

Investigations of Short-Pulse, Ultraviolet Laser Interactions with Gas Targets

A Thesis Submitted for the degree of Doctor of Philosophy

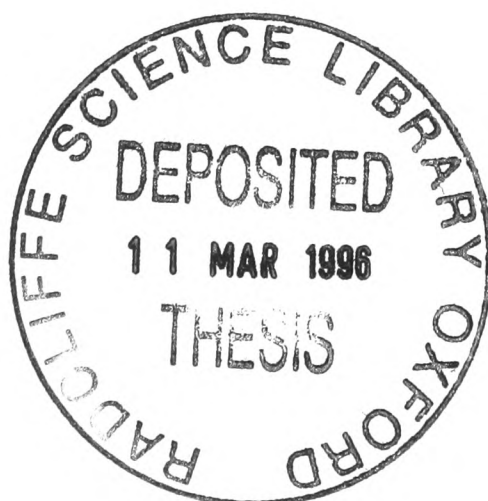
by

Stephen Gareth Preston

Linacre College

Oxford

Michaelmas Term, 1995



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Abstract

This thesis presents investigations of interactions of a 380 fs, 248.6 nm KrF laser beam focused to intensities up to 10^{18} W/cm² with gas targets of Helium and Neon. At these intensities, the targets are rapidly ionised by the process of optical field ionisation. The purpose of these investigations is two-fold. Firstly, we determine the relative importance of plasma heating mechanisms and investigate the scaling of electron temperature with density to ascertain whether an optically-ionised XUV recombination laser, utilising the 3s-2p transition in a Lithium-like ion, is viable. Such a laser requires, for the case of Neon, the production of very cold (<50 eV) electron temperatures at electron densities of the order of 5×10^{20} cm⁻³, and optical-ionisation to the Helium-like stage of Neon (Ne⁸⁺). The results are compared and contrasted to computer simulations of the electron temperature. Secondly, we investigate the process of harmonic generation. We show that, contrary to commonly held beliefs, it is possible to generate shorter harmonic wavelengths from ions using a short wavelength laser than from atoms using longer wavelengths. We also show high harmonic generation efficiencies from neutral Helium at lower intensities ($\sim 10^{15}$ W/cm²) away from the laser focus. The results are compared to, and found to be matched well by, simulations which predict the single atom/ion harmonic response.

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Chapter One

Introduction

What is a plasma? What is a laser? How does a laser produce a plasma? And how does a plasma produce a laser? These are some of the questions that will be considered in this thesis. We will review some of the ways in which a plasma can be used to produce coherent extreme ultraviolet (XUV) radiation: either by creating a laser in its own right; or by increasing the frequency of an optical laser by the non-linear process of high harmonic generation.

1.1. The Plasma State.

The plasma state has been called the fourth phase of matter. Plasmas are created when enough energy is supplied to a material to allow the electrons (normally bound to the nucleus by the Coulomb force) to escape the atomic potential. This results in the formation of a fluid of negatively charged electrons and positively charged ions that interact with each other through electrostatic forces. Over 99% of matter in the universe exists in the plasma state [1], primarily in stars and in the vast expanses of interstellar space. Plasmas on earth are less common, existing naturally in the ionosphere, in flames and in lightning.

Plasmas can be produced in the laboratory by depositing a large amount of energy into a material. This can be achieved by electrical methods, or by focusing a high power laser onto the material. Laser produced plasmas can reach densities greater than solid density

($>10^{23}$ ions/cm⁻³), and temperatures of tens of keV (over 100,000,000 Kelvin). A great many uses have been developed for these plasmas, of which we shall list just a few.

Laboratory produced laser-plasmas provide useful information about the conditions that occur in stars and the sun [2,3], helping us to understand the processes that occur in a stellar core by observing light emitted from the photosphere. It may take thousands of years for a photon produced in the core to escape from a star, and so a detailed knowledge of plasma opacities and radiative transfer are essential.

Stars are driven by nuclear fusion processes. Another line of laser-plasma research is the production of clean energy by producing controlled fusion of light nuclei at high densities and temperatures [4-8]. These high densities can be produced by inertial confinement of a pellet of fuel. In this process, a pellet is irradiated either directly by the laser, or with soft X-rays produced by laser interaction with a Gold target. The outer layers of the pellet are ablated which causes the pellet core to be compressed (equal and opposite reactions), leading to appropriate temperature and density conditions for fusion.

Plasmas heated to high temperatures become strong thermal sources of incoherent X-rays [9]. Alternatively, conditions can be created in the plasma so that it acts as a gain medium, producing coherent X-ray laser radiation [10,11]. These plasma X-ray sources can be used for many applications including the study of shock wave propagation in crystals [12]; the study of plasma structure using the extended X-ray absorption fine structure (EXAFS) [13,14]; and imaging (biological) samples using X-ray microscopy [15], X-ray holography [16,17], and X-ray interferometry [18].

Another source of coherent X-rays is the interaction of high power lasers with neutral gas targets, where the non-linear process of harmonic generation has been used to generate radiation down to about 70 Å [19,20]. The work in this thesis demonstrates the

generation of harmonics from ions in a plasma, producing wavelengths below 70 Å [21].

1.2. Laser development: higher powers, shorter pulses.

1.2.1. Increases in laser powers.

The field of laser-plasma research has evolved very rapidly over the last few years. This development has been primarily the result of a rapid increase in the laser intensities available to experimenters. Pulsed laser power has increased over the last few decades as a series of sharp jumps following the introduction of new technologies. This is shown schematically in figure 1.2.1. The earliest lasers utilised free-running cavities, in which laser action started as soon as the gain overcame cavity losses. This is typified by the first laser system, based on a flashlamp pumped ruby rod [22]. This could only produce peak laser powers of around a kilowatt per square centimetre of beam area. The method of Q-switching [23] was soon developed, which enabled large population inversions to build up before the cavity was 'activated' and lasing was allowed to occur. This took peak powers up beyond the megawatt/cm² level. In the early 1970s came mode-locking (which can be thought of as very high speed Q-switching) [24], taking powers to the gigawatt/cm² level. Finally, we have seen the introduction of chirped pulse amplification (CPA) [25], which is pushing laser powers into the terawatt/cm² and petawatt/cm² regime.

The intensity of a pulsed laser is dependent on three factors: the divergence of the beam, the pulse length, and the pulse energy. Beam divergence is a measure of the spot size we can focus the beam down to. Obviously, the smaller the focal spot, the greater the focused intensity. To generate a focal spot that is as small as possible we must produce a clean Gaussian (TEM₀₀) beam. Any additional modes in the Gaussian beam or phase

variations introduced by imperfect optical components in laser amplifier train will increase this spot size and lower the focused intensity. The radius, r , of a diffraction-limited focal spot size is given by $r = 1.22 f^\# \lambda$ where $f^\#$ is the f-number of the focusing lens and λ is the laser wavelength. Obviously, this radius reduces with decreasing wavelength, and so (with all else being equal) a diffraction limited 248 nm Krypton Fluoride laser will be brighter than a diffraction limited 1053 nm Neodymium:Glass laser.

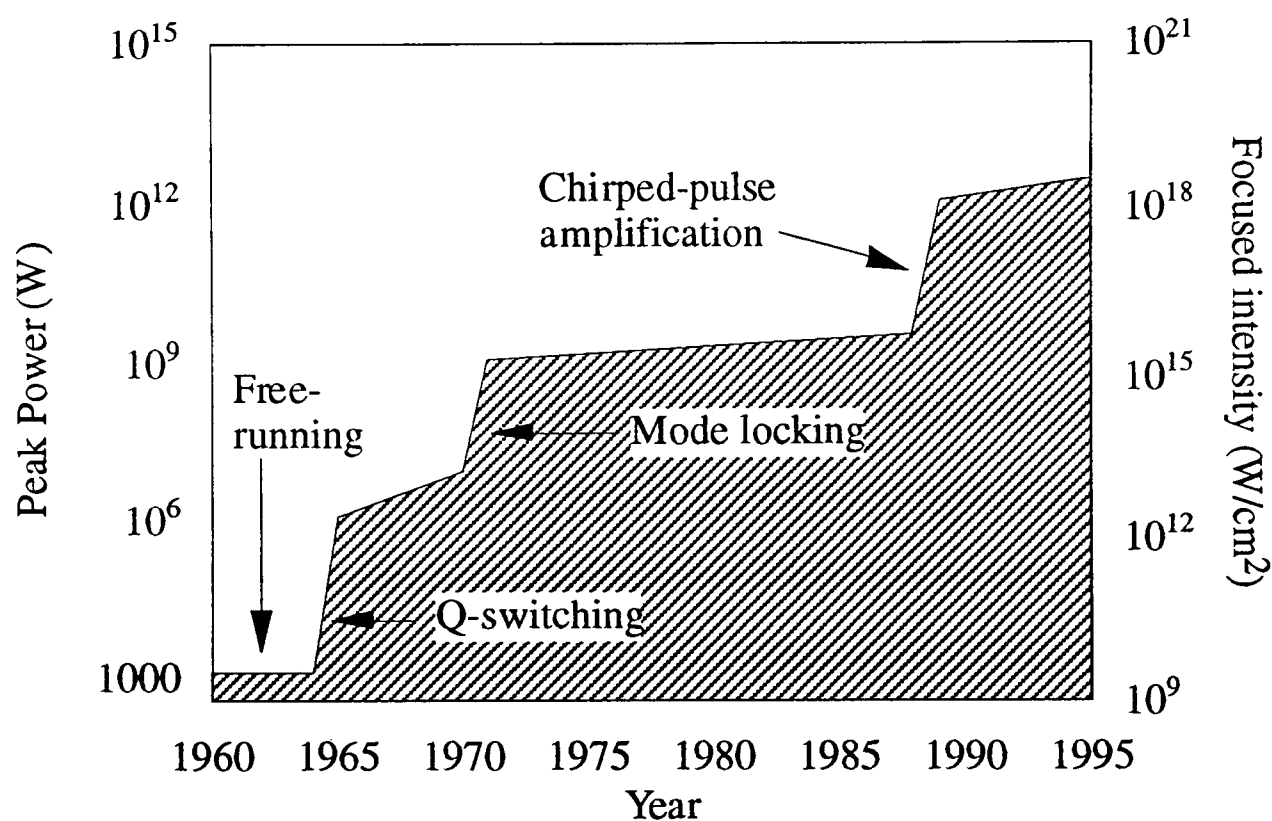


Figure 1.2.1. Schematic diagram of the development of pulsed laser power and focused intensities of a small-scale (1 cm^2) beam. The theoretical limit is slightly over 10^{21} W/cm^2 . (Taken from Ref. [19]).

The pulse length is ultimately limited by the gain bandwidth of the lasing medium. From the theory of Fourier transformation, we expect a medium with a gain bandwidth of $\Delta\omega$ to be capable of producing a transform limited pulse length of $\Delta\tau \approx 1/\Delta\omega$. When trying to propagate a short pulse through a laser amplifier, this theoretical pulse length will be increased by dispersion and non-linear effects (such as self-phase modulation) in the laser medium, and even by gain narrowing in the amplifier.

Moving to shorter and shorter pulses, we find that the extraction of energy from the laser becomes increasingly difficult. This is because the input fluence necessary for efficient extraction must be of the order of the saturation fluence, given by $F_{sat} = \hbar \omega / \sigma$ (where $\hbar$ is Planck's constant, ω is the laser frequency, and σ is the emission cross section). The saturation fluence in most solid state laser materials is between 1 J/cm² (Ti:Sapphire) and 6 J/cm² (Nd:Glass) and is 2×10^{-3} J/cm² for Krypton Fluoride (KrF) gas. Amplification of a picosecond or sub-picosecond pulse to such a fluence is not possible because of the intensity dependence of the refractive index ($n = n_0 + n_2 I$) in the laser material and optical components. This leads to filamentation and self-focusing of the laser pulse and the destruction of the laser amplifier.

1.2.2. Chirped pulse amplification.

Until the advent of chirped pulse amplification (CPA), laser energies were restricted by the limitations described above. The CPA process takes a very short (sub-picosecond), high bandwidth, low energy (nanoJoule) pulse from a mode-locked oscillator and stretches its pulse length using a grating pair (or a single grating and mirror configuration) to up to several nanoseconds. The gratings will have typically 3600 grooves/mm or more. The pulse can then be amplified to very high energies while keeping the intensity low enough to avoid non-linear effects in the components of the laser. The amplified pulse can then be recompressed to its original pulse length by another grating pair and focused on target. The limit to the intensities that can be achieved is set by the energy density that the recompression gratings can withstand. For example, the Sprite KrF CPA laser system [26] at the Rutherford Appleton Laboratory is capable of amplifying a chirped pulse to energies up to 2 or 3 joules, but the Aluminium coated compression gratings can only withstand around 600 mJ in a 10 cm diameter beam. It is possible to increase the size of the gratings and the beam, but this leads to problems with producing gratings cheaply with sufficient flatness that the beam

divergence is not degraded, especially when using 248 nm light. A fuller description of the Sprite laser CPA system can be found in section 4.1.2.

