$\sin^2\theta = \frac{[\eta_o(\omega)]^{-2} - [\eta_o(2\omega)]^{-2}}{[\eta_e(2\omega)]^{-2} - [\eta_o(2\omega)]^{-2}} \quad (V-10)$$

and is the angle between the optic axis and the direction of propagation. Figure V-3 shows the arrangement when phase matching occurs at $\theta=90^\circ$. In this configuration the phase match is least sensitive to angle, for the two ellipsoids share a common tangent, and this arrangement is therefore known as *noncritical phase-matching*. The angular variation due to tight focussing can often be sufficient to cause phase mismatching over large portions of the beam in angle tuned arrangements, and noncritical phasematching is therefore highly desirable. In practice some small adjustment of the wavelength for noncritical phasematching is possible by varying the temperature of the crystal. The temperature dependence of the refractive indices has to be fairly large, however, for any reasonable range to be achieved, and for most crystals in use the temperature tuning range is much less than that available through angle tuning.

Temperature tuning has its own drawback, for most crystals will absorb a proportion of the light passing through them, and this is dissipated as heat, leading to a thermal gradient across the beam. The high temperature coefficient now results in incomplete phase matching of the laser beam, and however weakly the beam is focussed the phase mismatch across the beam remains constant, for although the temperature gradient is reduced the beam width is proportionally greater. For high mean power lasers this can be a limiting mechanism, and it has been considered in some detail by Okada and Ieiri [V-9].

It is a general rule that the tunability of any method of phase matching is also its limitation: angle tuned systems are limited in angular acceptance, which is particularly bad if the source has poor beam quality so that the divergence is much greater than that of an equivalent Gaussian beam; and temperature tuned systems perform badly with high mean power lasers. Unfortunately, most high mean power lasers also have relatively poor beam quality, and few can be converted at the theoretical efficiency.

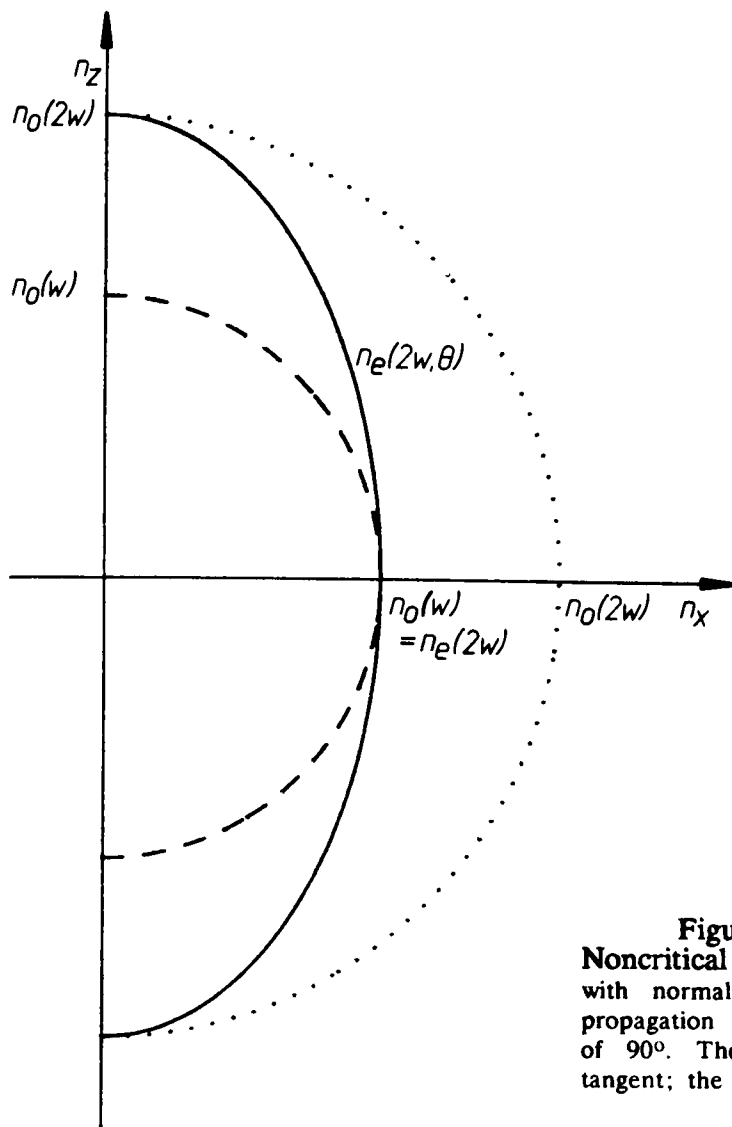


Figure V-3
Noncritical (90°) phasematching in a negatively birefringent crystal with normal dispersion. Phase matching occurs when the direction of propagation is along the x-axis, and therefore at a phase matching angle of 90°. The ellipse and circle no longer cross, but instead share a tangent; the phase matching is thus much less sensitive to θ .

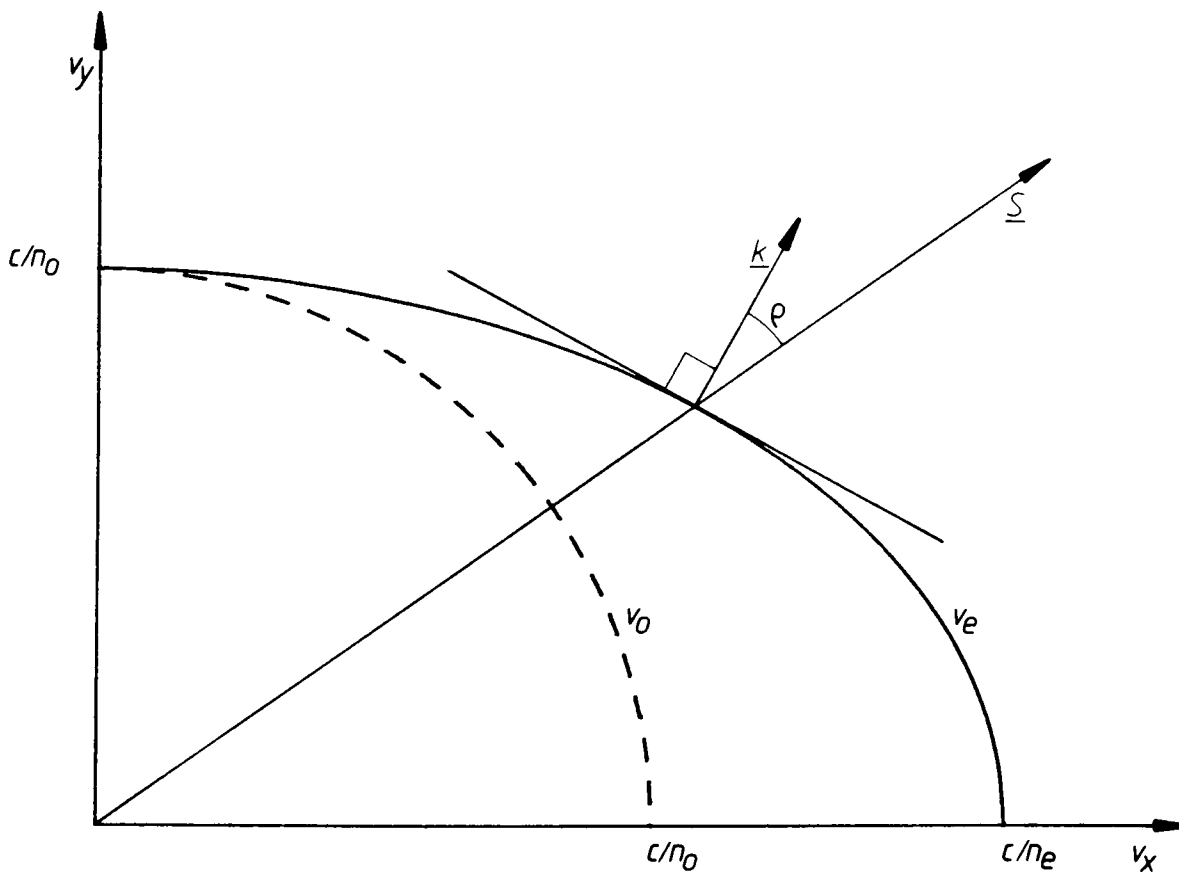


Figure V-4
The ray velocity surface describes the shape of the wavefront resulting from a point disturbance at the origin. The wavevector at each point is normal to the surface, and is therefore not generally coincident with the Poynting vector $\underline{S}$. This is the phenomenon of Poynting vector walk-off.

V-6. Secondary effect of birefringence: Poynting vector walk-off.

The ray velocity surface of figure V-4 shows how the refractive index of a birefringent crystal depends upon the direction of propagation of the wavefront. A Huygens-like construction of wavefronts emanating from pointlike sources, illustrated in figure V-5, demonstrates the propagation of a plane wave through a birefringent medium, and it is clear that the Poynting vector in general makes a small angle with the wave normal, the wavevector $\underline{k}$.

Focussed beams, however, cover a range of directions, and the variation of refractive index within this range will therefore have to be taken into account. Since the angular spread is usually small, however, we need only consider the variation within a small angle of the mean. This may be done by approximating the appropriate section of the ray velocity ellipse to a section of a circle, whose centre is offset from the origin in a direction perpendicular to the Poynting vector so that the two curves share the same tangent if not the same curvature. This approximation allows the birefringence to be modelled as a uniform refractive index superimposed upon a constant "drift" velocity perpendicular to the wavevector, so that the usual rules for wave propagation in isotropic media apply and the birefringence is included as a sideways drift. This is illustrated in figure V-6. Simple geometry shows that the angle between the Poynting vector and wavevector is given by the walk-off angle ρ , defined by

$$\tan(\Theta - \rho) = \left[\frac{\eta_o}{\eta_e} \right]^2 \tan \Theta \quad (\text{V-11})$$

and thus

$$\tan \rho = \frac{1 - \left[\frac{\eta_o^{(2\omega)}}{\eta_e^{(2\omega)}} \right]^2}{\cot \Theta + \tan \Theta \left[\frac{\eta_o^{(2\omega)}}{\eta_e^{(2\omega)}} \right]^2} \quad (\text{V-12})$$

where Θ is the usual phase matching angle defined above. When modelled as a constant refractive index with drift velocity, the Poynting vector walk off angle defines the ratio of drift velocity to ray velocity.

The walk-off angle describes the angular rate of change of refractive index, $d\eta/d\Theta$. This property may also be described by an acceptance angle, the angular range within which the phase mismatch between fundamental and harmonic is less than one cycle. The phase mismatch $\Delta k = 2k_1 - k_2$ at an angle $\Delta\Theta$ to the phase matched direction, where k_1 and k_2 are the wavenumbers of the fundamental and second harmonic, may be shown to be given by

$$\frac{\Delta k}{\Delta \Theta} = \frac{\pi \eta_1^3}{\lambda_2} \sin 2\Theta \left[\frac{1}{[\eta_e^{2\omega}]^2} - \frac{1}{[\eta_o^{2\omega}]^2} \right] \quad (\text{V-13})$$

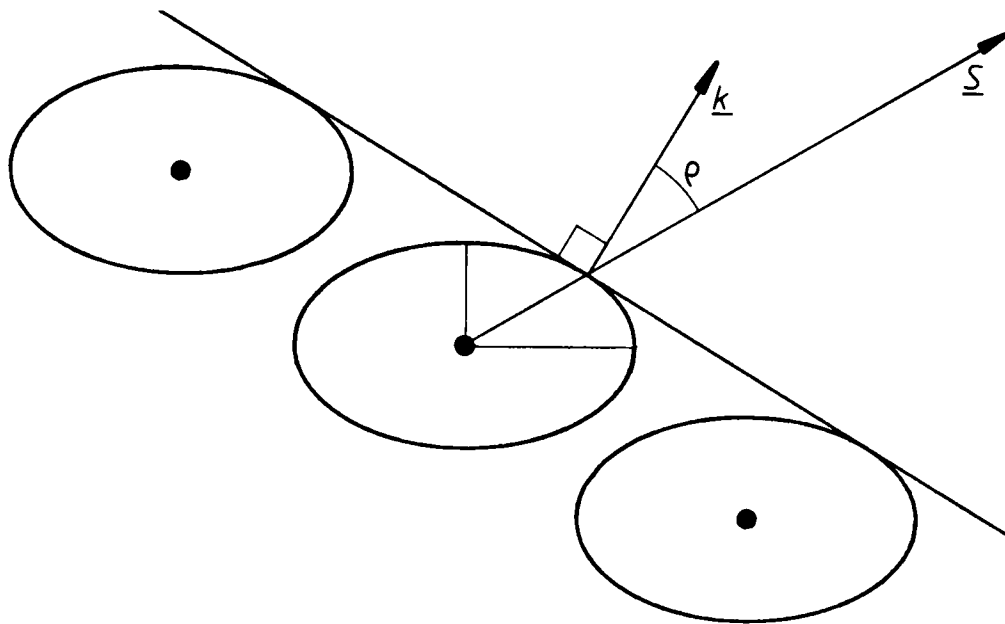


Figure V-5
Huygen's construction for propagation from a line source.

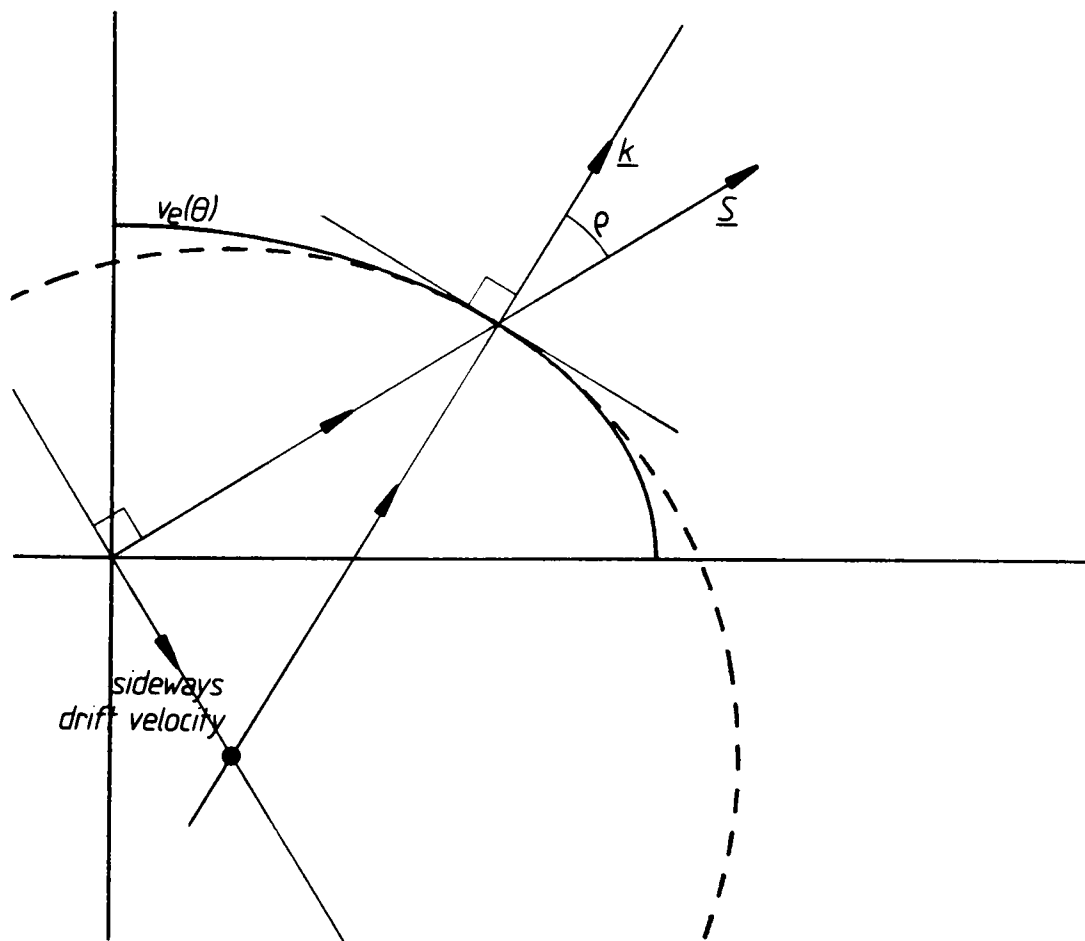


Figure V-6
"Sideways drift velocity" added to the spherical propagation characteristic of a non-birefringent medium approximates over small angular ranges to the true, birefringent, propagation.

The phase mismatch at $\Delta\theta$ is $l\Delta k$, where l is the crystal length. The acceptance angle θ_{acc} , at which $l\Delta k=2\pi$, is thus given by

$$\theta_{acc} = \frac{\lambda_1}{l\eta_1^3 \sin 2\theta} \left[\left[\frac{1}{\eta_e^2 \omega} \right]^2 - \left[\frac{1}{\eta_o^2 \omega} \right]^2 \right]^{-1} \quad (V-14)$$

V-7. Longitudinal coherence and bandwidth.

Second harmonic generation depends upon the longitudinal coherence or bandwidth of the laser source in two ways. The light must be coherent over the temporal range, caused by focussing in a birefringent medium, which contributes to a given instant of harmonic power; and the phase-matching orientation should be appropriate for the range of wavelengths present. The required coherence length thus depends upon the variation of refractive indices with angle, and the variation of refractive indices with wavelength.

When perfect phase matching occurs, the instantaneous harmonic intensity is dependent solely upon the accrued conversion of the corresponding single wavefront of the fundamental as it passes through the crystal, so longitudinally separated regions are relevant only if there is some phase mismatch. This may be seen explicitly by leaving the time dependence in the otherwise steady-state theory of chapter V. The mismatch results from the angular distribution present in a focussed beam and the variation in phase-matching within this angle, and there will be no such phase mismatch to first order, for example, when non-critical phase-matching is used. Longitudinal coherence is therefore required only over a distance of order $\Delta l = (\Delta k/k)l$ where Δk is the maximum phase mismatch acquired in the process, and k and l are the usual wavenumber and crystal length. In the later experiments with beta barium borate, this gives a coherence length of the order of $30\mu\text{m}$ and corresponds to a bandwidth of 5nm fwhm.

Dispersion of the birefringence, however, leads to a dependence of the phase matching angle upon wavelength. It is clear that there will thus be a limit to the bandwidth which may be converted efficiently, which will be of the order $\Delta\lambda = \theta_{acc}/(d\theta/d\lambda)$ where θ_{acc} is the acceptance angle of the crystal and which will impose another limit upon the bandwidth. For the sample used in the experiments of the next chapter, the dispersion $d\theta/d\lambda$ is around $0.12^\circ.\text{nm}^{-1}$ and the acceptance angle is 0.015° ; this suggests a limit to the bandwidth of only 0.1nm (100GHz , corresponding to a coherence length around 1mm). This bandwidth is rather narrower than that calculated above, but nonetheless far exceeds the copper vapour laser linewidth of around 3GHz .

V-8. Second harmonic generation with elliptical Gaussian beams.

The most rigorous derivation of the form of a Gaussian beam is that given by Yariv [V-1] and Marcuse [V-10] which shows that an assumed form of Gaussian beam is a solution to the electromagnetic wave equation in the slowly varying approximation. This has an advantage over the alternative derivation in terms of truncated spherical waves in that it is valid at the beam waist. It is easily applied to an elliptical form of Gaussian beam, and confirms the form of the elliptical Gaussian beam propagating along the z axis as

$$E = E_0 \frac{\exp(-ikz)}{[(1-i\tau_x)(1-i\tau_y)]^{\frac{1}{2}}} \exp \left\{ - \left[\frac{x^2/w_x^2}{1-i\tau_x} + \frac{y^2/w_y^2}{1-i\tau_y} \right] \right\} \quad (V-15)$$

where $\tau_x = 2(z-f)/k\omega_x^2$ and $\tau_y = 2(z-f)/k\omega_y^2$. The foci in the y - z and x - z planes coincide and are found at $z=f$, where the waist radii or ellipse semi-axes are w_x and w_y respectively. It is this form which will be used in the following calculations. The theoretical treatment of second harmonic generation from elliptical Gaussian laser beams is based upon the approach of Boyd and Kleinman [V-6] and maintains their notation throughout. However, in extending the treatment to allow the Gaussian beams to have an elliptical cross-section, x and y subscripts have been added where the elliptical asymmetry renders it necessary.

This is not the first attempt to treat elliptical focussing in second harmonic generation. Published work by Librecht and Simons [V-11] presents specific results for the example of a 2cm long crystal of ADP and suggests that the authors were aware of more general results. However, the treatment presented here leads to results of general significance, and reduces the relevant crystal properties to just one parameter from which the optimum circularly and elliptically focussed conversion efficiencies may readily be found. Curves relating the normalized conversion efficiency to this parameter have been calculated and form part of the recipe from which the performance of any uniaxial crystal may be predicted. Finally, in various limiting regimes, some physical interpretation of the results is possible, and this is discussed in section V-8.2.

For many nonlinear crystals, the birefringence is sufficient that an analytical calculation for the limit of high birefringence is a good approximation. The analytical treatment of section V-8.3, which reduces the calculation to tabulated functions, also follows that of Librecht and Simons, although it is appropriate not only in their extremes of strong and weak focussing but also in the interesting regime where the conversion efficiency is maximum.

V-8.1 Mathematical treatment of second harmonic generation.

The convention of Boyd and Kleinman [V-6], that an observer at $[x,y,z]$ sees harmonic generation from a range of points $[x',y',z']$ within the crystal, is maintained. The laboratory axes x,y,z are defined by the direction of propagation of the fundamental $[0,0,1]$ and the optic axis of the crystal $[x,0,z]$, and figure V-7 illustrates the relation between these laboratory axes and the crystal's own geometry.

It is worthwhile at this point to define the variables which will be used in this treatment. The table below summarizes this; amplification will be made as the variables are introduced.

ω_1, ω_2	=	fundamental and harmonic frequencies
k_1, k_2	=	fundamental and harmonic wavenumbers
Δk	=	$2k_1 - k_2$
w_x, w_y	=	waist radii (or ellipse semi-axes)
b_x, b_y	=	confocal parameters, $w_x^2 k_1, w_y^2 k_1$
l	=	crystal length
ϵ_x	=	focussing parameter, l/b_x
e	=	ellipticity, w_x/w_y
μ	=	position of focus in crystal, $= (l-2f)/l$ when the focus is at $z=f$
β	=	ratio of walk-off angle to Gaussian beam half-angle, $= \rho w_x k_1/2$

In the course of the calculation, two dimensionless coordinate systems will be used:

s, s'	=	angle made with axis in critical and non-critical planes, in units of Gaussian half-angle
$\tau_x, \tau_x', \tau_y, \tau_y'$	=	longitudinal dimension, in units of confocal parameter

The remaining variables are

σ	=	phase mismatch in critical direction, $= \frac{1}{2} b_x \Delta k$
σ'	=	$\sigma + 4\beta s$
K	=	$128\pi^2 \omega_1^2 d_{\text{eff}}^2 / c^3 \eta_1^2 \eta_2$
P_1	=	fundamental power
α_1	=	absorption coefficient for fundamental
α_2	=	absorption coefficient for harmonic
α	=	$\alpha_1 - \frac{1}{2}\alpha_2$
α'	=	$\alpha_1 + \frac{1}{2}\alpha_2$
κ	=	$\frac{1}{2}\alpha b_x$

We assume the form given for an elliptical Gaussian beam, so that at a point in the crystal $[x',y',z']$ the spatial variation of the fundamental electric field is given by equation (V-15). It follows that the harmonic polarization will be given by

$$P(x', y', z') = P_0 \frac{\exp(2ik_1 z' - \alpha_2 z')}{(1+i\tau_x')(1+i\tau_y')} \exp -2 \left[\frac{x'^2/w_x^2}{1+i\tau_x'} + \frac{y'^2/w_y^2}{1+i\tau_y'} \right] \quad (\text{V-16})$$

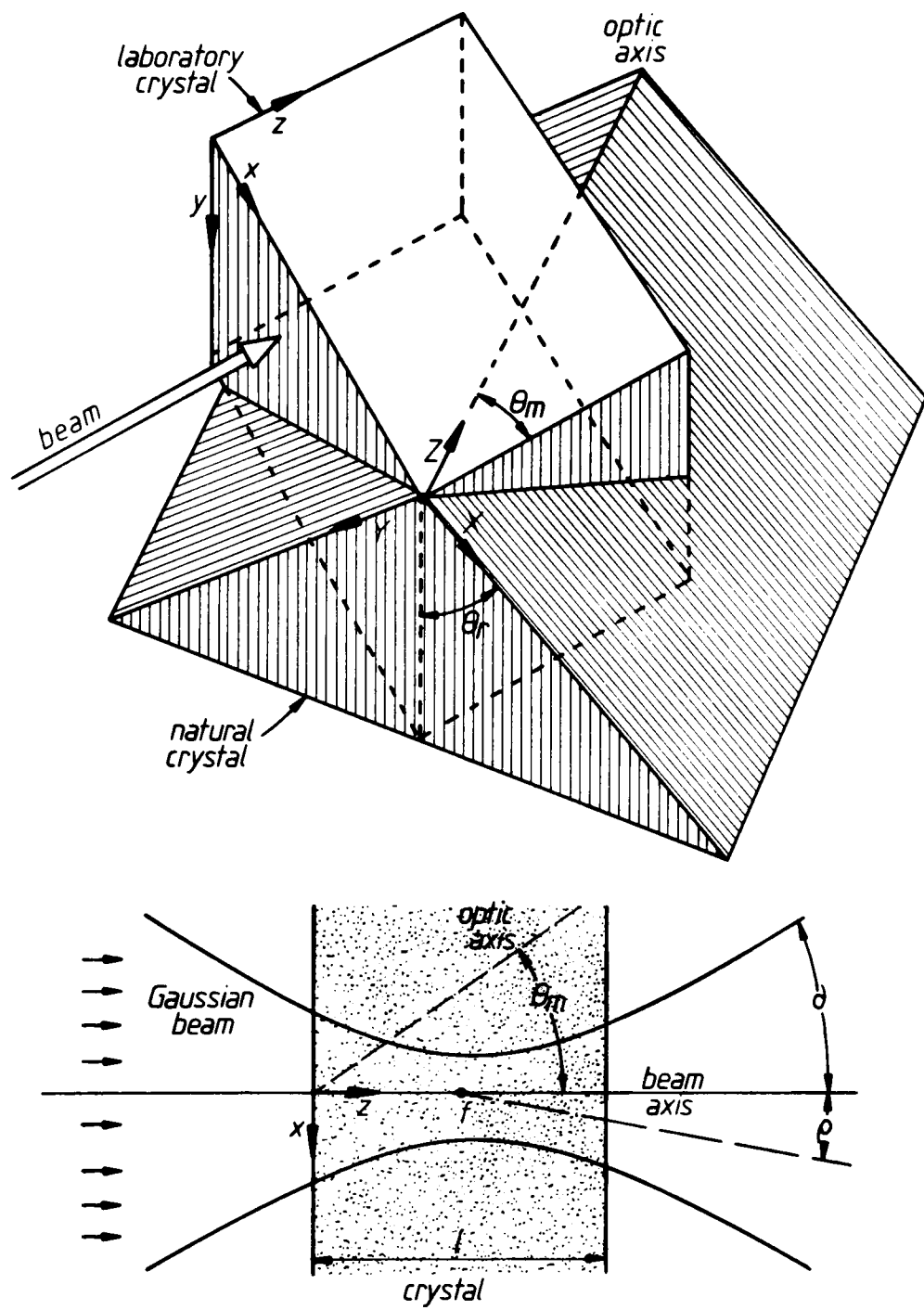


Figure V-7
Coordinate systems for second harmonic generation. The crystal is shown as a rectangular solid with the laboratory axes $\underline{x}, \underline{y}, \underline{z}$ parallel to the edges. For the purpose of visualizing the relation between $\underline{x}, \underline{y}, \underline{z}$ and the crystallographic axes $\underline{X}, \underline{Y}, \underline{Z}$, the latter are shown as the edges of a hypothetical "natural" crystal. The lower diagram shows the Gaussian laser beam focussed in the crystal at $x=y=0, z=f$. The divergence of the Gaussian beam is greatly exaggerated in the lower diagram. The origin of $\underline{x}, \underline{y}, \underline{z}$ is taken on the beam axis at the incident surface. Phase matching for a uniaxial crystal is determined by the matching angle $\theta_m = \angle(\underline{z}, \underline{Z})$. Rotation of the crystal about its optic axis is specified by $\theta_r = \angle(\underline{y}, \underline{Y})$. {From Boyd and Kleinman [V-6]}.

where $\tau_x' = 2(z' - f)/w_x^2 k_1$, etc. and confocal parameters may be defined by $b_x = w_x^2 k_1$, and so on. The increment in harmonic amplitude contributed by a slab of thickness dz' is

$$dA_2(x', y', z') = \frac{2\pi i \omega_2}{4\pi \epsilon_0 c \eta_2} P_{0x} \frac{\exp(i\Delta k z' - \alpha z')}{(1+i\tau_x')^{\frac{1}{2}}(1+i\tau_y')^{\frac{1}{2}}} dz' \cdot \frac{1}{(1+i\tau_x')^{\frac{1}{2}}(1+i\tau_y')^{\frac{1}{2}}} \exp -2 \left[\frac{x'^2/w_x^2}{1+i\tau_x'} + \frac{y'^2/w_y^2}{1+i\tau_y'} \right] \quad (V-17)$$

Now, the transverse dependence of this harmonic amplitude within the slice at z' looks like a slice through a Gaussian beam and will therefore propagate like one, so our technique is to calculate how all the beams from such slices will propagate down the crystal, and then to add them together coherently further downstream. The second harmonic oscillates with frequency 2ω and, being extraordinary, is subject to beam walk off. Propagation is thus described by allowing the transverse dependence to vary as would the appropriate Gaussian but including the transformation $x' = x - \rho(z - z')$, leading to an incremental amplitude at $[x, y, z]$ given by

$$dA_2(x, y, z) = \frac{2\pi i \omega_2}{4\pi \epsilon_0 c \eta_2} P_{0x} \frac{\exp \left[i\Delta k z' - \alpha_1 z' - \frac{1}{2}\alpha_2(l - z') \right]}{(1+i\tau_x')^{\frac{1}{2}}(1+i\tau_y')^{\frac{1}{2}}} \cdot \frac{1}{(1+i\tau_x)^{\frac{1}{2}}(1+i\tau_y)^{\frac{1}{2}}} \exp -2 \left[\frac{[x - \rho(l - z')]^2/w_x^2}{1+i\tau_x} + \frac{y^2/w_y^2}{1+i\tau_y} \right] \quad (V-18)$$

Integration over the length of the crystal, $z=0$ to l , gives the total harmonic amplitude at $[x, y, z]$,

$$E_2(x, y, z) = \frac{2\pi i \omega_2 P_{0x} \exp(-\frac{1}{2}\alpha_2 l + 2ik_1 z)}{4\pi \epsilon_0 c \eta_2 (1+i\tau_x)^{\frac{1}{2}}(1+i\tau_y)^{\frac{1}{2}}} \int_0^l \frac{\exp(-\alpha z' + i\Delta k z')}{(1+i\tau_x')^{\frac{1}{2}}(1+i\tau_y')^{\frac{1}{2}}} \cdot \exp -2 \left[\frac{[x - \rho(l - z')]^2}{1+i\tau_x} + \frac{y^2/w_y^2}{1+i\tau_y} \right] dz' \quad (V-19)$$

and hence the intensity at this point. The total harmonic intensity is then found by integrating the intensity over a transverse plane: for simplicity in the calculation, we perform this in the limit $z \rightarrow \infty$, when

$$\frac{1}{1+i\tau_x} = \frac{1-i\tau_x}{1+\tau_x^2} = \frac{1-i\tau_x}{\tau_x^2} \left[1 - \tau_x^{-2} + \tau_x^{-4} - \dots \right] \rightarrow \frac{1-i\tau_x}{\tau_x^2}$$

and similarly,

$$\frac{1}{1+i\tau_y} \rightarrow \frac{1-i\tau_y}{\tau_y^2}$$

For convenience later, we define

$$s = [x - \rho(l-f)]/w_x \tau_x \quad (V-20)$$

$$s' = y/w_y \tau_y \quad (V-21)$$

$$\text{and } \beta = \rho/\delta_x \quad \text{where } \delta_x = 2w_x/b_x \quad (V-22)$$

$$\text{so that } \frac{[x - \rho(l-z')]}{w_x \tau_x} = \left[s - \beta \frac{\tau_{x'}}{\tau_x} \right]^2$$

$$\begin{aligned} \text{and therefore } \frac{[x - \rho(l-z')]}{w_x^2 (1+i\tau_x)} &= \left[s - \beta \frac{\tau_{x'}}{\tau_x} \right]^2 (1-i\tau_x) [1 - \tau^{-2} + \dots] \\ &= s^2(1-i\tau_x) - 2i\beta\tau_{x'}s + [\text{order } \tau^{-1}] \end{aligned} \quad (V-23)$$

Equation (V-19) may now be written as

$$\begin{aligned} E_2(x, y, z) &= \frac{2\pi w_2 P_{0x}}{c\eta_2} \cdot \frac{i}{(1+i\tau_x)^{\frac{1}{2}}(1+i\tau_y)^{\frac{1}{2}}} \exp\left[-\frac{1}{2}\alpha_2 l + 2ik_1 z\right] \\ &\cdot \exp\left[-2\left(s^2(1-i\tau_x) + s'^2(1-i\tau_y)\right)\right] \cdot \int_0^l \exp\left[4i\beta\tau_{x'}s\right] \frac{\exp(-\alpha z' + i\Delta k z')}{(1+i\tau_{x'})^{\frac{1}{2}}(1+i\tau_{y'})^{\frac{1}{2}}} dz' \end{aligned} \quad (V-24)$$

and we note that as $\tau_{x,y} \rightarrow \infty$, $1/[(1+i\tau_x)^{\frac{1}{2}}(1+i\tau_y)^{\frac{1}{2}}] \rightarrow 1/i[\tau_x\tau_y]^{\frac{1}{2}}$. As a result, we reach

$$\begin{aligned} E_2(x, y, z) &= \frac{2\pi w_2 P_{0x}}{c\eta_2 (\tau_x \tau_y)^{\frac{1}{2}}} \exp\left[2ik_1 z - \frac{1}{2}\alpha_2 l\right] \exp\left[-2\left(s^2(1-i\tau_x) + s'^2(1-i\tau_y)\right)\right] \\ &\cdot \int_0^l \exp\left[4i\beta\tau_{x'}s\right] \frac{\exp(-\kappa\tau_{x'}) \exp(i\sigma'\tau_{x'})}{(1+i\tau_{x'})^{\frac{1}{2}}(1+ie^2\tau_{x'})^{\frac{1}{2}}} dz' \end{aligned} \quad (V-25)$$

Now define

$$H = \frac{1}{2\pi} \int_{-\epsilon_x(1-\mu)}^{\epsilon_x(1+\mu)} \frac{\exp(-\kappa\tau_{x'}) \exp(i\sigma'\tau_{x'})}{(1+i\tau_{x'})^{\frac{1}{2}}(1+ie^2\tau_{x'})^{\frac{1}{2}}} d\tau_{x'} \quad (V-26)$$

where

$$\sigma_x = \frac{1}{2}b_x\Delta k$$

$$\sigma' = \sigma + 4\beta s$$

$$\epsilon_x = l/b_x$$

$$\mu = (l-2f)/l$$

$$\kappa = \frac{1}{2}\alpha b_x$$

$$e = w_x/w_y = \text{ellipticity of Gaussian waist section}$$

Using $z' = \frac{1}{2}b_x\tau_{x'} + f$ and $d\tau_{x'} = [2/b_x].dz'$, H may be written as

$$H = \frac{1}{\pi b_x} \exp\left[\alpha f - i\Delta k f\right] \int_0^l \frac{\exp(-\alpha z' + i\Delta k z') \exp(4i\beta s \tau_{x'})}{(1+i\tau_{x'})^{\frac{1}{2}}(1+i\tau_{y'})^{\frac{1}{2}}} dz' \quad (V-27)$$

and may be substituted into (V-25) to give

$$E_2(x, y, z) = \frac{32\pi^2\omega_2 P_1 b_x}{\eta_1 \eta_2 c^2 w_x w_y (\tau_x \tau_y)^{\frac{1}{2}}} d_{\text{eff}} \exp\left[\frac{\alpha'}{2}l + \frac{\alpha l \mu}{2} - 2(s^2 + s'^2)\right] \cdot H \quad (V-28)$$

where we have used the expression $P_{0x} = d_{\text{eff}} E_0^2 = 16P_1 d_{\text{eff}} / [\eta_1 c w_x w_y]$ and have dropped the irrelevant phase term $\exp(-2i(s^2 \tau_x + s'^2 \tau_y)) \cdot \exp(2ik_1 + i\Delta k f)$. The second harmonic intensity, $S = [\eta_2 c / 8\pi] \cdot |E_2|^2$, is now given by

$$S(x, y, z) = \frac{128\pi^2 \omega_1^2}{c^3 \eta_1^2 \eta_2} d_{\text{eff}}^2 \frac{4\pi P_1^2 k_1^2}{\tau_x \tau_y} \left[\frac{w_x}{w_y} \right]^2 \exp \left[-\alpha' l + \alpha l \mu - 4(s^2 + s'^2) \right] |H|^2 \quad (\text{V-29})$$

and the total harmonic power is given by the integral of S over a transverse plane

$$\begin{aligned} P_2 &= \iint S \, dx \, dy = w_x w_y \tau_x \tau_y \iint S \, ds \, ds' \\ &= 4\pi K P_1^2 k_1^2 w_x w_y \left[\frac{w_x}{w_y} \right]^2 \exp \left[-\alpha' l + \alpha l \mu \right] \int_{-\infty}^{\infty} \exp(-4s'^2) ds' \int_{-\infty}^{\infty} \exp(-4s^2) |H|^2 ds \quad (\text{V-30}) \end{aligned}$$

where we have written $K = 128\pi^2 \omega_1^2 / [c^3 \eta_1^2 \eta_2]$. Using the standard result for the area under a Gaussian, we finally arrive at

$$P_2 = K P_1^2 l k_1 \exp(-\alpha' l) \left[\frac{\pi^2}{\epsilon} \exp(\mu \alpha l) \left[\frac{2}{\pi^{\frac{1}{2}}} e \int_{-\infty}^{\infty} \exp(-4s^2) |H|^2 ds \right] \right] \quad (\text{V-31})$$

where

$$H = \frac{1}{2\pi} \int_{-\epsilon_x(1-\mu)}^{\epsilon_x(1+\mu)} \frac{\exp(-\kappa \tau_x') \exp(i\sigma' \tau_x')}{(1+i\tau_x')^{\frac{1}{2}} (1+ie^2 \tau_x')^{\frac{1}{2}}} d\tau_x' \quad (\text{V-32})$$

This is our final expression for the second harmonic power. It will be noted that all the optimizable parameters are contained within the bracketted term which we designate h_m :

$$h_m = \left[\frac{\pi^2}{\epsilon_x} \exp(\mu \alpha l) \left[\frac{2}{\pi^{\frac{1}{2}}} e_g \int_{-\infty}^{\infty} \exp(-4s^2) |H|^2 ds \right] \right] \quad (\text{V-33})$$

h_m is a function of the parameters ϵ_x , e , β , σ , μ , l , α and κ . The last two characterize the absorption of the crystal, and may often be assumed to be zero. With negligible absorption, the optimum value of μ will be zero, and the dependence upon l drops out. σ represents the deviation from normal phase matching, and will in practice be optimized by fine adjustment of the crystal orientation, so only the optimum value of σ need be considered. This reduces the set of parameters necessary to determine h_m to ϵ_x , e , and β , and it is convenient to rewrite β in the form $B(\epsilon_x)^{-\frac{1}{2}}$, where B is completely defined by the physical properties of a given crystal through

$$B = \frac{1}{2} \rho [l k_1]^{\frac{1}{2}} \quad (\text{V-34})$$

In this way, the function h_m is defined by B , ϵ_x and e ; the first parameter describes the crystal, and the other two describe the focussing arrangement which is under our control. Equation (V-31) is consistent with the results of Boyd & Kleinman [V-6] for

circular focussing and Librecht and Simons [V-11] when there is no absorption and the focus is at the crystal centre.