The Vulcan 1053 nm Nd:Glass laser system at the Rutherford Appleton Laboratory has also been converted to CPA mode [27] and typically delivers about 15 - 20 J on target in 0.8 - 1.0 ps and at intensities up to 10^{19} W/cm². At these intensities the electron motion in the laser field is highly relativistic. Figure 1.2.2 gives a brief summary of some of the physical phenomena that occur at different focused laser intensities. The upper limit to focused intensities that are currently being produced are of the order of 10^{19} W/cm².

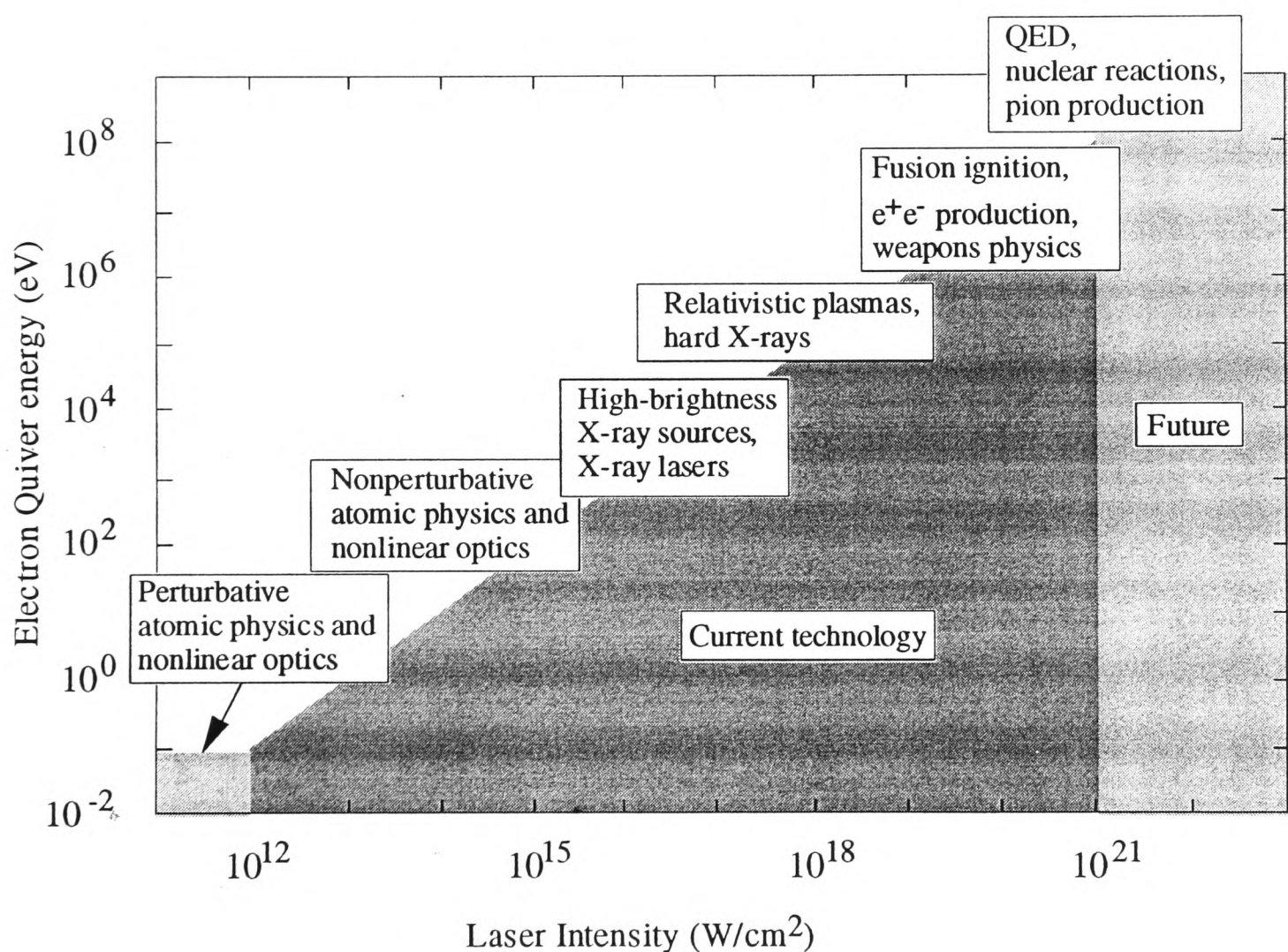


Figure 1.2.2. Schematic diagram of the electron quiver energy and accessible phenomena as a function of laser irradiance for a wavelength of 1053 nm. QED is quantum electrodynamics, e^+e^- is positron-electron production. (Taken from Ref. [19]).

1.3. The scope of this thesis.

As we have already discussed, laser intensities and powers have been constantly increasing since the development of the first laser in 1960 [22]. The shortest demonstrated laser pulse length is now of the order of 8 fs [28]. The available focused laser intensities have been increasing in step with pulse length developments, with intensities up to 10^{19} W/cm² now becoming available [26,27]. This rapid development is causing constant revision of what is considered to be a short pulse or a high intensity. Definitions also vary considerably from one field of research to another: those studying inertial confinement fusion (ICF) would consider 1 ns to be a short pulse, and 10^{16} W/cm² to be a high intensity; whereas in the field of optical ionisation 100 ps is a long pulse, and 10^{16} W/cm² is quite a low intensity. The experiments described in this thesis are conducted at pulse lengths of 200 - 400 fs, and at intensities mostly in the region of 10^{16} - 10^{18} W/cm². We can arbitrarily use the following approximate definitions:

Long pulse: 10 ps or longer.	Low intensity: less than 10^{16} W/cm ² .
Short pulse: 100 fs - 1 ps.	High intensity: 10^{16} - 10^{18} W/cm ² .
Ultra-short pulse: less than 100 fs.	Ultra-high intensity: 10^{18} W/cm ² or higher.

Plasma densities can also vary from greater than solid densities (in ICF for example) to the extremely low densities of interstellar plasmas. For laser-plasma interactions, we can classify the plasma as to whether the laser radiation we are using is able to propagate through the plasma. A plasma is underdense if the plasma resonant frequency (the electron plasma frequency - see chapter two) is less than the laser frequency: in this case the laser radiation can propagate. A plasma is overdense if the electron plasma frequency is greater than the laser frequency: in this case the refractive index becomes wholly imaginary and the laser radiation is absorbed over a very short scale length.

1.3.1. A brief note on units.

The units used in this thesis are something of a mixture. They are generally chosen to match the units that are used by the laser-plasma physics community. For example, we list below some of the more commonly used units:

Density - g/cm³.

Number density - cm⁻³.

Distance - microns (μm).

Intensity - W/cm².

Temperature - electron volts (1 eV = 11,594 Kelvin).

Energy - eV.

Wavelengths - nanometres (nm) for optical radiation, Ångstroms for XUV radiation (10 Å = 1 nm).

Pressures are expressed depending upon the units they were measured in during the experiments, so static gas fill pressures are in torr (760 torr = 1 bar), whereas gas jet backing pressures are expressed as bar gauge (gauge means above atmospheric pressure) or as psi gauge (15 psig = 1 bar gauge).

An attempt has been made to express equations in S.I. units wherever possible. Equations should be considered to be in S.I. unless it is specifically stated otherwise in the text.

1.3.2. The parameters for this thesis.

In this thesis we study plasmas with electron densities in the range from 10¹⁶ - 10²¹ cm⁻³. The critical electron density above which 248 nm light (the wavelength of

our laser) cannot propagate is $1.8 \times 10^{22} \text{ cm}^{-3}$ and so our plasmas are underdense. In the case of a plasma of eight times ionised Neon, the electron densities we work with would correspond to original gas pressures (at room temperature) of about 1 mbar to 100 bar.

A gas at room temperature has a temperature of $\sim 1/40 \text{ eV}$. The plasmas we will study will have electron temperatures that range from the order of 5 eV to over 1000 eV. In contrast, the ion temperatures of the plasmas produced by optical-ionisation do not exceed more than about 1 eV during the time of the laser pulse.

1.3.3. The role of the author.

This thesis contains a number computer simulations to support, and to help interpret, the experimental data that is presented. These simulation programs were not developed by the author, and so it is necessary to acknowledge here the work performed by others which is included in this thesis. The MEDUSA plasma simulation code used in chapter five was modified for the short pulse regime by Dr A. Djaoui of the Rutherford Appleton Laboratory, but the simulations presented were carried out by the author. The Thomson scattering simulation code used in chapters four and five was written by Dr J.S. Wark and modified by Dr W.J. Blyth of the Clarendon Laboratory. Again the actual simulations were performed by the author. The harmonic generation simulations presented in chapter six were performed, at the request of the author, by Dr A. Sanpera of the Clarendon Laboratory. However; the method of weighting the simulation to match experiment was developed by the author.

The experiments presented in this thesis were, of necessity, performed by a team. The author took part in the planning and running of the Thomson scattering experiment described in chapters four and five under the direction of Dr W.J. Blyth [29]. The data was shared, with analysis of data taken from gas jet targets being primarily the

responsibility of the author. Some of the Thomson scattered data shown in chapter five (particularly the Bohm-Gross fits in Helium and the Gaussian fits in Neon) were originally presented in the thesis of Dr W.J. Blyth [29]. It is necessary to present the data again to give a detailed account of the overall scaling of electron temperature over the whole range of electron densities studied. The other Thomson scattering data presented are a continuation of this work by the author. The author was responsible for the planning, running, and analysis of the harmonic generation experiment.

1.3.4. Thesis overview.

The work in this thesis is divided into seven chapters, including this introduction.

Chapter two discusses the physical processes involved in the production and evolution of an underdense plasma, including a description of how the plasma is formed by optical ionisation, how the propagation of the laser is affected by the presence of the plasma, and the mechanisms which lead to heating of the plasma.

Chapter three deals with methods of producing coherent XUV radiation. This includes a brief description of collisional and recombination X-ray laser schemes, and optically-ionised X-ray laser schemes. The non-linear process of harmonic generation is then discussed in detail.

Chapter four describes the experiments and diagnostics used to obtain the data presented in chapters five and six. It also discusses the role of Thomson scattering as a plasma diagnostic.

Chapter five presents calculations of the expected variation of electron temperatures with electron density in Neon. Electron temperature measurements in Helium and Neon

derived from Thomson scattering and stimulated Raman scattering data are shown and compared to the simulations. Results at the higher densities are supported by Faraday cup and X-ray spectra.

Chapter six presents simulations and experimental data of harmonic generation from Helium and Neon plasmas.

Chapter seven summarises the conclusions from the two experimental chapters and suggests ways of extending this work.

Chapter Two

Production and Evolution of an Underdense Plasma

The purpose of the work described in this thesis is to determine the viability of optically-ionised X-ray laser schemes and of harmonic generation from ions in a plasma as a means to produce coherent XUV radiation. For this reason it is necessary to determine the mechanism by which a plasma is created by optical ionisation, and how its interaction with the laser beam will affect the plasma at a later time. In this chapter we describe the production of a plasma from a gas using intense laser radiation. We will then go on to consider the interaction of the plasma with the laser field, how this affects the density and temperature of the plasma, and how it affects the propagation of the laser pulse in the plasma. Determining such an evolution is not trivial, as the state of the plasma and laser beam at any time are governed by the history of interactions between the laser and the plasma up to that time.

We will start with a brief overview of how waves propagate in a plasma during which some important plasma parameters will be introduced; such as the electron plasma frequency, the Bohm-Gross frequency and the Debye length. Then we will discuss the formation of a plasma by optical ionisation and consider the heating of electrons by above threshold ionisation (ATI), inverse bremsstrahlung (IB), stimulated Raman scattering (SRS), and ponderomotive heating. We will then discuss how the focusing of a laser is affected by the presence of a plasma (as opposed to a vacuum).