For the calculation of numerical results, we indeed set the absorption to zero and put the focus at the centre of the crystal, which is the best position when everything else is optimized. This makes the calculation slightly easier but mainly is chosen to give the results some general significance. The technique with the numerical calculations is to vary ϵ , β and σ so as to keep ρ and k constant. The result, as a function of β and σ , is then optimized with respect to σ , which has the same effect as fine-tuning the angle of the crystal to the laser beam. Further calculations may be made with different values of the ellipticity e . A listing of the program used is given in appendix V-1.

V-8.2 Numerical results and interpretation.

Curves showing the maximized h_m for spherical focussing and optimum elliptical focussing, as a function of crystal parameter B , are shown in figure V-8, and the corresponding focussing parameters are given in figures V-9 and V-10. An improvement in the conversion efficiency approaching a factor of 1.3 is predicted to accompany a change to elliptical focussing for crystals with appreciable birefringence. This improvement is probably large enough to be of interest in a number of commercial applications of second harmonic generation, but it is not proposed to attempt its use in the current copper laser based system: the experimental precision is insufficient to allow a proper verification of these results to be made, and in any case the interpretation of their significance for the temporally evolving copper laser pulse would not be trivial. Any useful increase in the power from the current copper laser system would be all but lost in reflections from the extra lens required to achieve elliptical focussing, and alignment is expected to be rather more difficult to achieve.

It was found that whilst h_m is a slowly varying function of both e and ϵ_x , the optimum value of ϵ_x varies swiftly with e ; the product $e\epsilon_x$ represents the waist area, and turns out to be rather less sensitive. The product $e\epsilon_x$ is therefore used to define the overall focussing strength.

Boyd and Kleinman's graph showing h_m as a function of both B and ϵ is reproduced here as figure V-11: some general features of this graph are justified intuitively in the following pages.

V-8.2.1 Very hard focussing without birefringence ($\epsilon \gg 1$, $B=0$)

Away from the beam waist, the harmonic intensity generated by a local interaction volume increases in proportion to the square of the fundamental intensity, which falls in inverse proportion to the beam cross-sectional area. Away from the

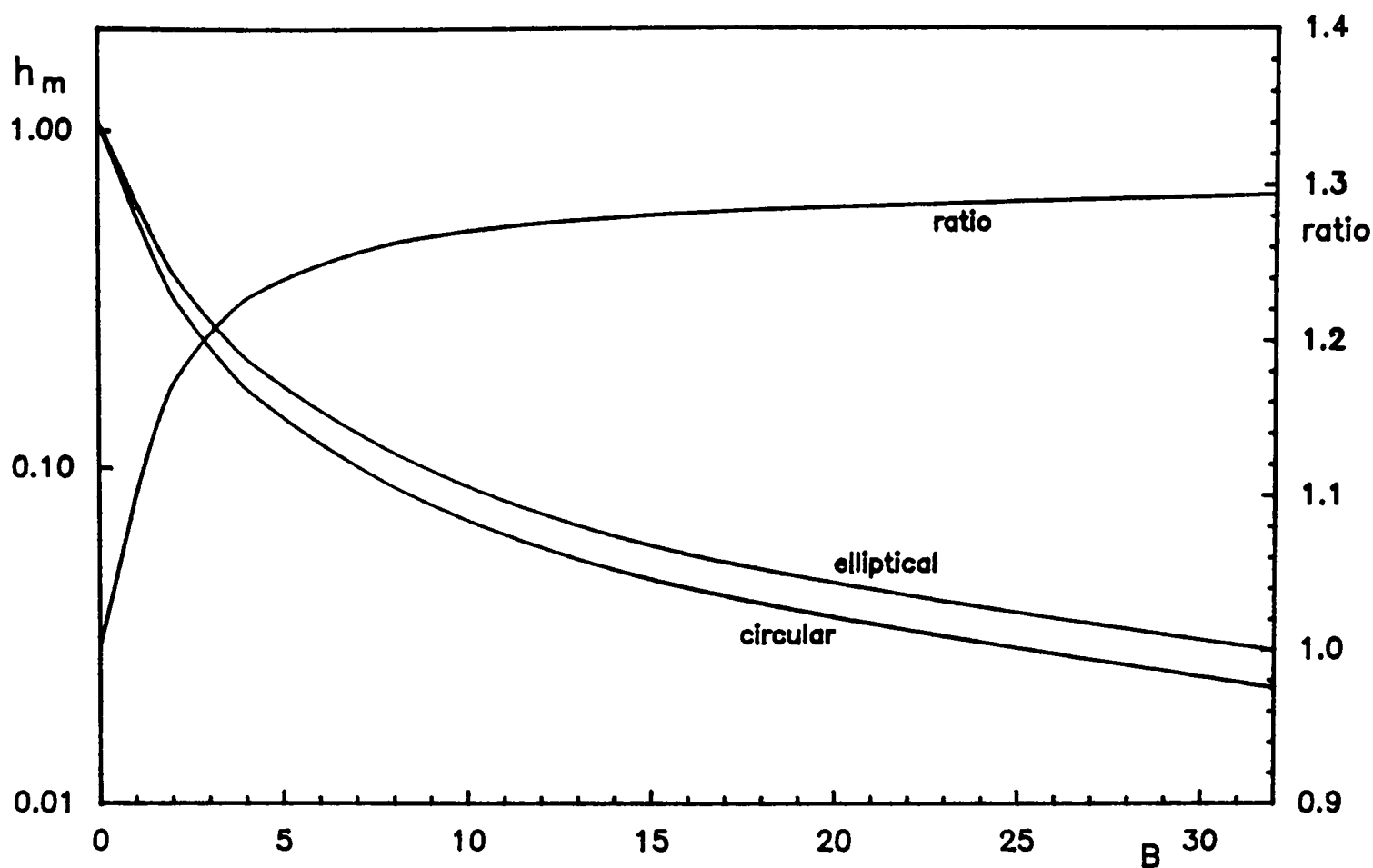


Figure V-8
Comparison of elliptical with circular focussing. The coefficients h_m are shown for the two cases as a function of the parameter B ; the relative improvement is highlighted by the plot of their ratio, and with the more birefringent crystals an enhancement by a factor of nearly 1.3 is predicted.

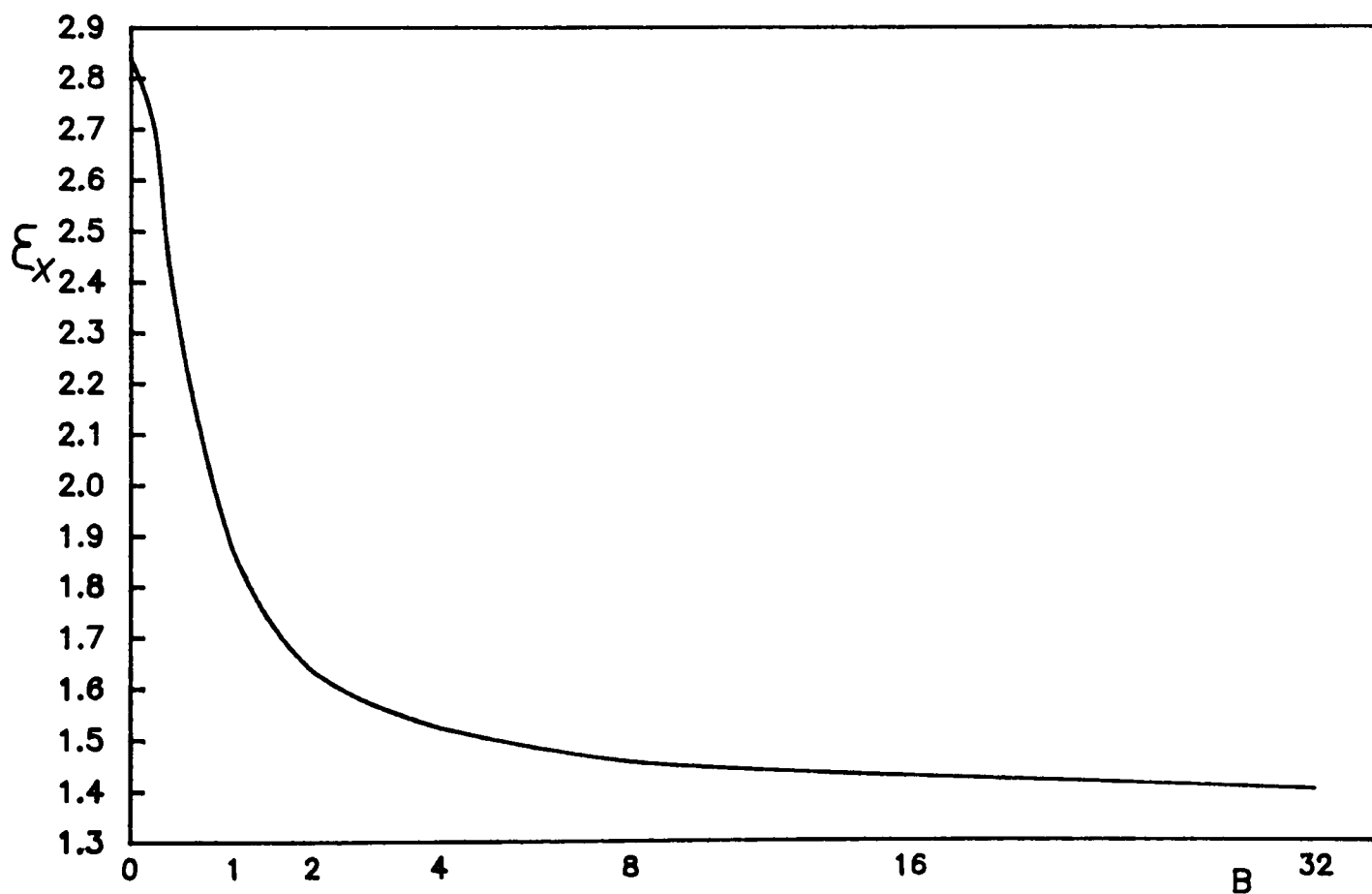


Figure V-9
Optimum circular focussing conditions are described by the parameter $\epsilon = l/b = l/(w^2 k)$. As B increases, ϵ approaches a value around 1.39.

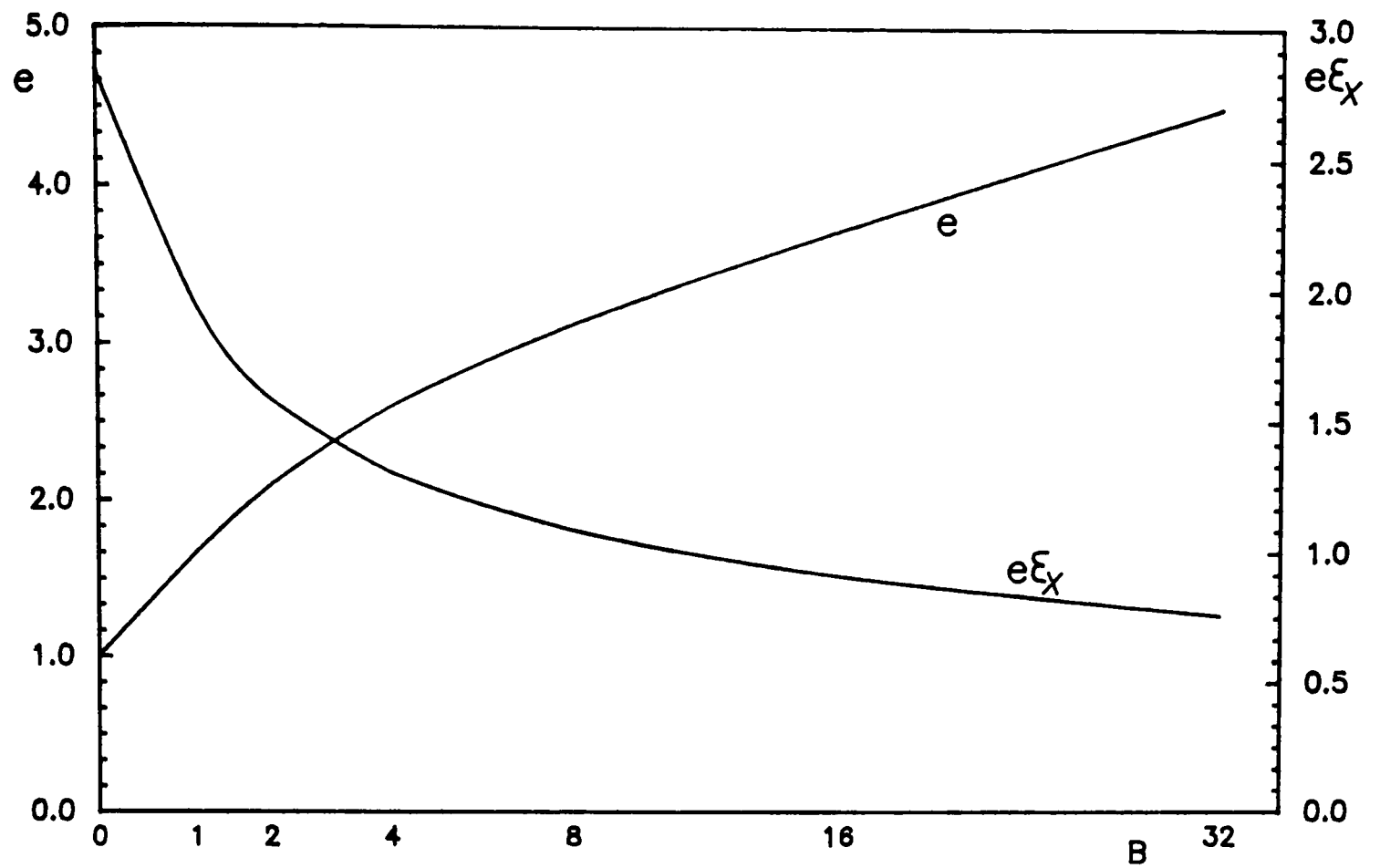


Figure V-10

Optimum elliptical focussing conditions are described by the parameter $\epsilon_x = l/(w_x^2 k)$ and the ellipticity $e = w_x/w_y$. Each is rather more dependent upon the other than upon the parameter B, and hence the relatively less sensitive function $e\epsilon_x$ is shown, from which ϵ_x may be derived.

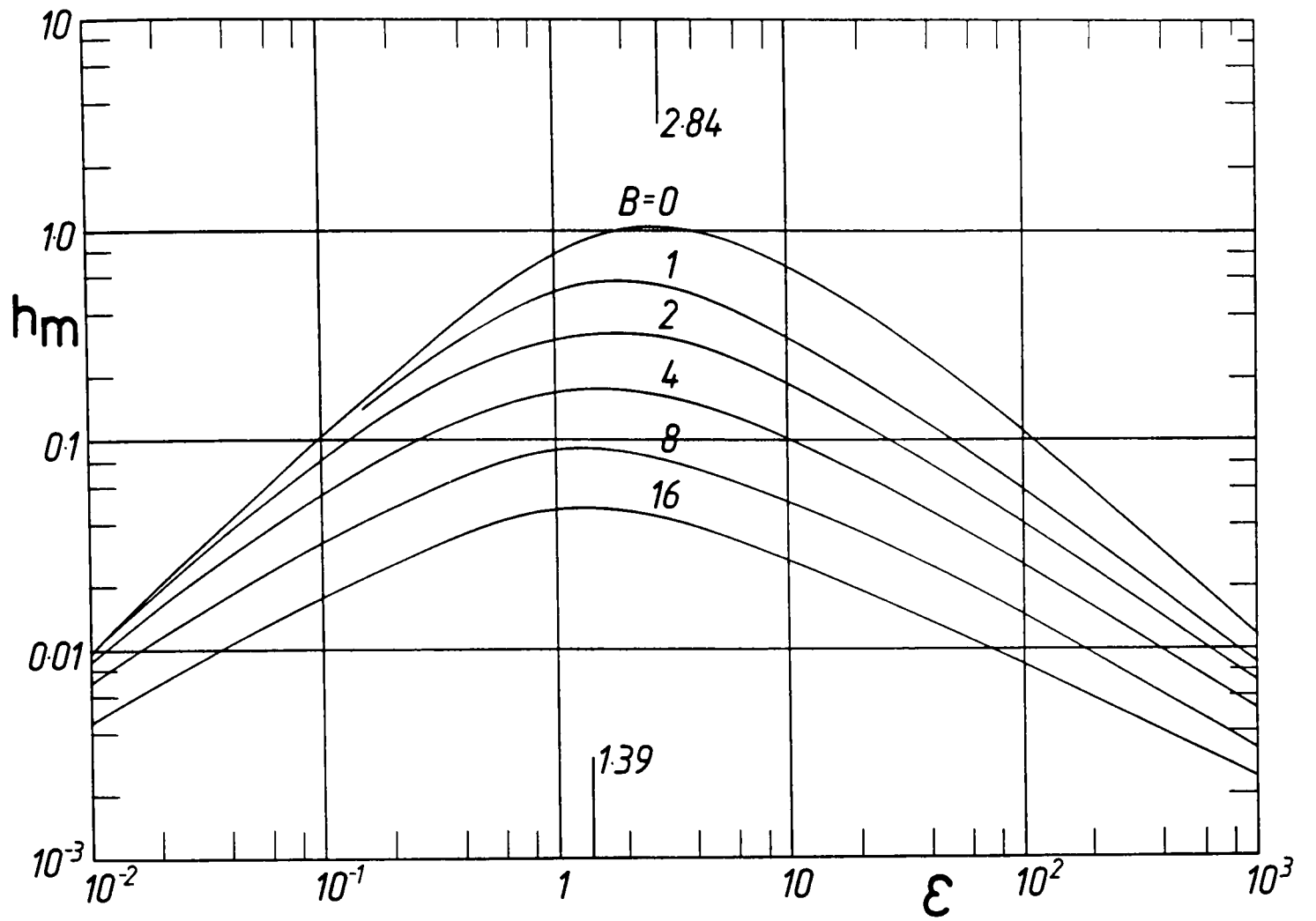


Figure V-11
Variation of harmonic power with circular focussing. The dependence of the harmonic power, represented by the function $h_m(B, \epsilon)$, upon the focussing parameter $\epsilon = l/b$ is shown for several values of the double-refraction parameter $B = \rho(lk_1)^{1/2}/2$. Vertical lines indicate the optimum focussing in the limits of small and large B .

focus, then, the increment to the total harmonic power will fall in proportion to the cross-sectional area, and will become a vanishingly small proportion of the total when the cross-sectional area is much greater than that at the beam waist. Thus with very strong focussing, the extremes of the crystal will make a negligible contribution.

With w fixed to define the focussing geometry, varying ϵ is equivalent to varying the crystal length. The harmonic power P_2 is known to vary in proportion to $l \cdot h_m$, and will be unchanged. Thus with strong focussing,

$$h_m \propto 1/l$$

$$\text{or } \log_{10} h_m = \text{const} - \log_{10} \epsilon.$$

V-8.2.2 Very weak focussing without birefringence ($\epsilon \ll 1$, $B=0$)

This is the case of bounded plane waves, and the harmonic power will increase with the square of the crystal length. This time, then, $h_m \propto l$, so for weak focussing

$$\log_{10} h_m = \text{const} + \log_{10} \epsilon.$$

V-8.2.3 With birefringence

Varying the crystal length without changing the beam focussing will cause a proportional change in ϵ , and a variation in B as $(l)^{1/2}$. Otherwise, the above arguments then apply, provided that an increase in ϵ is accompanied by a proportional increase in B^2 – and thus for constant B the asymptotic slopes will be less steep. With very low ϵ , the beams will propagate as plane waves, and the B dependence must then drop out; the curves will merge.

The variation in the optimum value of ϵ as a function of birefringence B is best seen by reference to the acceptance angle Θ_{acc} . Without birefringence, the optimum configuration corresponds to the highest intensity possible without a significantly reduced depth of focus. Birefringence, through the introduction of an acceptance angle, will reduce the effectiveness of the far-field regions (away from the focus), and thus there will be a tendency to reduce the focussing strength, and thus ϵ , as B increases.

An interesting phenomenon worthy of note is that the maximum harmonic power does not quite correspond to the traditional phase matching condition $\Delta k=0$, but rather Δk is found to take a small, positive value. This is explained by Boyd and Kleinman: the laser beam may be regarded as a diverging pencil of plane waves, and when $\Delta k > 0$ for the central portion of the beam, there still exist mixing processes for which $k_1' + k_1'' - k_2 = 0$; when $\Delta k < 0$, matching by mixing is not possible. When the birefringence is high, the angular range of the beams is reduced, and Δk tends to the expected limit.

As the birefringence increases, it becomes more appropriate to consider only the part of the Gaussian whose direction lies within the acceptance angle of the crystal. This corresponds to Boyd and Kleinman's case of high β [V-6] and reduces the

problem to maximizing the intensity within this region. In this regime, Boyd and Kleinman find that an analytic solution is possible; the same is true in the generalized case with elliptical Gaussian beams.

V-8.3 High birefringence case - an analytic solution

Once again ignoring absorption, we position the focus at the centre of the crystal and consider the regime where β is sufficiently large that $\exp(-4s^2)$ [$=\exp-4((\sigma'-\sigma)/4\beta)^2$] is essentially unity over the range of significant $|H|^2$. This latter condition means that within the crystal's acceptance angle the beam has uniform amplitude. h_m may now be written as

$$h_m = \frac{2\pi^{3/2}e}{\epsilon_x} \int_{-\infty}^{\infty} \exp -4 \left[\frac{\sigma' - \sigma}{4\beta} \right]^2 \left| \frac{1}{2\pi} \int_{-\infty}^{\infty} \exp i\sigma' \tau_{x'} f(\tau_{x'}) d\tau_{x'} \right|^2 \frac{d\sigma'}{4\beta} \quad (V-35)$$

with $f(\tau_{x'}) = \frac{1}{(1+i\tau_{x'})^{1/2}(1+ie^2\tau_{x'})^{1/2}}$. {aperture function bounded by $\pm\epsilon_x$ }

We now invoke Parseval's theorem, and find that

$$h_m = \frac{\pi^{1/2}}{4\beta\epsilon_x} \int_{-\infty}^{\infty} |f(\tau_{x'})|^2 d\tau_{x'} = \frac{\pi^{1/2}e}{4\beta\epsilon_x} \int_{-\epsilon_x}^{\epsilon_x} \left| \frac{1}{(1+i\tau)^{1/2}(1+ie^2\tau)^{1/2}} \right|^2 d\tau \quad (V-36)$$

The function of τ may be rewritten as

$$\left| \left[\frac{1}{(1+i\tau)(1+ie^2\tau)} \right]^{1/2} \right|^2 = \frac{1}{(1+\tau^2)^{1/2}(1+e^4\tau^2)^{1/2}} \quad (V-37)$$

and thus

$$\begin{aligned} h_m &= \frac{\pi^{1/2}e}{4\beta\epsilon_x} \int_{-\epsilon_x}^{\epsilon_x} \left[(1+\tau^2)(1+e^4\tau^2) \right]^{-1/2} d\tau \\ &= \frac{\pi^{1/2}}{2\beta\epsilon_x e} \int_0^{\epsilon_x} \left[(1+\tau^2)(e^{-4}+\tau^2) \right]^{-1/2} d\tau \end{aligned} \quad (V-38)$$

This is now an incomplete elliptic integral of the first kind,

$$F(\Phi \backslash \alpha) = a \int_0^x \left[(t^2+a^2)(t^2+b^2) \right]^{-1/2} dt \quad (V-39)$$

$$\begin{aligned} \tan\Phi &= x/b \\ \cos\alpha &= b/a \\ a &> b \end{aligned}$$

and is tabulated in the *Handbook of Mathematical Functions* by Abramowitz and

Stegun [V-12]. In this case, $a=1$, $b=1/e^2$ ($e>1$), and

$$h_m = \frac{\pi^{\frac{1}{2}} \epsilon_x^{\frac{1}{2}}}{2Be\epsilon_x} F(\tan^{-1} e^2 \epsilon_x \backslash \cos^{-1} \frac{1}{e^2}) \quad (V-40)$$

From the tabulated functions, with $B=16$, $e=1$, $\epsilon_x=1$, we obtain $h_m=0.04350$. This is in good agreement with the computed result of 0.04302, which is lower by 1%. In general, the approximation $\exp(-4s^2)=1$ will make the analytic value an over-estimate.

V-9. Crystal symmetry and nonlinear coefficients.

The nomenclature and theory of the symmetry properties of crystals and their significance for the nonlinear susceptibility tensor $\chi^{(2)}$ is well covered by Nye [V-5]; a more specific treatment for optical harmonic generation may be found in the review article by Franken and Ward [V-2], and a useful summary may be found in Yariv [V-1]. I am grateful to Malcolm Boshier in this laboratory for an early draft of those parts of his thesis which consider the theory of SHG with particular reference to beta-barium borate.

Neumann's principle is quoted by Nye as "*The symmetry elements of any physical property of a crystal must include the symmetry elements of the point group of the crystal*" and means that if the orientation of a crystal cannot be distinctly identified from a detailed knowledge of its layout, then it cannot affect its physical properties. The point group of a crystal describes the set of symmetry properties that the crystal follows. There are 32 different point groups, which fall into seven crystal systems: cubic, which exhibits no birefringence; hexagonal, tetragonal and trigonal, which will be uniaxial; and orthorhombic, monoclinic and triclinic, which will be biaxial.

By considering the effect of an inversion operation upon equation (V-2), it is easy to see that crystals possessing inversion symmetry cannot have non-zero $\chi^{(2)}$ components. Eleven of the 32 crystal classes have inversion symmetry; only the remaining 21 may be of use in second harmonic generation. The symmetry of equation (V-2) with respect to interchange of E_p and E_q requires that $\chi_{ijk}=\chi_{ikj}$, and thus suggests a shorthand by which the tensor is reduced to eighteen terms in a 3x6 array: the contracted form of the tensor is $\chi_{im}=\chi_{ijk}$, where $m=j$ for $\chi_{i11}, \chi_{i22}, \chi_{i33}$ and $m=9-(j+k)$ for the remaining terms. The expression $\underline{E} \cdot \underline{E}$ is now replaced by the tensor $[E_x^2 \ E_y^2 \ E_z^2 \ 2E_y E_z \ 2E_z E_x \ 2E_x E_y]$. This technique relies upon the indistinguishability of the two interacting electric fields, and is therefore invalid if, for example, they differ in frequency as in sum frequency mixing; the extent to which this is invalid is considered by Franken and Ward [V-2].

Kleinman [V-13] has further suggested that for certain materials χ_{ijk} may be

identical to χ_{jik} . It is the equivalent of this symmetry in the linear tensor, $\chi^{(1)}_{ij} = \chi^{(1)}_{ji}$, which leads to the description of the various crystal surfaces in terms of quadrics (which are then reduced to ellipsoids of rotation or whatever by reference to the further symmetry properties of the specific crystal systems). For many crystal classes relevant to second harmonic generation, the Kleinman symmetry holds.

For future use, we shall consider those uniaxial crystals for which Kleinman symmetry holds. Beta-barium borate, for example, is a negative uniaxial crystal of class 3, and ADP and KDP are of class $\bar{4}2m$.

V-10. The effective nonlinear coefficient.

With type I phase matching, the fundamental beam will be polarized in the ordinary direction and therefore in the x-y plane. If the propagation direction is defined by an azimuthal angle ϕ and inclination θ (see figure V-2), then the fundamental field may be written as

$$\begin{bmatrix} E_x & E_y & E_z \end{bmatrix} = E(\omega_1) \begin{bmatrix} \sin\phi & -\cos\phi & 0 \end{bmatrix} \quad (V-41)$$

and thus the $\underline{E} \cdot \underline{E}$ tensor $[E_x^2 \ E_y^2 \ E_z^2 \ 2E_y E_z \ 2E_z E_x \ 2E_x E_y]$ will be

$$\begin{bmatrix} \sin^2\phi & \cos^2\phi & 0 & 0 & 0 & -\sin 2\phi \end{bmatrix} E(\omega_1)^2 \quad (V-42)$$

The local induced harmonic polarization will thus be

$$\begin{aligned} \underline{P}(2\omega) &= \epsilon_0 E(\omega_1)^2 \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix} \begin{bmatrix} \sin^2\phi \\ \cos^2\phi \\ 0 \\ 0 \\ 0 \\ -\sin 2\phi \end{bmatrix} \\ &= \epsilon_0 E(\omega_1)^2 \begin{bmatrix} d_{11}\sin^2\phi + d_{12}\cos^2\phi - d_{16}\sin 2\phi \\ d_{21}\sin^2\phi + d_{22}\cos^2\phi - d_{26}\sin 2\phi \\ d_{31}\sin^2\phi + d_{32}\cos^2\phi - d_{36}\sin 2\phi \end{bmatrix} \end{aligned} \quad (V-43)$$

The direction of extraordinary polarization will be $[\cos\phi\cos\theta, \sin\phi\cos\theta, -\sin\theta]$ and thus the component of the induced harmonic polarization which may be phase matched will be

$$P_e(2\omega) = \epsilon_0 E(\omega_1)^2 \begin{bmatrix} \cos\phi\cos\theta & \sin\phi\cos\theta & -\sin\theta \end{bmatrix} \cdot \begin{bmatrix} d_{11}\sin^2\phi + d_{12}\cos^2\phi - d_{16}\sin 2\phi \\ d_{21}\sin^2\phi + d_{22}\cos^2\phi - d_{26}\sin 2\phi \\ d_{31}\sin^2\phi + d_{32}\cos^2\phi - d_{36}\sin 2\phi \end{bmatrix} \quad (V-44)$$

and hence, by the definition in equation (V-4a), the effective d coefficient is given by

$$d_{\text{eff}} = \cos\phi\cos\theta(d_{11}\sin^2\phi+d_{12}\cos^2\phi-d_{16}\sin2\phi)+\sin\phi\cos\theta(d_{21}\sin^2\phi+d_{22}\cos^2\phi-d_{26}\sin2\phi) \\ -\sin\theta(d_{31}\sin^2\phi+d_{32}\cos^2\phi-d_{36}\sin2\phi) \quad (\text{V-45})$$

This expression may now be simplified for a specific crystal by using a knowledge of its symmetry properties to deduce relations between the components of the d tensor. Fortunately, tables of the simplified d components exist for each of the crystal classes (see for example Zernike and Midwinter [V-14], Nye [V-5] or Byer [V-3]).

In chapter VI, results of second harmonic generation using beta barium borate (BBO) will be reported. The crystal is of class 3 (trigonal, with one three-fold rotation axis), and Kleinman symmetry applies. The simplified second order susceptibility tensor for BBO thus looks like

$$\begin{bmatrix} d_{11} & -d_{11} & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & -d_{11} \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix} \quad (\text{V-46})$$

so that the expression for d_{eff} may be reduced to

$$d_{\text{eff}} = (d_{22}\sin3\phi - d_{11}\cos3\phi)\cos\theta - d_{31}\sin\theta \quad (\text{V-47})$$

Using values for d_{11} , d_{22} and d_{31} originally measured by Chen [V-15] to be 1.6, 0 and 0.1 pm.V⁻¹ respectively, we obtain

$$d_{\text{eff}} = -1.6\cos3\phi\cos\theta - 0.1\sin\theta \quad \times 10^{-12}\text{m.V}^{-1} \quad (\text{V-48})$$

from which it is obvious that the highest nonlinearity will occur for $\phi=0$. At $\theta=45^\circ$, then, which is typical for copper vapour lasers, $d_{\text{eff}} \sim 1.2$ pm.V⁻¹.

Finally, it is worth mentioning that d coefficients are often defined in units of the d_{36} of KDP. Eimerl gives this as 0.39 pm.V⁻¹ [V-16], but has pointed out a significant discrepancy between published values of this standard. All experiments in which $d_{36}(\text{KDP})$ is measured by fitting the SHG conversion efficiency of highly collimated lasers give 0.39 ± 0.01 pm.V⁻¹, whereas the Landolt-Bornstein tables [V-17] quote 0.63 pm.V⁻¹.

V-11. Some practical examples.

Table V-1 lists a number of SHG parameters for a range of readily available crystals for use at the copper laser wavelengths. θ , the phase matching angle, and ρ , the Poynting vector walk-off angle, have been calculated from published coefficients for the Sellmeier expansion of refractive index dispersion. The B parameter then includes a typical crystal length which is usually 10mm. These data allow h_m and d_{eff}

to be computed, and from these a harmonic generation coefficient P_2/P_1^2 , the ratio of instantaneous harmonic power to the square of the fundamental power, can be found. Finally, the acceptance angle θ_{acc} is recorded.

Figure V-8 predicts that elliptical focussing will give an improvement in harmonic generation by a factor of at least 1.2 by all crystals for which $B > 3$, which is indeed the case for all the noncritically phasematched examples listed. The detrimental effect of birefringence upon harmonic generation is clearly apparent, for despite having low d_{eff} coefficients, both the noncritically phase-matched crystals (ADP for use with the green line and ADA for the yellow) have harmonic generation values around $0.5 \text{ mW} \cdot \text{W}^{-2}$ in comparison with the typical angle-tuned values below $0.1 \text{ mW} \cdot \text{W}^{-2}$. It thus appears that the noncritically phase-matched crystals should be the best for generating harmonics of copper laser radiation.

There are, however, two effects which have yet to be included. These are the constraint of a low damage threshold which prevents optimum focussing conditions being used with high power lasers, and the effect of absorption and thermal dephasing. Damage thresholds are generally well defined and are strongly dependent upon the material chosen; the disruptive effects of absorption, however, are not easy to calculate, but they are much more of a problem with the temperature-tuned materials which must have high temperature dependences. In many cases it is therefore the angle-tuned crystals which are best.

Table V-1 Crystals for second harmonic generation.

crystal class		KDP	KD*P	ADP	ADA	urea	BBO
d	pm/V	$\overline{4}2m$ $d_{36}=0.39(1)$	$\overline{4}2m$ $d_{36}=0.34(2)$	$\overline{4}2m$ $d_{36}=0.45(3)$	$\overline{4}2m$ $d_{36}=0.56$	$\overline{4}2m$ $d_{36}=1.35$	3 $d_{13}=0.07$
η		— 1.5099	— 1.5063	— 1.529	— 1.525	$d_{11}=1.6$ 1.575	1.6767 1.6710
θ		— 64.0	— 66.3	90 65.3	— —	63.8	50.5 41.9
ρ		— 2.2	— 1.2	0 1.43	— —	3.2	4.84 4.75
B		— 7.7	— 4.3	0 5.08	— —	12.4	16.05 ^c 14.78 ^c
h_m		— 0.0910	— 0.157(3)	0 ^a 1.069	0 ^b 1.069 ^a	0.0574(5)	0.0440(3) ^c
d_{eff}	pm/V	— 0.35(1)	— 0.31(2)	0.45(3) 0.41(3)	— —	1.21	1.1(3) 1.3(3)
$P_{P_1^2}^2$	$\mu W.W^{-2}$	— 15	— 20	414 830 ^a	— —	151	59(30) ^c 62(30) ^c
θ_{acc}	deg	— 0.06	— 0.10	1.86 1.86 ^a	— 0.06 ^a	0.03	0.03 ^c 0.03 ^c

The two values per column refer to fundamental wavelengths of 511nm and 578nm respectively.
All calculations are based on a crystal length of 10mm, except where otherwise indicated.

- ^a 20mm crystal length
- ^b temperature tuned to noncritical phase-matching
- ^c 7mm crystal length

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V-Appendix 1. Computer program SHG.FOR

```

C      SHG  VERSION 2.0    24.2.88          VAX FORTRAN
C
C      *****
C      Calculation of Second Harmonic Power with Boyd & Kleinman
C      theory modified to include Elliptical Gaussian Beams
C      *****
C      PROGRAM SHG
C
C      INTEGER*2 DI,DJ,DK
C      CHARACTER*8  TTIME
C      DOUBLE PRECISION      BETA,E, EPSILN,KAPPA,MU,SIGMA,ESQRD,FORBTA,
1      DELS,DELTAU,S1,S2,E4,PI,PISQRD,TWOPI,ANSWER,
1      B,X1,X2,X3,Y1,Y2,Y3,DELSIG,Q,R,XTP,TOL,DS1,DS2
C      COMMON  BETA,E, EPSILN,KAPPA,MU,SIGMA,ESQRD,E4,FORBTA,
X      DELS,DELTAU,S1,S2,PI,PISQRD,TWOPI,ANSWER
C
C
C      10  READ (8,*,END=999) B,E, EPSILN,KAPPA,MU,S1,S2,TOL
C      CALL TIME(TTIME)
C      CALL IDATE(DI,DJ,DK)
C      WRITE      (9,1001) DJ,DI,DK,TTIME
1001  FORMAT      (1X,'SHG.FOR      VERSION 2.0  ',10X,12.2,2('/ ',12.2)
,3X,A8)
C      WRITE      (9,1002) B,E, EPSILN,TOL
C      WRITE      (9,1002) KAPPA,MU,S1,S2
1002  FORMAT      (39X,4(1X,G9.3))
C
C
C      BETA      = B/DSQRT(EPSILN)
C      ESQRD     = E*E
C      E4        = ESQRD*ESQRD
C      FORBTA    = 4.0*BETA
C
C
C      DELS      = (S2-S1)/500.0
C      DELTAU    = 2.0*EPSILN/1000.0
C      WRITE      (9,1003) DELS,DELTAU
1003  FORMAT      (40X,'DELS=' ,E10.4,5X,'DELTAU=' ,E10.4)
C
C      PI        = 3.1415926544
C      PISQRD    = PI*PI
C      TWOPI     = 2.0*PI
C
C
C      Routine ISOL8MAX
C
C      X1        = 0.0
C      Y1        = -1.0
C      SIGMA     = 0.0
C      CALL      FCALC
C      X2        = SIGMA
C      Y2        = ANSWER
C      DS1       = PI/EPSILN
C      DS2       = 1.0
C      DELSIG    = (DMIN1(DS2,DS1))/2.0

```

```

C
100 SIGMA      = SIGMA+DELSIG
    CALL FCALC
    WRITE      (9,1004) SIGMA,ANSWER,X1,X2
    WRITE      (9,1005) Y1,Y2
1004     FORMAT (5X,G12.4,2X,G12.4,10X,G12.4,1X,G12.4)
1005     FORMAT (41X,G12.4,1X,G12.4)
    IF (ANSWER.GT.Y2) THEN
        X1     = X2
        Y1     = Y2
        X2     = SIGMA
        Y2     = ANSWER
        DELSIG  = DELSIG*1.4
        GO TO   100
    ELSE
        X3     = SIGMA
        Y3     = ANSWER
    ENDIF
C
C  Routine PARASCEND
C
200 Q      = (Y2-Y1)*(X2-X3)
    R      = (Y2-Y3)*(X2-X1)
C  IF R=Q THEN: A:DIVIDE BY ZERO WILL FOLLOW
C              B:QUADRATIC WOULD STOP AT X=X2-XTP
    XTP    = X2-((X2-X1)*R-(X2-X3)*Q)/((R-Q)*2)
    SIGMA   = XTP
    CALL FCALC
    WRITE   (9,1006) SIGMA,ANSWER,X1,X2,X3
    WRITE   (9,1007) Y1,Y2,Y3
1006     FORMAT (10X,G12.4,2X,G12.4,5X,G12.4,1X,G12.4,1X,G12.4)
1007     FORMAT (41X,G12.4,1X,G12.4,1X,G12.4)
    IF (ANSWER.GT.Y2) THEN
        IF (XTP.GT.X2) THEN
            X1     = X2
            Y1     = Y2
            X2     = XTP
            Y2     = ANSWER
        ELSE
            X3     = X2
            Y3     = Y2
            X2     = XTP
            Y2     = ANSWER
        ENDIF
    ELSE
        IF (XTP.GT.X2) THEN
            X3     = XTP
            Y3     = ANSWER
        ELSE
            X1     = XTP
            Y1     = ANSWER
        ENDIF
    ENDIF
    ERROR    = Y2-(Y1+Y3)/2.0
    IF (ERROR.LE.TOL) GO TO 300
GO TO      200

```

```

C
C   Acceptable value achieved
C
300 ANSWER      = ANSWER*2*PI*DSQRT(PI)*E
   WRITE        (9,1008) SIGMA,ANSWER
1008   FORMAT    (1X,G12.4,2X,G12.4)
   WRITE        (9,1009)
1009   FORMAT    (1X,79('*'))
C
   GO TO        10
C
C
999 CLOSE       (8,STATUS='KEEP')
   CLOSE       (9,STATUS='KEEP')
   STOP
   END

C      FCALC      VERSION 2.0      24.2.88      VAX FORTRAN
C      SUBROUTINE FCALC FOR SHG
C
C      SUBROUTINE FCALC
C
C      DOUBLE PRECISION      BETA,E,EPSILN,KAPPA,MU,SIGMA,ESQRD,FORBTA,
X      DELS,DELTAU,S1,S2,TAU,PI,PISQRD,TWOPI,E4,
X      MDLS(1002),ARG1(1002),BTAU(1002),ARG,HREAL,
X      HIMAG,RINT,IINT,HSQRD,HINT,S,TAUSQ,ANSWER
C
C      COMMON      BETA,E,EPSILN,KAPPA,MU,SIGMA,ESQRD,E4,FORBTA,
X      DELS,DELTAU,S1,S2,PI,PISQRD,TWOPI,ANSWER
C
C      Set up DO loop parameters to ensure positive integer constants
C
C      SS2      = (S2-S1)/DELS + 1
C      S loop now runs from 1 to SS2 in integer steps
C      S = (SS-1)*DELS+S1
C      Set S = S1 before loop; S = S+DELS at end of loop
C
C      TT2      = 2*EPSILN/DELTAU + 1
C      Tau loop now runs from 1 to TT2 in integer steps
C      TAU = (TT-1)*DELTAU+(EPSILN*(MU-1))
C
C      Calculate array of functions of TAU(TT)
C      TAU = EPSILN*(MU-1)
C      DO 100 TT=1,TT2
C          TAUSQ      = TAU**2
C          MDLS(TT)    = DEXP((-1.0)*KAPPA*TAU)
C          MDLS(TT)    = MDLS(TT)/DSQRT(DSQRT((1+TAUSQ)*(1+E4*TAUSQ)))
C          ARG1(TT)    = SIGMA*TAU-DATAN(TAU)/2.0-
X          DATAN(TAU*ESQRD)/2.0
C          BTAU(TT)    = FORBTA*TAU
C          TAU          = TAU+DELTAU
100   CONTINUE
C
C      HREAL      = MDLS(TT)*COS[ARG1(TT)+BTAU(TT)*S]
C      HIMAG      = MDLS(TT)*SIN[ARG1(TT)+BTAU(TT)*S]
C

```

```

C
C
C
C      Now start the real calculation ...
C
      S      = DBLE(S1)
      HINT    = 0.0
      DO 300 SS=1,SS2
C        Calculate /H/ **2
        RINT = 0.0
        IINT = 0.0
        DO 200 TT=1,TT2
            ARG      = ARG1(TT)+BTAU(TT)*S
            ARG      = DMOD(ARG,TWOPI)
            HREAL    = MDLS(TT)*DCOS(ARG)
            HIMAG    = MDLS(TT)*DSIN(ARG)
            RINT     = RINT + HREAL
            IINT     = IINT + HIMAG
200        CONTINUE
            HSQRD    = RINT**2 + IINT**2
            HINT     = HINT + HSQRD*DEXP((-4)*S**2)
            S        = S + DELS
300        CONTINUE
C
      ANSWER = HINT*DELTAU**2*DELS/(4*PISQRD*EPSILN)
      RETURN
C
      END

```

V-Appendix 2. Maxwell's equations with harmonic fields.

Maxwell's equations,

$$\underline{\nabla} \times \underline{H} = \frac{\partial \underline{D}}{\partial t} + \underline{J}$$

$$\underline{\nabla} \times \underline{E} = -\frac{\partial \underline{B}}{\partial t}$$

$$\underline{\nabla} \cdot \underline{D} = \rho$$

$$\underline{\nabla} \cdot \underline{B} = 0$$

are linear functions of $\underline{E}$, $\underline{D}$, $\underline{B}$ and $\underline{H}$, and thus for harmonic fields whose dependence upon time and position is given by $\exp i(\underline{k} \cdot \underline{r} - \omega t)$ the operators $\partial/\partial t$ and $\partial/\partial x$ etc. may be simplified to

$$\frac{\partial}{\partial t} = -i\omega$$

$$\frac{\partial}{\partial x} = ik_x \text{ etc.} \quad \text{so} \quad \underline{\nabla} = i\underline{k}$$

Maxwell's equations thus become

$$i\underline{k} \times \underline{H} = -i\omega \underline{D} + \underline{J}$$

$$i\underline{k} \times \underline{E} = \omega \underline{B}$$

$$i\underline{k} \cdot \underline{D} = \rho$$

$$i\underline{k} \cdot \underline{B} = 0$$

In dielectric media, therefore, $\underline{k} \cdot \underline{D} = 0$.

The practice of second harmonic generation.

VI-1. Introduction

VI-2. Second harmonic generation in beta-barium borate

2.1 Apparatus

2.2 Calibration

2.3 Results

2.4 Predicted harmonic generation

2.5 Generation of ultraviolet radiation at 255nm.

2.6 Doubling the yellow and mixing the green and yellow

2.7 Confirmation of phase matching angles

2.8 Thermal effects and laser damage

2.9 Developments elsewhere

VI-3. Second harmonic generation in temperature-tuned ADP

3.1 Apparatus

3.2 Results

3.3 Comment

VI-4. Conclusion

VI-5. Consideration of future systems

5.1 Improvements to current apparatus

5.2 High power second harmonic generation of 255nm radiation using BBO.

The practice of second harmonic generation.

VI-1 Introduction.

When this work was started, in 1986, second harmonic generation of ultraviolet radiation from a copper vapour laser had been reported twice – and only once in English. The first account was in a Russian paper from 1980 [VI-1]. Non-critical phasematching of a temperature-tuned crystal, ammonium dihydrogen phosphate (ADP), had been used with a 2W copper vapour laser: significant conversion could be achieved only transiently before the thermal influence of the laser was felt, and there was no prospect of repeating the best conversion efficiency of 8.5% at higher powers or for longer periods. The highest continuous uv power achieved was some 20mW.

A Chinese paper from 1984 [VI-2], however, reported early results from a crystal of beta-phase barium borate (BBO), a material which had been identified by workers at the Fujian Institute of Matter Structure in Fuzhou [VI-3] as being promising for nonlinear optics. This paper described the frequency doubling of a small copper laser by a crystal some 5.5mm long. Generation of the second harmonics of both the green and yellow lines was investigated, and the good performance achieved is comparable with the findings reported in this chapter. The Chinese had available only low laser powers, however, and their best conversion efficiency was thus no higher than 0.7%; the highest ultraviolet power achieved was 3.5mW.

In 1986, optical quality beta barium borate became widely available. It is used primarily in an angle tuned configuration and indeed displays remarkably little temperature dependence [VI-4]. Work described in this chapter has shown that the second harmonic of the copper laser green radiation may be generated with conversion efficiencies in excess of 10%, yielding mean powers up to 100mW of ultraviolet per W^2 of fundamental. Steady mean powers of 200mW at 255.3nm have been achieved from less than 2W of fundamental power, and there are no indications of immediate average power effects which would preclude scaling to higher power systems. Kuroda et al [VI-5] have published results obtained from a similar system at lower copper laser powers which entirely support the findings described in this chapter, and have recently reported generation of more than 220mW of ultraviolet at 8.9% conversion efficiency [VI-6]. Piper has similarly reported generation of 100mW, although only at an efficiency of 1.5% [VI-7].

The following pages cover the practical aspects of generating harmonics of the copper laser. Brief investigations using ADP have been performed and are described, but this chapter is primarily concerned with the extensive studies which have been made of second harmonic generation of ultraviolet radiation using beta-barium borate. The best conversion efficiency and optimum focussing conditions for frequency doubling of the green, 510.6nm, line are completely consistent with the predictions of the theory outlined in chapter V and the measured temporal development of the transverse coherence reported in chapter IV. The efficiency of second harmonic generation is found to vary little with the magnification of the unstable cavity of the copper laser over the experimental range from 15 to 150, for it is the pulse length and not peak power which is principally affected.

Similar performance has been measured for SHG with BBO using the yellow, 578.2nm, line whose superior coherence properties result in slightly better harmonic generation properties than the green, but whose lower fundamental power causes less efficient conversion overall. Sum frequency mixing of the two copper laser wavelengths has been studied briefly, and its poor performance is explained in terms of chromatic dispersion in the optical components and nonlinear crystal and relative timing of the green and yellow pulses. The damage threshold of beta barium borate has been indicated, and is consistent with recently published studies from low repetition rate lasers.

VI-2 Second harmonic generation in beta-barium borate.

VI-2.1 Apparatus.

Figure VI-1 shows an experimental arrangement typical of those used in these studies. The output of a copper laser, with an unstable cavity and intracavity polaroid, is filtered by two dichroic beamsplitters to leave only the green, 510.6nm, radiation which is focussed into the crystal of BBO by a lens of focal length f . The diverging radiation emerging from the crystal is collimated or slightly focussed by a silica lens and is dispersed by a 60° silica prism, allowing only the ultraviolet light to fall upon the radiometer which records its power. Measurements of the incident fundamental power are made by inserting a second radiometer into the beam between the first lens and the crystal. For this experiment, air tight beam path covers were used within the laser head box.

The crystal used for these experiments was cut for normal incidence at a nominal phase-matching angle of 45° , and measured around 4mm square by 7.2mm long. Beta barium borate is slightly hygroscopic, and the crystal was therefore supported in a sealed desiccator cell between a pair of silica windows. The E-field

polarization of the copper laser beam was horizontal, and that of the generated harmonic was vertical: the harmonic was thus readily reflected in a horizontal plane, and the majority of the prism losses may be attributed to the strong reflection of this polarization. The tortuous route from the laser before the first lens introduces sufficient optical delay (around 35ns) to isolate the various reflecting components from the laser cavity. Using this apparatus, the performance of second harmonic generation was investigated for a variety of lens focal lengths and cavity magnifications. In aligning each new cavity, attention was paid to the second harmonic power achieved as well as to the usual, but more subjective, features of copper laser beam profile and power. Initial alignment was performed using an apertured copper laser beam to avoid damaging the crystal when the focus was near the crystal ends.

The apparatus as drawn was used to investigate frequency doubling of the CVL green line yielding ultraviolet light at 255nm, this being the most promising process for generation of high ultraviolet powers. Sum frequency mixing of the two copper laser lines and second harmonic generation from the yellow line were also investigated, by replacing the dichroic beamsplitters of figure VI-1 with 45° reflectors, and using a single dichroic beamsplitter immediately afterwards to reject the green line when appropriate. Finally, the temporal dependence of the harmonic generation methods was investigated, using a Tektronix model 466 storage oscilloscope and the photomultiplier detector described in chapter IV.

VI-2.2 Calibration.

The power meter used for measuring the ultraviolet power was a Laser Instrumentation model 5103/15ST, which was calibrated at manufacture against standards traceable to the National Physical Laboratory. The copper laser power was measured mostly by a unit produced by Ophir, which was calibrated for the purposes of this experiment against the Laser Instrumentation meter. The transmission of the prism at the harmonic frequency was determined by filtering the beam after lens L2 with a Schott UG5 colour glass filter and comparing the measured power after the prism with that recorded in its absence. The ultraviolet transmission of the collimating lens was similarly measured, again using the copper laser harmonics as sources of the appropriate polarization and wavelength, and the transmissions of the prism and lens were thus found to be 74% and 93% respectively at both 255nm and 289nm. It should be noted that the ultraviolet transmission of the colour filter is rather poorer than advertised and tends to fall at first for a period of seconds; the transmitted ultraviolet power was therefore always allowed to settle before the prism transmission was measured. The results presented later in this chapter have been corrected to allow for these losses in the prism and collimating lens, and are derived from the

calibration of the Laser Instrumentation power meter.

No correction has been made for losses due to the crystal or its cell. Attenuation due to reflection at the desiccator cell windows must be at least 15% (assuming 4% per surface), and will account for a reduction in the u/g^2 ratio defined in the following section by a factor of 0.78; reflective losses at the crystal surfaces will introduce a further factor of 0.82. Pure reflection therefore lowers the u/g^2 ratio by a factor of 0.64. Losses due to scattering and absorption are likely to be dependent upon both wavelength and intensity and therefore cannot be established. They are nonetheless significant: the non-reflective losses in the crystal and cell at 510nm are around 35%.

VI-2.3 Results.

In order to interpret the measurements of fundamental and harmonic powers over the range of cavity magnifications and focussing parameters, it will initially be assumed that for a given arrangement the harmonic power u varies in proportion to the square of the fundamental power g , and thus that the ratio u/g^2 will indicate the performance of the harmonic generation irrespective of changes in laser power. This ratio will be referred to as the harmonic generation, and the validity of this assumption will be considered presently.

Figure VI-2 shows the variation of the green harmonic generation with cavity magnification for different lens focal lengths f . Lenses of focal lengths 0.15, 0.25, 0.5 and 1.0 metres were used, and the best results were clearly those obtained using the 0.25m lens. The harmonic generation may be seen to increase with cavity magnification, spanning a factor of two from a low magnification of 16 to the highest around 150. These results are presented as a function of focal length in figure VI-3, from which it appears that the best focal length to use is around 300mm.

Figures VI-4 and VI-5 show the achieved 255nm harmonic and green fundamental powers as a function of cavity magnification. With these figures it is the envelope of the points which is of interest, for poor alignment can only result in a reduction of the conversion efficiency and all data points must therefore lie below the true best conversion values. The highest harmonic power achieved was around 200mW at a conversion efficiency of 10.5%, using a cavity with a magnification around 40. Figure VI-6 illustrates the variation in efficiency over the range of cavities, and this is essentially constant.

Figure VI-7 shows the equivalent results for frequency doubling of the yellow copper laser line and for sum frequency mixing of the two lines together. Second harmonic generation from the yellow line shows a similar dependence to that illustrated in figure VI-3 for the green, but sum frequency mixing is found to exhibit

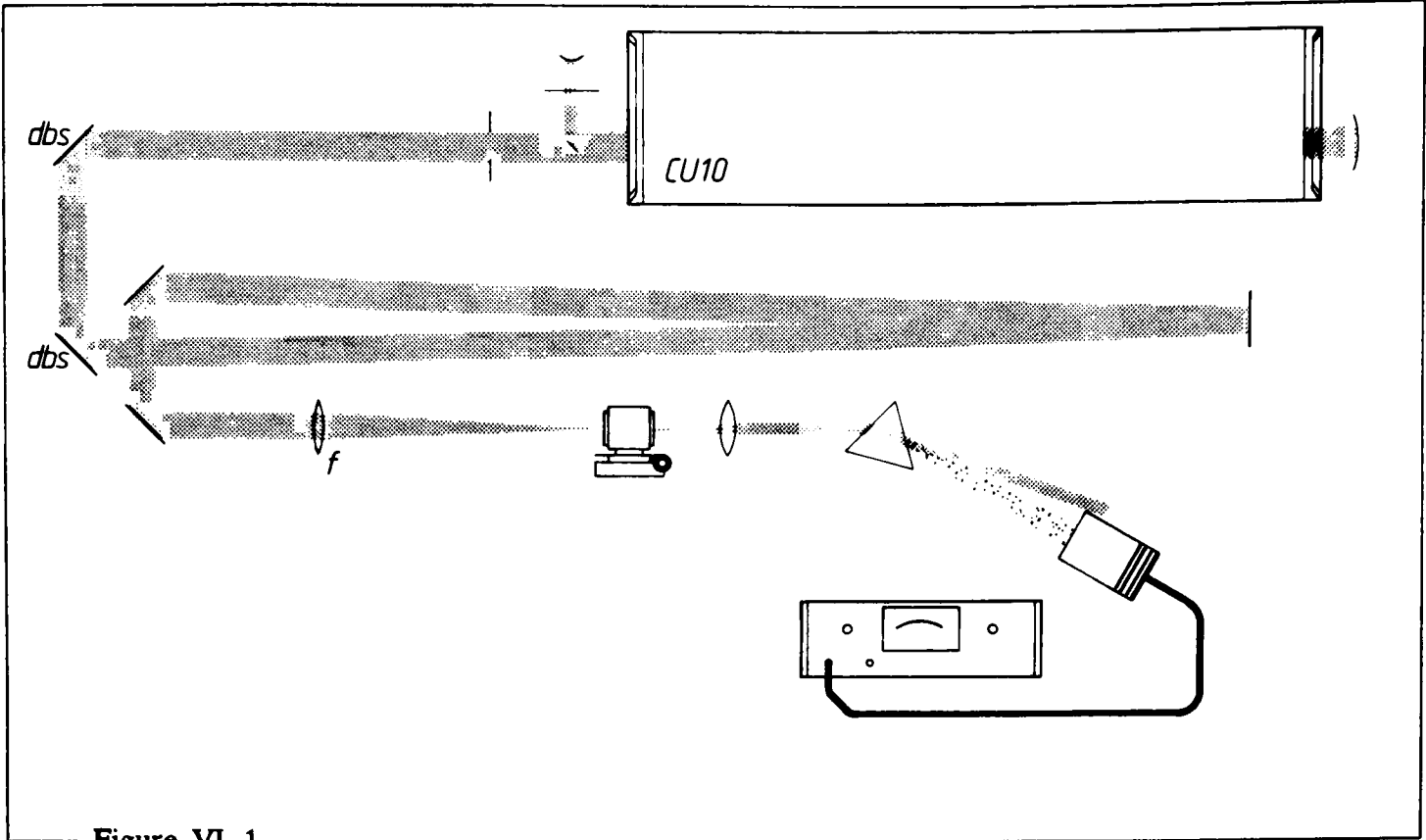


Figure VI-1
Experimental arrangement for investigation of BBO. The beam from the copper vapour laser is filtered by the pair of dichroic beamsplitters (dbs) to leave only the 510nm radiation, which is focussed by the lens of focal length f into the cell containing the BBO crystal. The emerging radiation is recollimated, and is dispersed by the prism before the ultraviolet power is measured using the radiometer.

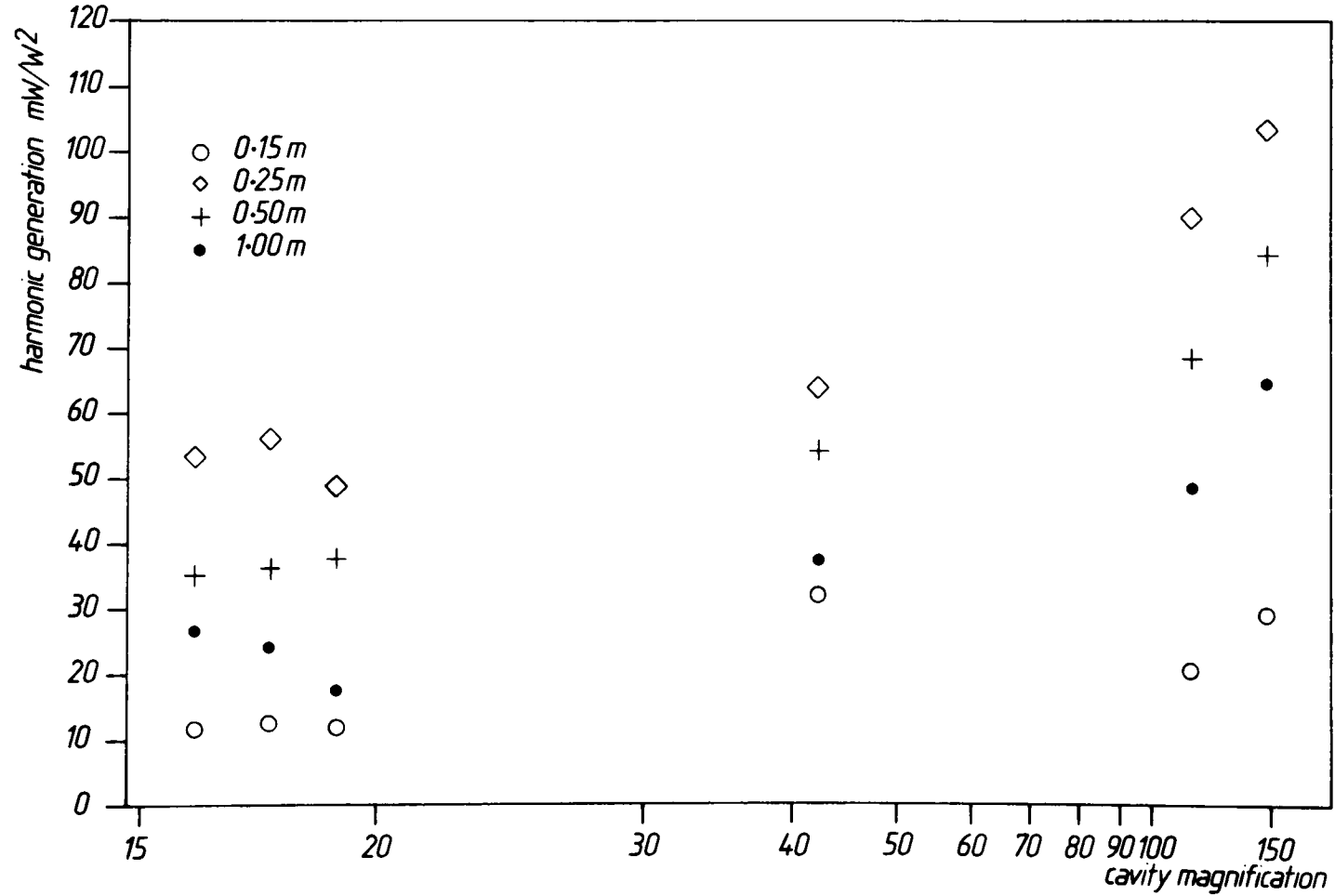


Figure VI-2
Harmonic generation of 255nm. The harmonic generation, the ratio of the harmonic power to the square of the fundamental, is shown as a function of cavity magnification for various strengths of focussing lens.

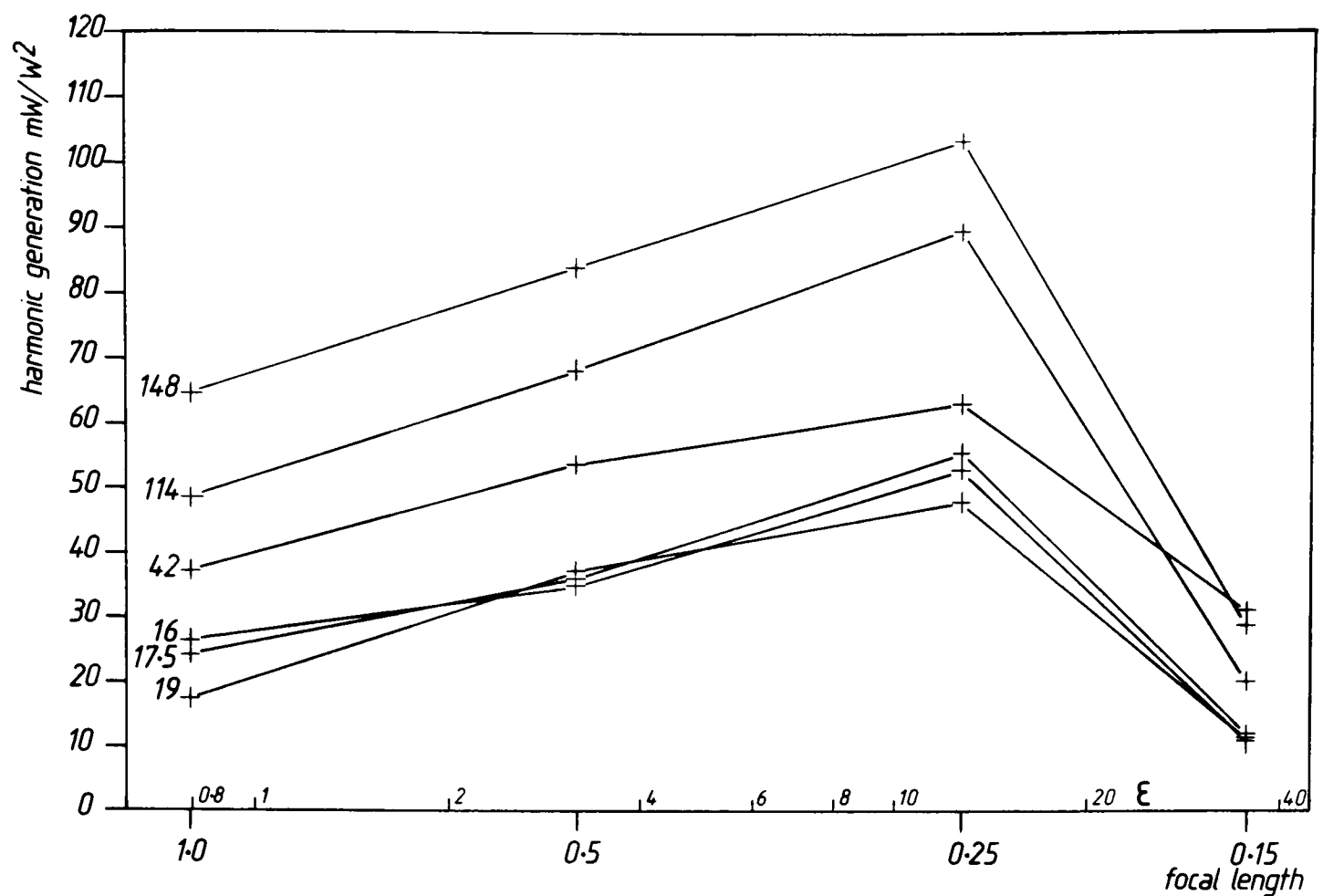


Figure VI-3
Harmonic generation of 255nm. The results of figure VI-2 are shown here as a function of the lens focal length. The secondary scale showing the parameter ϵ is based on a nominal laser beam radius of 10mm; in practice, occlusion of the laser aperture limits the vertical extent of the laser beam, and a different scale is found to be appropriate.

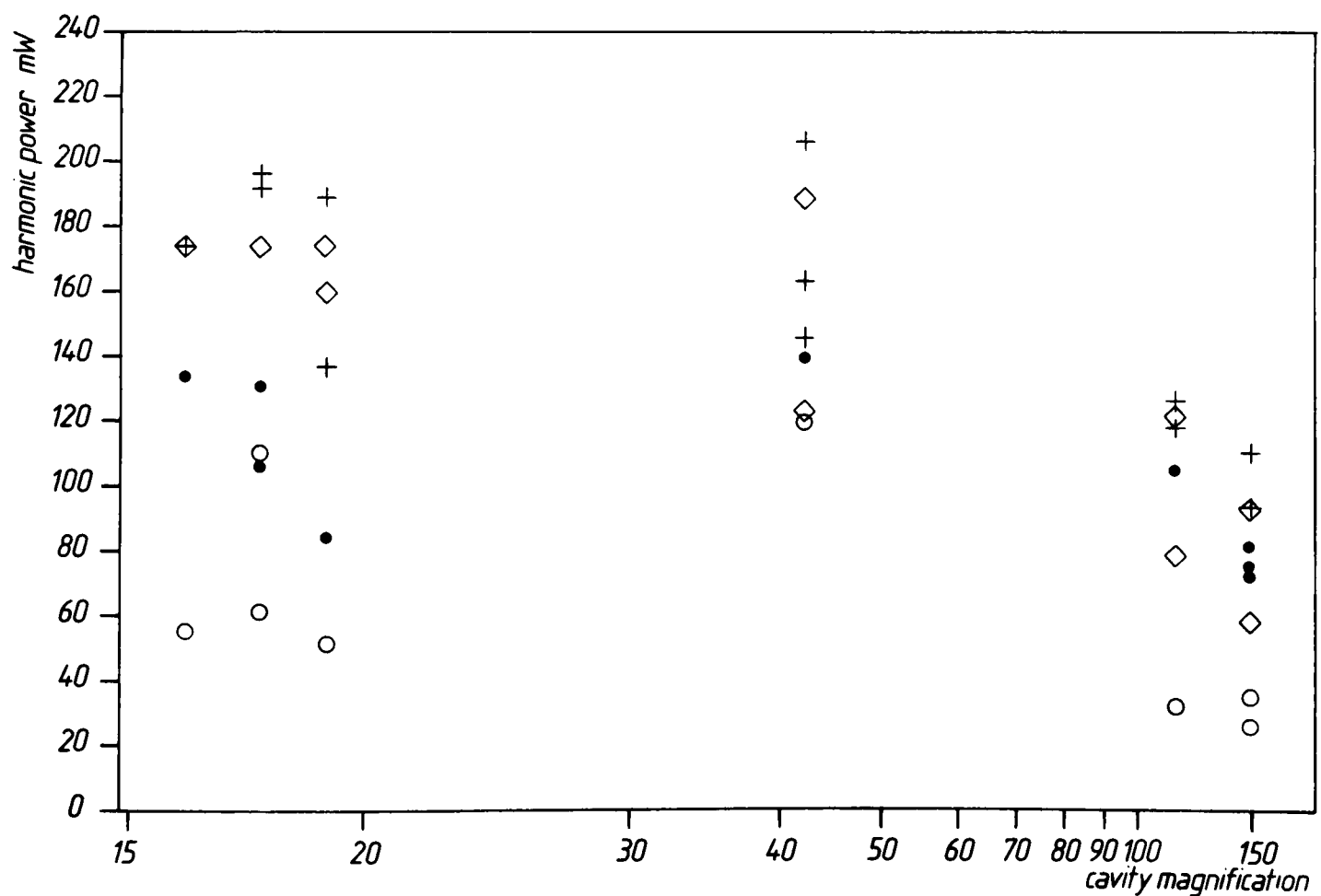


Figure VI-4
Achieved mean harmonic power, shown here as a function of cavity magnification for the range of lens focal lengths. As perfect alignment is difficult to achieve, it is the envelope of these points which is of interest.

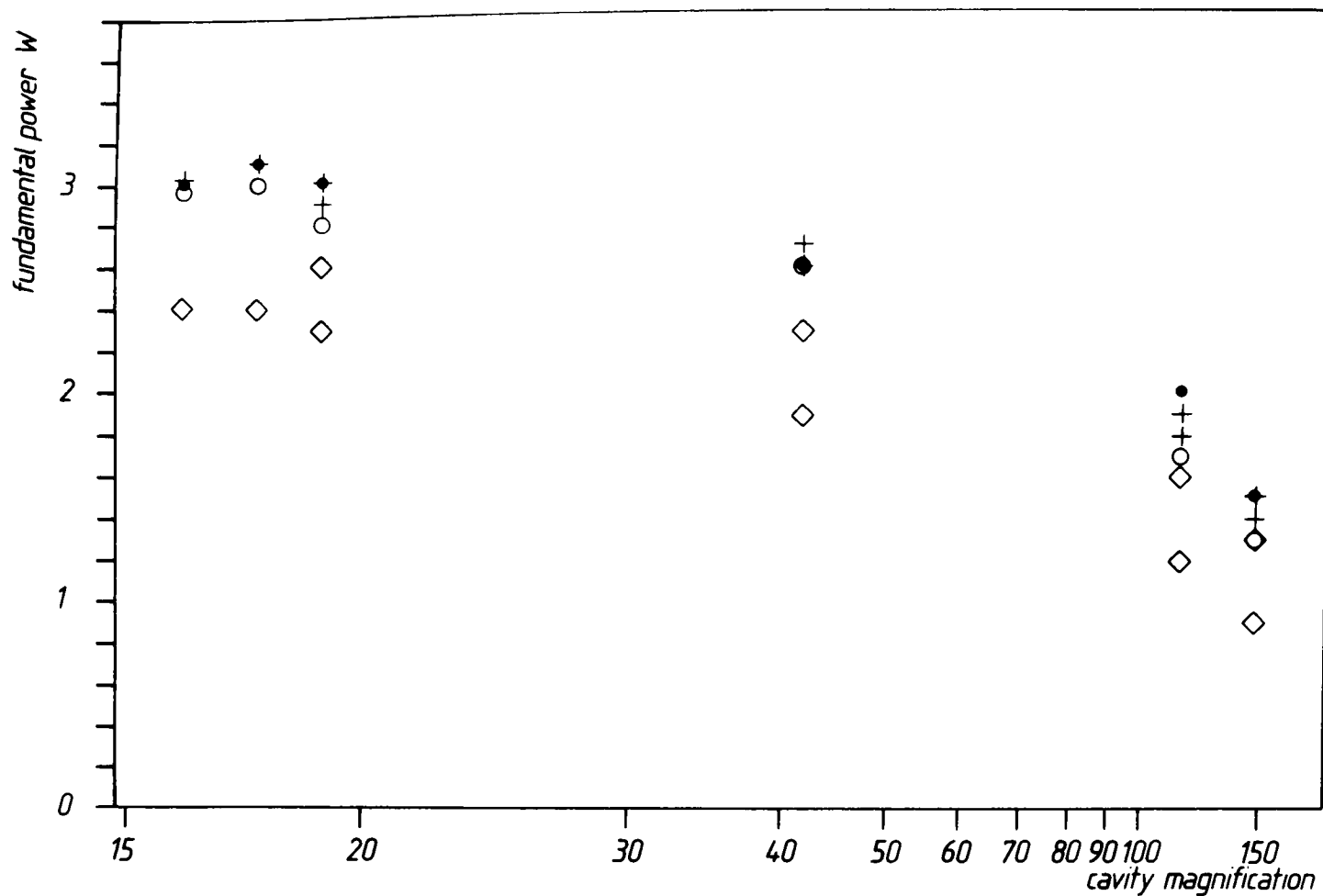


Figure VI-5
Mean fundamental power. An accompanying graph to figure VI-4. The peak laser power is in fact little altered by changing the unstable cavity, and it is primarily the variation in the length of the laser pulse which is seen.