2.1. Overview.

On a macroscopic level the behaviour of a plasma is that of a fluid in which electromagnetic or electrostatic waves can propagate. The dispersion relation of a transverse electromagnetic wave in a plasma is modified from that in a vacuum by the refractive index of the free electrons in a plasma that have a characteristic 'resonant' frequency: the plasma frequency, $\omega_{pe} = (Ne^2/\epsilon_0 m)^{1/2}$, where N is the electron density, and m is the electron mass. This modified dispersion relation is of the form

$$\omega_0^2 = \omega_{pe}^2 + k^2 c^2 \quad (2.1.1)$$

where ω_0 is the frequency of the electromagnetic wave, c is the speed of light, and k is the wavevector of the electromagnetic wave. This dispersion relation, plotted in figure 2.1.1.a, implies a plasma refractive index, given by

$$n = \left(1 - \frac{\omega_{pe}^2}{\omega_0^2} \right)^{1/2} \quad (2.1.2)$$

where in the limit $\omega_0 \gg \omega_{pe}$ the refractive index approaches unity. If $\omega_0 < \omega_{pe}$ however, the refractive index becomes purely imaginary and no light can propagate in the plasma. The electron density at which the refractive index becomes zero and above which it would become imaginary defines the critical plasma electron density,

$$N_{cr} = (\epsilon_0 m / e^2) \omega_0^2 \quad (2.1.3)$$

Electron or ion density fluctuations in the plasma produce longitudinal plasma waves. Since the electrons and ions are charged particles, these plasma waves may interact

strongly with electromagnetic waves. The electron plasma waves follow a dispersion relation (shown in figure 2.1.1.b) given by

$$\omega_{BG}^2 = \omega_{pe}^2 + \left(\frac{3\kappa T_e}{m} \right) k^2 \quad (2.1.4)$$

where k refers to the wavevector of the electron plasma wave, T_e is the electron temperature, κ is Boltzmann's constant, and ω_{BG} is referred to as the Bohm-Gross frequency of the electron plasma wave. The second term in equation (2.1.4) is a temperature correction to the electron plasma frequency, $(\kappa T_e/m)^{1/2} = v_{th}$ - the thermal electron velocity. (It is necessary to include this thermal correction, because a finite electron velocity results in information being carried through the plasma, i.e. we have a travelling wave rather than a standing wave). At very high electron temperatures the thermal term dominates and the velocity of the electron plasma wave asymptotically approaches $\sqrt{3}v_{th}$. Ion waves propagate with much lower frequencies than electron waves. This is because of the much greater mass of the ion than an electron. For this reason, ion waves in the plasma are referred to as ion-acoustic waves. This slow response time of the ions is the reason that we need not correct the electron plasma wave dispersion for the motion of the ions. This is not true for the ion-acoustic wave dispersion which is affected by motion of the electrons. The ion-acoustic wave dispersion, shown in figure 2.1.1.c, is given by

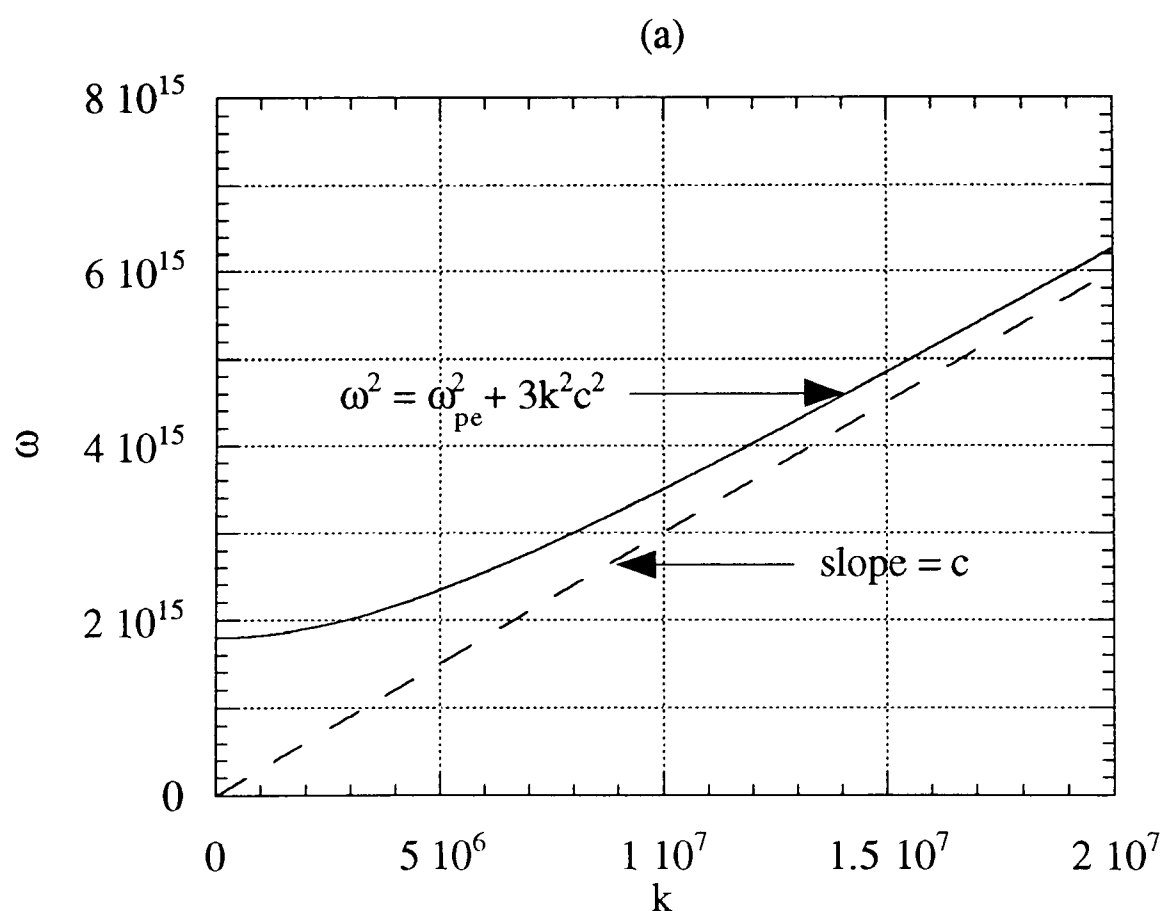
$$\left(\frac{\omega_{ki}}{k_i} \right)^2 = \frac{Z\kappa T_e}{m_i(1 + k_i^2 \lambda_D^2)} + \frac{3\kappa T_i}{m_i} = v_{ia}^2 \quad (2.1.5)$$

where ω_{ki} is the ion-acoustic frequency, k_i is the ion-acoustic wavevector, m_i is the ion mass, and v_{ia} is the ion-acoustic velocity. As the wavevector of the ion-acoustic wave increases the ion-acoustic frequency approaches a fixed value of $\omega_{pi} = (ZNe^2/\epsilon_0 m_i)^{1/2}$.

Like electromagnetic waves, plasma waves can be expressed in terms of either classical waves or in terms of particles. The plasmon is the quantised 'particle' of a plasma wave, just as the photon is the quantised 'particle' of an electromagnetic wave, or a phonon is a quantised 'particle' of a normal mode of vibration. We use the wave description when we are describing a process classically. We use the particle description when considering quantised processes.

The term $\lambda_D = \left(\frac{\epsilon_0 k T_e}{N e^2} \right)^{1/2}$ (2.1.6)

is the Debye length, and defines a shielding length in the plasma. If a positively charged particle is introduced into the plasma, the electrons will be attracted to it and will shield its charge. The Debye length is the distance over which the electric field of a positively charged particle will be reduced to $1/e$ of the magnitude it would be in a vacuum.



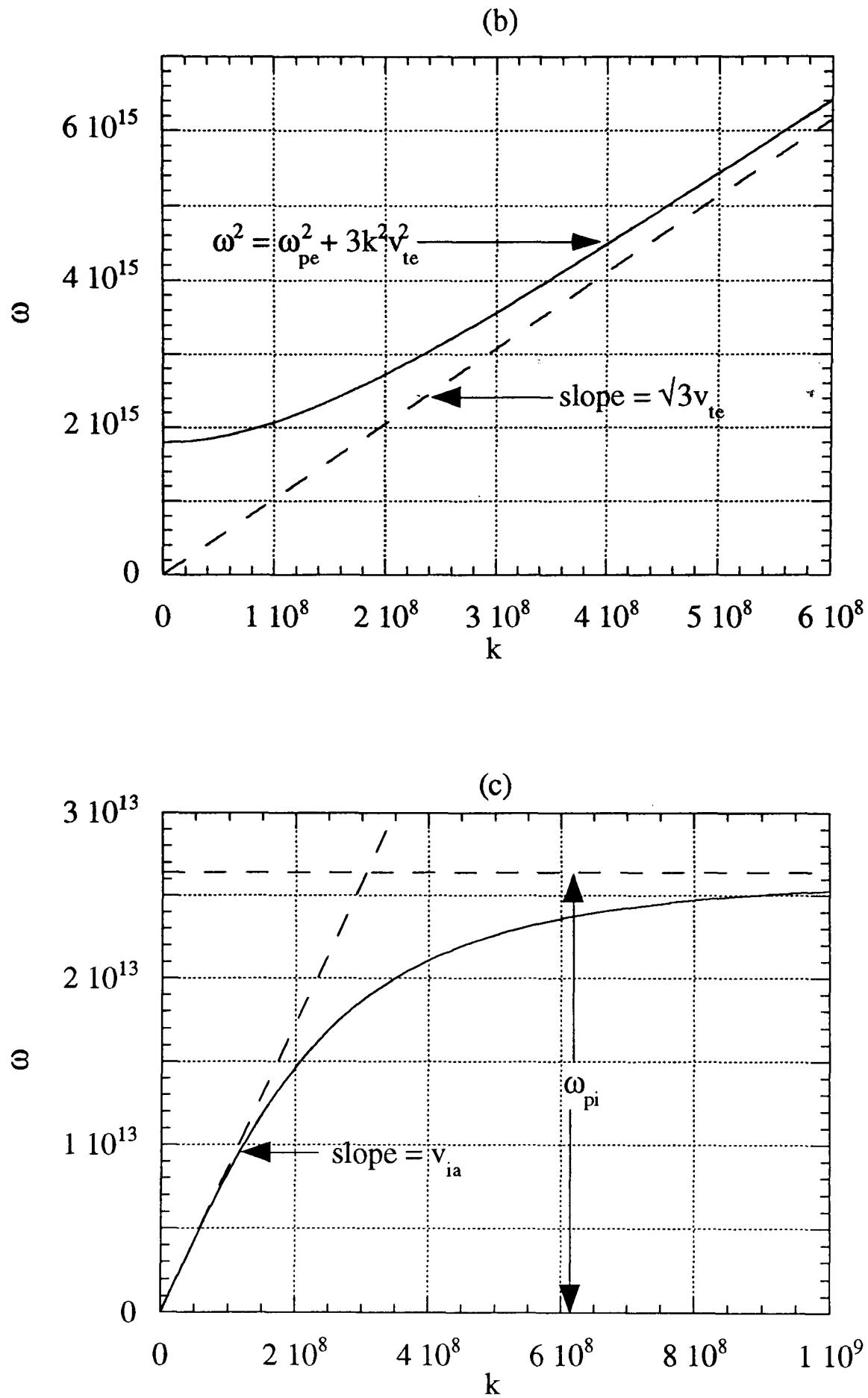


Figure 2.1.1. Dispersion curves for (a) electromagnetic waves, (b) electron-plasma waves and (c) ion-acoustic waves in a plasma of He^{2+} with $N = 10^{21} \text{ cm}^{-3}$, $T_e = 200 \text{ eV}$, $T_e/T_i = 100$.

2.2. Optical ionisation.

There are two methods by which we may create a plasma with a laser: optical ionisation and collisional ionisation. In the intensity and density regime we are considering (linearly polarised laser light at intensities greater than about 10^{16} W/cm² interacting with gas densities of 10^{16} - 10^{20} atoms/cm³), optical ionisation is the dominant ionisation mechanism for any charge state that has a threshold intensity for ionisation which is below the maximum laser intensity. However, if free electrons in the plasma gain sufficient energy from inverse bremsstrahlung, Landau damping of plasma waves, or from optical ionisation using circularly-polarised light, then collisional ionisation and excitation of the higher ionisation stages can occur. This will be shown in section 5.5.2.

Optical ionisation occurs when the force exerted on an electron by the electric field of the laser reaches the same order of magnitude as, or becomes greater than, the Coulomb force binding the electron to the atom. Consider a laser of frequency ω_0 and a ponderomotive potential (or time averaged energy of an electron oscillating in the laser field - see section 2.3.1) of $U_P = e^2 E_0^2 / 4m\omega_0^2$ (where e is the charge on an electron, E_0 is the magnitude of the laser electric field, and m is the mass of an electron) interacting with an atom with an ionisation potential of U_I . As the laser field increases in intensity, optical ionisation passes through three regimes [30] :

- (i). $U_I > \hbar\omega_0 \gg U_P$, where multiple order perturbation theory may be used, and we may look on the ionisation as being due to the absorption of a number of photons by the electron. In this case the laser field is a small perturbation on the atomic field.
- (ii). $U_I > U_P > \hbar\omega_0$, where perturbation theory breaks down and we must develop a theory of quantum mechanical tunnelling. Here the laser field is of the same order as the atomic field.

(iii). $U_P > U_I \gg \hbar\omega_0$, the regime of classical tunnelling, or 'over the top' ionisation. The laser field is now greater than the atomic field, and electrons are free to move out of the atomic potential over the top of the suppressed atomic potential which is now lower than the (formerly bound) energy level of the electron.

2.2.1. Tunnel ionisation.

Ionisation of the systems we will study is dominated by tunnel ionisation (either classical or quantum mechanical). Such a situation is depicted in figure 2.2.1 which shows an instant in the laser cycle when the laser field is non-zero. Here we see a Coulomb potential modified by the electric field of the laser. In the classical regime (A) the atomic potential is suppressed far enough that the electron is free to leave the atom. In the tunnelling regime (B) the electron must tunnel through the finite potential barrier. When the laser field is strong enough to reach the classical tunnelling regime then this mechanism will dominate.

As a simple estimate of the threshold intensity for optical ionisation we may equate the ionisation potential, U_I , of an atomic state to the potential of the saddle point in a hydrogenic potential modified by the laser field. This defines a critical electric field below which 'over the top' ionisation will not occur, given by

$$E_c = \frac{U_I^2}{4Ze^3} \cdot (4\pi\epsilon_0) \quad (2.2.1)$$

where Z is the charge state of the final state ion, and E_c is the critical laser field. This corresponds to a critical optical intensity of

$$I_c = \frac{U_I^4}{32Z^2e^6} \cdot (4\pi\epsilon_0)^2 \epsilon_0 c \quad (2.2.2)$$

For the case of Hydrogen, $I_C = 1.4 \times 10^{14} \text{ W/cm}^2$.