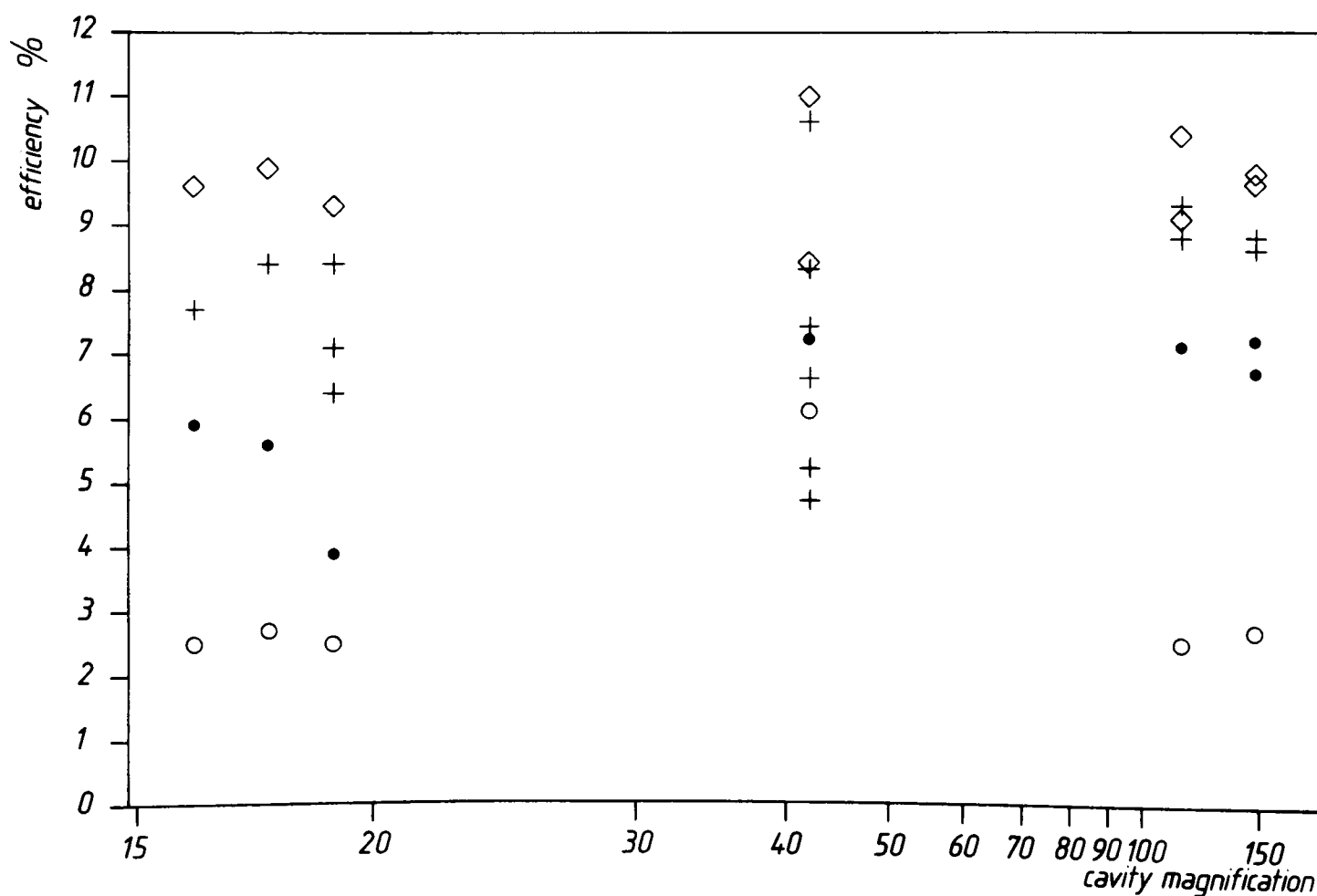


Figure VI-6
Efficiency of harmonic generation. The leading portion of the laser pulse makes little contribution to the second harmonic, and its removal results in an increase in the conversion efficiency. The efficiency is otherwise fairly constant, for the peak power is little affected by the unstable cavity. Nonetheless, there is some reduction in peak power with magnification, and to the right of the graph the efficiency does indeed decrease.

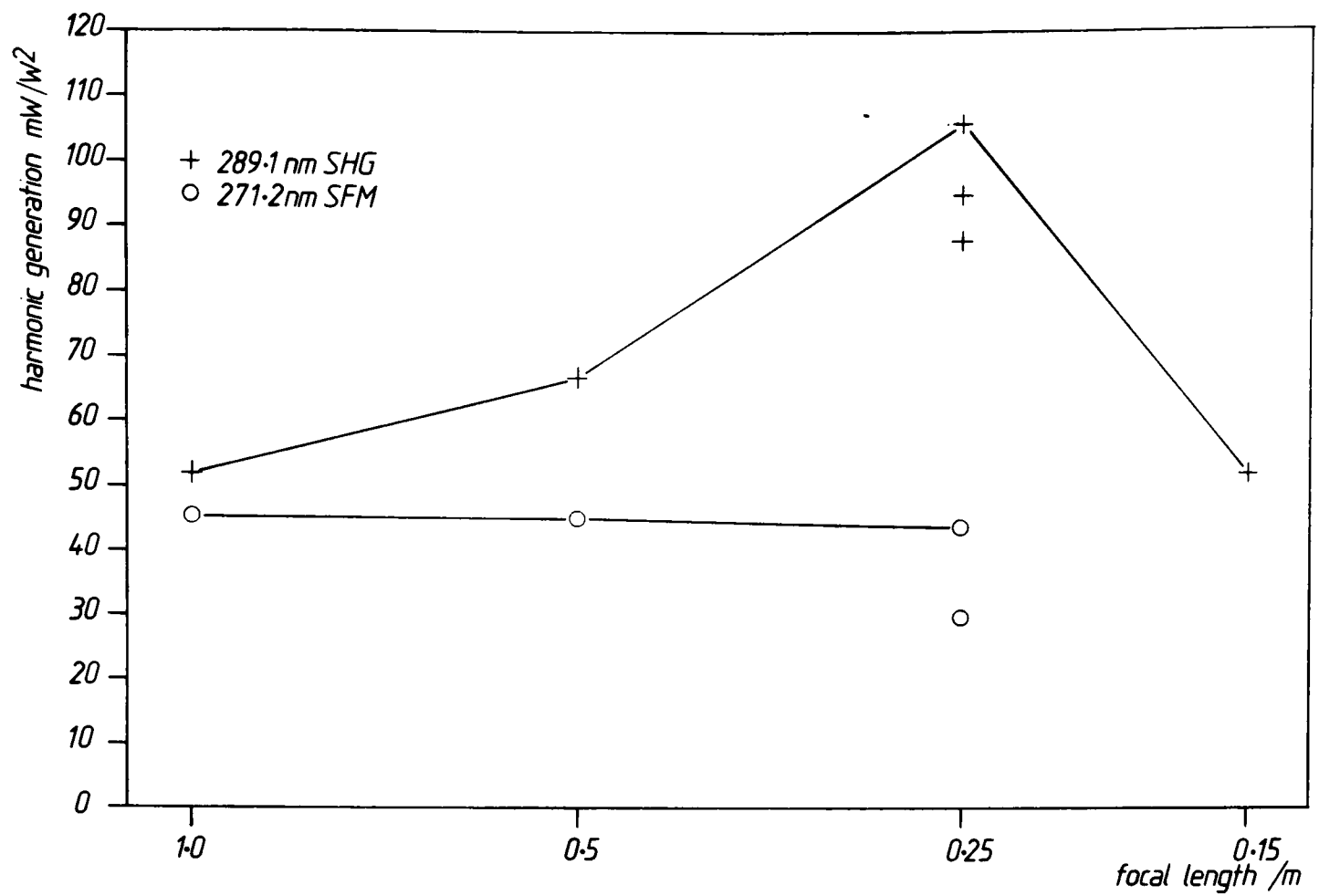


Figure VI-7
Harmonic generation of 289nm and 271nm by second harmonic generation from the CVL yellow line and sum frequency mixing of the pair of wavelengths. The behaviour of SHG follows that shown in figure VI-3 for the green line. Poor performance from SFM is attributed principally to dispersion in the optical components, leading to a spatial separation of the two wavelengths.

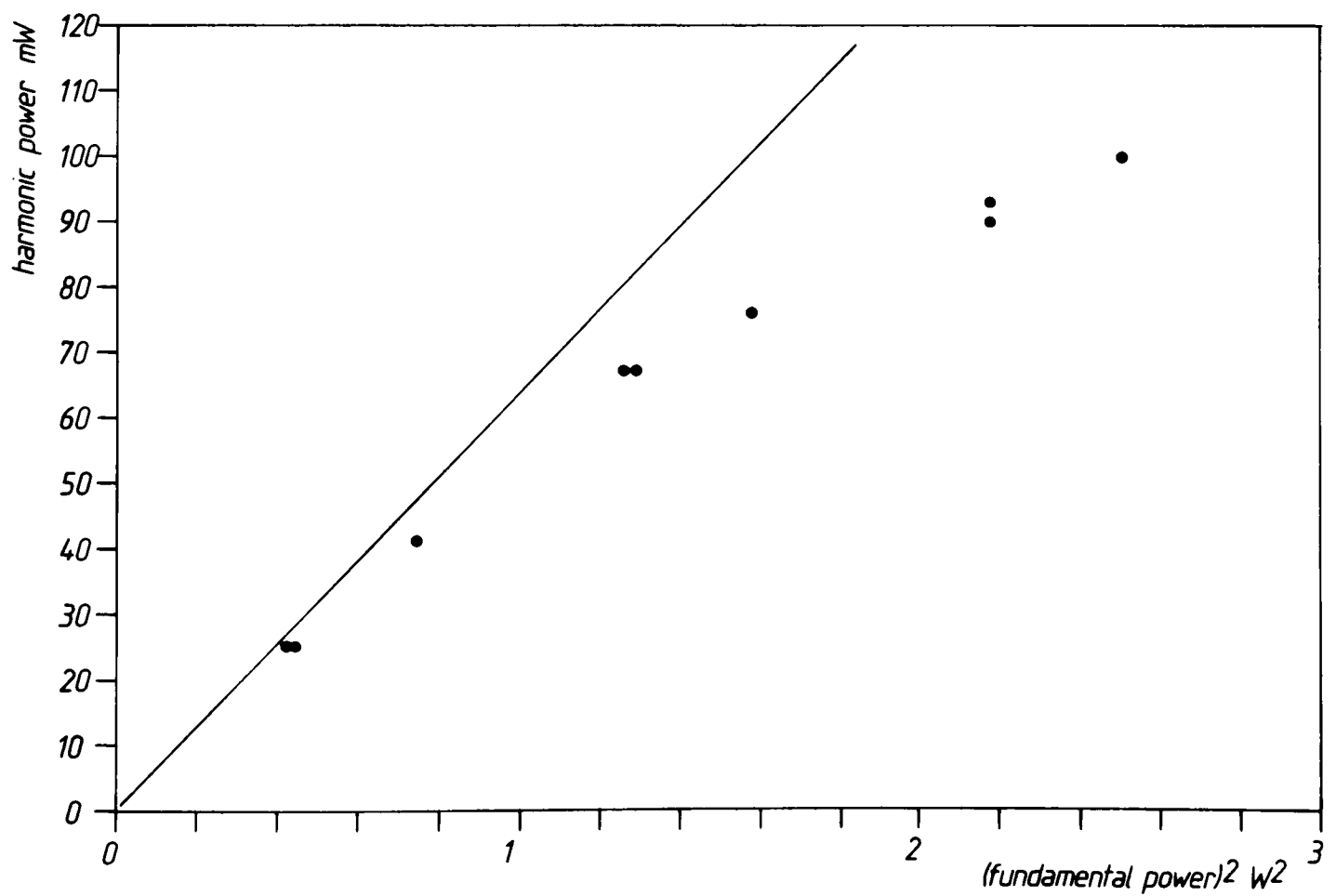


Figure VI-8
Deviation from low power behaviour. As the proportion of the fundamental converted to the harmonic increases, depletion of the fundamental becomes significant and the relation between fundamental and harmonic powers deviates from a quadratic.

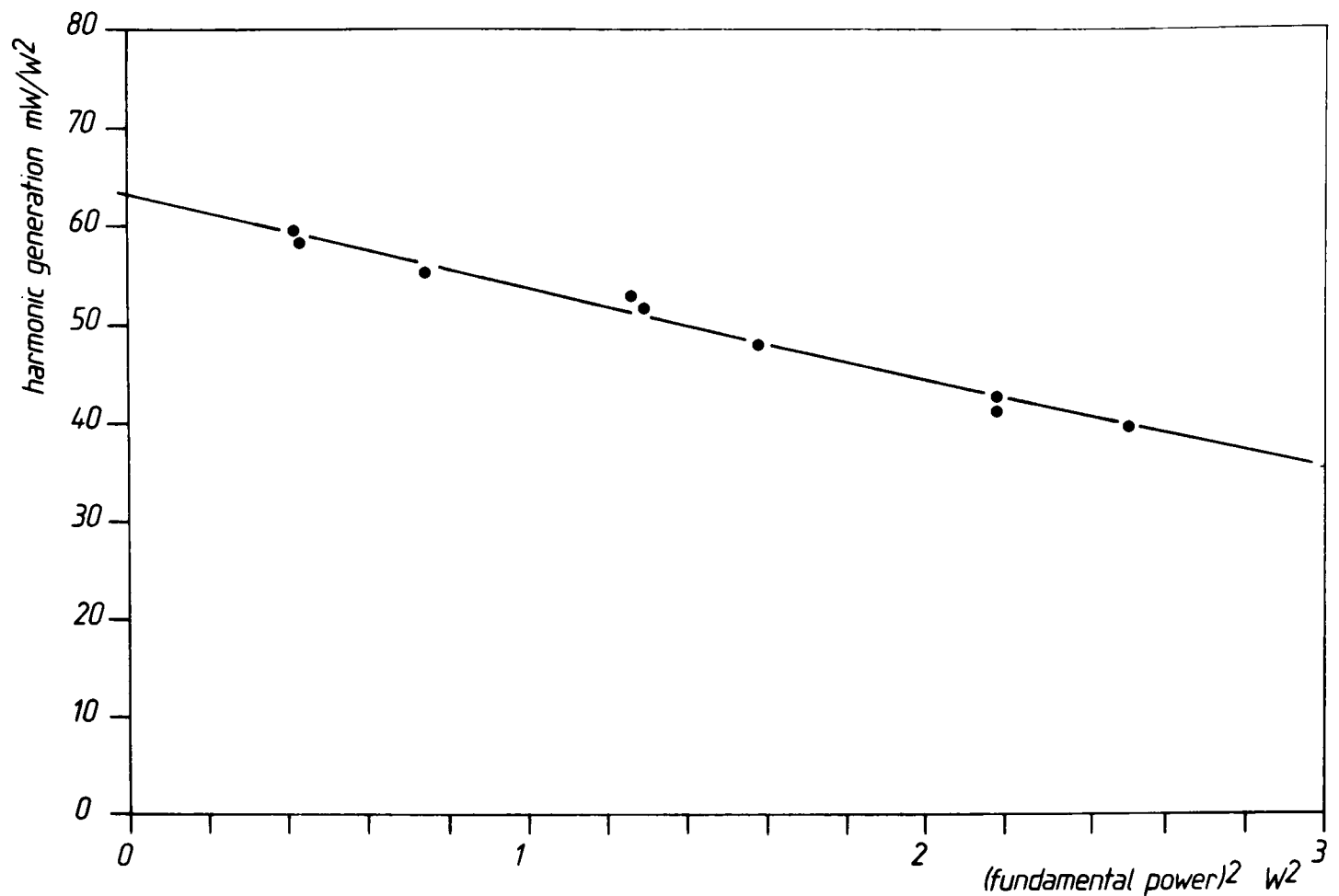


Figure VI-9

Deviation from low power behaviour: the results of figure VI-9 replotted as harmonic generation. The deviation from the low efficiency value is seen to vary with the square of the fundamental power. In the case of plane waves a linear dependence is predicted. Here focussed beams are used. Nonlinear absorption will also be manifest as a deviation from the low efficiency behaviour.

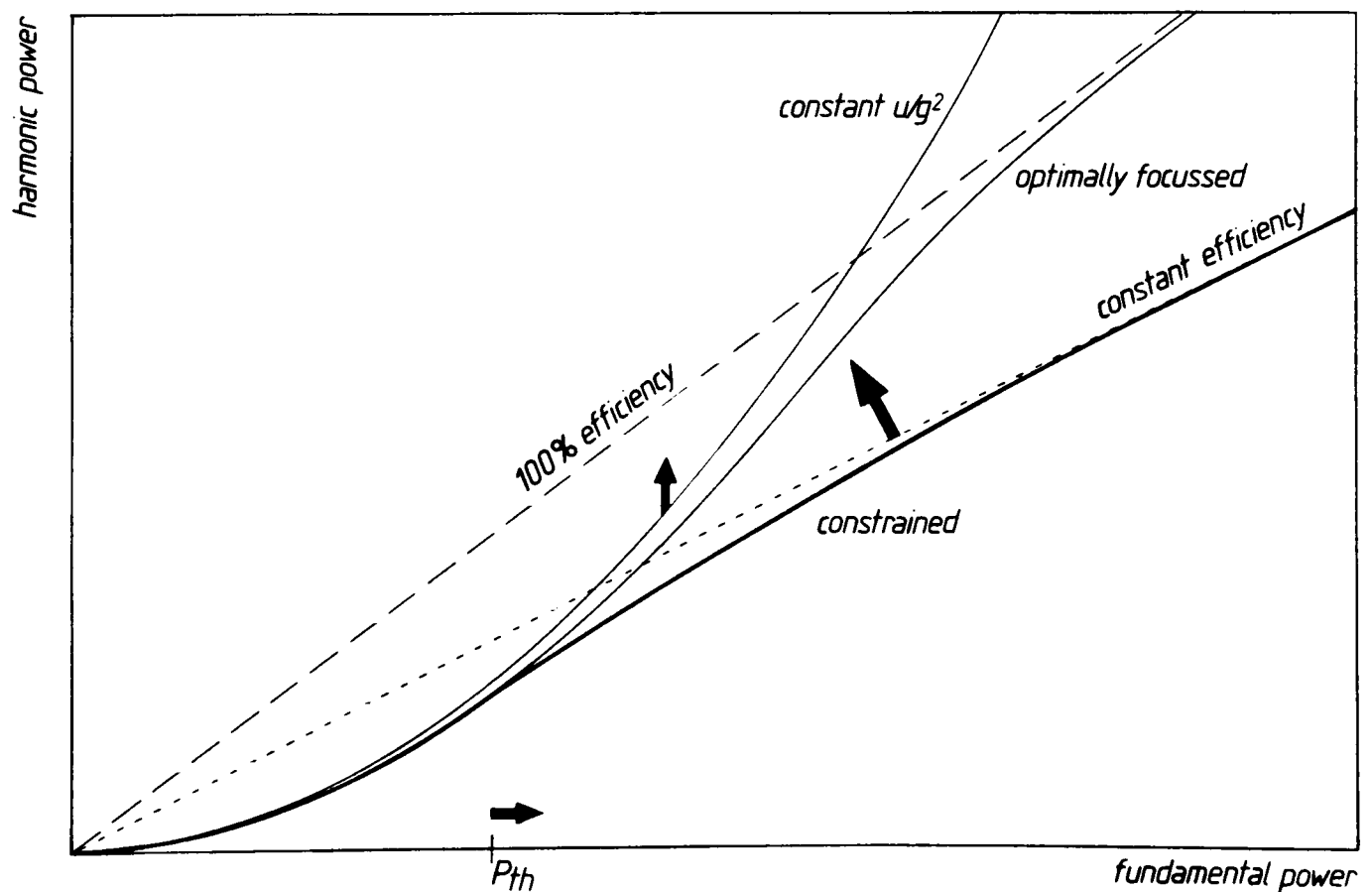
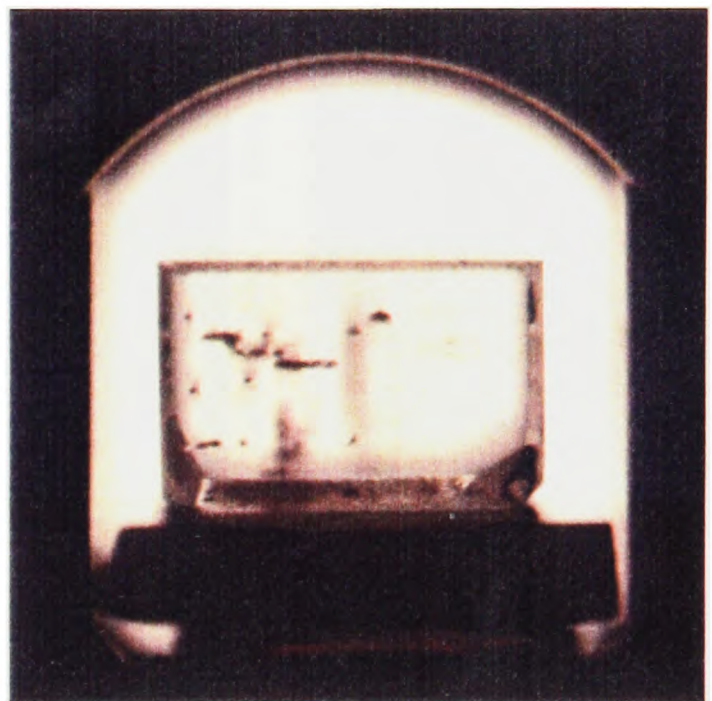
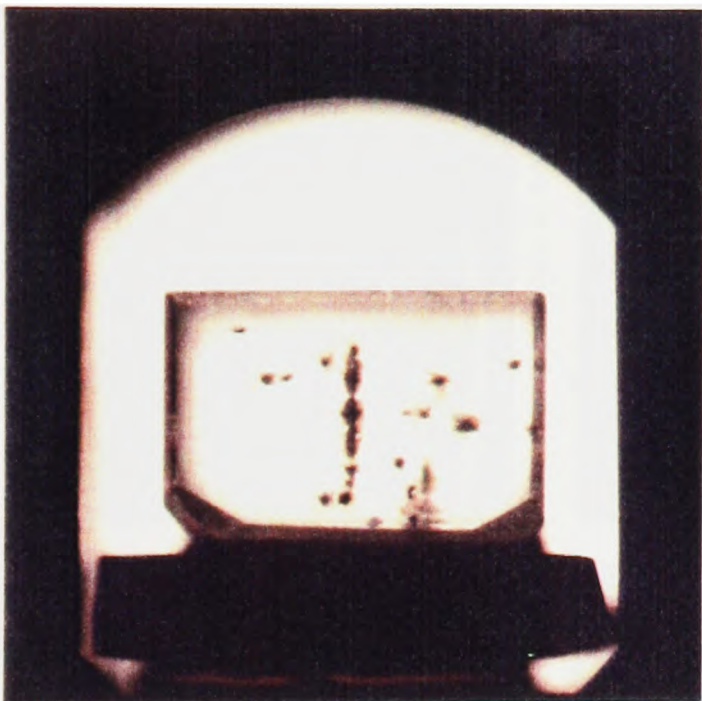


Figure VI-10

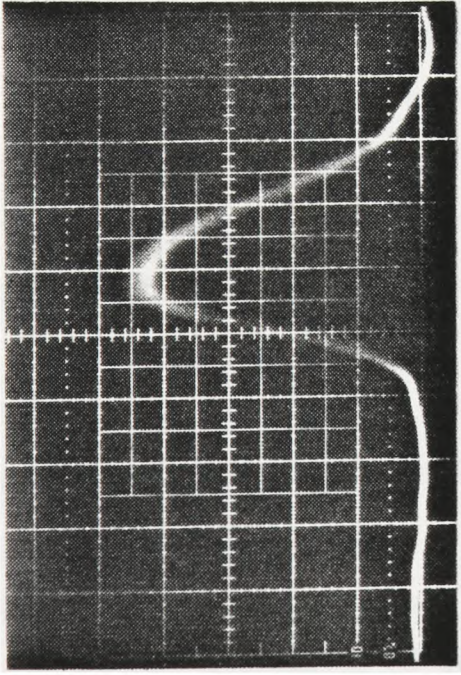
Schematic high power performance showing the predicted variation in harmonic power with fundamental power as the efficiency increases and the damage threshold is reached. Ultimately, the best efficiency approaches that for plane waves incident at just below the damage intensity for the material. In practice, high average laser powers may introduce a significant perturbation as thermal distortion of the beam path; this is not indicated on this diagram. The effect of increasing the crystal length is indicated by the bold arrows. P_{th} is the power which, when optimally focussed, corresponds to the crystal damage threshold intensity.



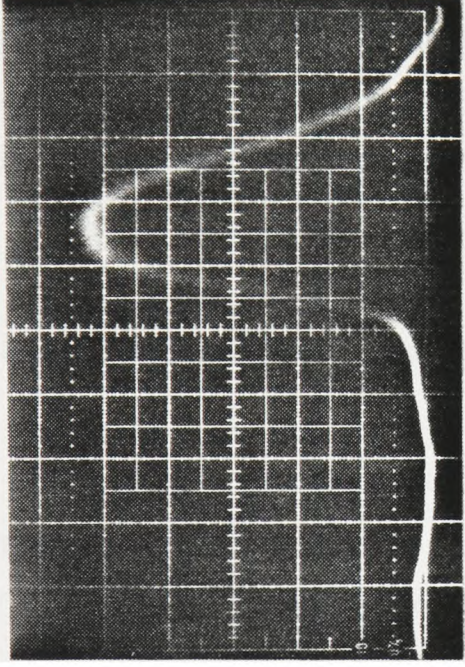
Plate VI-1. Dispersed harmonics. A piece of card in front of the power meter shows the fundamental beam and blue fluorescence from its second harmonic. By appropriate positioning of the recollimating lens, an image of the crystal is produced showing the path of the laser beam through the sample.



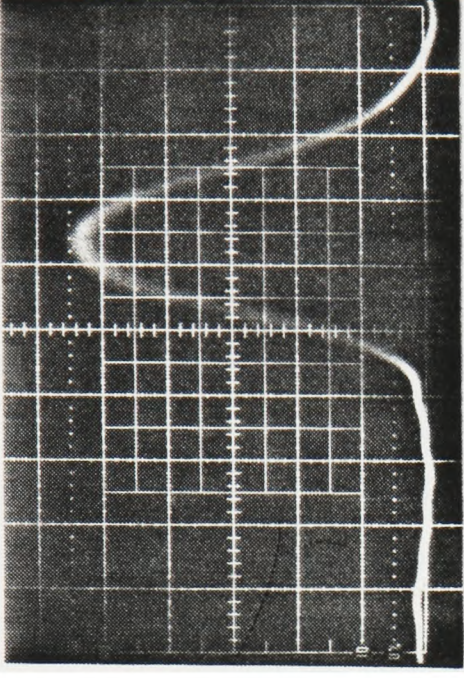
Plates VI-2. Laser damage to beta-barium borate at the input face (a, top left), exit face (b, above) and a view from the exit showing damage propagation within the crystal (c, left). The crystal measured $3.9 \times 5.7\text{mm}$, and was 7.2mm long.



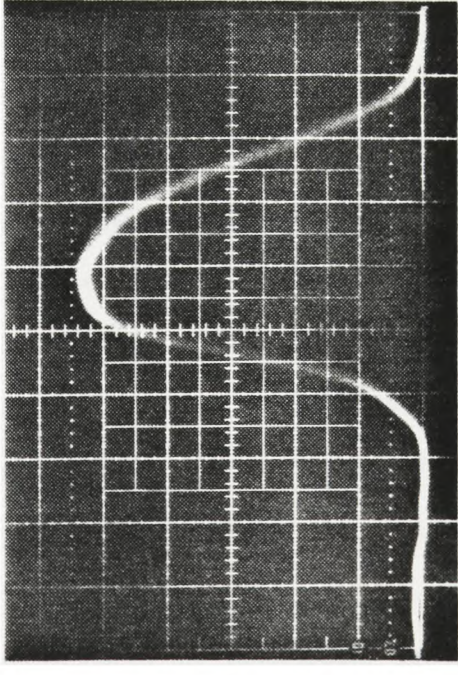
a 255nm, harmonic of 511nm copper laser line



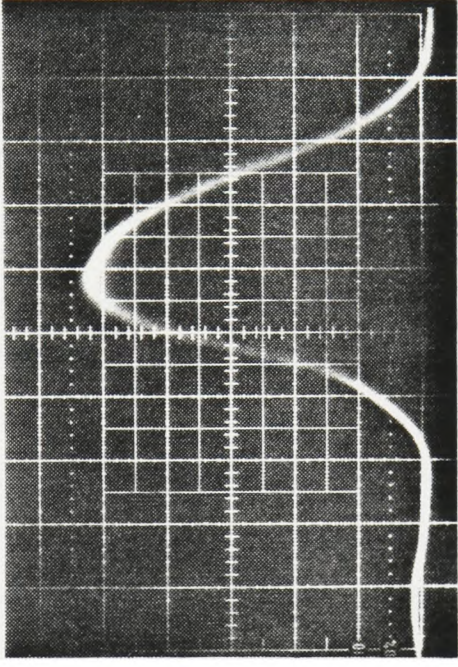
b 289nm, harmonic of 578nm copper laser line



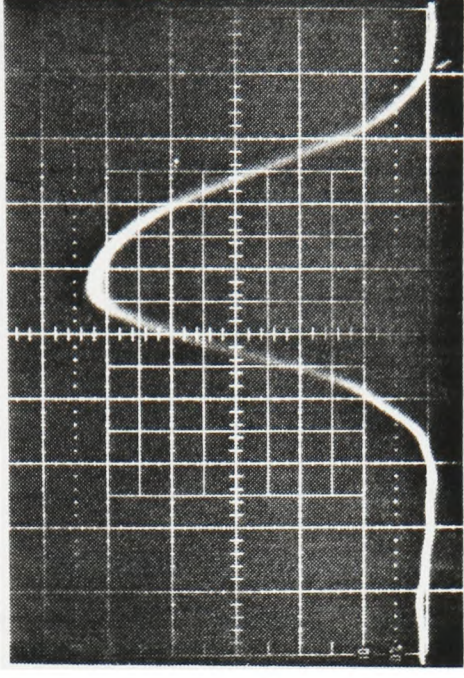
c 271nm, frequency sum of 511nm and 578nm



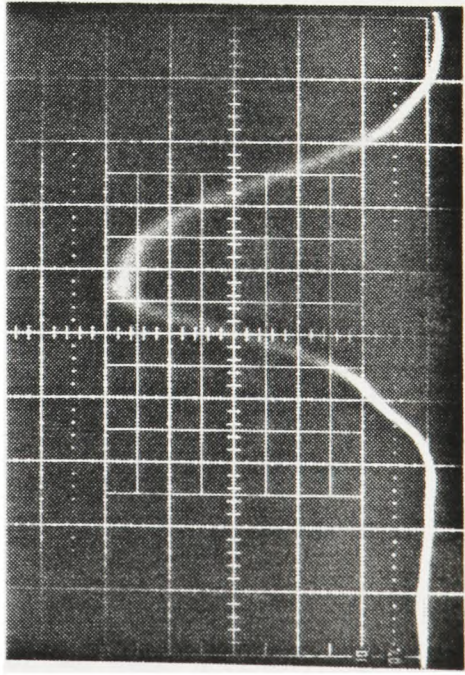
d combined 511nm and 578nm reference



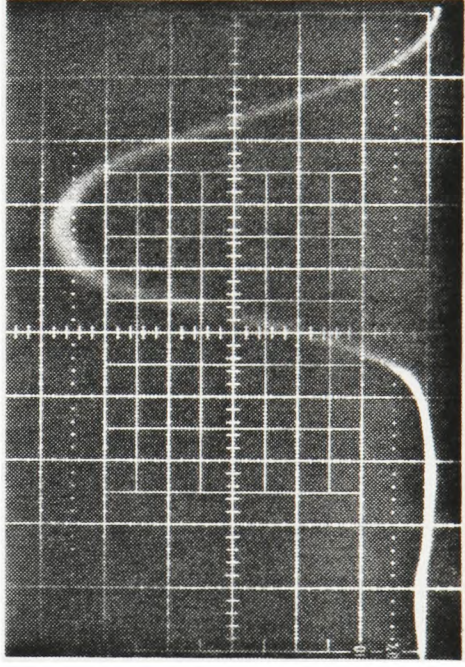
e combined 511nm and 578nm reference



f combined 511nm and 578nm reference



g green copper laser pulse only



h yellow copper laser pulse only

Plates VI-3. Temporal dependence of harmonic generation. Oscilloscope traces taken from a Tektronix model 466 at $50\text{mV.div}^{-1} \times 10\text{ns.div}^{-1}$, with a cavity magnification of 14.

All traces are intended to have the same timing relative to the laser trigger pulse, but traces d, e and f which immediately followed a, b and c respectively allow a direct comparison to be made. The green laser pulse g shows a slightly anomalous upper leading edge shape; further investigations show the leading edge of the combined green and yellow traces to be more representative.

little variation over the range of lens focal lengths investigated.

The oscilloscope traces of plate VI-3 allow the differences in temporal dependence of the various fundamental and ultraviolet beams to be determined. It is apparent that the green pulse precedes the yellow pulse by around 5ns, and is a little shorter in duration. The second harmonics fall in sympathy with their respective fundamentals, but are both delayed by some 5ns at the start. The green pulse shows a slightly anomalous upper leading edge shape; further investigations show the leading edge of the combined green and yellow to be quite representative.

For all the results presented so far, the laser was fitted with the foam beam path covers mentioned in chapter IV, which had been rendered more airtight by wrapping in thin plastic 'Cling-film'. Earlier experiments performed at the same time as those of chapter IV used the foam guards alone and some beam wander occurred, typically reducing the harmonic generation to half the values given here; the dependence upon focussing, however, was identical. It was predicted in chapter IV that the inclusion of the beam path covers would increase the mean harmonic generation by a factor of around 2.5, and that this would itself be only half the value appropriate to a stable beam; with the Cling-film wrapped around the foam a stable beam results, and both these factors have indeed been measured.

VI-2.4 Predicted harmonic generation.

The theory outlined in chapter V gives the instantaneous efficiency of second harmonic generation, for a 7mm crystal with circular Gaussian beams, as around $60 \mu\text{W.W}^{-2}$, the conversion being 5% higher at 578nm than it is at 510nm. At 6.4kHz, a rectangular pulse of width 32ns has a duty cycle of .0002; the peak power is thus 5,000 times the mean power, and the mean harmonic power will be around 300 mW.W^{-2} . The copper laser pulse is not rectangular, however, and for comparison we should consider a triangular pulse with the same energy and half-width; the conversion efficiency will be reduced by a factor of 2/3 to some 200 mW.W^{-2} . The copper laser pulse is somewhat flatter than a triangle, and the conversion efficiency for a Gaussian copper laser pulse should thus be somewhere between 200 and 300 mW.W^{-2} . At high powers, however, as the efficiency approaches 100%, significant depletion of the fundamental beam will occur, and the effectiveness of harmonic generation will be reduced: this sensibly limits the conversion efficiency to below 100%.

A proper evaluation of the reduction in harmonic generation at high powers with Gaussian focussing presents a complex problem and will not be attempted. However, it may be shown (see appendix VI-2) that in the case of harmonic generation with plane waves, the initial departure of the harmonic generation from its low power

behaviour takes the form

$$\frac{I(2\omega)}{I(\omega)^2} = \frac{I(2\omega)}{I(\omega)^2} \bigg|_0 \left[1 - \frac{2}{3} \frac{I(2\omega)}{I(\omega)^2} \bigg|_0 I(\omega) \right] \quad (\text{VI-1})$$

This equation strictly applies to instantaneous power: the same form holds for mean powers provided that the value 2/3 is multiplied by a factor

$$\frac{\int I(\omega) dt \int I(\omega)^3 dt}{\left[\int I(\omega)^2 dt \right]^2} \quad (\text{VI-2})$$

which is 1 for a rectangular pulse and 9/8 for a triangular pulse. We therefore predict a 10% departure from the low power limit at fundamental powers above 0.5–0.8W. It must be remembered, however, that this calculation is based upon the interaction of phase-matched plane waves; when perfect phase-matching does not occur, the behaviour may be expected to be different.

The size of the focussed copper laser beam at the focus will be related to the coherence area of the copper beam – the coherence function defines the proportion of the amplitude which is coherent, and it is thus each coherence area which propagates in a Gaussian fashion (this is the Van Cittert–Zernike theorem). For a given focussing arrangement, therefore, the effectiveness of harmonic generation will be that for a Gaussian beam characterised by the coherence area, scaled according to how many such beams lie within the acceptance angle of the crystal. Since the process of harmonic generation is nonlinear, it is the suitability of the appropriate Gaussian beam which will be the more important factor, and a coherence area which corresponds to a non-optimum Gaussian will not be converted efficiently.

It follows that if the transverse coherence evolves rapidly until it is limited by the laser tube dimensions – as is suggested in chapter IV – then the optimum focussing arrangement for harmonic generation using copper lasers will match only the near-diffraction-limited portion of the pulse with any efficiency. This enables both the temporal characteristics of the harmonic generation and its dependence upon the cavity magnification to be predicted, for the harmonic pulse will start only with the high coherence part of the pulse, and any energy in the low coherence portion will be wasted. At low cavity magnifications, the first 5ns or so of the pulse has poor spatial coherence; the harmonic will thus start around 5–10ns after the fundamental, and there will be a corresponding reduction in the harmonic generation u/g^2 . With higher cavity magnifications, even the early part of the pulse has good coherence, and thus the whole of the pulse will be used.

The theory of chapter IV predicts that the highest conversion efficiency for beta barium borate will occur with a value of epsilon around 1.4, and thus for a 7mm crystal with a beam radius w_0 of around $16\mu\text{m}$. For a Gaussian beam, the 1/e amplitude radius w_0 is related to the radius, w_0 , at the focussing lens by

$$w_1 = f\lambda/\pi w_0 = 2 f/kw_0 \quad (\text{VI-3})$$

and with a circular aperture of radius a the corresponding expression is

$$a = 2.58f/kw_0 \quad (\text{VI-4})$$

For a circular beam of radius 10mm, then, the optimum focal length will be around 750mm.

The focussing parameter ϵ , $= 1/w_0^2 k$, may thus be written as

$$\epsilon = a^2 k / (2.58)^2 f^2 \quad (\text{VI-5})$$

This expression has been used with the rather arbitrary value $a=10\text{mm}$ to define the secondary scale of figure VI-3.

VI-2.5 Generation of ultraviolet radiation at 255nm.

Achieved power and optimum focussing conditions

The predicted harmonic conversion for a Gaussian beam with a temporal profile similar to that of a copper vapour laser is $200\text{--}300\text{mW.W}^{-2}$; the effect of reflective losses in the crystal cell is to reduce this to between 100 and 200mW.W^{-2} . Given that crystal absorption has not been taken into account, the best measured harmonic conversion around 100mW.W^{-2} is quite consistent with the predicted values.

The agreement between the calculated and measured optimum focussing conditions appears to be rather poorer, with $\epsilon=1.4$ and $\epsilon=8$ respectively. However, the ϵ axis of figure VI-3 takes its calibration from a nominal laser beam radius of 10mm, and whilst at the focussing lens the beam indeed had a horizontal diameter of around 18mm, it was severely obscured in the vertical dimension by the combined effects of molten copper and sagging of the tube. The central height of the beam was thus limited to around 12mm. It is indeed the vertical dimension which is appropriate to the focussing of the laser beam for second harmonic generation, for it is in this direction that the phase-matching varies. There is in effect a small degree of ellipticity in the focussing of the laser beam; this ellipticity, around 1.5, is well below the value recommended in chapter V for a crystal with $B=16$, and the entire width of the beam should thus be useful. The focussing dependence will therefore be upon the vertical dimensions. When recalculated accordingly, the peak achieved harmonic power was found to correspond to ϵ between 1.15 and 4.6: this is quite consistent with the predictions of the previous chapter.

The extent to which the measured harmonic generation and the predicted performance are consistent suggests that most of the copper laser beam contributes usefully to the harmonic power, and thus has a high degree of coherence. This supports the belief, expressed in chapter IV, that the copper laser reaches diffraction limited performance on a single shot basis.

Accuracy of the quadratic dependence.

It has been assumed so far that a low efficiency regime applies, in which the fundamental beam is not significantly attenuated by passage through the crystal, and that the harmonic power thus varies in quadratic relation to the fundamental. Some departure from this behaviour must occur when a significant proportion of the fundamental radiation is converted to the harmonic, however, if only to limit conversion efficiencies to below 100%. To investigate the validity of this assumption the fundamental beam was attenuated with a range of reflective neutral density filters and the effect upon the harmonic power was recorded. The variation of harmonic power with fundamental is illustrated in figure VI-8, and the same results are shown as u/g^2 in figure VI-9; it is from the low power limit of figure VI-9 that the straight line of figure VI-8 has been derived.

There is certainly a departure from quadratic behaviour at the higher conversion efficiencies, and the 10% departure from the low power limit indeed occurs at the top of the predicted range 0.5–0.8W. However, the dependence of the harmonic generation upon fundamental power appears to follow a quadratic relation, rather than the linear form predicted. This discrepancy could be due to the use in practice of focussed beams with birefringence but figure VI-8 will also be sensitive to nonlinear absorption within the crystal, and in many materials the primary mechanism for attenuation of ultraviolet beams is two photon absorption.

Over the range of experimental parameters reported here, the deviation from quadratic behaviour never exceeds 30%, and the harmonic generation u/g^2 is thus a useful and valid measure of the effectiveness of SHG. Nonetheless, this phenomenon will be shown to be important in a complete interpretation of the results of this chapter, and its extension beyond this range will be of value in the consideration of higher power systems.

Temporal dependence

The oscilloscope traces of plate VI-3 show the temporal characteristics of the fundamental and harmonic beams for the best conversion with a cavity magnification of 14. Under these conditions the u/g^2 ratio, for conversion of the 510nm laser line, is around 55mW.W⁻², which is half of that obtained with higher cavity magnifications.

It has been predicted that only the part of the pulse with a high coherence radius will be converted to the second harmonic with any efficiency, and the measurements of chapter IV show that at these low cavity magnifications this condition is not satisfied until at least 5ns after the start of the pulse. A similar delay is thus expected between the start of the fundamental and that of the harmonic, and this is indeed apparent when photographs VI-2a and VI-2d are compared. Perhaps 15–20% of the pulse energy is contained within this leading section of the fundamental pulse: this will reduce the u/g^2 ratio by at least a factor of 1.3 below that measured for the

higher magnifications. The departure from quadratic behaviour of the harmonic generation process suggests that a change from a useful fundamental power of around 1W (typical of experiments with high cavity magnifications) to around 2W (at lower cavity magnifications) will further reduce the ratio u/g^2 by a factor of around 1.4. The harmonic generation values for high and low magnification cavities should thus differ by a factor of 1.8.

Figure VI-2 shows that the harmonic generation with high cavity magnifications is indeed nearly twice the value measured at low magnifications. To within the precision of these calculations, the variation in u/g^2 with magnification is completely accounted for.

VI-2.6 Doubling the yellow and mixing the green and yellow.

Figure VI-7 shows the variation in performance of sum frequency mixing of the two copper laser lines and second harmonic generation from the yellow. The mean fundamental powers for the green and yellow lines throughout were around 1.8W and 1.0W respectively. The ratio u/g^2 is again used to characterize second harmonic generation, and is replaced by u/g_1g_2 for sum frequency mixing where g_1 and g_2 are the powers of the two mixed wavelengths. The highest conversion achieved to 289nm was 77mW.W^{-2} ; the dependence upon focal length is the same as is shown in figure VI-3 for generation of 255nm radiation, for which the corresponding value was around 55mW.W^{-2} at similar fundamental powers.

With the cavity magnification of 14 used here, then, the harmonic generation of the yellow copper laser line is larger than that of the green line by a factor of 1.4. This is primarily due to the better spatial coherence of the yellow line, as demonstrated by the time-averaged measurements reported in chapter IV, and confirmed by the timing in the photographs of plate VI-3. The performance of harmonic generation from the two wavelengths may therefore be expected to converge at higher magnifications as both assume a high beam quality throughout the pulse. The different pulse lengths will, however, cause some difference to remain and the shorter green laser pulse is likely to be better in this case.

Sum frequency mixing of the green and yellow laser lines gave rather poorer results. This may be attributed to dispersion in the focussing lens, resulting in a displacement of the focal regions of the two fundamental wavelengths, and to a temporal separation between the green and yellow pulses. The scale of the spatial separation will depend upon the focal length and material of the lens, and it is perhaps for this reason that the dependence of sum frequency mixing upon focal length is rather different from that of second harmonic generation.

VI-2.7 Confirmation of phase matching angles.

The phase matching angles for SHG of 255 and 289nm may be predicted using the theory of chapter IV with published Sellmeier coefficients. Using data from various sources, the following angles are derived:

<u>Table VI-1.</u>	Predicted phase-matching angles		
reference	SHG(255.3)	SHG(289.1)	difference
Chen[VI-2]	50.19	41.23	8.96
Chen[VI-9]	50.69	41.73	8.96
Eimerl[VI-8]	50.46	42.23	8.23
Kato[VI-10]	50.65	42.46	8.19

The crystal used for these experiments was cut for normal incidence to give a nominal phase-matching angle of 45°. The measured angles relative to normal incidence for the three ultraviolet generation processes studied were as listed below. Beta barium borate has a refractive index which varies from 1.677 at the green wavelength to 1.671 for the yellow; these refractive indices have been used to derive the internal angles of propagation for phase matching.

<u>Table VI-2.</u>	Measured phase-matching angles relative to normal to polished face		
	absolute	incidence	internal
255nm	- 3.0	- 2.5	- 1.5
SFM	+ 4.3	+ 4.8	+ 2.9
289nm	+10.5	+11.0	+ 6.6
normal	- 0.5		

The measurement precision of these angles was 0.1°. The interval between green and yellow phase matching angles is thus measured, using published refractive index data, to be 8.1(1)°, which is consistent with the phase matching angles calculated from the data of Eimerl [VI-8] and Kato [VI-10] and at variance with the results of Chen [VI-2, VI-9]. The implication is that true normal incidence angle for the sample was around 49.0°.

VI-2.8 Thermal effects and laser damage.

At no time during the course of these experiments has there been any indication of thermal perturbations upon the process of harmonic generation. Chen [VI-11] has reported a temperature bandwidth for second harmonic generation using BBO at 1064nm of 55 C, and certainly the measured harmonic powers reported here have been sustained for periods of hours without significant variation.

The photographs of plate VI-2 show laser damage to the crystal. Degradation of both input and output faces is apparent, and filaments within the crystal can be seen to connect with damage to the output face. Marking of the input face can only have been caused by the fundamental, and the extended nature of this damage illustrates the way that laser burning can be 'dragged' along the surface when the beam is moved slowly across the crystal face from an initial area of increased absorption, such as a crystal defect or burnt area. It is for this reason that initial alignment is best carried out at low powers.

The damage to the output face and connected bulk damage could have been caused by either the fundamental or the harmonic beams, but the similarity with the front surface burns suggests that again it is the fundamental which is responsible. The filamentary marks will almost certainly have been caused by the propagation upstream of initial damage at the output face, due to the increased absorption of the burnt areas. Degradation of the output face is a little less likely than that of the input face, for the fundamental will be attenuated to some extent by the crystal absorption and, under phase matching conditions, second harmonic generation. Nonetheless, bulk damage is permanent and, unlike front surface burning, the output surface damage cannot simply be polished away.

The star-like damage apparent at the bottom-left of the input face (plate VI-2a) is an example of damage which highlights the cleavage planes of the crystal, as reported by Nakatani et al [VI-12]. Some marks may be seen entirely isolated within the bulk of the crystal; it is possible that this was present originally as a crystal defect.

In general, no damage was sustained to the crystal after the focussing had been optimized. Nonetheless, damage could occur for quite small misalignments when the 0.15m lens was used with the highest laser powers. The combination of tight focussing, high laser powers, and misalignment suggests that surface damage occurs when the corresponding peak focal intensity, of order 4GW.cm^{-2} , is achieved at the crystal surface.

Recently published measurements of the multiple-shot damage threshold of BBO by 8ns pulses at 532nm from a doubled Nd:YAG laser [VI-12] show that the crystal damage is probabilistic in nature: a crystal is likely to survive a single pulse provided that the intensity is below 50GW.cm^{-2} but will probably be damaged by a thousand shots unless the intensity is kept below 30GW.cm^{-2} . Unfortunately, these experiments were limited to only 1800 shots, which corresponds to around a third of a second in front of a copper laser. Nakatani comments that the surface damage threshold lies perhaps an order of magnitude below the bulk damage values given above, and also that the damage tends to run along the crystal mirror planes. The damage threshold intensity for copper laser radiation appears to be around 4GW.cm^{-2} , which is rather lower than the range of values given by Nakatani; however, the majority of the

copper laser damage appears to have been to the crystal surfaces, and far higher numbers of pulses will in general have been accrued before the damage was incurred.

VI-2.9 Developments elsewhere.

Beta barium borate possesses a quite remarkable combination of properties for harmonic generation: its wide tuning range and transmission into the far ultraviolet allow phasematched harmonic generation of wavelengths as short as 200nm, and high nonlinear coefficients lead to good conversion efficiency at the high intensities afforded by excellent thermal and mechanical qualities. In the generation of short wavelength radiation alone it is quite unique, and indeed, the phase-matching angle at these wavelengths approaches 90°. It has even been considered worth-while extending this range by thermal tuning, by cooling the crystal to cryogenic temperatures.

The first reported use of beta barium borate to generate harmonics of the radiation from a copper laser [VI-2] highlighted the potential of the material for use at powers as low as a few watts. Zhang et al reported that, with apparatus similar to that described in this chapter, the harmonic generation u/g^2 for the green and yellow copper laser lines were 21 and 44 mW.W^{-2} respectively, and the achieved conversion efficiencies were below 1% only because the lasers used were particularly feeble. A small copper chloride laser, producing 0.5W of green or a little over 100mW of yellow light at some 16kHz, was operated with an unstable cavity of magnification 7.5 and used with a 5.5mm long crystal of BBO. The harmonic generation values are around three times smaller than are reported in this chapter for a longer crystal and side-spot unstable cavity of magnification 14; the results therefore appear consistent, and there is no doubt that Zhang would have found improved performance from a better copper laser.

More recent work in Japan by Kuroda et al [VI-5, VI-6] has demonstrated conversion of 2.55W of green radiation from a copper vapour laser with unstable cavity magnification of 60 to give 226mW at the second harmonic. This corresponds to a harmonic generation of 35 mW.W^{-2} and a conversion efficiency of 8.9%: values which are a little below those that the findings of this chapter might predict, but which are nonetheless essentially consistent given that Kuroda's full laser parameters are not known. The work presented in this thesis, then, supported by the published results of Zhang and Kuroda, appears to provide a coherent description of the use of BBO with copper vapour lasers.

Very recently, Piper [VI-7], using a 6.8W copper vapour laser with a $\times 16$ unstable cavity, has generated 100mW of the second harmonic of the copper green line with an efficiency of 1.5%. Sum frequency generation is claimed to be superior to SHG of either copper laser line, with efficiencies above 5% being possible.

However, Piper's efficiencies of SHG are substantially lower than would be expected while his frequency mixing, at several times the fundamental power, is only twice as efficient as is reported in this chapter. It is likely that Piper's was a wide bore laser and that the low magnification unstable cavity had a central reflecting spot: this would lead to particularly poor transverse coherence, which would be exacerbated by any thermal beam distortions. The true reasons for Piper's poor performance are not clear, but such results do not contradict the conclusions of section VI-2.8 regarding the relative merits of SFM and SHG.

Since the conclusion of the work described in this chapter, experiments have been conducted by Graham Naylor of Oxford Lasers Ltd. to investigate harmonic generation of light from copper vapour lasers of rather higher powers. The best arrangement yielded 2.25W of ultraviolet radiation at 255nm from 33W of green fundamental produced by an oscillator-power amplifier arrangement; this corresponds to an efficiency of 7% and a harmonic generation value of 2.1mW.W^{-2} . The threat of crystal damage limited the range of usable focussing parameters and prohibited the use of optimum conditions, and beam wander was found to be a particularly serious problem. In addition, some retuning of the crystal was necessary after a period of running, indicating a significant increase in the crystal temperature. Experiments with 25W of green light from an injection controlled 100W CVL suffered badly from similar problems and yielded at best some 400mW of ultraviolet. These results are in general reminiscent of those reported recently by Piper [VI-7].

Thermal beam distortion is a significant problem with the higher power lasers which use large beam diameters, for convection occurs within the beam path even if effective beam guards have been fitted. The convection is driven largely by the hot laser windows, and investigations are being made to try to reduce these effects. The gross thermal detuning of the crystal further suggests that there may be some thermal distortions introduced within the crystal itself, although any such effect has so far been hidden by the wander of the fundamental beam. The limit upon the strength of focussing which is imposed by the crystal damage threshold prevents optimum focussing conditions being used and thus reduces the harmonic generation which may be achieved. At the time of writing, no serious attempt has been made to optimize the harmonic generation under these conditions.

VI-3. Second harmonic generation in temperature-tuned ADP

VI-3.1 Apparatus.

The experimental arrangement for the studies of ADP was identical to that used for BBO, except that the crystal was enclosed in a temperature controlled housing to allow temperature-tuned non-critical phase matching. ADP is predicted to be phase matched for frequency doubling of the copper laser green line at around -30°C ; we note that the equivalent condition for the yellow line may be achieved with ammonium dihydrogen arsenate (ADA) at some 20°C .

A schematic diagram of the temperature controlled housing constructed for these experiments is shown in figure VI-11. The crystal, which measures some 18.4mm long and which was cut for 90° phasematching, was supported in a brass mounting, which was attached to the cold face of a Peltier effect cooler, the hot side of which adjoined a water cooled brass block. The Peltier cooler, crystal and a platinum resistance thermometer to monitor the crystal mount temperature were enclosed within a vacuum jacket, sealed by quartz windows. The vacuum provided thermal isolation of the cooled mount and ensured that no water vapour was present which might be absorbed by or condense upon the hygroscopic crystal.

Control and stabilization of the crystal mount temperature was achieved under computer control, using the SC84 microcomputer and a specially developed program (LaSCAMP). This used the low frequency input and output interface to monitor the platinum resistance thermometer and calculate the required power to be supplied to the Peltier cooler. The program was mainly written in machine level code and used the computer's interrupt structure to provide correct timing and to allow the program's execution to be essentially transparent to other software. A small piece of FORTRAN code was included, however, to contain a three term (differential, integral, proportional) algorithm for temperature control and calibration. This FORTRAN code may be altered and is readily linked to the rest of the program when the calibration or algorithm are to be modified.

The LFIO interface provided a digital output of the required Peltier power, and this was connected to a specially made Peltier controller, illustrated in plate VI-4, which contains a pulse-width modulated high current power supply. The computer, software and controller were all capable of operating two independent crystal cells, and could provide if required some heating of the crystal by driving current through the Peltier cooler in the reverse direction, thereby using it as a heat pump.

Temperature calibration was achieved by calibrating the computer analogue interface against a resistance box and assuming the carefully determined calibration of the resistance thermometer to be valid with the three terminal connection used here (there being insufficient space in the cell for full four terminal wiring). Overall

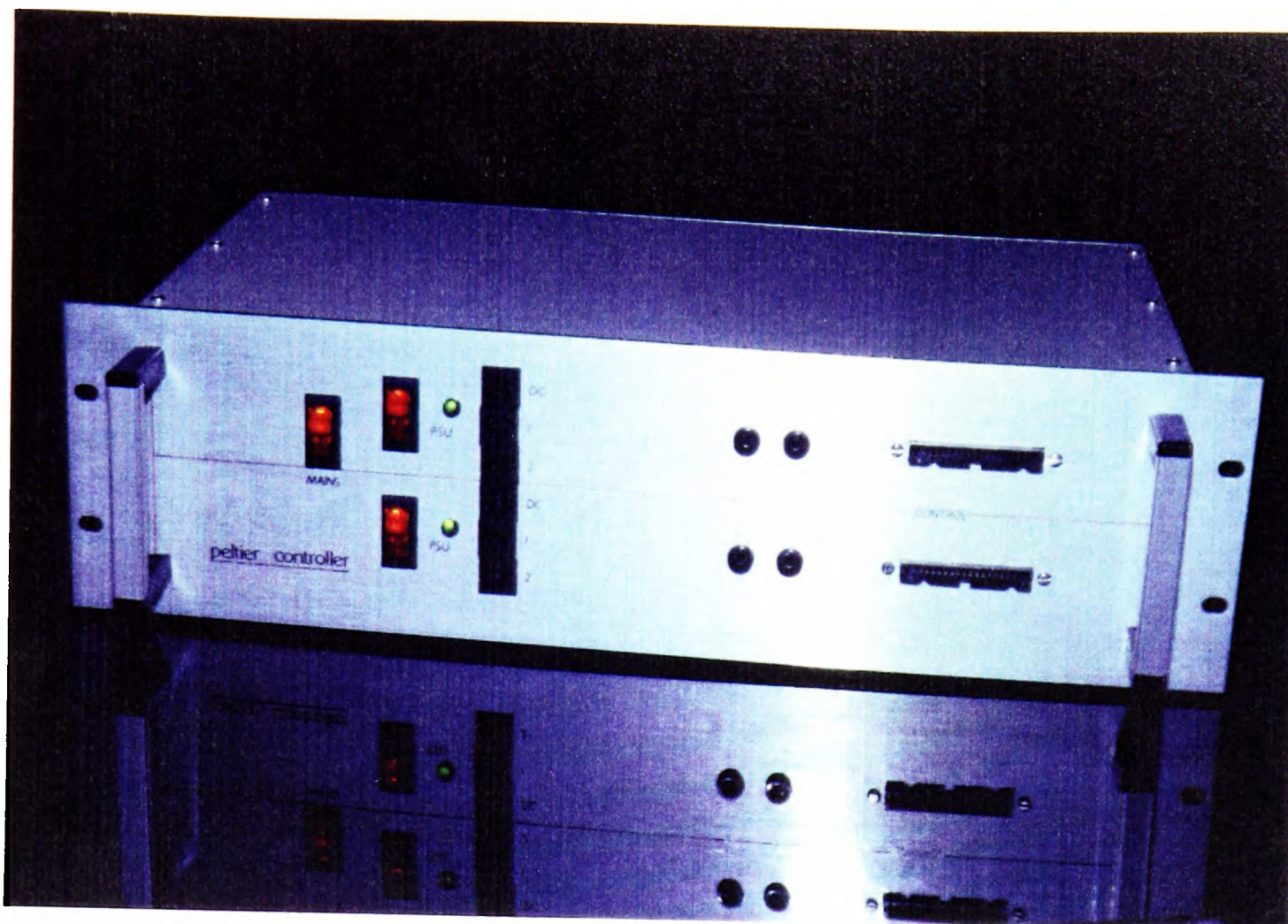


Plate VI-4. Peltier heat pump controller contains high current low voltage power supplies and associated electronics to interface the unit to a microcomputer, to which the cell's thermometer is connected to complete the feedback loop.

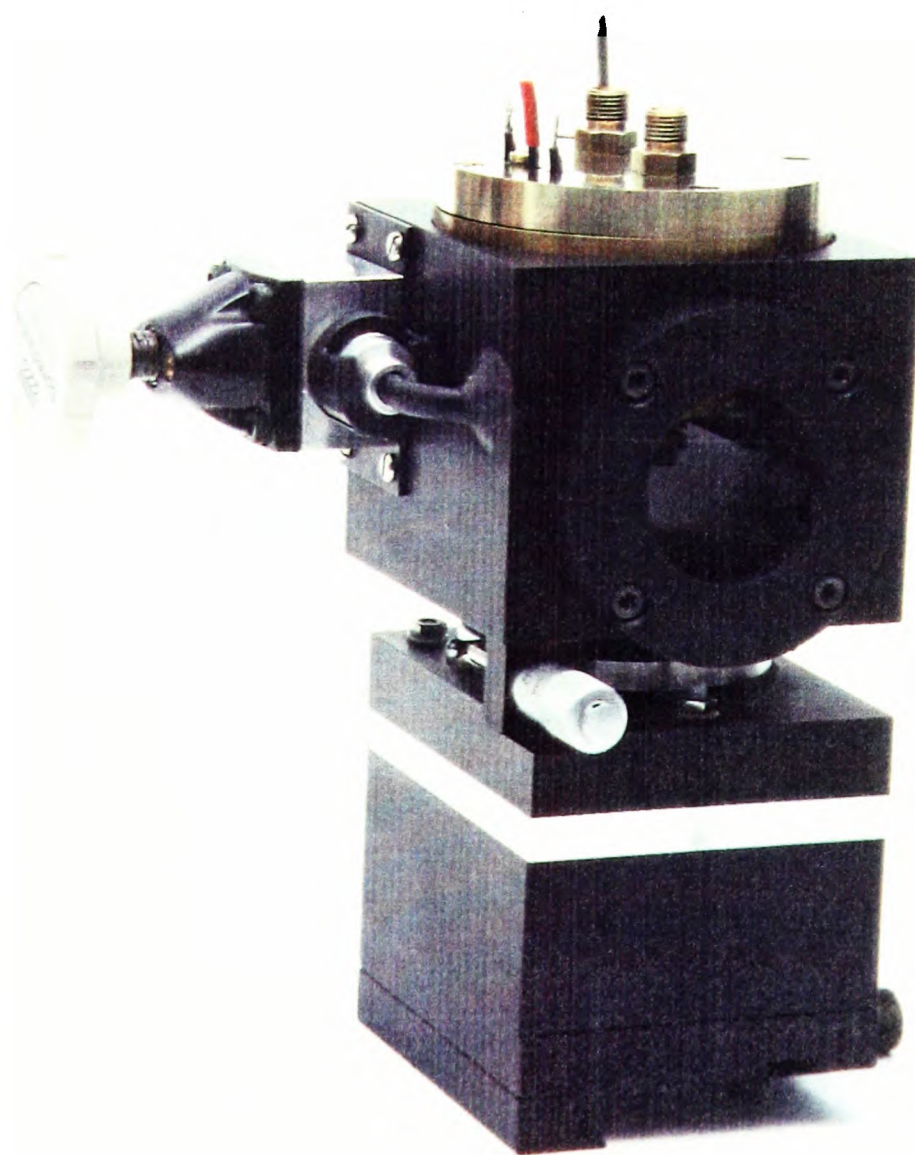


Plate VI-5. Temperature controlled housing.

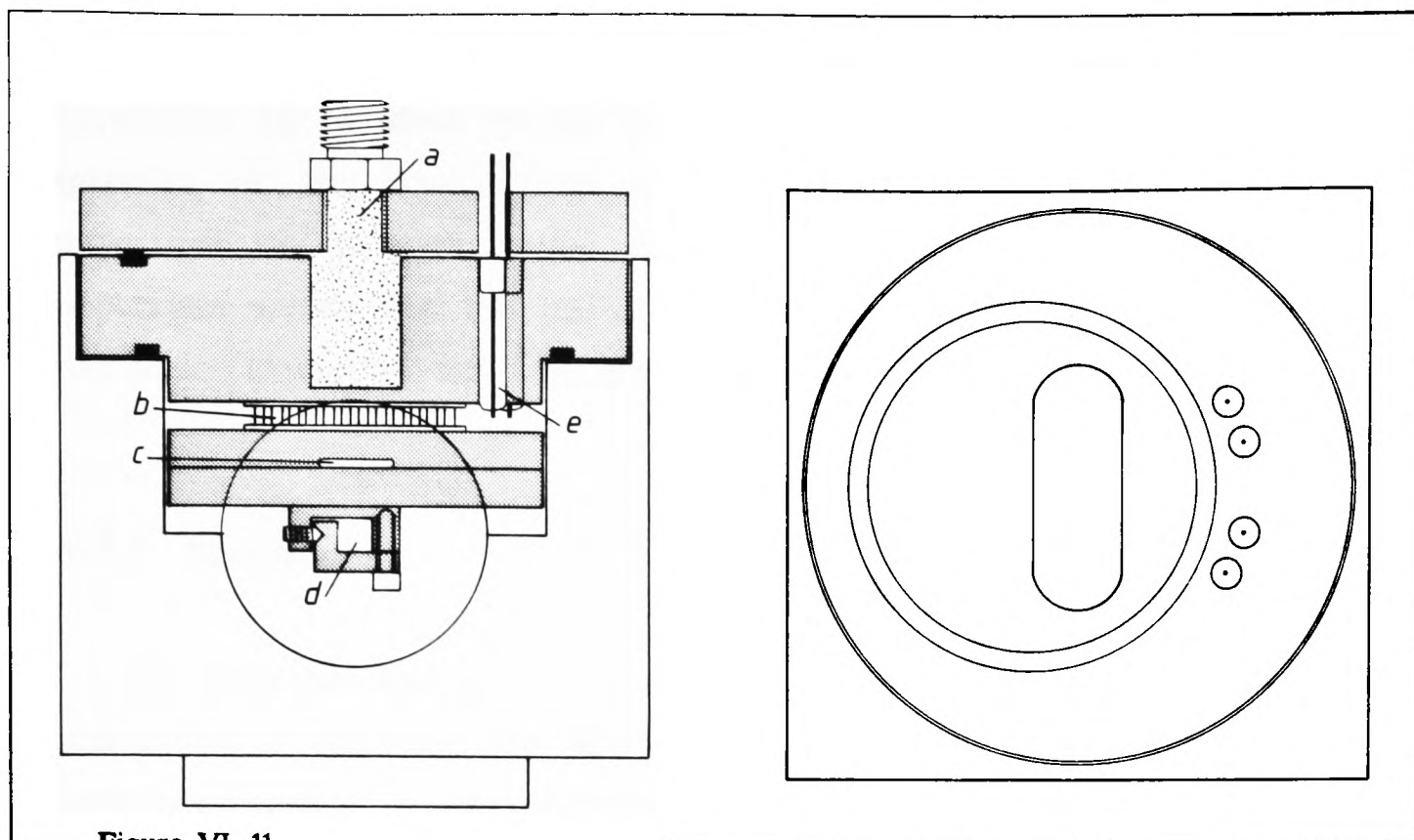


Figure VI-11

Temperature controlled housing. Viewed, left, along the laser beam path. Flowing water (a) removes heat from the output side of a Peltier effect heat pump (b), which cools the mount for the crystal (d). The mount temperature is monitored by a platinum resistance thermometer (c). Connections to the Peltier device and thermometer are made via vacuum feed-throughs (e). The plan view shows the O ring water seal, water chamber and electrical feed-throughs.

temperature stability was rather better than the indicated resolution of 0.1 C, and was rapidly achieved with only slight initial overswing using the original guessed algorithm parameters. Nonetheless, sudden laser heating of the crystal would cause a rise in temperature for a short period until the system regained control, and the small separation of the crystal from the thermometer meant that the crystal surface temperature was to some extent unknown. At full cooling power and with a water temperature around 9 C the coldest achieved temperature was around -37 C, although laser power absorption was sufficient to raise this by up to 2 C.

VI-3.2 Results.

The principal finding of the experiments using ADP was of the problems and uncertainties of operating with ADP at high mean laser powers. The average harmonic power was found to be time-dependent at all power levels investigated, including those achieved when the fundamental beam was interrupted by a chopper wheel with a duty cycle around 1/12. Surface damage to the crystal was also sustained. Steady ultraviolet powers up to around 15mW were achieved, but severe time-dependent distortion of the harmonic beam profile was apparent and proper optimization was impossible. The limited results achieved were consistent with the findings of Isaev et al [VI-1], and there was little danger of achieving steady high conversion efficiencies at useful laser powers.

VI-3.3 Comment.

Ammonium dihydrogen phosphate appears to be a totally unsuitable material for second harmonic generation from copper laser radiation. The wide range of temperature tuning requires a high variation of refractive index with temperature for any typical dispersion behaviour, and the temperature gradients resulting from partial absorption of the high mean copper laser powers thus produce severe phase mismatching across the beam section.

The optimum length of a crystal for frequency doubling of the copper laser, is worthy of discussion. Table V-1 estimates the harmonic generation of noncritically phasematched ADP as around $830 \mu\text{W.W}^{-2}$ for a 20mm crystal length (which corresponds to a mean power conversion around 4W.W^{-2}). For noncritical phasematching, the harmonic generation should be directly proportional to the crystal length. A similar harmonic generation coefficient to that for our sample of BBO would thus require a crystal less than 1.5mm long, although the otherwise optimum focussing for such dimensions would far exceed the crystal damage threshold. Thus, to

achieve efficient conversion in a system in which the focussing is severely limited by the focal and incident intensities, longer crystal lengths are normally used. However, it has been seen in chapter V that the phasematching term in all calculations is the product $\Delta k.l$, and the longer crystal length thus exaggerates any mismatching of the refractive indices of the crystal.

The advantages of ADP are thus its high theoretical conversion efficiency, and its low cost, which makes the use of large crystals possible. The disadvantages are its low damage threshold and high temperature dependence. Work has been carried out, at the Lawrence Livermore National Laboratory in the USA, on systems designed to overcome such problems incurred when frequency doubling very high mean power Nd:YAG lasers up to 100kW [VI-13]. In these systems, damage thresholds prohibit the focussing of the fundamental laser beams, and the crystal is cut into a succession of thin slabs, between which gaseous coolants are flowed. Such arrangements are considerably more complex than have been investigated here, but thermal problems are largely removed by the longitudinal heat flow to the coolant, and long effective crystal lengths allow weak focussing to be useful. Given the low cost of ADP and its analogues (KDP, KD*P), such a system might prove attractive for harmonic frequency conversion of the higher power copper vapour lasers.

Finally, we consider the possibility of repeatedly passing a weakly focussed beam through a thin section of crystal, by containing the fundamental within a Herriott cell. This allows a single piece of crystal to be used a number of times, and is of particular use if the fundamental absorption is low, so that enough passes for high conversion efficiency may be made. By walking the beam around the crystal from one pass to another, the cross-sectional area for thermal absorption (and thus over which thermal gradients occur) can be much larger than that of a single beam, and the temperature variation across a given beam can thus be quite small. Furthermore, the longitudinally cooled thin slab geometry could be used in such an arrangement; in this case the system is essentially similar to that used by Eimerl and described in the previous paragraph, except that some focussing of the beams is possible and that the phasematching crystal length is limited to the thickness of just one slab. Both these effects will be of advantage; the added complexity of the delay-line cavity, however, may prove unacceptable.

VI-4. Conclusion.

Before this work was started, both BBO and ADP were expected to suffer drawbacks for use with the copper vapour laser: ADP was known to be sensitive to thermal detuning, and BBO suffered high birefringence and hence a small acceptance angle. These are disadvantages which automatically accompany any reasonably large tuning range for phasematching, and as pulsed metal vapour lasers were generally felt to provide high mean powers with poor beam quality, the two materials were expected to yield equally poor results.

This work has shown that with the right cavity design, even copper lasers may be characterized by good transverse beam quality; yet during this period the mean power available from these devices has been increased to over 100W. Angle-tuned beta barium borate, with its higher damage threshold, is superior, and the advantage of the higher effective nonlinear coefficient of ADP is completely lost. The results reported in this chapter have confirmed the predictions of the theory of chapter V and the interpretations of chapter IV, and show that beta barium borate can generate the second harmonic of radiation from the copper vapour laser with almost the full theoretical conversion efficiency. Until copper lasers reach such high powers that liquid cooled slab geometries become worthwhile, BBO will remain the better material.

The remainder of this chapter will consider a number of small improvements which might be made to the current apparatus; finally, the extension of the results of section VI-2 to higher power systems will be considered.

VI-5 Consideration of future systems.

VI-5.1 Improvements to current apparatus.

The work reported in this chapter suggests a number of improvements which might be made to the existing system of CU-10 copper vapour laser and beta-barium borate. Some are of specific relevance to this combination, whereas others merely reflect standard practice. Yet there are other standard techniques which are not appropriate to this specific system.

The laser cavity.

The unstable resonator design requires little improvement. The best results have been achieved with unstable resonator magnifications between 20 and 60, and with the small reflector of the unstable cavity set off from the axis of the laser tube so that it intercepts the earlier laser action at the edge of the beam. By using a side-arm

geometry, the inclusion of polarizing components within the laser cavity is made simple. Although the polymer polaroid used in these experiments introduced no adverse effects upon the beam quality, some gradual deterioration of the polaroid did occur, and a prism polarizer would be preferable.

Recently, "graded reflectivity" spot mirrors have been developed, whose reflectivity falls smoothly at the edges, perhaps following a Gaussian function. This results in an apodized focal intensity distribution with the laser power concentrated in the centre. With high magnification cavities appropriate to the copper laser, however, the mirror occupies only a small proportion of the beam area, and the power in the side lobes of the diffraction pattern is therefore low. The use of graded reflectivity mirrors with the copper laser is therefore unnecessary.

Reflection.

Much can be done to reduce the losses incurred by reflection in the current optical arrangement: the use of antireflection coatings, for example, could almost double the harmonic power. Phase matching fluid within the crystal cell would further reduce reflective losses, and might also raise the surface damage threshold of the nonlinear material (although there can be complications if the phase matching fluid absorbs or can be photo-degraded by the laser beam or harmonic). Certainly a Brewster angle prism should be used, for not only are the reflective losses of the harmonic reduced, but the rejection of the orthogonally polarized fundamental is enhanced. Finally, the collimating lens might be built into the crystal cell as the output window, whereby tidying up the arrangement and completely removing a pair of reflecting surfaces.