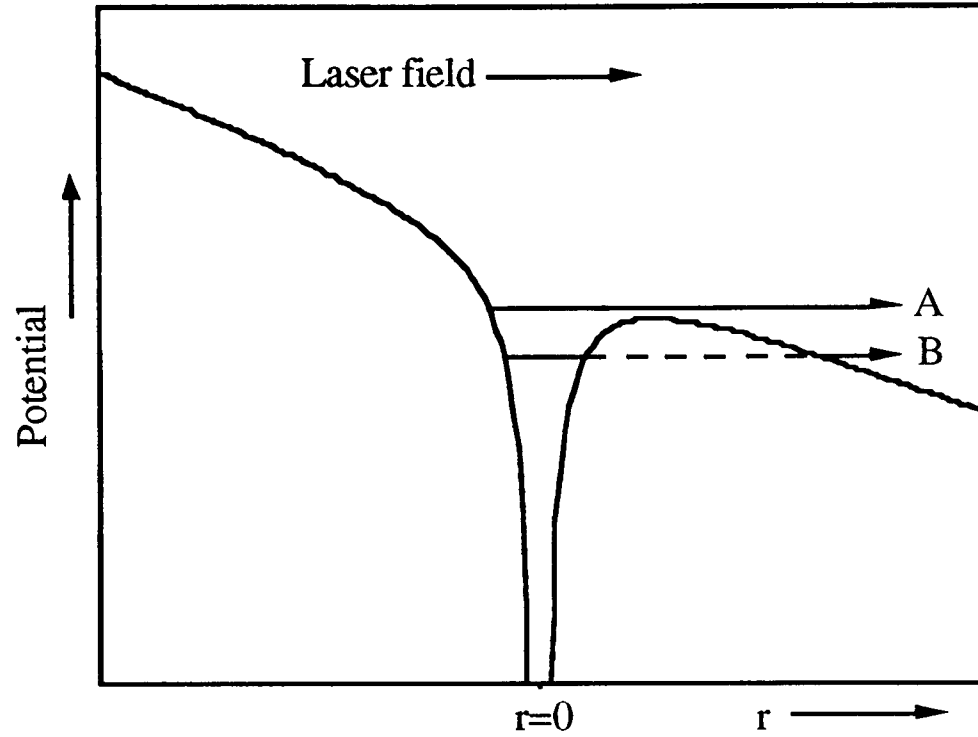


Figure 2.2.1. Schematic diagram of classical (A) and tunnelling (B) ionisation.

2.2.2. The Ammosov equation.

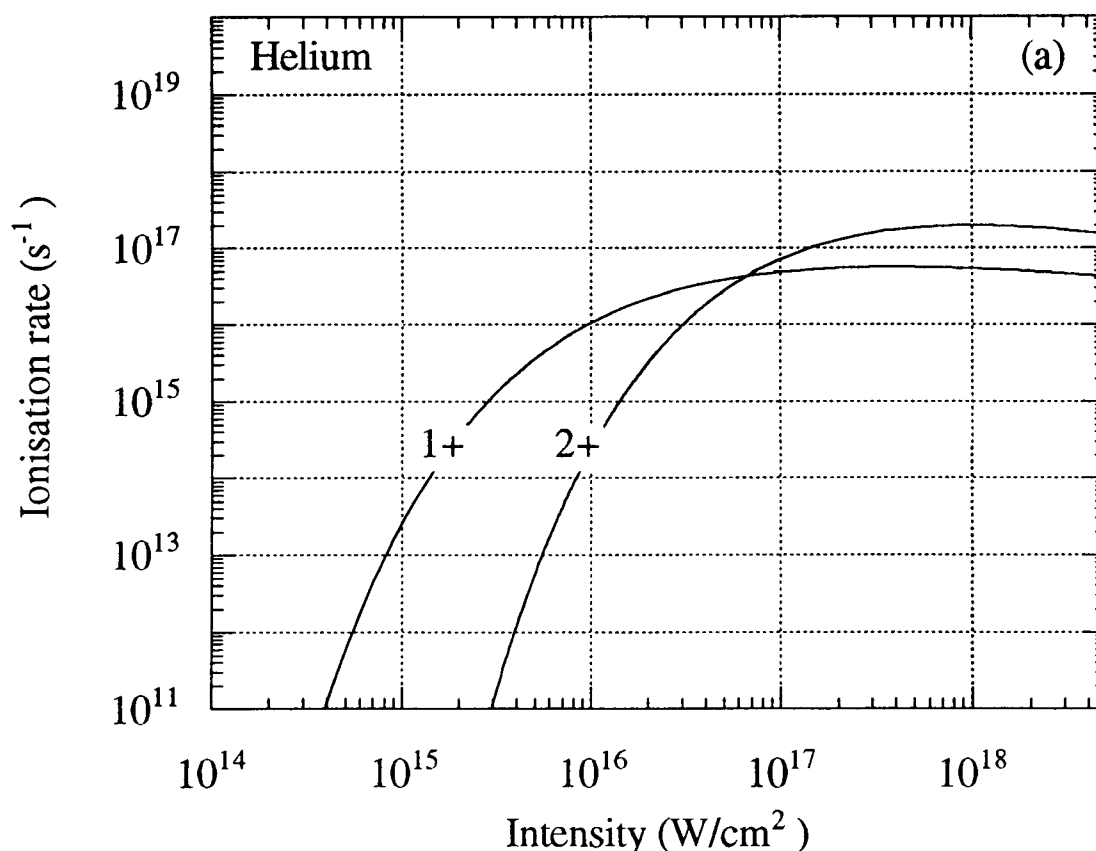
A more accurate expression for the cycle averaged ionisation rate in a laser field which agrees well with experiment [31,32] is that derived by Ammosov et al. [33]. This is given (following the form of Rae [34]) by

$$\begin{aligned}
 R = & 1.52 \times 10^{15} U_I \left(\frac{5.44}{n^*} \right)^{n^*} \left(\frac{1}{2\pi n^*} \right)^{1/2} \frac{(2l+1)(l+|m_l|)!}{2^{|m_l|} (|m_l|)! (l-|m_l|)!} \\
 & \times \left[7.47 \times 10^6 \frac{U_I^{3/2}}{I^{1/2}} \right]^{2n^* - |m_l| - 1} \exp \left[-2.49 \times 10^6 \frac{U_I^{3/2}}{I^{1/2}} \right]
 \end{aligned} \tag{2.2.3}$$

Where l and m_l are the quantum numbers for the initial state, $n^* = 3.69Z(U_I)^{1/2}$ is the effective principle quantum number, U_I is the ionisation potential (in eV) of the atom or

ion concerned, I is the laser intensity (in W/cm^2), and Z is the ionisation stage of the ion produced.

Applying this equation to the cases of Helium and Neon, which are relevant to the experimental work described in this thesis, gives the ionisation rates shown in figure 2.2.2. A simple way to obtain an approximate intensity at which a given laser pulse length will cause complete ionisation of a particular charge state is to find the laser intensity at which the ionisation rate is equal to the inverse of the laser pulse length. This shows that Helium is fully ionised in a 380 fs pulse at intensities greater than $7 \times 10^{15} \text{ W}/\text{cm}^2$ and that the maximum ionisation stage reached in Neon is Ne^{8+} for intensities up to $10^{18} \text{ W}/\text{cm}^2$.



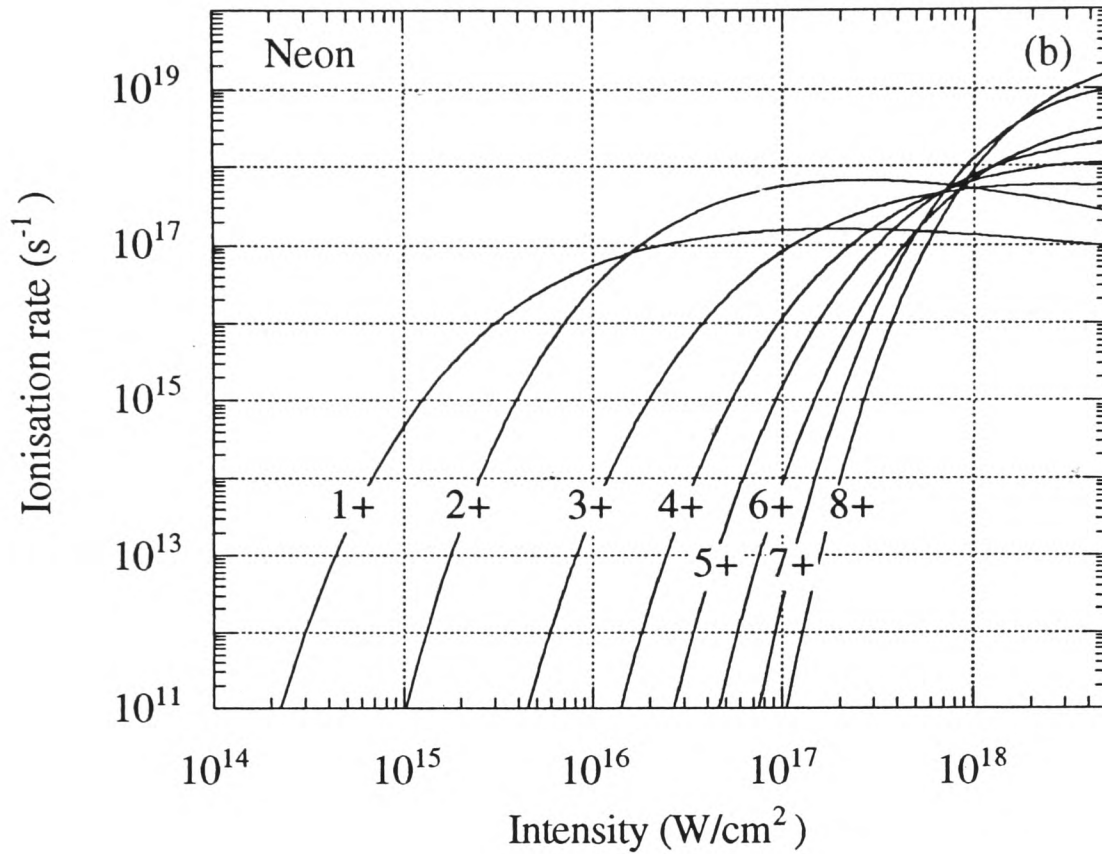


Figure 2.2.2. Ionisation rates in (a) Helium and (b) Neon calculated from the Ammosov formula. The labels refer to the ionisation state of the residual ion.

Optical ionisation is a sequential process at laser pulse lengths and intensities currently available, but it should be noted that the Ammosov model will break down if the laser intensity rises quickly enough that lower ionisation stages cannot fully ionise. In the case of Neon, for example, if the laser is instantaneously switched on at an intensity of 10^{18} W/cm², figure 2.2.2.b predicts that the first ionisation stage we meet will be Ne⁷⁺. This is obviously incorrect. For such a rapid onset of intensity we would expect the ionisation to become non-sequential. This extreme example emphasises that the Ammosov equation is only valid in the tunnelling regime.

2.3. Above threshold ionisation.

2.3.1. The effect of the laser field on free and bound electrons.

Before considering how optical ionisation can impart energy to the electrons as they are ionised, let us consider the effect of the laser field on free and bound electrons [30]. A free electron in the laser field possesses a ponderomotive (or quiver) energy due to its motion as it is driven by the oscillating electric field of the laser. A free electron in an electric field of magnitude $E = E_0 \sin \omega_0 t$ has a time dependent velocity, $v(t) = -v_0 \cos \omega_0 t$ where $v_0 = eE_0/m\omega_0$ (the quiver velocity). This gives an instantaneous kinetic energy of $(e^2 E_0^2 / 4m\omega_0^2)(1 + \cos 2\omega_0 t)$. Hence the time averaged kinetic energy, or ponderomotive energy, is $U_P = e^2 E_0^2 / 4m\omega_0^2$. This ponderomotive energy (although it is obviously a kinetic energy) does not contribute to the time averaged motion of the electron. In many situations it may simply be regarded as a potential energy that depends on the local laser intensity. The total energy of a free electron can then be divided between the ponderomotive energy and its energy of directed motion

$$U_{total} = U_P + \frac{1}{2} m \langle v \rangle^2 \quad (2.3.1)$$

where the brackets denote a time average over one laser cycle.

For a long laser pulse, an electron which is optically-ionised with a certain initial velocity $\langle v \rangle$ will travel out of the laser beam before there is any significant change in the laser electric field amplitude. During its passage the electron's total energy is conserved and so, as the electron leaves the laser focus, its ponderomotive energy is converted to directed kinetic energy. For a short laser pulse, however, the laser field has vanished before the electron has time to escape the focus. In this case the ponderomotive energy is returned to the photon beam (via the process of stimulated scattering).