Elliptical focussing.

Substantial occlusion of the laser tube by molten copper fortuitously gave the laser beam a useful ellipticity for the polarizations of fundamental and harmonic used here. The more obvious arrangement, in which the crystal is rotated about a vertical axis, would instead have been adversely affected by the beam ellipticity. For small bore lasers where occlusion is substantial, rotation about a horizontal axis is therefore appropriate. However, if an elliptical laser plasma tube were used with its major axis in the stronger, vertical orientation, a vertical axis of crystal rotation would be necessary and this orientation is indeed preferable as it allows the use of a Brewster angle prism whilst keeping all the beams within a horizontal plane.

Beam path covers.

The use of airtight beam path covers has been essential in achieving efficient harmonic generation with the copper vapour laser. Even within good beam path covers, however, turbulent distortion can be significant, especially when the laser tube

bore is large. It has always been an advantage with elemental metal vapour lasers to extend the laser tube beyond the discharge region, thereby reducing the rate of sputtering of the metal onto the laser windows. A most effective method of eliminating convective distortion of the beam path would therefore be to extend the laser vacuum chamber to cooler regions at the ends of the laser head box, using active cooling of the tube ends if necessary. At the high voltage end at least, the tube extension will have to be electrically insulating, but this arrangement will work irrespective of the laser tube bore.

Intra-cavity crystals.

A common way of improving the efficiency of second harmonic generation of special value with low gain c.w. lasers is to enclose the nonlinear crystal within an optical cavity. Frequently this is the laser cavity itself, in which case the crystal sees the full intracavity energy flux, and the harmonic and fundamental are separated by an intracavity prism or the dichroic nature of one of the laser mirrors. Crystal losses may be significant, however, and in these circumstances an external cavity may be used (and is effectively a high finesse etalon).

There is no point in attempting intracavity frequency doubling of the copper laser, for the high laser gain allows external intensities almost as high as those within the laser cavity; in any case, an unstable cavity is essential for good beam quality. Similarly, the storage time of an external cavity must be less than the coherence time of the laser. Since this was found in chapter III to be around 10^{-10} s, any reasonably high finesse cavity would have to be rather shorter than the crystal.

If depletion of the fundamental is low, repeated passes through the crystal may be used to increase the extracted energy, and this may be achieved by the use of a non-resonant arrangement such as a Herriott cell [VI-14], which in practice is made by misaligning an external cavity. A narrow beam of light then follows a "Cat's Cradle" route through the crystal by making multiple reflections between the two slightly off-axis mirrors, so that each pass is made in a slightly different direction. This arrangement is thus of particular interest with beams of poor spatial quality when only limited areas of the beam, characteristic perhaps of the coherence area, are converted on each pass. An attractive feature of this arrangement is that all the harmonic radiation will emerge within a small angle of the phase matched direction. Some mirror arrangements, suitable for temperature tuned crystals, will allow each transit to pass through a different portion of the crystal, so that the area in which the laser power is absorbed is enlarged. A confocal geometry, however, will permit all the transits to pass through the same region of crystal, so that spatial as well as angular confinement of the harmonic beam is achieved.

A more compact arrangement.

The apparatus for second harmonic generation occupies an area which is comparable with that of the copper laser head itself, but which is determined principally by the focal length of the focussing lens. With low power copper lasers this is some 300mm, but with higher power and larger bore lasers the focal length will increase. An obvious solution is instead to focus the laser beam with a pair of lenses in a telescopic arrangement. It was noted at the end of chapter IV, however, that some of the effects of beam wander may be eliminated by arranging that the perturbing region coincides with the effective focal point of the lens combination; this may be difficult to achieve, and may indeed prevent a reduction in the space required for the focussing system.

VI-5.2 High power second harmonic generation of 255nm radiation using BBO.

The results reported in this chapter show conversion efficiencies around 10% from mean fundamental powers of 2W. Depletion of the fundamental beams is becoming significant, and the peak intensities at the crystal are approaching the level at which damage occurs. As higher laser powers are used, therefore, it will become necessary to weaken the focussing of the laser beam both to make full use of the available crystal length and in order not to damage the crystal. The focussing will no longer be defined by the predicted optima of chapter V but will instead tend to produce a constant intensity at the crystal, and the behaviour of the harmonic generation will approach that for plane waves. Provided that there are no effects from the increasing mean power dissipated within the crystal, then, the efficiency of harmonic generation should become constant.

In the low power regime, an increase in the crystal length both increases the interaction length and enhances the effect of birefringence. The harmonic power thus varies according to the square root of the crystal length l , and the weaker optimum focussing for longer crystals will permit a higher power before the damage intensity is reached. In the high power regime where the focussing parameters are independent of the crystal length, however, the power may be expected to show the full l^2 dependence.

Figure VI-10 shows schematically the form of the dependence of harmonic power upon fundamental power as the departure from low powers occurs. An initial quadratic dependence begins to approach complete conversion but is limited instead to a lower constant conversion efficiency by the constraint of the damage level of the crystal. With a longer crystal, the low power conversion is more efficient and the power at which crystal damage becomes likely is somewhat higher. Bold arrows indicate the movement of the curve as the crystal length is increased.

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VI-Appendices.

VI-Appendix 1 Focal plane distribution for circular aperture.

The focal plane intensity distribution from the copper laser beam will take the form

$$I(r) = I_0 \left[2 \frac{J_1(kar/f)}{kar/f} \right] \quad (\text{VI-A1})$$

where J_1 is the first Bessel function, a is the radius of the laser beam, and f is the focal length of the focussing lens. The amplitude 1/e point of this function is at

$$kar/f = 2.5838$$

and for the purposes of comparing experiment with theory we shall compare this 1/e radius with w_0 , the 1/e of the Gaussian amplitude distribution.

VI-Appendix 2 Departure from quadratic behaviour.

The dependence of the harmonic power for plane wave harmonic generation in the case of high conversion efficiency (see Byer [V-3]) is given by

$$I(2\omega) = I_{\text{inc}} \tanh^2 \Gamma l \quad (\text{VI-A2})$$

where

$$(\Gamma l)^2 = \frac{2\omega^2 |d_{\text{eff}}|^2 l^2 I_{\text{inc}}(\omega)}{\eta^3 c^3 \epsilon_0}$$

With low efficiencies, the conversion efficiency approaches the form

$$\frac{I(2\omega)}{I(\omega)} = \Gamma^2 l^2 = \left. \frac{I(2\omega)}{I(\omega)} \right|_0 \quad (\text{VI-A3})$$

where the subscripted 0 denotes the low conversion efficiency limit. We now expand

$$\tanh^2 x = x^2 \left[1 - \frac{2x^2}{3} \right] + O[x^6] \quad (\text{VI-A4})$$

and with a little rearrangement find

$$\frac{I(2\omega)}{I_{\text{inc}}^2(\omega)} = \left. \frac{I(2\omega)}{I_{\text{inc}}^2(\omega)} \right|_0 \left[1 - \frac{2}{3} I_{\text{inc}} \left. \frac{I(2\omega)}{I_{\text{inc}}^2(\omega)} \right|_0 \right] \quad (\text{VI-A5})$$

This result describes the relation between the instantaneous fundamental and harmonic intensities. By integrating this equation and writing the result in terms of the total fundamental and harmonic intensities P_ω and $P_{2\omega}$, we arrive at

$$\frac{P_{2\omega}}{P_\omega^2} = \left. \frac{P_{2\omega}}{P_\omega^2} \right|_0 \left[1 - \frac{2}{3} P_\omega \left. \frac{P_{2\omega}}{P_\omega^2} \right|_0 \frac{\int I dt \cdot \int I^3 dt}{[\int I^2 dt]^2} \right] \quad (\text{VI-A6})$$

The extra term here introduces a factor of 9/8 for triangular pulse shapes. With rectangular pulses, the expression is unchanged.

VI-Appendix 3 Measurement of focal lengths of small convex mirrors.

The unstable cavity for the measurements reported in this chapter was based around a 1600mm focal length concave mirror at the back of the copper vapour laser and a range of convex mirrors around 4mm in diameter. In order to determine the focal lengths of these mirrors, each was incorporated into a microscope arrangement whose magnification was determined, whilst viewing a ruler, by the use of a travelling microscope. The mirrors all had focal lengths much greater than their apertures (small f numbers), and for this reason a number of more obvious techniques were unsuitable. The mirrors had large depths of field, for example, and the focal lengths could not therefore be determined from a measurement of the component separations necessary to produce a sharp image; similarly, parallax methods were quite unsuitable, and the expanding beam produced by reflecting the emission from a He-Ne laser disclosed no precise information.

The geometry of the exercise is shown in the figure below. A ruler lies in front of the convex lens which forms a microscope together with the convex mirror to be measured. The image formed by the microscope is measured using the small travelling microscope, and the magnification of the microscope is thus determined. From a knowledge of the focal length of the lens and the relative positions of the object, lens and mirror, the focal length of the mirror may be found using the easily derived expression

$$F = \frac{M x_1 (f_1 - x_3)}{M (x_1 + x_3 - f_1) - f_1} \quad (\text{VI-A7})$$

The results for the set of six mirrors available for this experiment are given below.

label	focal length	R of C	magnification
27	10.8(2)	21.7(4)	148(3)
60	37.9(5)	75.8(10)	42.2(5)
80	13.9(2)	27.8(4)	115(2)
166	82(2)	164(4)	19.5(5)
200	90(2)	180(4)	18.8(4)
300	106(2)	211(3)	15.1(3)

Chapter VII.

Narrow bandwidth pulsed dye lasers.

VII-1 Introduction

VII-2 Dyes, bonds, absorption and fluorescence

VII-3 Broad band dye lasers

VII-4 Refined narrow bandwidth dye laser cavities

VII-5 Current designs

5.1 The Lambda Physik FL series

5.2 The Lumonics Hyperdye SLM

5.3 The Ewart laser

Narrow bandwidth pulsed dye lasers.

VII-1 Introduction

Organic dyes have performed as laser media since the mid-1960's. They are used almost exclusively in solution, and show high optical gain over bandwidths of a few tens of nanometres. Dyes have been used quite successfully as image amplifiers, although the active volume of solution is usually only a few millimetres long and fractions of a millimetre in cross-section; nonetheless, the usual use of the dye laser is as a tunable source of laser radiation, and dye lasers are operated in all the usual configurations – pulsed, cw, modelocked, and with ring cavities. They are also used to generate and amplify ultrashort pulses, which may be limited in brevity only by the transform limit of the dye bandwidth.

The introduction of narrow bandwidth dye lasers opened a new door to spectroscopists, who suddenly had the high spectral intensity of laser radiation available in tunable form and, very shortly afterwards, in very short duration pulses if required. Whole areas of weak or nonlinear effects could now be studied, and the resolution available from cw systems went far beyond what had hitherto been possible. Tiny atomic phenomena could now be measured directly.

In time, other applications for dye lasers evolved. High power dye laser chains are used in the separation of fissile uranium from its natural mixture by selective ionization of the lighter isotope, and the same principle is used on a smaller scale in the particularly sensitive detection and identification of trace elements either directly or in conjunction with a mass spectrometer. Remote detection of resonant fluorescence can be used to make measurements of atmospheric constituents (Light Detection and Ranging, or LIDAR), and as tunable, easily controllable sources of intense radiation dye lasers have found applications in photochemistry and even medicine.

VII-2 Dyes, bonds, absorption and fluorescence

The laser dye has been defined as an organic molecule containing conjugated double bonds – a chain which might be drawn as alternately single and double bonds but which truly exists only as a whole. The usual, σ , bonding occurs between all atoms of the chain, but the remaining electrons form ' π ' bonds, whose eigenfunctions have nodes at the plane of the molecule and which thus correspond to charge

densities running the length of the conjugated bond chain but displaced from it, below and above the plane of the molecule, by perhaps half the atomic spacing. The pi-electrons are thus contained within a region which runs the length of the conjugated chain and which has essentially a uniform cross-section. The eigenfunctions are well modelled by those of free electrons in a uniform potential well of similar dimensions [VII-1]. In the ground state, the N electrons forming the bond fill the first $N/2$ levels of the potential well, and the first excited state is reached by a transition from the highest filled level of the ground state to the lowest empty level. The energy of this transition depends upon the length of the potential well, in much the same way that the note of an organ pipe depends upon its length. The highest excitation energy, that of the shortest bonds, corresponds to a wavelength around 200nm. At this level, photodecomposition is an effective alternative to radiative decay for the photon energy is comparable with the bond energies of the molecule, and practical laser dyes are limited to wavelengths above about 250nm.

Figure VII-1 shows schematically the energy levels of a dye molecule. The electronic levels S_0 , S_1 etc. described by the potential well are split by the vibrational structure of the molecule, which is further coupled to rotational sublevels. Rapid collisional and electrostatic perturbations then broaden this fine structure into a continuum which extends upwards from each vibronic ground state, and within which thermalization to a Boltzmann distribution occurs in the order of a picosecond. Optical transitions thus occur from the Boltzmann distribution at the bottom of each band to any level in the destination, and population in the excited electronic states has an automatic inversion with respect to all sublevels of the lower states which are not thermally populated. Also as a result of the virtually instantaneous thermalization within each band, the absorption spectrum is the mirror image of the fluorescence spectrum, as reflected about the energy of the purely electronic transition.

Apparent on the right of the figure are the electronic triplet levels of the dye, in which the states of the unpaired electrons have the same spin and thus antisymmetric spatial eigenfunctions. The triplet levels lie below the corresponding singlet states, and the lowest triplet level is metastable with respect to decay to the (singlet) ground state. The two unpaired electrons of the excited state give the dye a highly reactive, biradical chemical character, and most of the population which reaches the triplet state will decay nonradiatively by chemical decomposition. The stability of the dye is thus to a large extent determined by the thermal population of the chemically reactive triplet levels, which is in turn related to the long-wavelength absorption limit: if the energy of the triplet levels is high enough for the dye lifetime to be useful, then the shortest wavelength for absorption may be calculated. Typically, this will be around 1 μm .

The degeneracy of the thermally populated levels of the singlet states with the higher lying triplet structure results in ready population of the triplet bands, which are

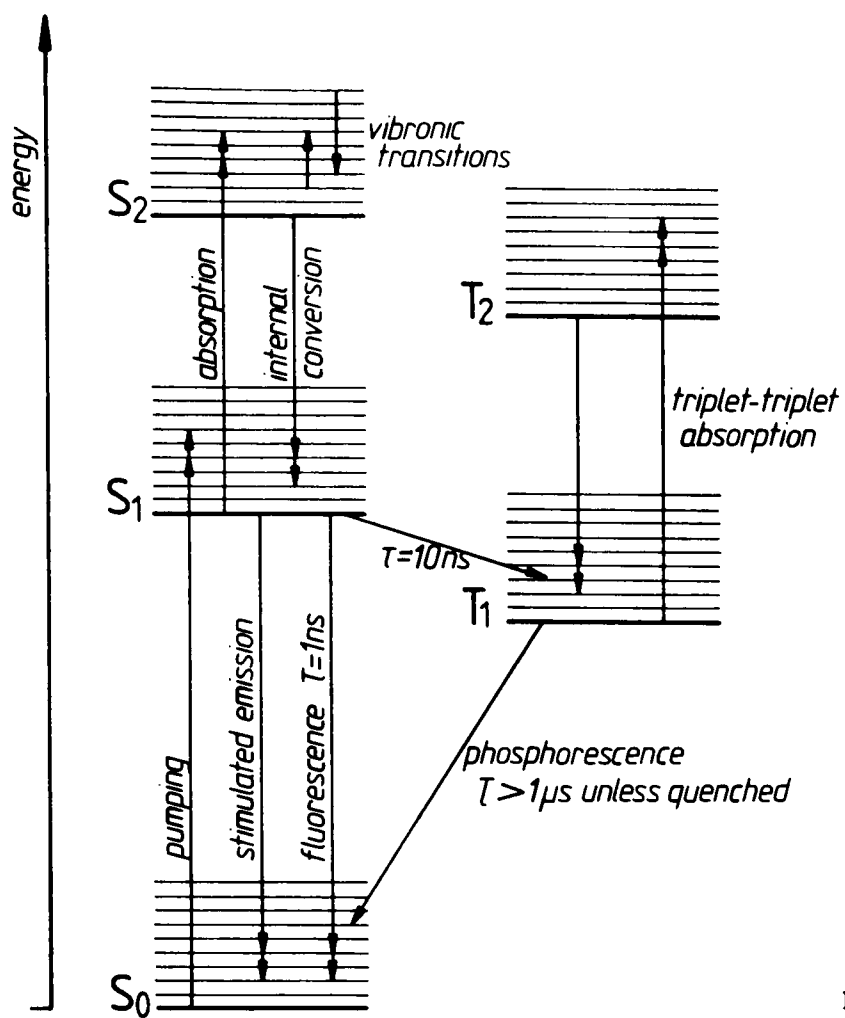


Figure VII-1
Energy levels of a laser dye.

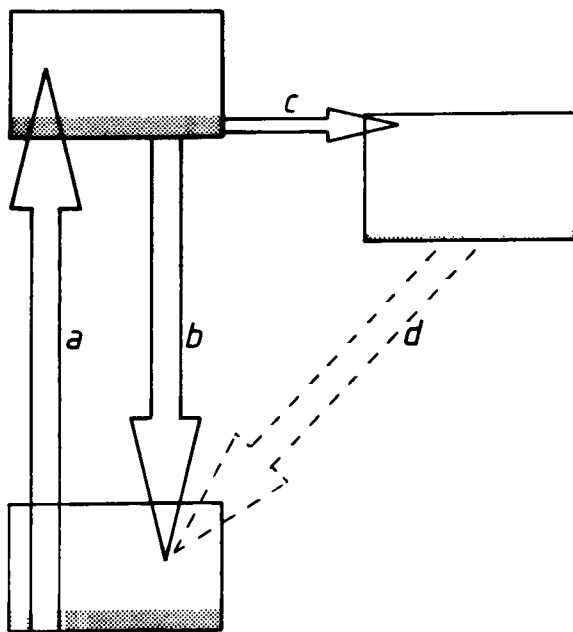


Figure VII-2
Dynamics of a dye laser. Dye molecules are optically pumped from the thermally populated region of the ground state S_0 by route a to the upper laser level S_1 , and rapidly thermalize to the lower regions of this band. Laser emission then returns the population to the ground state band, and thence by further collisional thermalization to within $\Delta E = kT$ of the bottom. Population in the upper laser level may readily transfer to the degenerate levels of the first triplet band T_1 . Decay from the lower levels of this band, however, is slow and this route is one of the primary loss mechanisms in laser dyes.

quickly thermalized and trapped in the metastable levels. The triplet levels thus provide a drain upon the useful excited population of the singlet series as well as an absorption structure through transitions from triplet to triplet. Quenching of the triplet levels is thus desirable, and the most common method is by energy transfer to an excited state of dissolved molecular oxygen, which is usually present at sufficiently high concentrations from contact with the atmosphere. "It is not known whether the triplet quenching of oxygen is due to its low-lying excited singlet states or to its paramagnetic properties." [Schafer, 158], but the same process is used to excite molecular oxygen optically, via a dye derived from blood products, in the medical treatment "photodynamic therapy" [VII-2], where the excited oxygen is sufficiently reactive to destroy cancerous tumours in which the dye has been localized.

VII-3 Broad band dye lasers

The dynamics of a dye laser are indicated in figure VII-2. The thermally populated lower levels of the electronic ground state are excited optically to the vibronic levels of the first excited electronic singlet state S_1 by narrow band laser radiation or the broader bandwidth emission from a flashlamp. The excited population quickly distributes itself within the band according to the Boltzmann thermal distribution, and there is an automatic population inversion with respect to all but the thermally populated levels of the ground state band S_0 .

Gain will be apparent over a range of wavelengths which correspond to lower laser levels throughout the ground state band S_0 - typically, the gain of a dye extends over around 30nm. The rapid thermalization between vibronic sublevels means that this gain is essentially homogeneously broadened. The earliest dye lasers, in which a dye solution was enclosed between a pair of plane reflecting surfaces and excited by a pulse of radiation from a ruby laser, thus operated with a bandwidth of a nanometre or so, at a wavelength which could be tuned by changing the dye concentration, solvent, or the resonator mirror reflectivity [VII-3].

By replacing one of the cavity mirrors with a diffraction grating, spectral narrowing to around 50GHz was achieved and the first easily tuned, narrow band dye laser was born [VII-4]. This single oscillator operated with a peak efficiency of 15% and had a tunable range of 40nm. For many biological, medical and industrial applications, this performance is quite adequate and this original system is quite representative of a number of dye lasers now in commercial production. In order to realise the high resolution theoretically possible from dye lasers, however, some refinements were necessary.

VII-4 Refined narrow bandwidth dye laser cavities.

Where Soffer and McFarland had simply used a Littrow diffraction grating as the dispersive element (fig. VII-3a), Hansch [VII-5] added a beam expanding telescope to increase the dispersion and introduced an etalon to further enhance the selectivity of the cavity, shown in figure VII-3b. This yielded a bandwidth of only 330MHz, but the combination of selective elements made continuous mechanical scanning of the laser wavelength difficult, and Hansch opted for the limited tuning range accessible by pressure tuning of the instrument. Where Hansch had used a telescope to expand the beam emerging from the dye cell, Hanna [VII-6] tried a prism arrangement (fig. VII-3c); Shoshan [VII-7,8] instead used the grating at grazing incidence thereby combining beam expansion and dispersion in the same component (fig. VII-3d), albeit at the penalty of higher grating losses.

By shortening the cavity of a grazing incidence dye laser – thereby increasing the longitudinal mode separation and the number of round trips possible during the gain period – Littman [VII-9,10] was able to operate dye lasers on a single longitudinal mode, with a bandwidth below 1 GHz. A double grating design produced a single shot bandwidth of below 300MHz, and an elegant modification of this configuration by Hung [VII-11] replaced the tuning grating with an off normal mirror, demonstrating the possibility of bandwidths below 400MHz from a single mode, single grating cavity, shown dotted in figure VII-3d. Apart from the more usual advantages of a simple system, the single grating cavity reduces the number of frequency selective elements to two: the grating angle and the cavity length. By judicious positioning of the grating pivot [VII-12,13], these may in principle be scanned together. Incorporating his specific design formula of [VII-12] into the grazing incidence grating dye laser, and using a shorter cavity length and longitudinal pumping, Littman has further reduced the time-averaged bandwidth below 150MHz in the device shown in figure VII-3e, which gave near TEM_{00} spatial beam quality and a tuning range of 15 cm^{-1} without hopping modes [VII-14].

In general, the performance of properly designed dye lasers of any geometry tends to be determined by the transform limit of the pulse length. Highly complex cavities incorporating beam expanding prisms, etalons and numerous gratings are capable of no narrower operation than the grazing incidence grating designs with their single dispersive element (although they may not suffer such high cavity losses). As an example, a novel design developed by Paul Ewart et al in the Clarendon Laboratory, which used a variable bandwidth amplifier to select one of a comb of longitudinal modes from a short plane-plane cavity oscillator, yielded bandwidths of 300MHz or below [VII-15] with pulse energies of around $10\mu\text{J}$. This performance is quite similar to that achieved by the copper laser pumped dye laser reported in the next chapter.

With the realisation of cw dye laser operation, by a combination of high pump

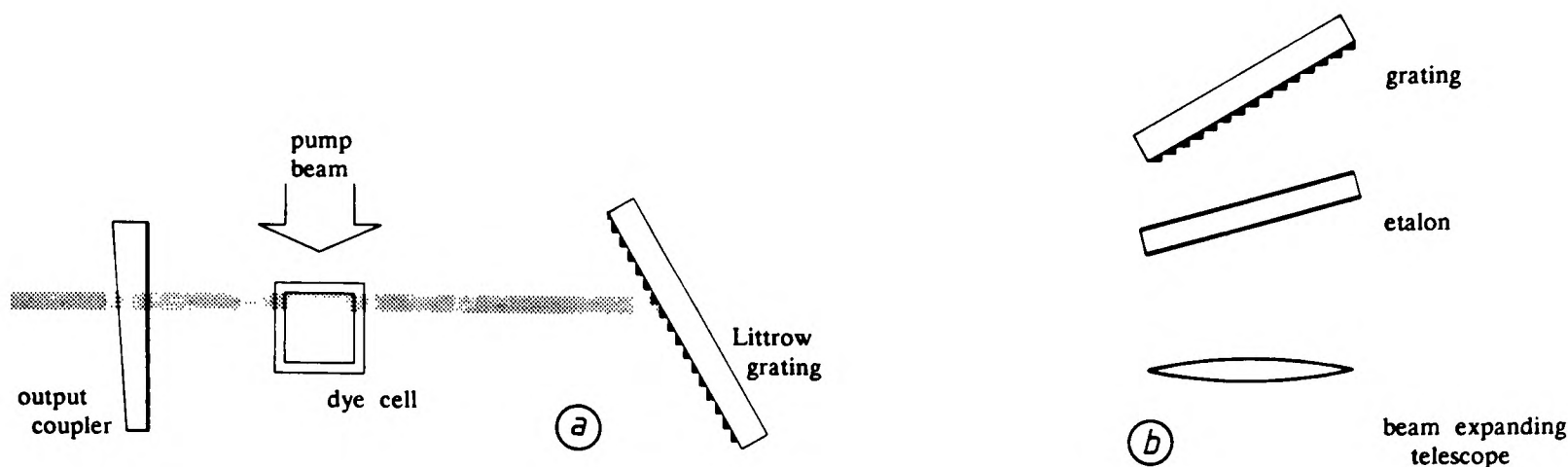
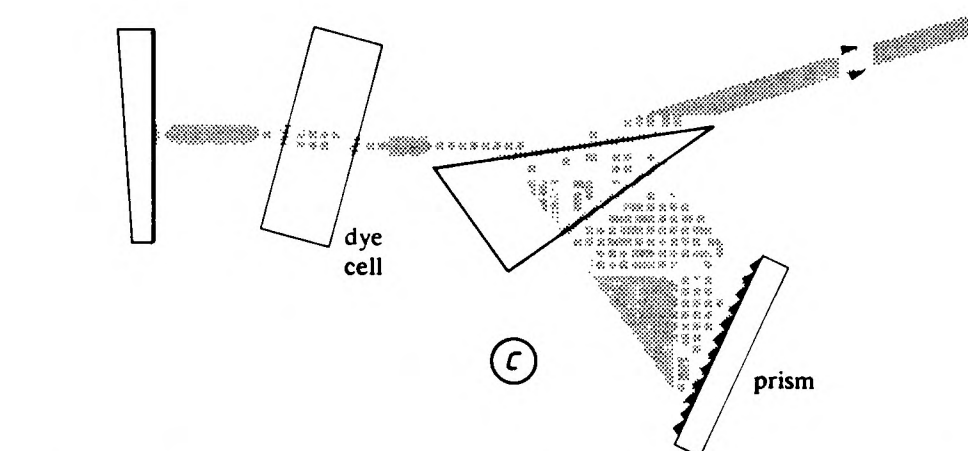


Figure VII-3. Tunable narrow-band dye lasers. The first tuned dye laser (a, above), made by Soffer and McFarland [VII-4], used a Littrow grating in place of one laser mirror. This design was developed (b, right) by Hansch [VII-5], who introduced an etalon and a beam-expanding telescope to enhance the grating dispersion. Hanna et al [VII-6] instead achieved beam expansion using a prism (c, below) and used the prism reflection to couple light out of the laser.



An alternative form of beam expansion (d, right) uses a grating at grazing incidence and a normally incident tuning mirror (Shoshan et al, [VII-7]), laser output being the zeroth order grating reflection. Hung [VII-11] proposed an alternative mirror angle (shown dotted) which introduces an extra, Littrow, grating reflection into the cavity and further increases the dispersion. With a short cavity of this design, using a carefully chosen pivot point which is the common intersection of the grating and mirror planes, Littman [VII-12] achieved wide tunability of operation on a single longitudinal mode. The figure below right, e, shows Littman's configuration with an extra degree of freedom which allows equal but opposite mirror displacements.

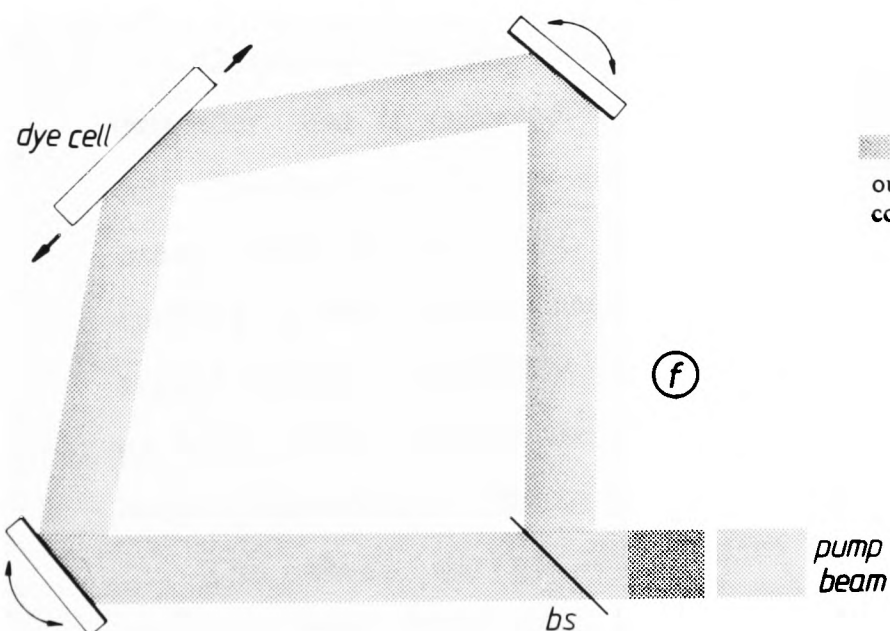
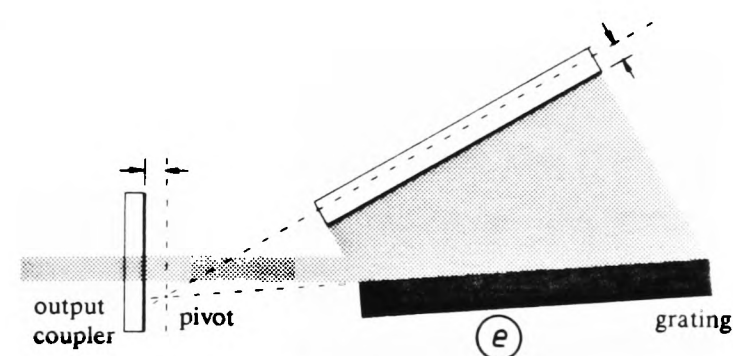
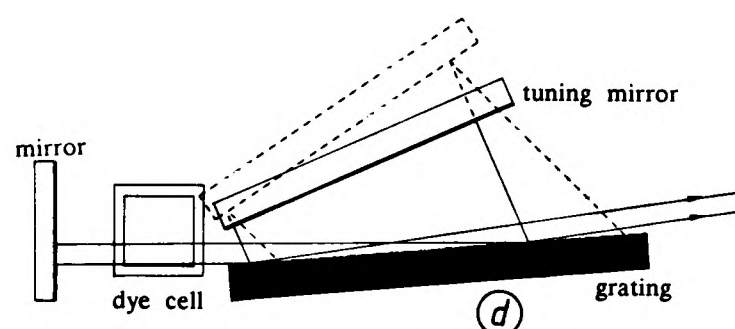


Figure f, left, is the distributed feedback dye laser, in which two portions of the pump beam, separated by the beam splitter bs, interfere and spatially modulate the dye gain. Bragg reflection resulting from this modulation provides the optical feedback for the laser, and the laser is tuned by varying the pump wavelength or the angle made by the two pump beams.

powers, effective quenching agents and high flow rate dye jets, dye lasers of particularly narrow linewidths were possible. Most continuous wave dye lasers in commercial production now use ring resonators incorporating a number of prisms as the dispersive elements, and active stabilization allows linewidths well below a megahertz to be achieved. The final configuration worthy of note is the distributed feedback dye laser (fig. VII-3f). This has no specific cavity elements but instead uses the periodic variation in gain which results from interfering two pump beam components at an angle. The Bragg reflections which result lead to the selection of a single dye laser wavelength which is determined by the pump beam wavelength and angles, but which allows particularly compact tunable, narrow bandwidth, lasers to be constructed. A similar device in which the Bragg planes are made permanently in a piece of Perspex by the interference beforehand of a pair of high power ultraviolet beams [VII-16] demonstrates the somewhat rare use of dyes in the solid state - in this case the dye is suspended in the Perspex.

VII-5 Current designs

In the following section, some details of three current designs of narrow linewidth pulsed dye lasers will be given. Under consideration are the Lambda Physik FL series, the Lumonics Hyperdye SLM, and the Ewart design of [VII-15].

VII-5.1 The Lambda Physik FL series [VII-17]

A schematic diagram of the Lambda Physik FL 2002 dye laser is shown in figure VII-4. The full system comprises a narrow-band oscillator, a preamplifier stage, and a final dye laser amplifier without which the unit is sold as the FL 2001. The oscillator uses a diffraction grating in the Littrow arrangement, with prismatic beam expansion. The inclusion of an intra-cavity etalon then completes the narrow bandwidth version and adds a suffixed E to the model number. The oscillator is housed in a vacuum tight chamber which may be pressure-scanned.

Output of the dye laser oscillator is taken as the reflection from the prism beam expander, and is reflected by the grating through a different region of the same dye cell. This acts as the pre-amplifier and is pumped by a delayed portion of the pump beam. With the FL 2002, the preamplified beam is telescopically expanded before entering a final power amplifier. With an intra-cavity etalon, the usual bandwidth around 0.2cm^{-1} (6GHz) is reduced to 0.04cm^{-1} (1.2GHz). The further inclusion of a confocal etalon between the preamplifier and power amplifier allows Fourier transform limited linewidths as low as 0.009cm^{-1} (0.27GHz) to be achieved.

The above performances, which strictly apply only when pumped by low repetition rate, high pulse energy sources such as excimer or nitrogen lasers, are

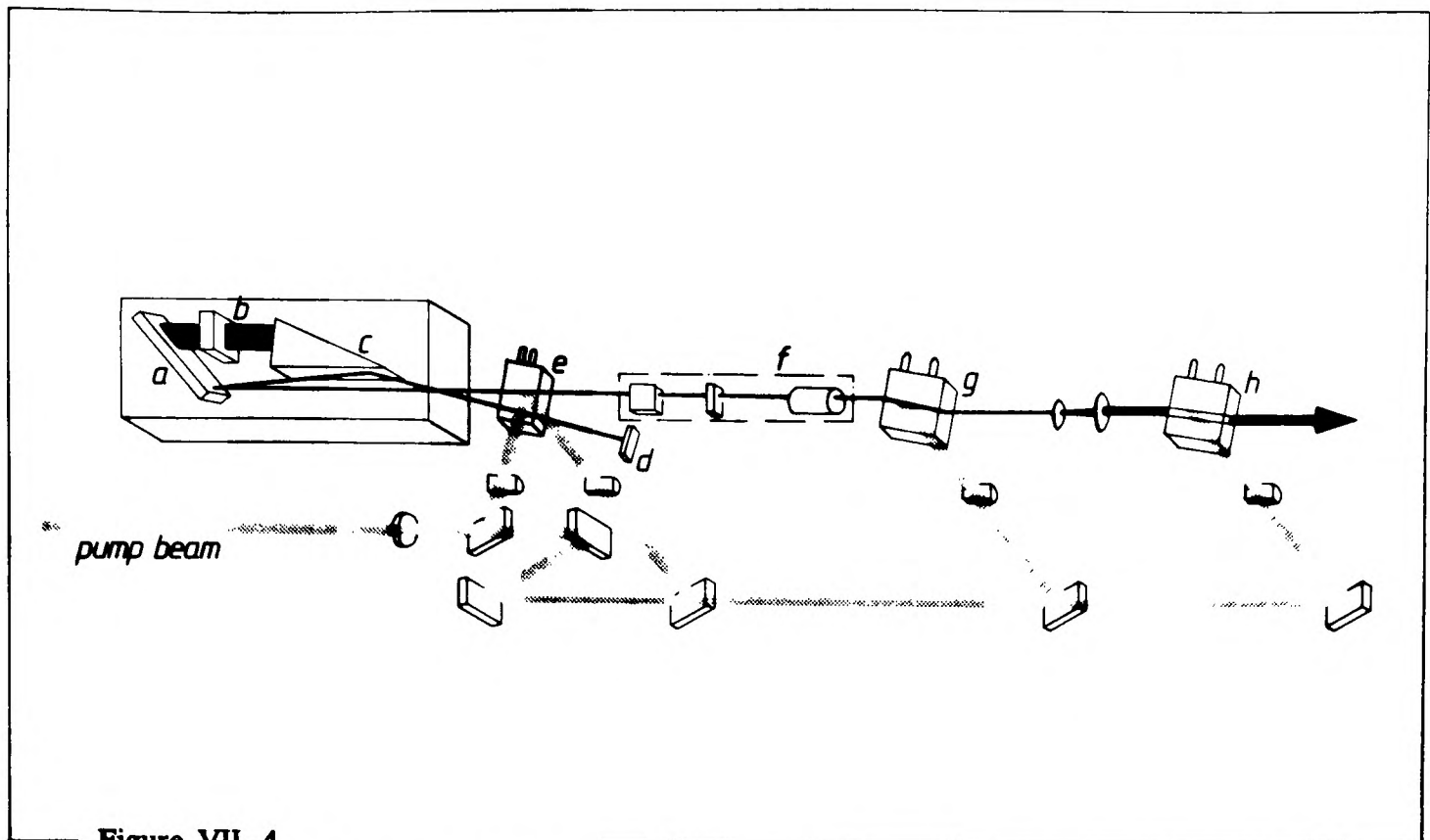


Figure VII-4
Lambda Physik FL series dye laser. Grating a, prism beam expander c, mirror d and the optional etalon b form the dye laser oscillator about the dye cell e. The oscillator output, the otherwise wasted reflection from the prism, passes through a second excited region of the dye giving initial amplification. Further portions of the pump beam excite the preamplifier and amplifier stages g and h. A confocal etalon f is included when the narrowest linewidths are required.