High intensity laser fields not only affect free electrons, but also electrons bound in atoms. In general, all energy levels of an atom will be shifted to some degree by the a.c. Stark shift [30]. The magnitude and direction of this shift for a given electronic state depends on the intensity and frequency of the laser radiation. The atomic energy levels will obviously oscillate with the laser field. When referring to an energy level we really mean the time averaged energy level. The total energy of a high lying Rydberg state will be increased by approximately U_P , whereas the quiver energy of a low lying (deeply bound) state with a characteristic frequency ω_c in a laser of frequency ω_0 will be reduced from that of a free electron by ω_0/ω_c . The net result of this effect is that the ionisation potential of a bound state is increased by approximately U_P . This can be explained by considering that to ionise an electron we must take it from its bound state to a state quivering in the laser field with an energy U_P . The ionisation potential has therefore become $U'_I = U_I + U_P$ [30].

In long pulse experiments the ponderomotive increase in the ionisation potential decreases the initial directed energy of the electron, but this is countered by the conversion of the electron ponderomotive energy into directed energy as the electron travels out of the laser focus. Hence the shift in the ionisation potential is difficult to detect. The energy spectrum of the optically-ionised electrons can be predicted by an extension of the Einstein photoelectric formula to m photons of energy $\hbar\omega_0$,

$$U_{electron} = m\hbar\omega_0 - U_I \quad (2.3.2)$$

In short pulse experiments, however, the shift in ionisation potential is not masked, as the ponderomotive energy is not converted into directed motion. In this case the energy spectrum of the electrons is given by

$$U_{electron} = m\hbar\omega_0 - U'_I \quad (2.3.4)$$

where U_I' will vary across the laser focus.

2.3.2. Heating by above threshold ionisation.

Let us now consider how much energy can be imparted to an electron as it is optically-ionised. The experiments discussed in this thesis are performed in a regime in which classical suppressed barrier ionisation dominates. When the electron tunnels out of the Coulomb potential, it appears (classically) as a point particle with some initial (small) velocity close to the atomic core (we approximate this position to be at the atomic core, $z = 0$). A distribution in electron velocities is caused by either a spread in the initial velocities, or else a spread in the time of ionisation relative to the phase of the laser field. Consider again the case of a simple oscillating electric field, $E = E_0 \sin \omega_0 t$. An electron tunnels out of the atom into the field at time t_0 , with initial velocity v_i . It can be shown that its velocity at a later time is given by [30]

$$v(t) = -\left(eE_0/m\omega_0\right)(\cos \omega_0 t - \cos \omega_0 t_0) + v_i \quad (2.3.5)$$

The time averaged kinetic energy $\bar{U}$ of the electron in the electric field covers a continuous range

$$\bar{U} = U_p + \left(\sqrt{2U_p} \cos \omega_0 t_0 + \sqrt{\frac{m}{2}} v_i \right)^2 \quad (2.3.6)$$

which, if we neglect the initial velocity (v_i), has a maximum of $\bar{U}_{\max} = 3U_p$. This maximum ATI energy corresponds to the electron being ionised when the laser field is very close to zero. At this time in the laser period the probability of ionisation is almost negligible. The most probable time for ionisation is at the peak of the laser field, where $\omega_0 t_0 = \pm \pi/2$, and the time averaged kinetic energy of the electron will be $U_p + mv_i^2/2$.

This is as we would expect from an electron liberated from an atom with a light-induced ionisation potential shift. The energy U_P is due to the electron quiver in the electric field. In the case of a very short linearly polarised laser pulse this quiver energy will not be converted into electron thermal velocity, but instead will be returned to the radiation field. Hence, the most probable electron kinetic energy after the laser pulse has ended will be $mv_i^2/2$. If the electron is emitted at some other time, t_0 , then the extra kinetic energy is due to the work done by the electric field during the first fraction of a cycle following the appearance of the electron. This is known as above threshold ionisation (ATI). Free electrons created by optical ionisation with a circularly polarised laser pulse have no fixed phase relation between each other or with the laser pulse. This means that the ponderomotive energy of the electron will not be returned to the laser field, and the most probable electron kinetic energy after the laser pulse has ended will be $U_P + mv_i^2/2$.

The experiments presented in this thesis use a laser with a wavelength of 248 nm and intensities up to 8×10^{17} W/cm², producing a maximum of eight times ionised Neon. We can make a simple estimate of the maximum ATI energy of each ionisation stage of Neon by calculating the critical intensity (given by equation 2.2.2) for each ionisation stage and then calculating $3U_P$ for this intensity. These values are shown in table 2.3 below. It should be noted that the ATI energies shown in this table are the maximum possible, the mean ATI energy will be much lower than this.

<i>Final ionisation stage</i>	<i>Ionisation potential (eV)</i>	<i>Critical intensity (W/cm²)</i>	<i>Maximum ATI energy (eV)</i>
Ne ⁺	21.56	8.7 x 10 ¹⁴	14.9
Ne ²⁺	41.07	2.9 x 10 ¹⁵	49.8
Ne ³⁺	63.5	7.2 x 10 ¹⁵	123.7
Ne ⁴⁺	97.02	2.2 x 10 ¹⁶	377.9
Ne ⁵⁺	126.3	4.1 x 10 ¹⁶	704.3
Ne ⁶⁺	157.91	6.9 x 10 ¹⁶	1185.4
Ne ⁷⁺	207.21	1.5 x 10 ¹⁷	2576.9
Ne ⁸⁺	239	2.0 x 10 ¹⁷	3435.8

Table 2.3. Critical ionisation intensities and maximum ATI energies for Neon and its ions. For a real laser pulse, the typical ATI energy of a given ionisation stage will be much lower than the upper limits shown (see text).

Figure 2.3.1 shows the results of a simulation of the contribution of ATI to the average electron temperature in Neon at the intensities listed in table 2.3, and at a neutral gas density of 1.25×10^{20} atoms/cm³. The MEDUSA hydrodynamic code used for this simulation is described in chapter 5. The figure also shows the average ionisation of the plasma, confirming that the critical intensities calculated from equation 2.2.2 are approximately correct. The average ATI temperatures are much lower than the maxima listed in table 2.3. This confirms that very few electrons are ionised at a time in the laser pulse when the electric field is close to zero.

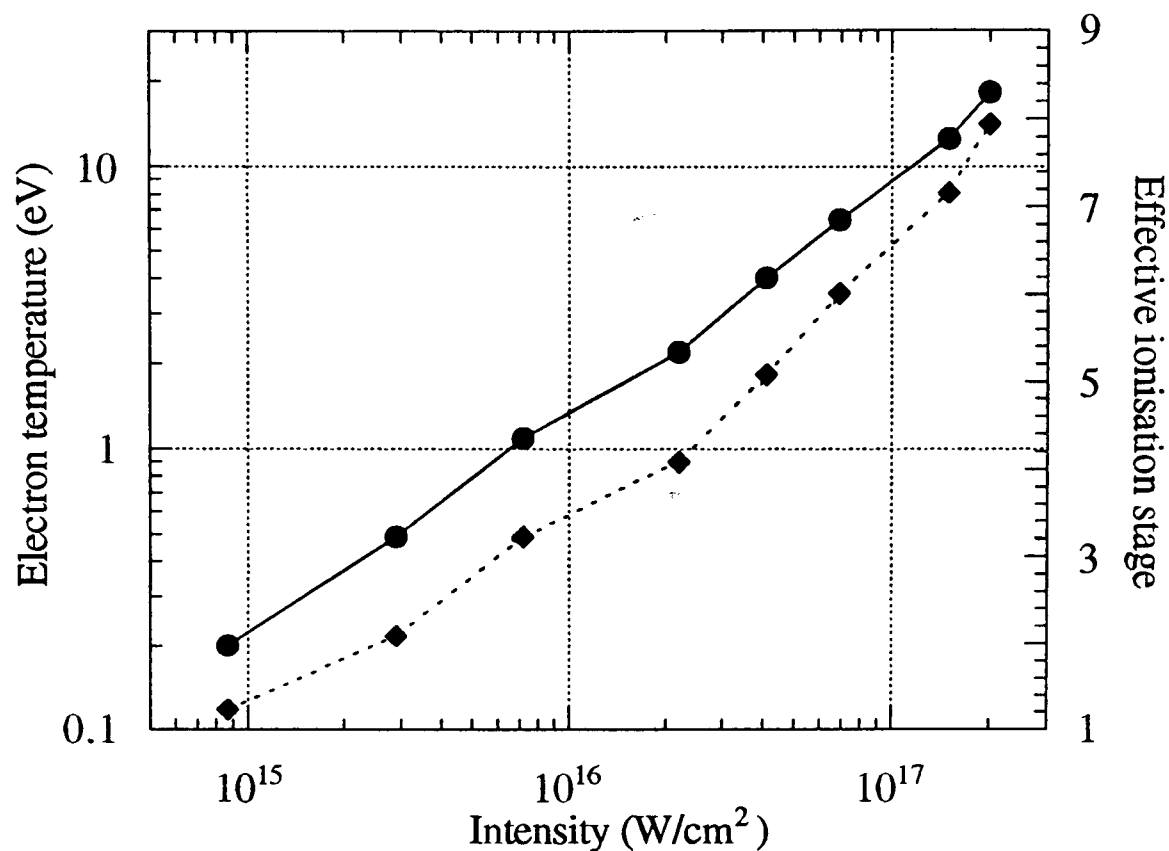


Figure 2.3.1. The variation of the average contribution of ATI heating to the electron temperature (circles, solid line, left axis), and the average ionisation stage (diamonds, dotted line, right axis) of Neon as a function of intensity for a 380 fs, 248 nm laser pulse.

2.4. Inverse Bremsstrahlung.

Inverse bremsstrahlung (IB) is one of the major heating mechanisms in laser produced plasmas, especially at low intensities. Bremsstrahlung, or 'braking radiation', is the emission of radiation by an electron as it decelerates. This deceleration is caused when the electron interacts with an atom or ion via the Coulomb field. Inverse bremsstrahlung, as its name suggests, is the opposite of this, i.e. it is the process whereby the ordered motion of an electron oscillating in the laser field is converted into disordered thermal motion by the scattering of the electron from an ion (caused by interaction with the Coulomb potential). So, in this case, a photon is absorbed by an electron in the presence of the Coulomb field of an ion.

The cycle averaged power dissipated from the laser beam in a plasma by inverse bremsstrahlung is given by

$$P = 2U_P \nu_{ei} N \quad (2.4.1)$$

where U_P is the ponderomotive energy, N is the electron density, and ν_{ei} is the electron-ion collision frequency (or the inverse of the time taken for an electron to scatter through 90° as a result of many small angle collisions), given by [35,36]

$$\nu_{ei} = \frac{4\sqrt{2}\pi}{3} \frac{ZNe^4 \ln \Lambda}{(4\pi\epsilon_0)^2 \left[1 - \frac{\omega_{pe}^2}{\omega_0^2}\right]^{1/2} m^2 v_{th}^3} \quad (2.4.2)$$

where Z is the average ionisation state, N and N_i are the electron and ion density respectively. Λ is the ratio of b_{max}/b_{min} , the maximum and minimum distance respectively of the electron from the ion in a scattering event. The maximum distance is given approximately by the Debye length, λ_D . The minimum distance is given by the larger of the classical distance of closest approach, or the de Broglie wavelength of the electron, $b_{min} = \max\left[\left(Ze^2/4\pi\epsilon_0 m v_{te}^2\right), (h/mv_{te})\right]$. This leads to an expression for the IB heating rate at low intensity and high frequency, given by [35,37]

$$J = \frac{4}{3} \left[\frac{\pi}{2}\right]^{1/2} \frac{Z^2 N N_i e^4 v_0^2 \ln \Lambda}{(4\pi\epsilon_0)^2 \left[1 - \frac{\omega_{pe}^2}{\omega_0^2}\right]^{1/2} m v_{th}^3} \text{ J s}^{-1} \text{ m}^{-3} \quad (2.4.3)$$

The electron-ion collision frequency gives us a rough measure of how quickly the electron temperature distribution in the plasma will equilibrate to a Maxwellian distribution. Table 2.4 lists the electron plasma frequency and the electron-ion collision frequency at an electron temperature of 40 eV. At an electron density of 10^{17} cm^{-3} the

time for an electron to scatter through 90° is of the order of 17 ps. This indicates that the plasma cannot have thermalised during the laser pulse length of 380 fs. Even at an electron density of 10^{19} cm^{-3} the electron-ion collision time is still about 277 fs, which is still of the same order as the laser pulse length. At high laser intensities, the electron quiver velocity in the laser field can be much greater than the electron thermal velocity. This will greatly reduce the electron-ion collision frequencies (and hence the IB heating rates) listed in table 2.4. Electron-electron collision frequencies are given by [36]

$$v_{ee} = 2.2 \times 10^5 \frac{N \ln \Lambda}{v_{th}^3} \quad (2.4.4)$$

The electron-electron collision frequencies are also included in table 2.4. Even at an electron density of 10^{20} cm^{-3} , electron-electron collision times are of the order of 300 fs. From this we infer that (for all but the highest densities in our experiments) the electrons in the plasma created by optical ionisation will not have time to equilibrate to a Maxwellian velocity distribution during the time of the laser pulse.

$N (\text{cm}^{-3})$	$\omega_{pe} (\text{s}^{-1})$	$v_{ei} (\text{s}^{-1})$	$v_{ee} (\text{s}^{-1})$
10^{17}	1.78×10^{13}	5.73×10^{10}	7.39×10^9
10^{18}	5.63×10^{13}	4.66×10^{11}	6.02×10^{10}
10^{19}	1.78×10^{14}	3.61×10^{12}	4.66×10^{11}
10^{20}	5.63×10^{14}	2.57×10^{13}	3.30×10^{12}
10^{21}	1.78×10^{15}	1.55×10^{14}	1.94×10^{13}

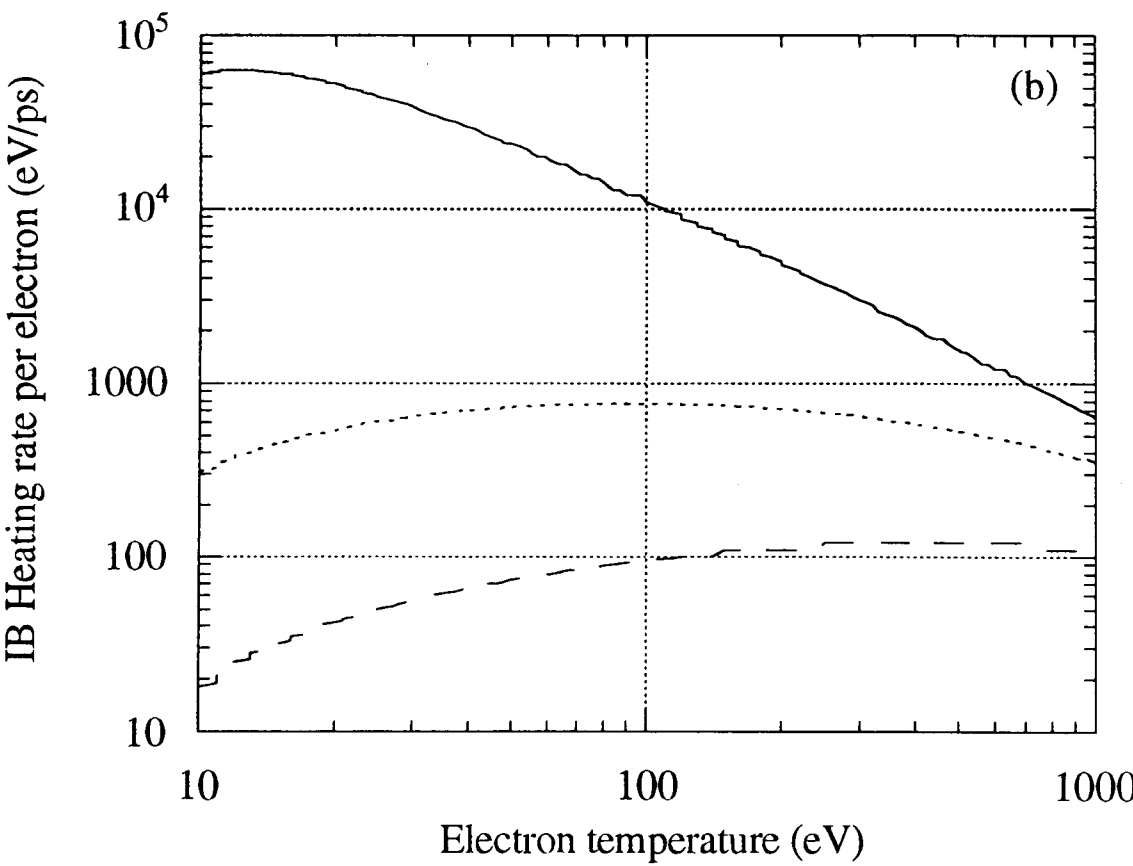
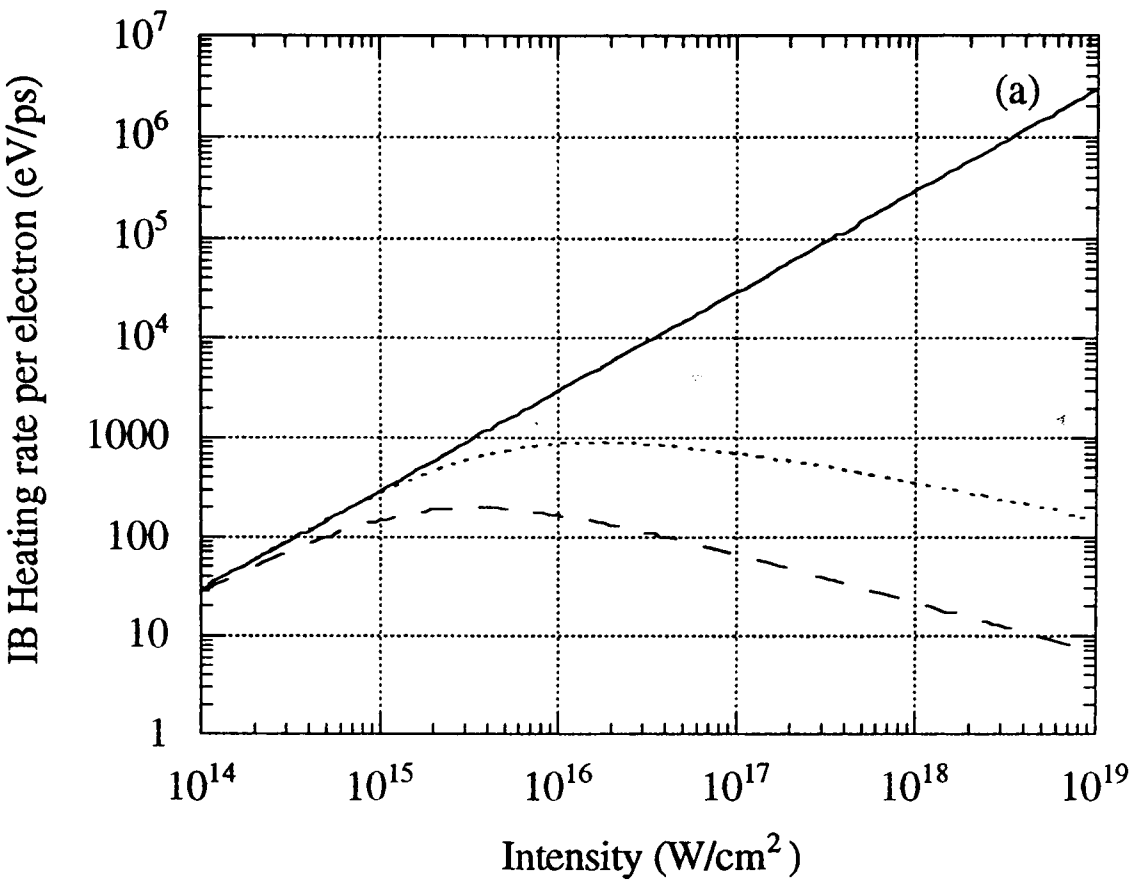
Table 2.4. The electron plasma frequency, the electron-ion collision frequency and the electron-electron collision frequency at a range of electron densities and an electron temperature of 40 eV.

2.4.1. High intensity corrections to the inverse bremsstrahlung rate.

Various corrections to the IB heating rate (equation 2.4.3) for high intensities have been proposed. Here we shall consider just two. The first is to simply replace v_{th}^2 by $v_{th}^2 + v_0^2$ [38] which obviously reduces to the low intensity expression at low intensities (as the quiver velocity, v_0 , approaches zero). This form takes into account the quiver velocity of the electron, which may be much greater than the thermal velocity, v_{th} at high intensities. Such a correction to the electron-ion collision frequency at a laser intensity of $3 \times 10^{17} \text{ W/cm}^2$ would reduce the values quoted in column 3 of table 2.4 by a factor of over 2000. An alternative, more recent approach proposed by Polishchuk et al [39] takes account of modifications to the $\ln \Lambda$ term by the laser field by replacing $\ln \Lambda / v_{th}^3$ by

$$\frac{\frac{1}{4} \ln^2 \left[1 + \frac{v_0^2}{v_{th}^2} \right] + \ln \left[\frac{v_0^2}{v_{th}^2} + \exp \left(\frac{1}{3} \sqrt{\frac{\pi}{2}} \right) \right] \ln \Lambda}{\frac{1}{3} \sqrt{\frac{\pi}{2}} (v_0^2 + v_{th}^2)^{3/2}} \quad (2.4.5)$$

This equation is a special case of a more rigorous calculation presented by G.J. Pert [40] which, for simplicity, we shall not consider here. (For the conditions of our experiments the difference is small). The above three forms of the IB heating rate predict significantly different absorption. For comparison we present, in figure 2.4.1, the scaling of the three forms of the single electron IB rates in a plasma of Ne^{8+} (a) with intensity at a constant temperature of 40 eV and density of 10^{20} cm^{-3} ; (b) with temperature at a constant intensity of 10^{17} W/cm^2 and density of 10^{20} cm^{-3} ; and (c) with density at a constant temperature of 40 eV and an intensity of 10^{17} W/cm^2 .



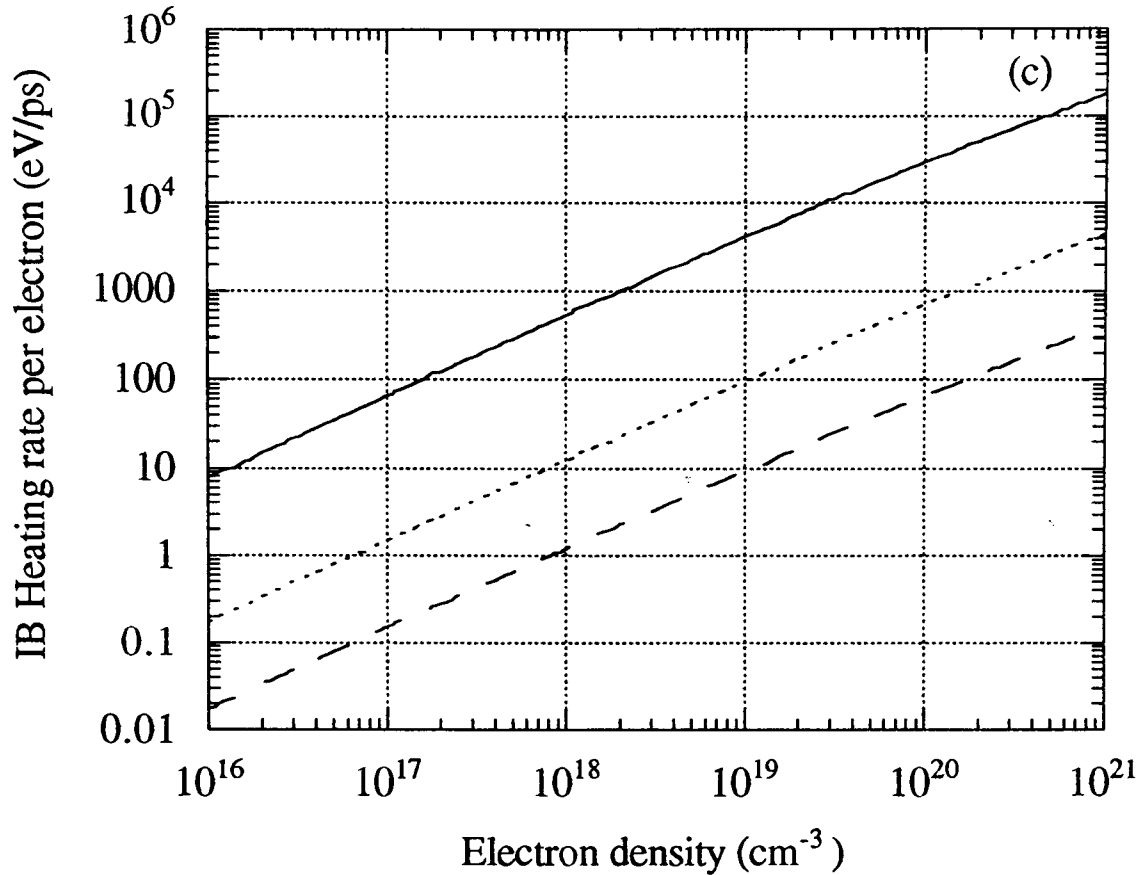


Figure 2.4.1. Variation of the inverse bremsstrahlung heating rate for Ne^{8+} with (a) laser intensity, (b) electron temperature, and (c) electron density. When not variable, the intensity, temperature and density are fixed at 10^{17} W/cm^2 , 40 eV, and 10^{20} cm^{-3} respectively. The three lines correspond to the standard formula (solid), the $v_{th}^2 + v_0^2$ form (dashed), and the Polishchuk form (dotted).

As can be seen in the above figure, the standard form gives much higher heating rates than the two high intensity forms. The IB heating rate predicted by the Polishchuk correction, however, is not as low as that predicted by the standard high intensity correction. In fact, using the Polishchuk form, rather than the more usual high intensity correction, increases the IB heating rate by about one order of magnitude. This will obviously lead to higher electron temperatures than previously expected. The MEDUSA [41] plasma simulations in this thesis, described in section 5.1, use the Polishchuk form for the IB heating rate.

To compare the relative magnitudes of ATI and IB heating we show, in figure 2.4.2, a plot of MEDUSA simulations of the ATI and IB energy at the peak of the laser pulse as a function of the electron density in an optically-ionised Neon plasma. The peak laser intensity for these simulations is $3 \times 10^{17} \text{ W/cm}^2$ and the laser pulse length is 380 fs. We see that the ATI contribution to the electron temperature is independent of electron density, whereas the IB contribution has a strong density dependence. IB heating dominates ATI at electron densities above about $2 \times 10^{19} \text{ cm}^{-3}$.