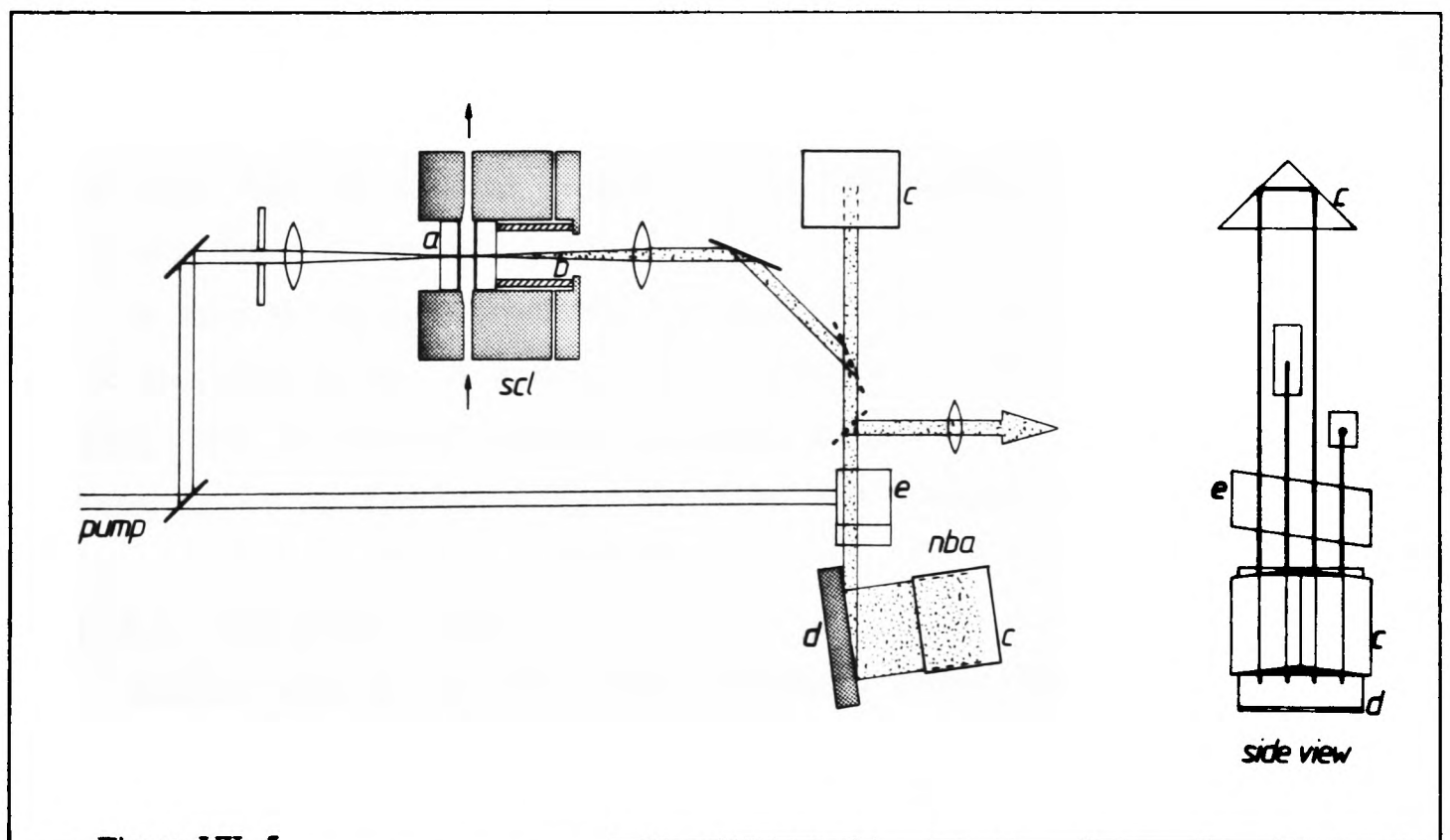


Figure VII-5
Ewart dye laser. The short cavity laser, scl, is composed of two closely spaced mirrors a which form the windows of a cell through which a dye solution may be flowed. The spacing of the mirrors is adjusted by applying a voltage to the piezoelectric spacer b. One of the resulting comb of longitudinal modes is then amplified by the narrow band modeless amplifier nba formed by prisms c, grating d and dye cell e.

nonetheless repeated when pumped by a copper vapour laser. The threshold power for the FL 2001 using rhodamine 6G at a pulse repetition frequency of 6.5kHz is 1.7W, and a maximum conversion efficiency of 26% has been reported [VII-18]. However, Lambda Physik regard CVL pumping of the FL series dye lasers as rather courageous, and recommend the CVL as a pump source only where its high repetition rate and duty cycle necessitate it and high peak powers are not required.

VII-5.2 The Lumonics Hyperdye SLM [VII-19]

The Lumonics Hyperdye laser is a grazing-incidence-grating arrangement with four stage prism beam expansion, and was designed for excimer and Nd:YAG laser pumping. The specified typical linewidth is 0.06cm^{-1} (2GHz), and the maximum repetition rate is given as $>500\text{pps}$. A single amplifier stage provides the conversion efficiency. The Hyperdye SLM (single longitudinal mode) is marketed as the narrow linewidth version of the range, but it is in fact the commercial development of the synchronously tuned grazing incidence grating arrangement of Littman [VII-14], to which a dye amplifier has been added. The oscillator therefore lacks the intra-cavity beam expansion of the basic model.

The Hyperdye SLM uses a short cavity, with a round trip path length around 8cm, to ensure that a reasonable number of round trips is possible within the gain period of the dye, which may only be 2ns when the laser is pumped by an excimer source. The specified time-averaged bandwidth is then less than 500MHz. Nd:YAG laser pumping yields dye laser pulses some 6ns long, and reduces the time-averaged bandwidth to 280MHz, with single pulse linewidths of 200MHz being possible. The dye laser oscillator is enclosed within a thermally isolating case and is mounted on a single solid substrate. The dye solution temperature needs to be stabilized to ± 0.1 C. The laser, like the original Littman design, uses slightly off-axis longitudinal pumping of a thin layer of circulating dye.

It may be inferred from the Lumonics literature that alignment of the Hyperdye SLM is a little tricky. A built-in He-Ne laser is required, and a tool for aligning the grating with the axis of rotation is subject to patent applications. There is no report of the use of the Lumonics Hyperdye SLM with a copper vapour laser pump source.

VII-5.3 The Ewart Laser.

Recent work in the Clarendon Laboratory under Paul Ewart has resulted in a novel geometry of tunable, single longitudinal mode dye laser, shown in figure VII-5. The closely but adjustably spaced faces of a dye cell form the plane-plane cavity of a simple multi-mode oscillator whose small dimensions result in a large frequency interval between longitudinal modes. One of these modes is then selectively amplified by a narrow bandwidth amplifier based upon the variable bandwidth dye laser developed by Ewart et al over recent years: in this case, the bandwidth is adjusted so

as to reject all but the desired mode. Tuning of the laser is accomplished by the synchronous adjustment of the oscillator cavity length and the amplifier tuning prism, and a transducer-limited range of 1 THz has been reported without mode hopping. With a cavity length around 2.6mm, the oscillator gave some 130 modes, each with an energy of around 100nJ per pulse. The amplifier stage increased the energy of the selected mode to around 10 μ J. A further stage of broad band amplification yielded up to 1mJ without noticeable effect upon the bandwidth. Throughout, the Ewart laser was pumped by a 10Hz Nd:YAG laser, of which some 250 μ J longitudinally pumped the oscillator and 5mJ provided the amplifier excitation.

The 300MHz time-averaged bandwidth was found to be determined exclusively by fluctuations in the pump laser and dye flow system: the chosen oscillator dye concentration absorbed only half of the pump energy in order that fluctuations in pump pulse energy should not affect the optical length of the cavity, and in the course of development a flowing dye solution in methanol was replaced by a stationary sample with a solvent (ethylene glycol and water) carefully chosen to give high thermal conductivity. It is fair to assign all these problems to the predictably high sensitivity to absolute cavity length of the oscillator, and it is clear that such a system would be unsuited to operation at the higher repetition rates and thus mean powers characteristic of copper laser pump sources.

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A CVL pumped narrow bandwidth dye laser.

VIII-1 Introduction

VIII-2 Pumping a dye with a copper vapour laser

VIII-3 Design of the high resolution copper laser pumped dye laser

3.1 Detailed design

3.2 Tuning mechanisms

3.3 Mechanical considerations

3.4 Dye cell

3.5 Sensitivity to mechanical perturbations

VIII-4 Practical operation of the copper laser pumped dye laser

4.1 Alignment

4.2 Performance

4.3 Suggested improvements

VIII-5 A copper laser pumped Ti:sapphire laser: a case study

5.1 Gain of copper vapour laser pumped Ti:sapphire

5.2 Bulk thermal damage

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5.4 Self-focussing in sapphire

VIII-6 Conclusion

A CVL pumped narrow bandwidth dye laser.

VIII-1. Introduction.

The principal driving force behind development of the copper vapour laser itself has been its use in the enrichment of nuclear fuels. ^{235}U , which forms about 0.7% of naturally occurring uranium, must be present at concentrations around 3% in order to fuel many nuclear reactors, and the traditional method of fuel enrichment by gaseous diffusion is sufficiently expensive to encourage investigation of alternative methods [VIII-1]. High speed centrifuges which have been developed run with remarkably high reliability, but the enhancement in isotopic concentration at each stage is small and long chains of centrifuges are needed. The method favoured for future commercial use is therefore that of *laser isotope separation*, LIS, in which narrowband dye lasers selectively excite and ionize the fissile isotope, whose electronic energy levels are displaced by the isotope shift. Electrostatic collection then allows substantial enrichment in a single stage of the process [VIII-2].

High laser powers are required if the LIS process is to proceed at a reasonable rate, and all the present industrial LIS systems therefore use chains of copper vapour lasers to pump a further array of dye lasers. A master dye laser oscillator provides perhaps a watt of light at the start of the line of dye laser amplifiers, and the overall system yields mean laser powers of tens of kilowatts. The laser bandwidth must be sufficiently narrow to resolve the isotope shift in the element of interest, but bandwidths below the Doppler width associated with the vapourized material are of no advantage.

At more conventional laser powers, the techniques used in laser isotope separation have a diagnostic role. Resonant ionization mass spectrometry, RIMS, combines isotopically selective ionization with a mass spectrometer, hence providing a particularly sensitive method of trace element analysis, with a detection limit below 10^8 atoms [VIII-3, VIII-4]. Here, the copper vapour laser is a popular dye laser pump source as it provides high intensity pulses of light at relatively high repetition rates; vapour samples tend to remain within an interaction region for only a short time, and thus a high duty cycle increases the sensitivity of the instrument. The high repetition rate may equally be of use when the dye laser is scanned across a spectrum: in this case, it is the rate of acquisition of data which is increased.

There are a number of spectroscopic techniques to which copper vapour pumped dye lasers can contribute a higher rate of data acquisition than other lasers of comparable pulse power. Many of these require, or can use, laser bandwidths rather narrower than the Doppler widths of the samples. This chapter describes the design and initial operation of a CVL pumped narrow bandwidth dye laser built for general spectroscopic use.

VIII-2. Pumping a dye with a copper vapour laser.

The only narrow band commercial dye lasers to have been used frequently with a copper vapour laser pump are the Lambda Physik FL series [VIII-3] described in chapter VII. Pumped by a CVL, these dye lasers typically give bandwidths below 5GHz which can be reduced to around 1GHz by the inclusion of an intra-cavity etalon. Peak conversion efficiencies without the etalon approach 25%, and tuning ranges of 30nm with a single dye seem characteristic. However, with the intracavity etalon installed, tuning is somewhat clumsy, and even when pumped by a 40W copper vapour laser the FL dye lasers are operating near threshold, for the series was originally designed for use with excimer lasers of much higher peak power.

The Ewart dye laser, also described in the previous chapter, was designed to be pumped by a Nd:YAG laser of comparable pulse energy to that of the CVL. However, in order to achieve frequency stability, a static dye solution has to be used. At the higher mean power and repetition rate of the copper vapour laser, dye circulation is essential, and any perturbations upon the very short cavity length result in large variations in the laser frequency. The Ewart design of dye laser therefore appears unsuitable for use with a copper vapour laser.

Both the Lambda Physik FL series dye laser and that designed by Ewart use the pump laser solely to provide excitation power for the dye. With a distributed feedback dye laser, however, the pump laser defines the dye laser output wavelength by the spacing of the induced grating pattern, and thus any changes in the frequency of the pump laser will be reflected in variations in the dye laser wavelength. It will therefore be impossible to reduce the bandwidth of a copper vapour laser pumped distributed feedback dye laser below 2GHz. The same will be true of any attempt to pump a parametric oscillator with the copper vapour laser.

It has therefore been the aim of the work reported in this chapter to design a simple copper laser pumped dye laser with a bandwidth below 1GHz. The chosen configuration is that initially reported by Littman [VIII-5], employing a single grazing incidence diffraction grating in a geometry capable of synchronous tuning of cavity length and grazing angle.

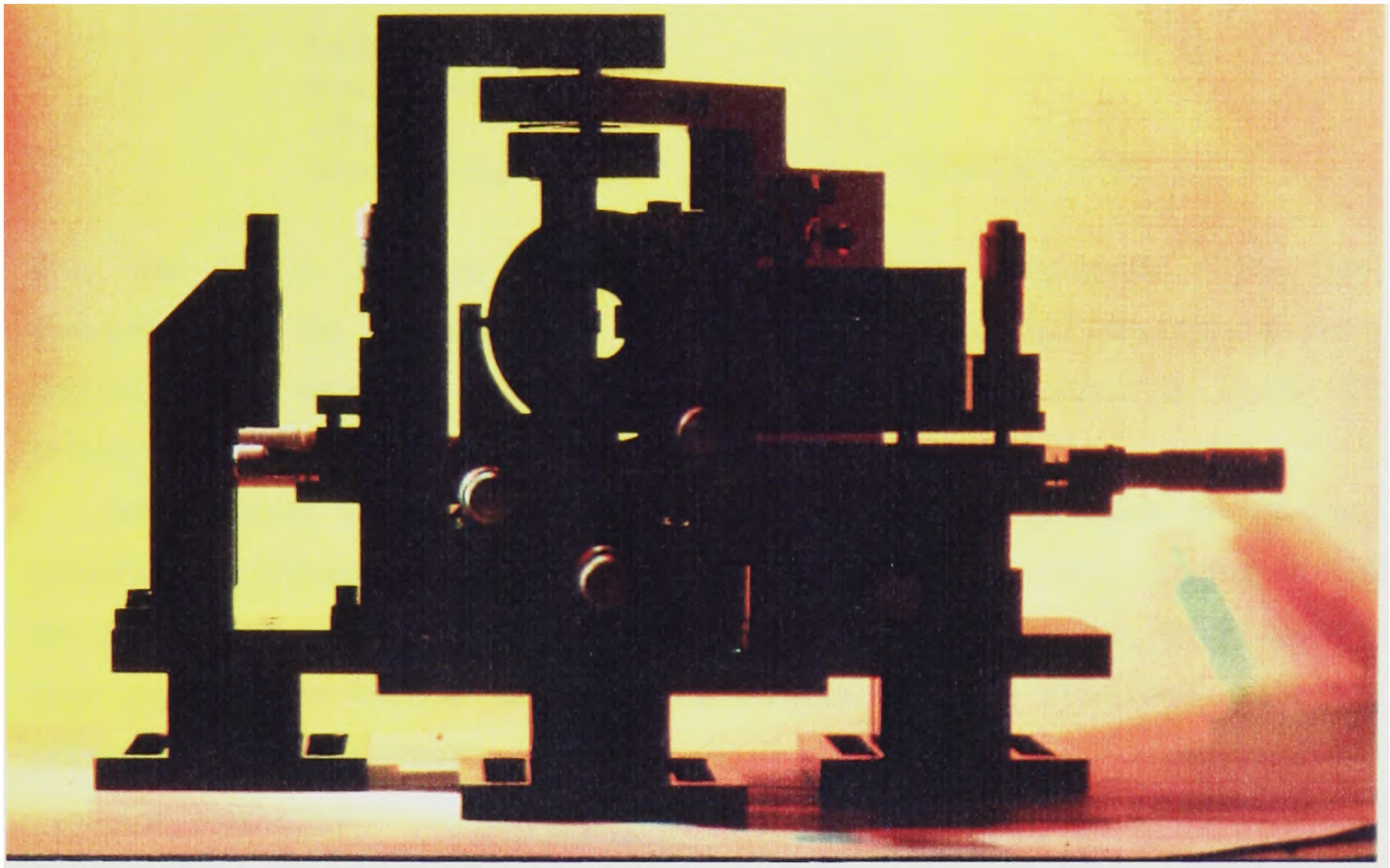
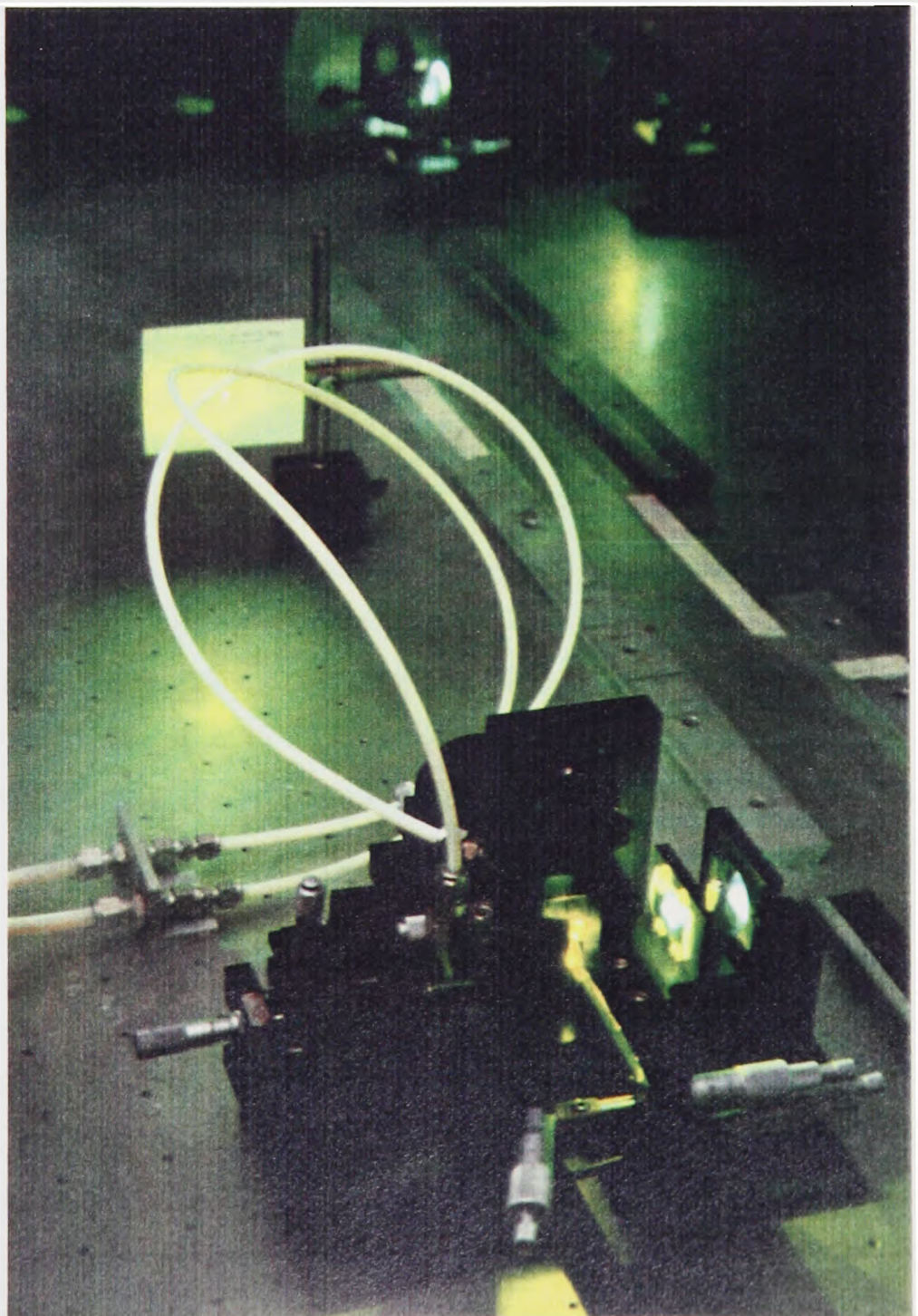


Plate VIII-1 High resolution dye laser shown assembled, above, and working, below.



VIII-3. Design of the high resolution copper laser pumped dye laser.

The original specification for a copper vapour laser pumped dye laser system was

bandwidth	< 1GHz
tuning range	20nm
efficiency	20%
beam quality	near diffraction limit

This was to be achieved by an oscillator–amplifier combination. The oscillator determines only the bandwidth and tuning range of the system; efficiency is principally defined by the amplifier, and the beam quality may be improved by spatial filtering between the two stages.

Such narrow linewidths can only be achieved by the inclusion in the dye laser resonator of diffraction gratings or multiple Fabry–Perot etalons. Tuning is eased if the number of components to be adjusted is minimized, and for the copper vapour laser pumped dye laser it was therefore decided to use a diffraction grating as the principal dispersive element.

The single–round–trip bandwidth of any resonator based on a diffraction grating is ultimately determined by the maximum path length variation introduced by the grating, and thus to the grating length. For a single–pass bandwidth of 0.1cm^{-1} , for example, a path difference of 10cm must be possible, and the grating must therefore be at least 5cm long.

In order to illuminate a sufficient length of diffraction grating for narrow bandwidth to be achieved, some form of beam expansion is required, for the use of spatial dispersion to determine the wavelength imposes an upper limit upon the width of the gain medium and thus upon the beam width. Beam expansion may be achieved either by imaging using a system of lenses, or geometrically by using prisms and the like. The telescopic system is preferable, for it allows a beam waist in the dye region to be imaged to a parallel beam at the grating. However, if the lensless method of beam expansion is used in an optimum configuration so that the grating lies one Rayleigh length from the beam waist, the limiting fundamental resolution of the grating is only increased by a factor of $2^{\frac{1}{2}}$ [VIII-6].

Beam expansion using lenses or prisms allows the grating to be used at angles around 45° , at which angles they are relatively efficient, and the simple Littrow geometry may be used over a wide tuning range. Alternatively the cavity may be completed by a further tuning mirror, so that the grating presents a constant profile to the incident beam, and in this case the grating may be rotated towards grazing incidence. Beam expansion is now effectively performed by the grating alone, and although its efficiency will be much reduced at such shallow angles, considerable simplicity and shortening of the laser cavity can result. The high gains offered by laser dyes make the lensless grazing incidence diffraction grating resonator an

attractive solution for narrow bandwidth tunable dye lasers, and it is this configuration which has been chosen for the device reported in this chapter.

The design of the high resolution dye laser is illustrated in figure VIII-1. The 25mm diameter copper laser beam is focussed to a 10mm long horizontal stripe by the spherical lens a and cylindrical lens b. This focus coincides with the front of the dye channel c, which is 10mm in width and around 1mm thick. The dye flows upwards in order to ease the purging of air from the system. The excited dye region is thus a 10mm long horizontal channel; emission is reflected from the output coupler d and, to the right, by the combination of grazing incidence diffraction grating e and tuning mirror f.

The geometry of the high resolution dye laser has been chosen to achieve two aims. The first is that the overall cavity length be small, so that a large number of round trips can be made in the short duration of the copper laser pulse. The longitudinal mode spacing of the laser will also be correspondingly large. Secondly, the diffraction grating and tuning mirror should share a common pivot in the plane of the grating, so that the synchronous scanning of cavity length with grating angle can be achieved [VIII-5]. A further aim, to match the minima of the grating transmission function to the longitudinal modes adjacent to that which is lasing, is in fact impossible for this configuration of cavity for it would require the grating to continue as far as the pivot, leaving no room for the dye cell; this may only be achieved in the Littrow case, where the grating need occupy only half the cavity length. In the design reported here, the grating transmission function is confused by the divergence of the incident laser beam and the comparable grating and Rayleigh lengths, and it has simply been the aim to maximize the proportion of the cavity length which may be filled by the grating. Considerable attenuation of longitudinal modes adjacent to that which is lasing should nonetheless be offered.

VIII-3.1 Detailed design.

The entire high resolution dye laser is mounted on a base plate, measuring 280×315mm. The total height of the dye laser is 215mm. As with all optical components used with the copper laser, the beams lie 130mm above the table, and the top of the base plate is accordingly 45mm above the table surface. If necessary, the fixed height jacks may be replaced by micrometers to allow the base plate to be levelled. The dye laser uses a number of commercial translation and rotation stages.

Micrometer adjustment of most degrees of freedom of the dye laser has been provided. The transverse positions of both pump beam focussing lenses can be changed by moving the components around a breadboard of tapped holes in the base plate, and fine adjustment of the longitudinal position and tilt of the cylindrical lens are provided by small micrometers. The pitch and yaw angles of the output coupler

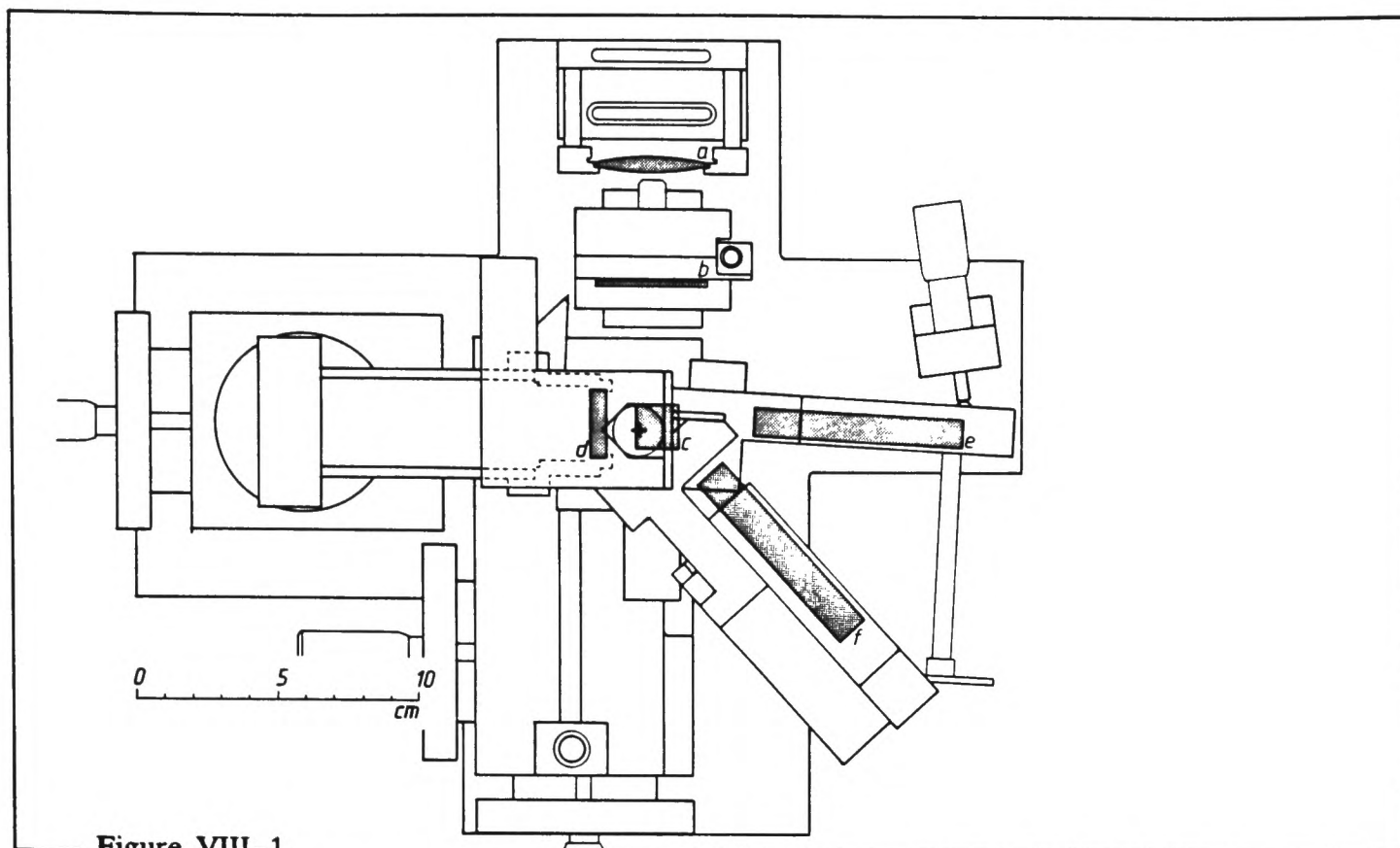


Figure VIII-1.
The high resolution dye laser, viewed from above. The copper laser beam is focussed by the spherical lens **a** and cylindrical lens **b** to a 10mm wide stripe at the dye cell **c**. The optical cavity is formed by the output coupler **d**, diffraction grating **e** and tuning mirror **f**. Note that only the spindle of the Inchworm micropositioner has been drawn, the body having been omitted for clarity.

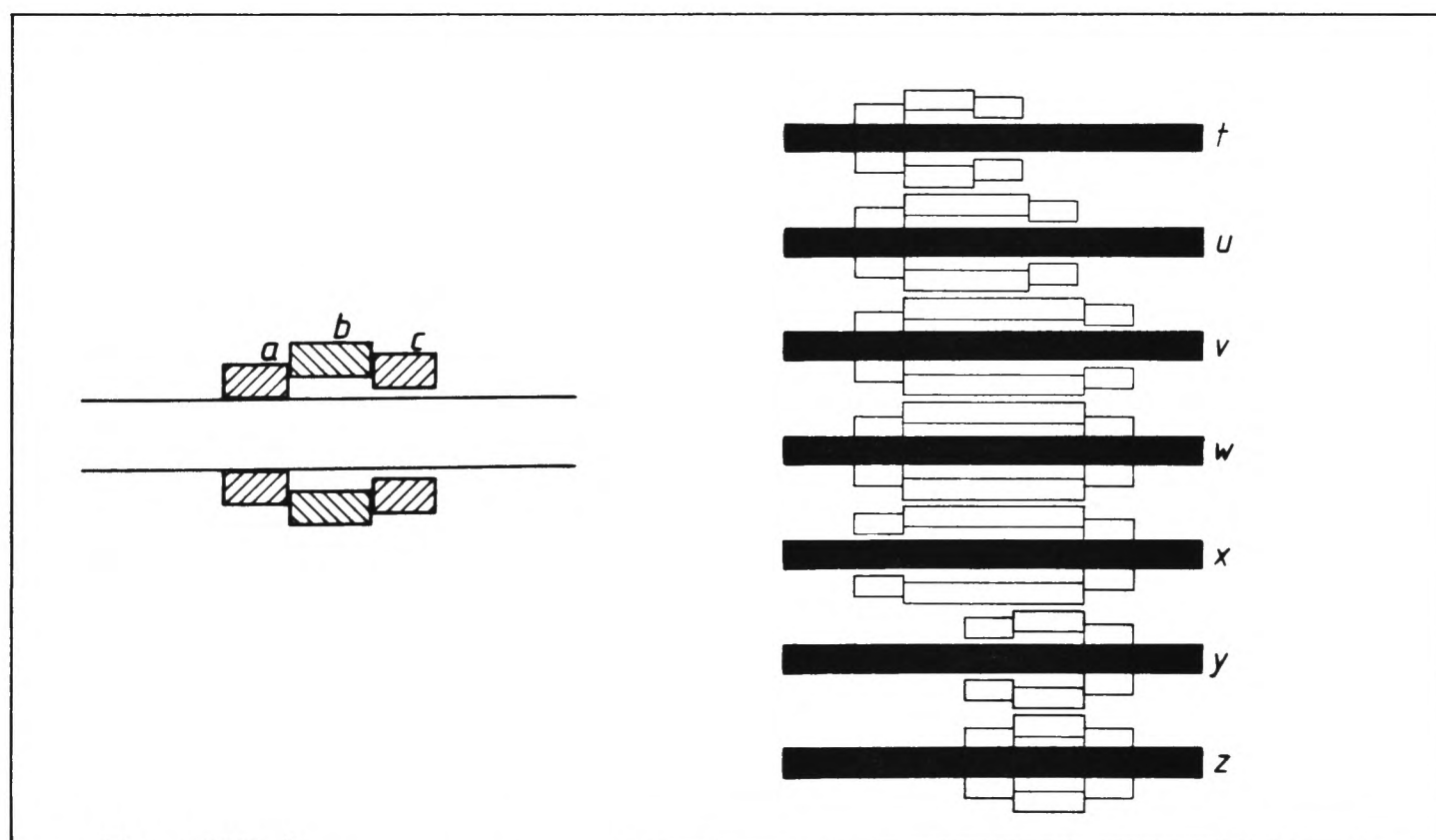


Figure VIII-2.
The Burleigh Instruments "Inchworm" micropositioner. Three rings of piezoelectric material, **a**, **b** and **c**, support a precision ground shaft. Rings **a** and **c** are designed to grip the shaft when activated, and their spacing depends upon the voltage applied to sleeve **b**. By anchoring one end of the assembly until full extension has been achieved, swapping the grip to the other end, and then contracting the sleeve **c** again, a worm-like progression along the shaft may be achieved, as is indicated by the sequence **t-z**. In practice, some 2000 increments in the extension are made, each giving a movement of 4nm. The total travel is limited only by the length of the shaft.

and its longitudinal displacement are similarly adjustable, as are the position of the dye cell and its pitch. The grating incidence angle is controlled by a large micrometer towards the righthand end of the base plate.

It is a prime requirement for the synchronous scanning arrangement that the pivot lie in the plane of the diffraction grating. The provision of a method of adjustment of the position and pitch of the grating would provide an accuracy no better than can be achieved absolutely by careful machining, and there is furthermore no simple method of measuring the grating misalignment with an appropriate precision. The grating is therefore merely pressed up to a machined surface, and no adjustment is possible. For the same reason, the pitch of the tuning mirror is similarly fixed. In practice, nonetheless, any slight misalignment could be corrected by varying the pressure on the clamping plates for these components, or in some cases it could be compensated for by adjusting the output coupler. Adjustment of the grating yaw has been provided so that the grating rulings may be aligned with the vertical before the clamp plate is finally tightened.

Some longitudinal movement of the tuning mirror is afforded by a set of spacer plates which fit between the mirror arm and the mirror mount itself. This allows dead space between the mirror and grating to be eliminated, and the output coupler may then be moved back to allow more space for the dye cell. When this space is large, there is room for an intracavity pinhole, and the dye cell supports have been machined to accommodate a pair of micrometers for this purpose.

VIII-3.2 Tuning mechanisms.

Tuning a high resolution dye laser over a wide tuning range presents something of a problem, for the ratio of tuning resolution to range is difficult to achieve. A typical micrometer has a travel only 2000 times its resolution (eg. 20mm, 10 μ m), whilst the original specification for this dye laser requires a factor some ten times higher. The high resolution dye laser has therefore been designed for use with the Burleigh Instruments "Inchworm" [VIII-7], which moves in 4nm steps over a travel of 50mm. A resolution of 10MHz is thus possible with a tuning range in excess of 100nm. The Fourier transform limit for a 30ns pulse is 60MHz.

A schematic diagram of the Inchworm is shown in figure VIII-2. A precision ground shaft lies between two piezoelectric sleeves, a and c, which are separated by a spacer b, also of piezoelectric material. The electrically induced strain in one of the sleeves causes the shaft to be gripped, and the position of the free sleeve is then adjusted by varying the voltage applied to the spacer: in the Inchworm, a range of 2 μ m is achieved in this manner. By causing the second sleeve to clamp at the full extension and then relaxing both the first clamp and then the spacer, however, the assembly can be made to move relative to the shaft in 2 μ m steps. The instrument

resolution of 4nm is determined by the voltage increments applied to the spacer, and the travel of 50mm depends upon the length of the precision shaft. Any slip which occurs when the Inchworm "changes hands" is apparently reproducible.

VIII-3.3 Mechanical considerations.

In designing this instrument, it has been intended that little or no active stabilization should be required, and some effort has therefore been put into minimizing the effects of stress and temperature. Precision bearings have been used in the main pivot, and provision has been made for counter-weights to be added to the mirror and grating support arms. The Inchworm is supported directly upon the grating arm so that the mirror-grating angle is fixed, and it is the axle itself, rather than the main bearing structure, which is the primary reference object.

Recesses have been included throughout the dye laser to permit the addition of temperature stabilization components. These should be 4W resistors (RS 155-396) and 100k Ω thermistors (RS 151-243), which may be used to maintain a stable laser structure temperature a few degrees above the ambient. Thermal strains are around 20ppm.C⁻¹. Mechanical stability has inspired the use of Alumax (cast, rather than rolled, aluminium alloy) for the grating and mirror support arms and other critical components.

VIII-3.4 Dye cell.

Although the dye cell eventually used in this instrument was taken from an Oxford Lasers broadband dye laser, initial designs were for a cell of slightly different dimensions which would have left more free space in the laser cavity and which would have used PTFE O-rings at the ends rather than the less reliable gasket seals currently fitted. Drawings exist for the alternative design. The dye channel in each case measures 1mm thick by 10mm wide.

The dye cell is supported at an angle to the laser beam, so that its faces cannot form a cavity around the gain region in preference to or in competition with the proper resonator. In addition, some tilt towards the pump beam is possible, to remove a resonator path involving grazing reflections off the dye channel surfaces.

VIII-3.5 Sensitivity to mechanical perturbations.

As an indication of the sensitivity of the dye laser to mechanical perturbations, the table below gives the changes necessary in various parameters in order to cause a shift in wavelength or mode position of one longitudinal mode spacing. The high resolution dye laser uses a 75mm long grating, with 2400 rulings per millimetre of its

length. The tuning range is centred about a 30° tuning mirror angle, which corresponds to a cavity length of 113mm. The operating wavelength, when used in the normally reflecting, first order configuration, varies with the mirror angle by 6.3nm.degree⁻¹. The full tuning range is intended to extend from 20° to 40°, and corresponds to 80% of the Inchworm travel, although the free range of the tuning mirror extends beyond. The longitudinal mode spacing is around 1.3GHz.

Table VIII-1. Sensitivity to mechanical perturbations

path length	0.3μm
mirror angle	4.4μrad
sine bar drive (Inchworm)	0.55μm
dye solution density	20ppm
overall temperature	0.1C
temperature differential across arm	0.04C [†]
A longitudinal displacement of the output coupler by 10μm causes a deviation from the synchronous scanning geometry sufficient to give one mode hop over a 20nm tuning range.	

These predictions show the sensitivity of the instrument to mechanical perturbations to be substantial. Problems may thus be expected over short timescales with vibration and dye flow instabilities, as well as longer term drift resulting from changes in temperature.

VIII-4. Practical operation of the copper laser pumped dye laser.

VIII-4.1 Alignment.

The assembled high resolution dye laser is shown in plate VIII-1. The output coupler is a wedged silica flat, and the tuning mirror owes its broadband reflectivity to an aluminium coating. Initially, a plane silica plate output coupler had been fitted, and, as has been reported by Saikan [VIII-8], interference between similar reflections from the two surfaces periodically inhibited laser action as the wavelength was scanned. All results reported in the following section refer to solutions of the dye rhodamine 6G (also known as rhodamine 590) in methanol.

Alignment of the dye laser is quite straightforward. Initially, the output coupler is rotated to prevent emission from the dye being returned through the gain medium, and the grating is positioned away from the broad beam of dye fluorescence. The pump beam focussing is then adjusted to make the fluorescence beam horizontal. In practice, the beam at the output side will also be set to the correct height above the optical table (in this case, 130mm). It should be noted that because of the tilt of the

[†] This corresponds to a power throughput of around 5W, which is about twice the blackbody power radiated by one of the faces.

dye cell, the stripe of focussed copper laser beam will quite correctly not be horizontal, and the parallel fluorescence beams may be at different heights on the two sides of the dye cell.

The grating is then rotated to the chosen grazing incidence angle, and transverse adjustment of the dye cell is made to allow full grating illumination. The yaw of the diffraction grating – the angle in the plane of the grating between the rulings and the vertical – may now be adjusted by observing the heights of the various orders of diffracted beam. This adjustment is made by tightening the three screws which push the grating onto a curved piece of springy metal, and the grating clamp plate is then tightened. The reflection from the output coupler may now be aligned with the gain region.

This is an appropriate point at which to adjust the focus of the pump beam. Fluorescence from the dye will show a divergence which depends upon the width of the gain region, and a sufficiently wide gain volume will allow laser action between the two dye cell faces to occur. In general, the sharpest focus of the pump beam should be achieved. At this stage, the longitudinal position of the dye cell may also be adjusted.

The tuning mirror is now moved to its operating position and laser action should occur. Peak emission from typical concentrations of rhodamine 6G occurs around 580nm. Once oscillation has been achieved, the focussing and output coupler can be adjusted to maximize the tuning range. With the aid of a suitable etalon, the bandwidth may similarly be maximized. Finally, the longitudinal displacement of the output coupler is adjusted to maximize the tuning range possible without mode-hopping.

VIII-4.2 Performance.

The power available from the copper vapour laser during these experiments was rather low, and the dye laser was consequently pumped with less than a watt of 510nm radiation. Typical dye concentrations of 3mmol were used, although little change in performance was found over a variation in concentration from less than 1mmol to 5mmol. The copper laser pulse repetition frequency was 6.5kHz. A tuning range in excess of 20nm, from as low as 570nm to above 590nm, was measured with a single dye concentration, and operation down to 560nm has been observed with weaker dye solutions. The dye laser pulses were typically 13–14ns long, and the output beam quality appeared to be good.

The spectrum of the laser emission was observed using a CCTV camera and a variable spacing etalon with a finesse rather lower than 10. After initial investigations, a plate spacing of 42mm was used ($\text{fsr}=3.6\text{GHz}$). Single mode operation was initially not observed, the dye laser preferring to oscillate simultaneously on a pair of

longitudinal modes. The tuning mirror was therefore rotated to use the arrangement described by Hung [VIII-9], whereby the beam strikes the tuning mirror at an angle and the cavity has a dispersion equivalent to that of a double grating arrangement. In this configuration, the dye laser showed clear single mode operation, as indicated by the upper photograph of the etalon fringe pattern in plate VIII-2. Much movement of the etalon fringes occurred, however, and the time averaged bandwidth is around 3GHz. As the variation occurred, mode hopping and double mode operation were observed.

No single pulse measurements have been possible which might have allowed the single shot linewidth to be determined. The upper photograph of plate VIII-2 clearly demonstrates single mode operation, however, and the average bandwidth over the exposure time of 1/30s is no more than 1GHz. In the lower photograph of plate VIII-2, simultaneous oscillation of two longitudinal modes may be observed.

VIII-4.3 Suggested improvements.

The single shot performance of the high resolution dye laser is encouraging, for the demonstration of confinement to a single longitudinal mode suggests that linewidths below 1 GHz, approaching the transform limit, might be achieved. The output of around 10mW from the low pump powers available for these experiments would be quite sufficient to seed a single stage amplifier, which itself could be adequately pumped by the extra power available when the CVL is operating properly.

The wavelength instability, which broadens the time-averaged bandwidth to match the basic Lambda Physik FL series lasers, may be attributed to vibration of the laser structure originating at the copper vapour laser, and to uneven dye flow. Mechanical isolation of the dye and copper lasers should be quite straightforward, but it is likely to be the dye flow instability which is the more significant.

A high dye solution velocity is required in order to ensure that fresh dye is available for each pulse from the copper laser, and this flow rate can only be achieved by a large circulator pump and the use of wide bore hoses, which do little to dampen out the disturbances associated with such machinery. A similar problem is routinely solved with cw jet dye laser systems either by covering a length of the dye hosing in sand or lead shot, or by plumbing in a pneumatic damper - a bag of high pressure air which deforms to absorb pressure changes in the fluid.

A further, and perhaps more important, result of the high dye flow rate is the introduction of turbulence and formation of bubbles. Bubble formation - cavitation - is encouraged by extreme turbulence, although it might be suppressed by the use of a less volatile solvent and by reducing the concentration of dissolved gases in the solution. Turbulence results from high flow velocities and sharp changes in the flow path, and thus it may to some extent be eliminated by careful design of the dye cell

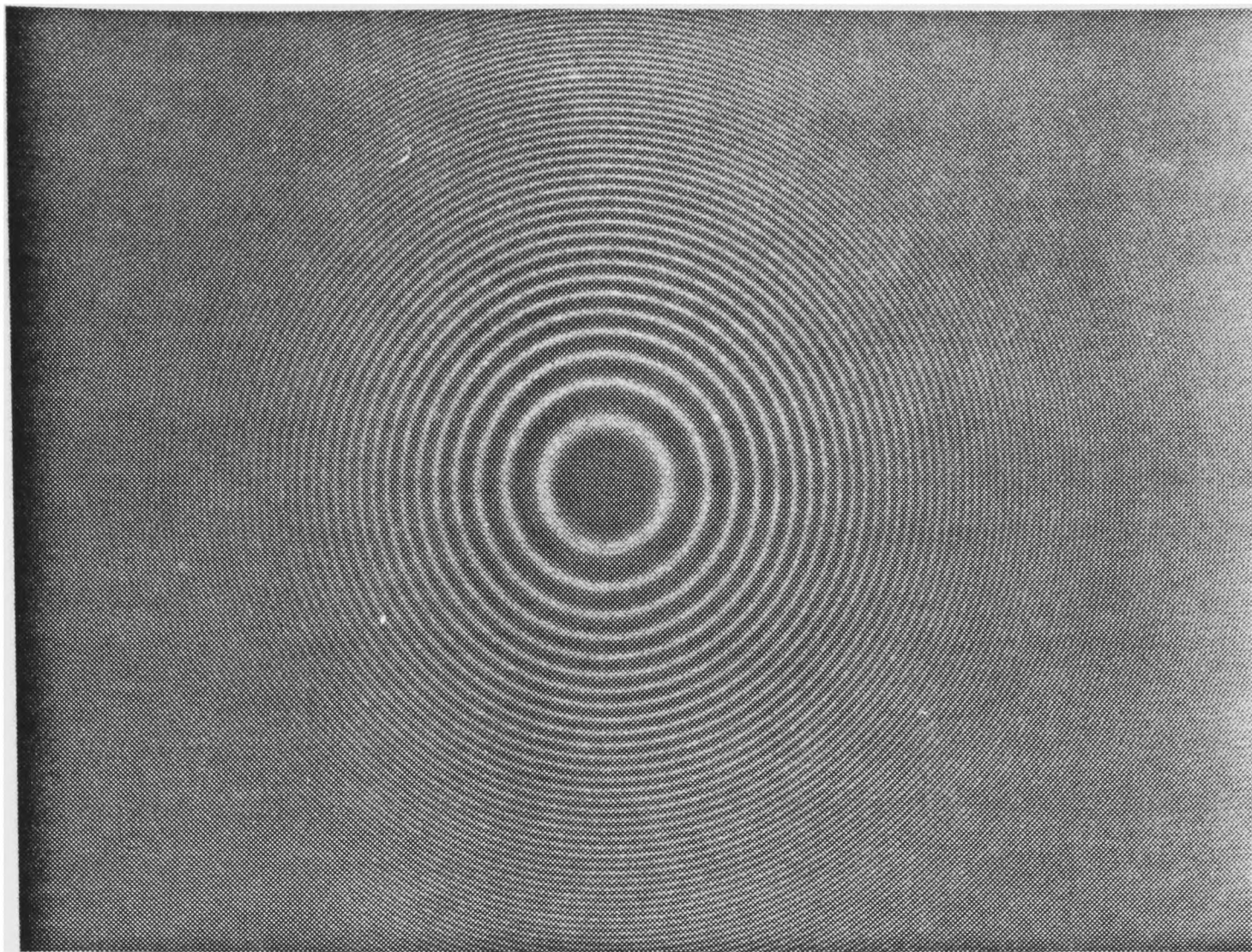
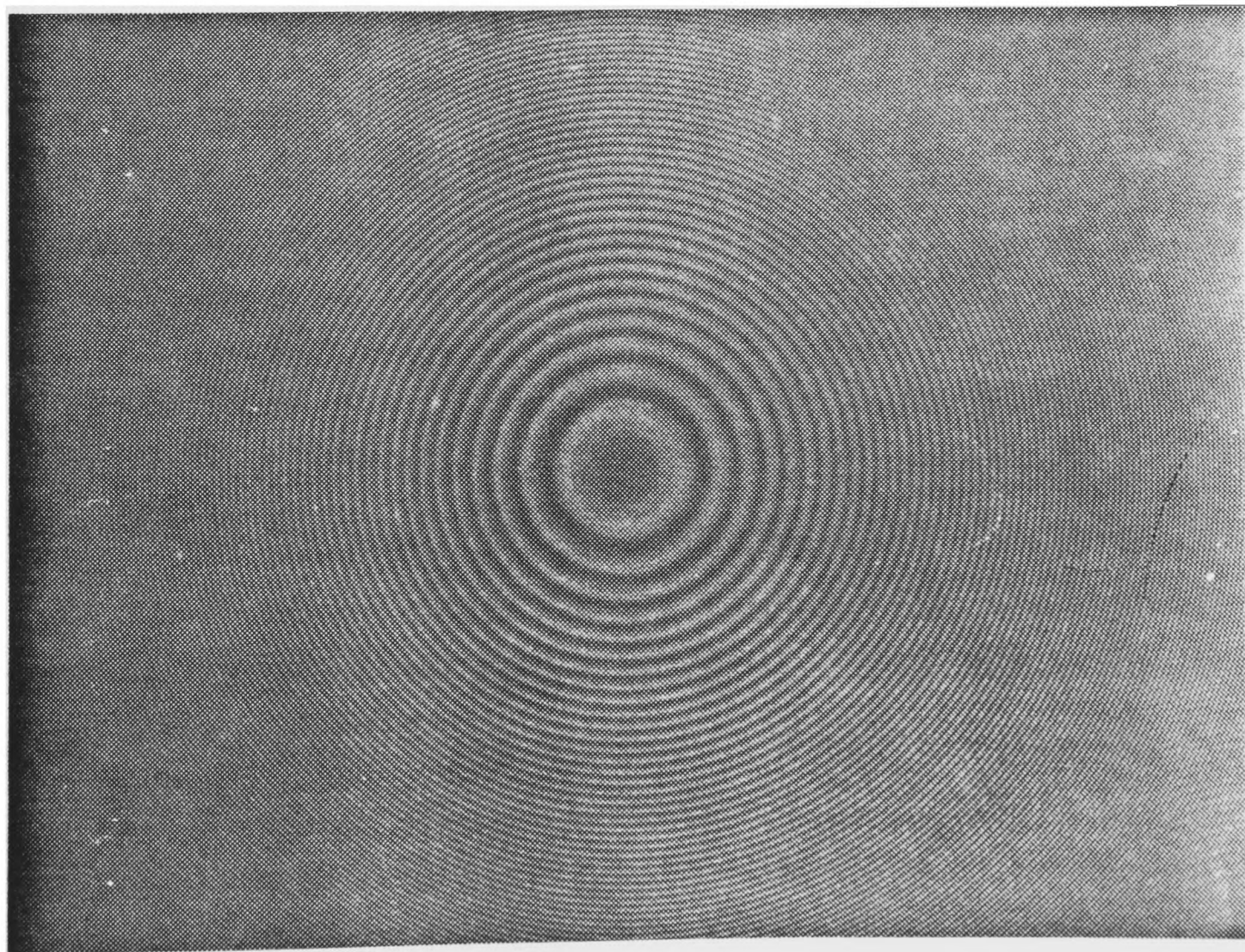


Plate VIII-2. Etalon fringes demonstrating single longitudinal mode (a, above) and multi-mode (b, below) operation of the high resolution dye laser. The speckle pattern was eliminated by using a rotating diffusing screen as the extended source for the air spaced etalon, which had a plate spacing of 42mm.



and associated pipework. It is an irritating feature of transversely pumped dye lasers, however, that the region of pump beam absorption is at the edge of the dye channel where laminar flow will be slowest; turbulence which distorts the optical path may therefore be necessary to agitate the fluid. This suggests that longitudinal pumping, whereby the copper laser beam enters along or close to the axis of the dye laser, may be appropriate for stability of this laser. A rather shorter dye channel will now be required, and indeed the same dye cell might be used after a 90° rotation. The dye concentration will have to be reduced to ensure excitation of the whole dye length. Should problems still persist, the dye cell could be replaced by a dye jet system. Longitudinal pumping is in fact quite easy to arrange with this dye laser, for the low reflectivity of the output coupler will offer little attenuation to the copper laser beam. The emerging dye laser radiation may be selected by using one of the copper laser dichroic beamsplitters.

If, in addition to the above measures, some active stabilization of the dye laser should be required, then this may be achieved by including a piezoelectric spacer in the output coupler translation stage, allowing fine adjustments of the cavity length to be performed. Changes in the grating angle are already made using the Inchworm. A monitoring signal to determine the required correction may be derived from the fringe pattern observed through a fixed etalon which might be required to maintain an absolute measure of the dye laser wavelength. If no such etalon is needed, then an alternative method would be to listen, using a fast photodiode, for the beating of multiple longitudinal modes, at a frequency around 1.3GHz but depending upon the dye laser wavelength. This would in principle be a simple and cheap solution, but the usual problems of detecting short bursts of small signals in a noisy environment would be encountered.

VIII-5. A copper laser pumped Ti:sapphire laser: a case study.

During the course of this work, considerable interest has developed in a recently discovered laser medium for use in the near infra-red. Sapphire, Al_2O_3 , doped to concentrations typically around 0.1% (w/w) with Ti^{3+} ions, has been shown to oscillate over a wavelength range stretching from 660nm to $1.12\mu\text{m}$, around ten times larger than that available from a dye. With a particularly high gain cross-section and remarkably low excited state absorption, Ti:sapphire is characterized by high efficiency – up to 80% of the quantum efficiency – and its long-lived upper laser level can store population inversion for a number of microseconds. The material has all the good thermal and mechanical characteristics of ordinary sapphire, including a high optical damage threshold, and large volumes are readily grown with excellent crystal quality. The table overleaf summarizes some relevant properties.

Table VIII-2. Properties of Ti:sapphire

peak gain cross-section	$3 \times 10^{-19} \text{ cm}^2$
peak absorption cross-section	$6.4 \times 10^{-20} \text{ cm}^2$
peak absorption wavelength	495 nm
half-maximum range	455–577 nm
upper laser level lifetime	2.9 μs at room temperature 3.9 μs below 200K
figure of merit [†]	up to 140
typical Ti^{3+} concentration	0.1% w/w $\approx 3.3 \times 10^{19} \text{ ions.cm}^{-3}$
corresponding peak absorption	2 cm^{-2}
refractive index η	1.77
temperature dependence $d\eta/dT$	$+13 \times 10^{-6} \text{ K}^{-1}$
thermal conductivity	33–35 $\text{W.m}^{-1}.\text{K}^{-1}$
coefficient of linear expansion	$5\text{--}6.7 \times 10^{-6} \text{ K}^{-1}$
Young's modulus	300 GPa
modulus of rupture	2880–380 MPa

Laser action was first observed in 1982 by Moulton [VIII-10], and interest picked up properly with a spate of publications in 1986. Continuous-wave Ti:sapphire lasers are now available commercially to cover the range from 700–1000nm, and offer both a higher output power and a wider bandwidth than any currently available dye for this region of the spectrum. Pulse energies as high as 2J have recently been reported [VIII-11]. Ti:sapphire has been used in place of the dye in a grazing incidence grating laser similar to that described in this chapter [VII-12], and indeed most common configurations of laser have been tried and proven using this laser medium. The absorption spectrum, meanwhile, makes the copper laser an ideal source of excitation, and CVL pumping of a Ti:sapphire laser was reported earlier this year [VIII-13].

The following calculations consider the feasibility of a copper laser pumped Ti:sapphire laser for use where high single-pass gains are required, as in a grazing-incidence grating laser. In order to achieve adequate gain the copper laser will be focussed to give a confocal parameter comparable with the crystal length – as with non-birefringent SHG, the mean squared intensity in the crystal is maximized. A typical Ti^{3+} concentration of 0.1% will be assumed.

VIII-5.1 Gain of copper vapour laser pumped Ti:sapphire.

A crystal length of 1cm will be chosen. This gives an absorption of 85% at 511nm, and 65% at 578nm. If a 10W total power copper vapour laser is used, then, the absorbed power will be around 8W, or an energy of 1mJ per pulse at 8kHz. The concentration of Ti^{3+} ions is around $3.3 \times 10^{19} \text{ cm}^{-3}$, and the volume illuminated by the focussed beam with a waist radius of $30 \mu\text{m}$ will be roughly 10^{-4} cm^3 . The majority of the Ti^{3+} ions are therefore excited. With the gain cross-section given in

[†] The figure of merit is the ratio of the pump beam absorption coefficient to the absorption at the wavelength of emission. Note that different manufacturers values can apply to different wavelengths of measurement.

table VIII-2, the single pass gain should be around e^{10} , or 20,000. This is the maximum gain possible from a crystal of the assumed concentration and length, for the entire population is assumed excited, and the use of a copper laser therefore imposes no constraints on the use of this material other than upon the total energy which may be extracted. The same working allows us to calculate that for a gain of 5% per pass, which may be the threshold for a low loss cavity, a pump energy of $5\mu\text{J}$ is required. These results are consistent with a more rigorous calculation based on the theoretical analysis of Moulton [VIII-14], and with the threshold for cw operation reported by Sanchez [VIII-15]. An input energy of 50nJ should thus be sufficient to extract the stored power in a single pass; a lone photon will require four passes to achieve a similar goal.

The predicted gain is sufficient to allow the use of copper laser pumped Ti:sapphire with any laser configuration. It may be used in place of the dye in the high resolution dye laser, or in short pulse amplification systems where the long storage time will allow maximum power extraction with minimum a.s.e.

VIII-5.2 Bulk thermal damage.

We assume that 5W of copper laser radiation is absorbed in a 1cm length of sapphire, in a beam of radius $30\mu\text{m}$. The surface area of the beam is thus 2mm^2 , and since the thermal conductivity of sapphire is $33\text{--}35\text{W.m}^{-1}\text{.K}^{-1}$, the temperature gradient at this surface will be 700K.cm^{-1} . The coefficient of linear expansion is around $5\times 10^{-6}\text{K}^{-1}$. It follows that the thermal strain will be 10^{-5} , and using a Young's modulus of 300GPa , the thermal stress should be around 3MPa . The reported modulus of rupture in bending is in the range $280\text{--}380\text{MPa}$, which is two orders of magnitude higher than is possible here.

VIII-5.3 Surface crystal damage.

Moulton [VIII-16] has reported laser action in Ti:sapphire at surface energy densities of 70J.cm^{-2} from a Nd:YAG laser without incurring crystal damage. By contrast, a 1mJ CVL focussed to a $30\mu\text{m}$ radius spot can deliver only 35J.cm^{-2} . The peak intensity delivered by the CVL will be proportionally still lower than from the shorter pulse YAG laser. Surface damage of the Ti:sapphire crystal should not therefore occur.

VIII-5.4 Self-focussing in sapphire.

The maximum radial temperature gradient in the crystal was calculated above to be around 700K.cm^{-1} . The variation in refractive index with temperature is some $13\times 10^{-6}\text{K}^{-1}$, and a radial refractive index gradient of 0.009.cm^{-1} will thus develop. The 10mm crystal length will thus have an effective focal length around 0.6m .

VIII-6. Conclusion.

The high resolution dye laser has shown encouraging early performance, with linewidths below 1GHz and a frequency variation around 3GHz. Little work has yet been done to develop the dye laser to its full potential, and it is hoped that instabilities due to vibration and uneven dye flow might be removed, allowing single-mode operation at bandwidths approaching the transform limit to be achieved. A number of solutions to likely problems have been suggested, and the implementation of a longitudinal pumping arrangement has been considered. Such a configuration could then be adapted to use titanium-doped sapphire in place of the dye as the gain medium.

The copper vapour laser is an ideal pump source for titanium doped sapphire. A small CVL, such as the CU10 used in the experiments of this thesis, would be well matched for use with a typical crystal of 0.1% Ti^{3+} , 1cm in length, using longitudinal focussing to a waist of $30\mu\text{m}$. No surface or bulk crystal damage should be incurred. Single pass gains of around 10^4 are predicted, which allows all common configurations of laser cavity to be used, and a long upper laser level lifetime together with the wide gain bandwidth gives much potential for short pulse amplification applications.

VIII-References.

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Chapter IX.

Conclusion.

IX-1 Summary

IX-2 Suggestions for future work

IX-3 A novel CVL-pumped femtosecond pulse source

Conclusion.

IX-1. Summary.

Before this work was started, copper vapour lasers were characterized by poor transverse coherence which rendered them unsuitable for harmonic generation and which presented numerous problems in other applications. The investigations of chapter IV have demonstrated that the use of an unstable resonator can improve the transverse coherence until the laser beam divergence approaches the diffraction limit.

The cavity of the copper laser has been modelled as an unfolded chain of lenses, alternately converging and diverging, which acts like a series of telescopes to reduce the solid angle subtended by the initial, spontaneous, emission. The apparent size of this source at any time defines the transverse coherence of the laser beam, and the rate of increase of the radius of coherence is therefore dependent upon the magnification of the unstable cavity. In the case of a plane-plane resonator, the lenses are not present and the effective cavity magnification is unity: a typical copper laser tube would then require several microseconds for diffraction limited divergence to be achieved. Unfortunately, emission from the copper laser lasts only 30ns or so.

The rise in coherence radius as the pulse develops has been measured using a technique based upon Young's double slit experiment, and the variation with cavity magnification has been investigated. By comparison with a simple theory from geometrical optics, it can be seen that the spontaneous source for the copper laser pulse occurs some 10ns before the main pulse power can be extracted, and a judicious choice of the cavity magnification should thus allow the whole of the main laser pulse to have good transverse coherence. In practice, the use of a high magnification cavity also further delays the start of the laser pulse, so that the available build-up period is extended, and any resonator with a magnification greater than 50 will give the majority of the laser energy a high spatial coherence.

The approach of single pulses to diffraction-limited divergence has been confirmed by the performance of second harmonic generation using the copper vapour laser as well as by observation of clear fringes from widely spaced Young's pinholes, but it is apparent from results averaged over a number of pulses that there is some variation in the shape of the wavefront from pulse to pulse. Whilst the optical path is static over short timescales, slow changes result from thermal distortions to the beam path both within and beyond the laser tube. The largest effect comes from convection

around the laser head, and results in a beam wander an order of magnitude greater than the single pulse divergence; by introducing baffles to limit currents of hot air, considerable improvements in the transverse coherence have been achieved.

Second harmonic generation from the green line of the copper laser has been extensively investigated. By varying the strength of focussing into a nonlinear crystal and altering the magnification of the unstable cavity around the copper laser, the process of ultraviolet generation has been optimized, and conversion efficiencies in excess of 10% have been achieved, yielding steady mean powers of 200mW from less than 2W of fundamental laser power. Conversion rates of 100mW of harmonic power per W^2 of fundamental have been achieved, and compare with the predicted performance for Gaussian laser beams. Low cavity magnifications give the initial part of the laser beam poor transverse coherence, and the efficiency of second harmonic generation is correspondingly reduced. The primary effect of increasing the magnification is to trim the pulse length, so that over a wide range of cavity magnifications the efficiency of second harmonic generation is constant. At the highest magnifications, however, some reduction in the peak laser power also occurs and the efficiency eventually falls. No change in the falling edge of the laser pulse is observed. These findings in themselves further suggest that the peak laser power results from hard saturation of the laser transition so that the emission depends directly upon the rate of population of the upper laser level. The unstable cavity is now seen to slow the approach to saturation.

A limited study of second harmonic generation from the copper laser yellow line has shown similar performance to that observed for the green line, whilst poor performance of sum frequency mixing may be attributed to spatial dispersion of the two copper laser emission wavelengths. The variation in the green line's second harmonic generation performance with fundamental power has been investigated with powers below 2W, and a reduction in the harmonic generation ratio has been seen to occur as the power increases, due to increasing depletion of the fundamental. At a fundamental power of 1W the reduction in harmonic generation is a little above 10%, and is consistent with calculations based upon a plane wave theory. When combined with the knowledge of transverse coherence and pulse length, this understanding gives a complete explanation of all the observed features of nonlinear conversion of the copper vapour laser lines.

In investigating second harmonic generation, an established theory accounting for the conversion of Gaussian laser beams has been extended to allow the focussing of the laser beam a degree of ellipticity. Most crystals used for harmonic generation will convert efficiently only radiation which enters within a limited angular range which is narrower in one dimension than in the other, and it has therefore been suggested that matching the angular variation of the focussed laser beam to the acceptance of the nonlinear crystal might offer a useful increase in conversion efficiency. Calculations

based upon the extended theory indeed predict a small increase by a factor of 1.3 in the harmonic power. An analytic approximation to the numerical results has been derived which is valid for many common practical cases. The predicted improvement in ultraviolet generation has not yet been investigated experimentally.

As a complement to the studies of transverse coherence, the longitudinal coherence of the copper vapour laser has been considered. By applying the Wiener-Khintchine theorem to a theoretical copper spontaneous emission lineshape, the longitudinal coherence function has been estimated. Experimental investigations have then found that the actual coherence function lies mid-way between the theoretical spontaneous emission function and that determined by Doppler broadening of a single component alone. Under the conditions prevailing at the time of the experiment, the measured coherence length (hwhm) was 40mm. The copper vapour laser may therefore be used as a source for the holographic recording of objects with dimensions as large as 100mm.

The final sections of this thesis have considered the design of a high resolution dye laser oscillator for use with the copper vapour laser. Using a configuration chosen for its simplicity of alignment and tuning, a prototype dye laser has indeed been constructed, and in limited initial testing the required performance has been achieved. The laser is particularly compact and easy to align, and its performance compares well with commercially available dye lasers used with copper lasers. However, frequency instability of the order of 3GHz has been observed and is attributed to vibration and uneven dye flow. Neither of these problems has yet been addressed.

IX-2. Suggestions for future work.

The principal thrust of this work, towards high power ultraviolet generation, is already being extended to use with higher power copper vapour lasers, and uv powers approaching 2.5W have been achieved with an efficiency of 7%. Significant weakening of the focal intensity is required to avoid crystal damage, and these results are therefore obtained with a cautious focussing arrangement which has not yet been optimized. Some mean power effects have already been seen, for the crystal needs to be retuned after a period of illumination, and the importance of beam path covers even outside the laser head has been demonstrated. Nonetheless, a full investigation of the dependence of harmonic generation upon laser power and focussing arrangement has still to be carried out.

In the high power regime of second harmonic generation, where a reduction in the strength of focussing is required, the use of elliptical focussing may prove particularly valuable, and theoretical, computational and experimental investigations might all be attempted. In the optimally focussed case, experimental verification of the

predictions of chapter V has yet to be performed.

Holography using copper vapour laser illumination is as yet untried, except for preliminary attempts at holographic replication which is in any case particularly tolerant of poor coherence. It has been demonstrated that the longitudinal coherence of the copper laser should allow holograms of objects measuring a few inches in diameter to be made, and the high repetition rate of the CVL offers great potential for multiple exposure holograms or even holographic "movies" for diagnostic applications.

The high resolution dye laser remains only partially developed. The demonstration of transform limited bandwidths with similar lasers using lower repetition rate sources should provide a spur to the elimination of dye and vibration induced drift and the investigation of longitudinal pumping. The further possibilities of using titanium-doped sapphire as the gain medium should also be explored, and indeed the applications of copper laser pumped dye and Ti:sapphire lasers for resonance ionization mass spectrometry have already become the subject of a collaborative study involving the Clarendon Laboratory Laser Group.

IX-3. A novel CVL-pumped femtosecond pulse source.

As a final, and rather speculative, suggestion, a possible source of millijoule femtosecond pulses at around 800nm and kilohertz repetition rates is proposed. The system, illustrated in figure IX-1, is based upon the soliton laser of Mollenauer [IX-1], whereby short pulses from a colour centre laser were resolved into solitons by a length of optical fibre. A single soliton then seeded the next pulse from the laser. The brevity of the resulting pulses was given by the Fourier transform of their bandwidth, and was thus limited by the bandwidth of the colour centre laser; the pulse shape followed the classic sech^2 function of the soliton.

In this application, the broad bandwidth of Ti:sapphire is used to permit pulse durations which may in principle fall below 1fs, and the pulse repetition frequency of the copper vapour laser is matched to the round trip time of the soliton through the optical fibre. The lifetime of the upper laser level and high single pass gain of titanium-doped sapphire then allow the whole pulse energy to be extracted in a single pass of the soliton pulse. However, for solitons to form in the optical fibre, negative group velocity dispersion must be shown. For silica fibres, this restricts the radiation in the fibre to wavelengths beyond $1.5\mu\text{m}$, and a nonlinear crystal is therefore used to introduce a factor of two in frequency between the beams in the laser and in the fibre. A doubling in frequency is performed as second harmonic generation; a halving occurs through optical parametric amplification. A relatively short pulse of an appropriate frequency from a separate Ti:sapphire laser provides the high pump

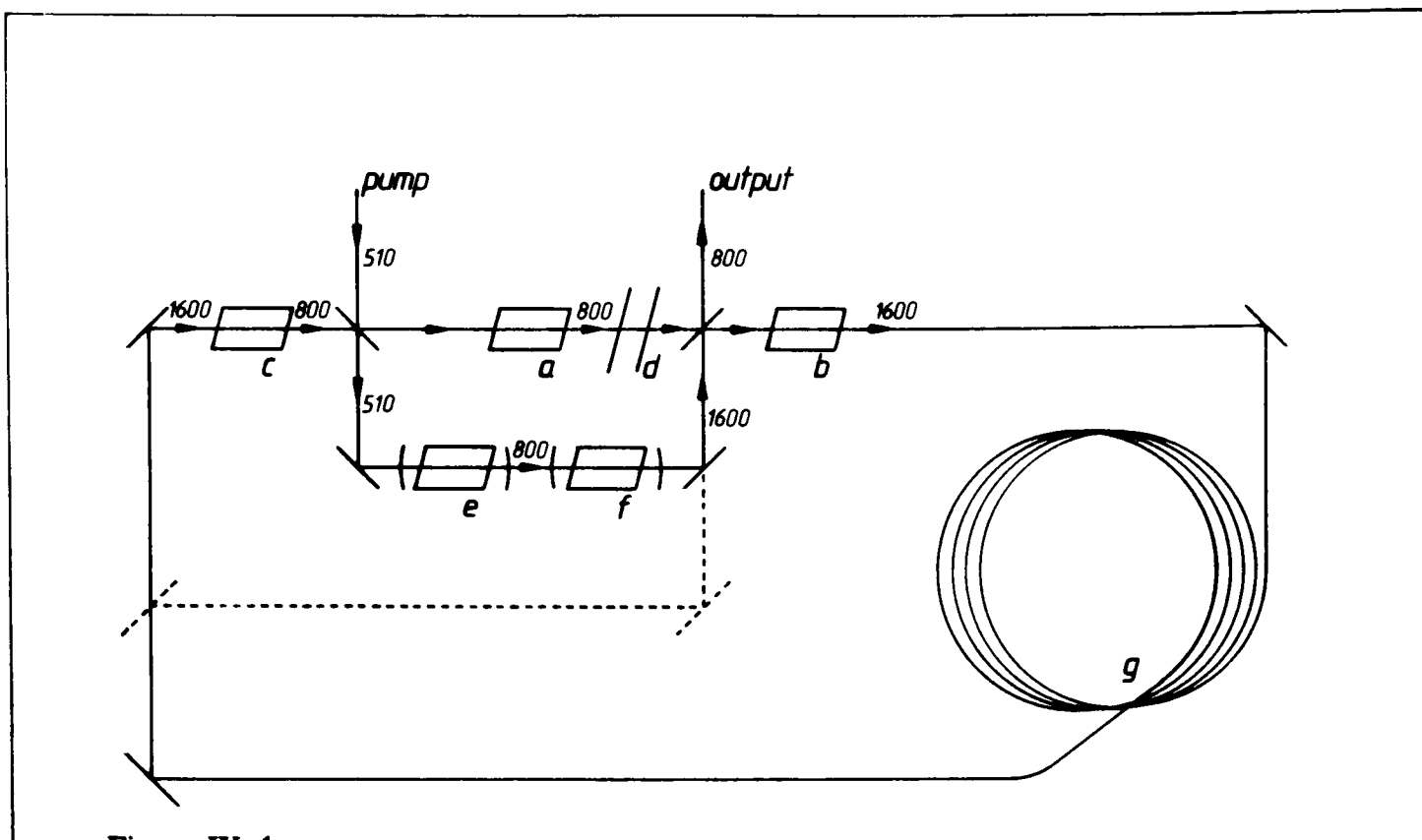


Figure IX-1.

Proposed source of millijoule femtosecond pulses with kilohertz repetition rate. A rod of Ti:sapphire, **a**, is longitudinally pumped by the copper laser, yielding pulses of light at around 800nm which are parametrically converted to 1600nm by the nonlinear crystal **b** before entering the loop of optical fibre **g**, where the long pulse is dispersed into solitons. These are then converted back to 800nm by second harmonic generation by the second nonlinear crystal **c**, to be amplified at **a**. Appropriate adjustment of the period between successive pump pulses allows a single soliton to be amplified. The second Ti:sapphire laser **e** and optical parametric oscillator **f** provide a source of long pulse length radiation at 1600nm as an idler for the optical parametric amplifier **b**. Dispersion compensation components **d** will also be required.

intensity for the parametric amplifier and allows any self-frequency-shift of the pulse in the fibre to be cancelled. Compensating components correct any dispersion introduced.

IX-References.

1. L F Mollenauer, R H Stolen, "The soliton laser," Opt Lett 9 (1) 13-15 (1984);
L F Mollenauer, "Soliton optical fibres: an experimental account," 8th National Quantum Electronics conference, St. Andrews, 1987.