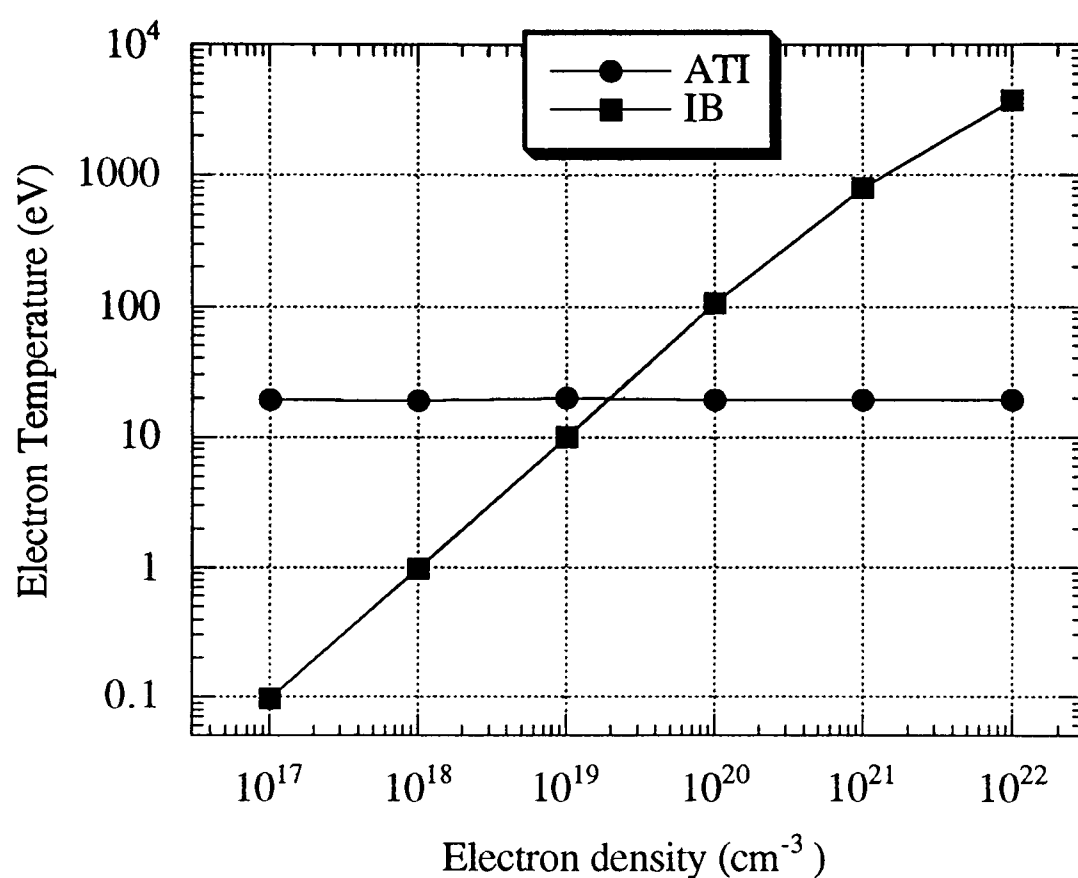


Figure 2.4.2. MEDUSA simulation of the variation of inverse bremsstrahlung and ATI heating with electron density in an optically-ionised Neon plasma. The peak laser intensity is $3 \times 10^{17} \text{ W/cm}^2$, and the laser pulse length is 380 fs.

2.5. Stimulated Raman and Brillouin scattering.

2.5.1. Parametric instabilities.

Stimulated Raman scattering (SRS) is a parametric decay process where a transverse electromagnetic wave decays into a scattered transverse electromagnetic wave and an electron plasma wave (a longitudinal electrostatic wave). Stimulated Brillouin scattering (SBS) is an equivalent process, but this time the plasma wave is an ion-acoustic wave. In both processes a positive feedback mechanism results in exponential growth of a plasma wave as energy is pumped from the electromagnetic wave at one frequency into the plasma wave at another frequency. This will be demonstrated below (following the argument of Hughes [42]).

For any scattering process the Manley - Rowe relations must be followed, i.e.

$$\omega_0 = \omega_1 + \omega_2 \quad (2.5.1)$$

$$\text{and} \quad \mathbf{k}_0 = \mathbf{k}_1 + \mathbf{k}_2. \quad (2.5.2)$$

where ω is a frequency and k is a wavevector, and the subscripts 0, 1, and 2 refer to the incident electromagnetic wave, the plasma wave (electron or ion-acoustic) and the scattered electromagnetic wave respectively. The dispersion relations for these waves are shown in section 2.1.

If the plasma has an existing mode of oscillation at ω_1 , either for an electron or an ion wave (which is initially only at noise level), beats will occur between the two waves and sidebands are generated which oscillate at $\omega_2 = \omega_0 - \omega_1$ and at $\omega_3 = \omega_0 + \omega_1$. If ω_2 is a scattered electromagnetic wave, it will beat with ω_0 at $\omega_1 = \omega_0 - \omega_2$ and at

$\omega_4 = \omega_0 + \omega_2$. The beating electromagnetic waves will generate a ponderomotive force that drives oscillations of the charged particles in the plasma and enhances the mode at ω_1 , which will beat more strongly with ω_0 to give a stronger ω_2 mode. This process then continues, the positive feedback loop resulting in greatly enhanced (or stimulated) scattering. This is shown schematically in figure 2.5.1. If the driven modes are pumped more strongly than they are damped, they will grow. There is a threshold pump wave amplitude at ω_0 for the instability to grow, which is set by the damping constants Γ_1 and Γ_2 , and by the strengths of coupling of the pump wave to the driven modes, c_1 and c_2 . In general the threshold will be lowest when one of the driven modes is resonant in the plasma.

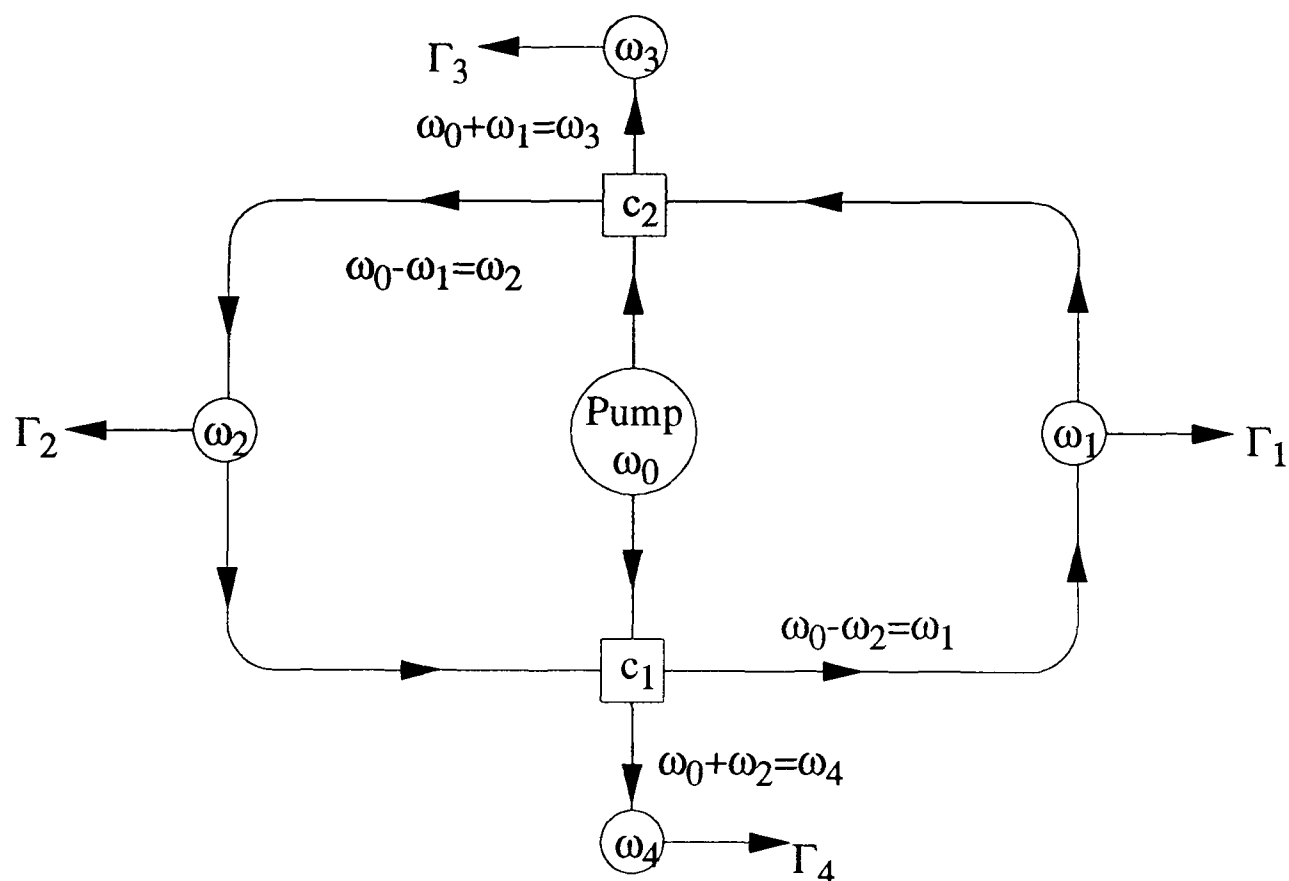


Figure 2.5.1. Schematic diagram showing the growth of a parametric instability. (From ref. [42]).

2.5.2. Growth rates for stimulated Raman (and Brillouin) scattering.

The threshold intensity, I_{th} , for absolute SRS growth is the intensity above which the gain of the stimulated scattering process overcomes losses due to damping of the electromagnetic and electrostatic waves. It is given by Forslund [43] as

$$I_{th} = \epsilon_0 c \left(\frac{m \omega_0 c}{e} \right)^2 \frac{\Gamma_p \Gamma_-}{\omega_{pe} \omega_0} \quad (2.5.3)$$

where ω_{pe} is the electron plasma frequency, ω_0 is the laser frequency, Γ_p is the plasma wave damping rate (Landau damping for example), and Γ_- is the damping rate on the electromagnetic wave caused by collisional absorption which is given by $\Gamma_- = (v_{ei}/2)(\omega_{pe}/\omega_0)^2$ (where v_{ei} is the electron-ion collision rate defined in equation 2.4.2) [43]. Growth occurs in three distinct regimes. These are when the pump intensity is just above threshold, or well above threshold, or when the laser and plasma are strongly coupled. The latter regime is not relevant to our experimental conditions. The growth rate just above threshold is given by

$$\gamma = \left(\frac{\omega_{pe}^2}{\omega_k(\omega_0 - \omega_k)} \frac{k^2 v_0^2}{8} - \Gamma_p \Gamma_- \right) (\Gamma_p + \Gamma_-)^{-1} \quad (2.5.4)$$

where ω_k is the plasmon frequency, k is the plasmon wavevector, $v_0 = eE_0/m\omega_0$ is the quiver velocity of an electron in the laser field of frequency ω_0 and electric field strength E_0 . This shows that the growth rate is directly proportional to the intensity ($v_0^2 \propto I$). When the intensity is well above threshold the growth rate varies as the square root of the intensity

$$\gamma = \frac{k v_0}{2\sqrt{2}} \frac{\omega_{pe}}{\sqrt{\omega_k(\omega_0 - \omega_k)}} \quad (2.5.5)$$

In our experiment the collisional damping rate for the electromagnetic wave, $\Gamma_.$, will not rise above about 10^{12} s^{-1} (the value for an electron density of 10^{21} W/cm^2 and an electron temperature of 40 eV) which gives a threshold intensity of about $3 \times 10^{12} \text{ W/cm}^2$ for the onset of SRS. Hence, for our experimental intensities of $8 \times 10^{17} \text{ W/cm}^2$, we will be well above the threshold intensity defined in equation 2.5.3, and SRS growth will be governed by equation 2.5.5. If we assume that the thermal correction to the Bohm-Gross frequency is negligible then $\omega_k = \omega_{pe}$, and if we consider light scattered at 180° to the direction of laser propagation (the direction for which the growth rate is highest), then $k \approx 2k_0$. The above growth rate (equation 2.5.5) then becomes

$$\gamma = \frac{\omega_0}{\sqrt{2}} \frac{v_0}{c} \left(\frac{\omega_{pe}/\omega_0}{1 - \omega_{pe}/\omega_0} \right)^{1/2} \quad (2.5.6)$$

The corresponding growth rate for SBS well above threshold is given by

$$\gamma_B \approx \frac{v_0 \omega_{pi}}{2\sqrt{2}c} \quad (2.5.7)$$

where ω_{pi} is the ion-acoustic frequency. For the typical conditions we will encounter in our experiments (for example those depicted in the dispersion curves of figure 2.1.1) the SBS growth time ($1/\gamma$) will be of the order of a picosecond or more, and the SRS growth time will be a few femtoseconds. Since the experimental laser pulse length has been measured to be about 380 fs we do not expect SBS to occur, but there will be plenty of time for SRS to grow. Therefore, we will not consider the SBS instability any further.

For an infinitely long laser pulse, the plasma waves will grow until they non-linearly saturate. The fractional density fluctuation, ΔN caused by a plasma wave must always be less than unity, since at the point of rarefaction in longitudinal wave there can never be less than zero particles. For a finite pulse length, however, the plasma waves may never

reach the point of saturation. Eder [44], shows that in such a situation the growth rate is modified to $\gamma' = \sqrt{2}/3 \gamma$. The growth of the SRS instability (in the absence of saturation or significant damping) can be written in terms of the final scattered power, S_f , compared to the initial power, S_i , which is Thomson scattered from thermal fluctuations, by

$$S_f = S_i \exp\left(\frac{\sqrt{2}}{3} \gamma \tau\right) \quad (2.5.7)$$

where τ is the laser pulse length. The levels of Thomson scattering which initiate the SRS scattering are discussed in section 4.4.

2.6. Damping of plasma waves.

Having established how small fluctuations in the plasma density can grow to large amplitudes by the process of SRS, we now consider how the energy in these plasma waves is converted into thermal energy in the plasma. The two processes by which this can happen are Landau damping and wave breaking.

2.6.1. Landau damping.

Landau damping is the process by which electrons with velocities that are resonant with the phase velocity of a plasma wave absorb energy from the wave, which is later converted to thermal energy by collisions in the plasma. Consider the schematic representation of a Maxwellian electron velocity shown in figure 2.6.1. The phase velocity, v_p , of a plasma wave is indicated by the solid line. Electrons that have a velocity slightly less than the plasma wave will be accelerated by the wave and will draw energy from it in a similar way to a surfer riding on a water wave. Electrons with velocities slightly greater than the plasma wave will be decelerated and will transfer

energy into the wave. Since the velocity distribution is Maxwellian there will always be more electrons with velocities less than the phase velocity than with velocities that are greater. This means that the result will always be a net damping of the wave [1,42].

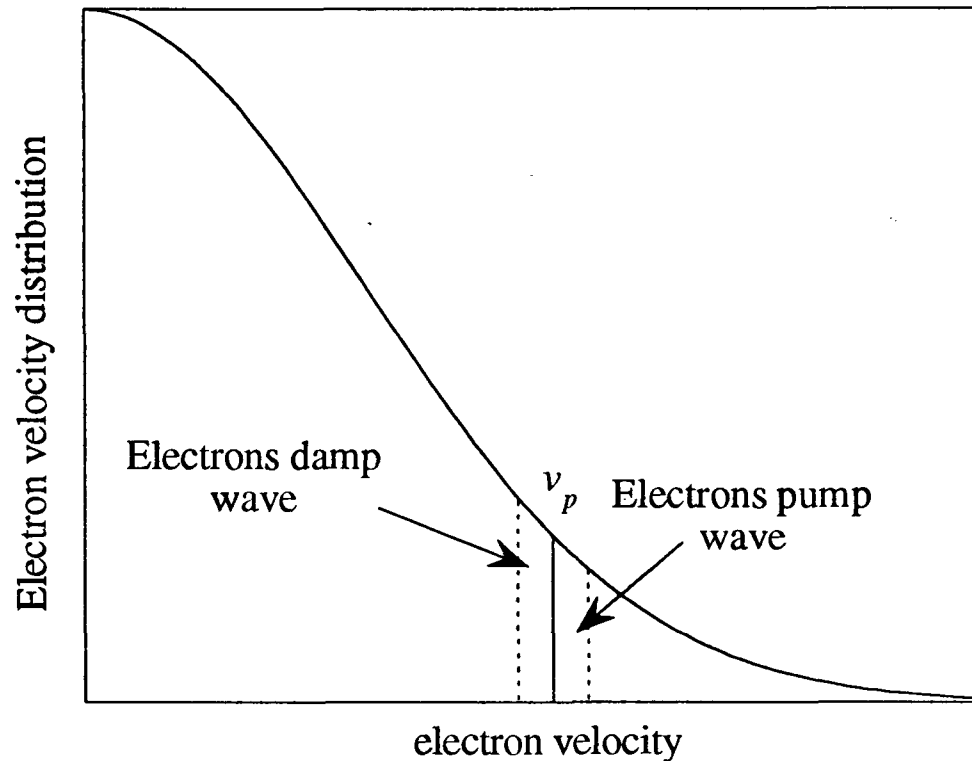


Figure 2.6.1. Schematic diagram of Landau damping of a plasma wave. v_p is the plasma wave phase velocity.

Another name for this process is particle trapping, as electrons with certain velocities are trapped and carried along by the wave. Should Landau damping occur at the same phase velocity for a long period of time the electron distribution will become non-Maxwellian and the damping will be stopped. This is shown schematically in figure 2.6.2. The acceleration of the electrons has now caused a 'hump' in the electron distribution. Conditions are now right for the pumping of a plasma wave with a phase velocity greater than the original wave. In this way we would expect the frequency of plasmons created by SRS to increase during a laser pulse.

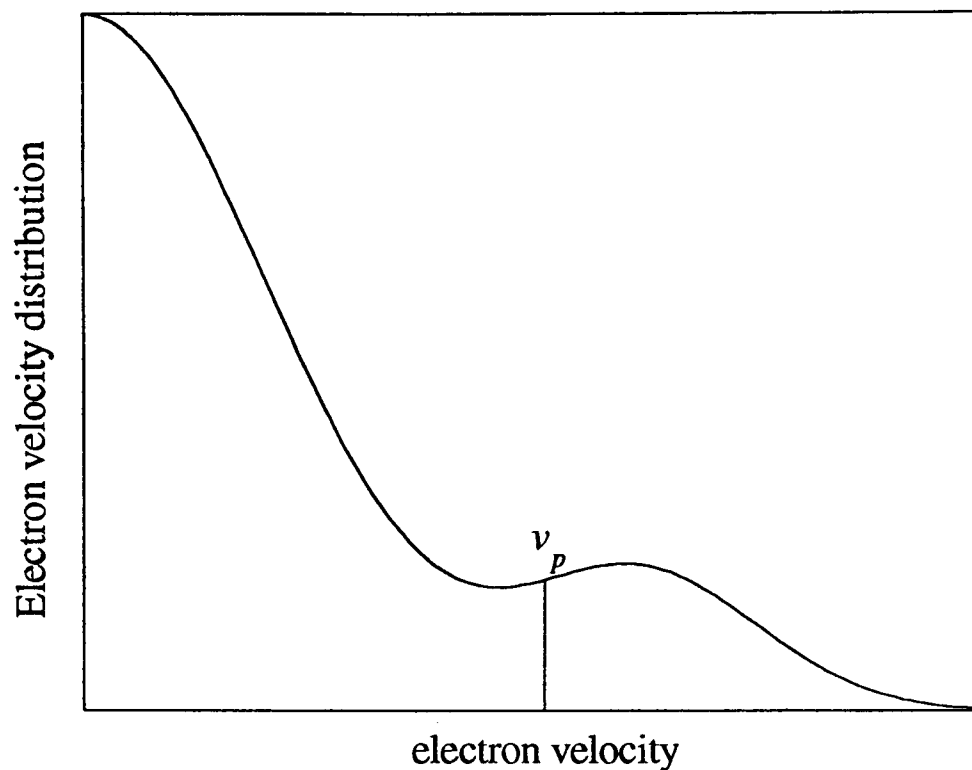


Figure 2.6.2. The result of Landau damping - a non-thermal electron distribution.

2.6.2. Wave breaking.

If the amplitude of a longitudinal wave (of wavevector $\mathbf{k}$) becomes comparable to its wavelength, the order of particles along $\mathbf{k}$ is disturbed and the wave destroys itself. This process is known as 'wave breaking', or 'phase space mixing' [42].

Consider the one-dimensional problem of a plane electron plasma wave travelling along a direction x in an infinite plasma [42]. In the wave field, an electron oscillates about a mean position x_0 , the displacement from x_0 being $X(x_0)$. In moving a distance X , the electrons pass (fixed) ions with charge NeX per unit area, N being the unperturbed electron density. If the ordering of the electrons is maintained, an excess positive charge of NeX will occur on the $(-x)$ side, and vice-versa. Using Gauss's theorem we see that

$$E = \frac{Ne}{\epsilon_0} X \quad (2.6.1)$$

The equation governing electron motion is

$$m \frac{d^2 X}{dt^2} = -eE = -\frac{Ne^2 X}{\epsilon_0} \quad (2.6.2)$$

which gives the normal oscillatory motion at the plasma frequency

$$\omega_{pe} = \left(\frac{Ne^2}{m\epsilon_0} \right)^{1/2} \quad (2.6.3)$$

This is valid provided that the change in X due to Δx_0 is greater than $(-\Delta x_0)$.

Let us now suppose that the electrons are perturbed by a wave in which

$$X = A \sin(kx_0) \cos(\omega t) \quad (2.6.4)$$

So at time $t = 0$: $X = A \sin(kx_0)$, $E = (Ne/\epsilon_0) A \sin(kx_0)$, and $x = x_0 + X$. In terms of E and A , $X = \epsilon_0 E / Ne$ and $\sin(kx_0) = \epsilon_0 E / NeA$.

Writing $NeA/\epsilon_0 = E_0$, and $E/E_0 = y$, so that $-1 \leq y \leq 1$ we derive the function

$$kx = \sin^{-1} y + (kA)y \quad (2.6.5)$$

Figure 2.6.3 shows y as a function of kx . If $kA > 1$, y becomes double-valued near $kx = (2n+1)\pi$, which is physically impossible, i.e. the wave has broken. Thus the limiting value of E_0 is $E_{\max} = Ne/\epsilon_0 k$.

Hence
$$\frac{eE_{\max}}{m\omega} = \frac{Ne^2}{m\epsilon_0 k\omega}, \quad (2.6.6)$$

i.e. $v_{0\max} = \frac{\omega_{pe}^2}{k\omega} \approx v_p$ provided that $\omega \approx \omega_{pe}$. So, the wave breaks if $v_0 > v_p$.

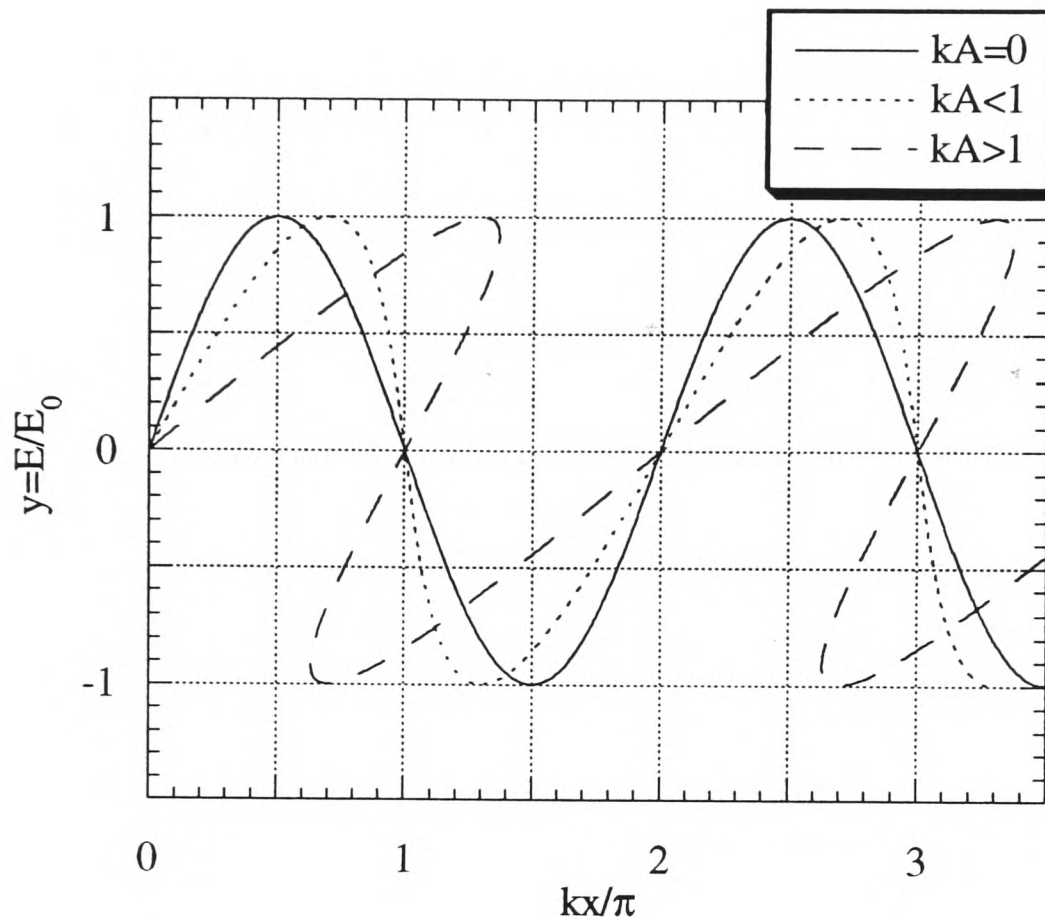


Figure 2.6.3. Breaking of a longitudinal plasma wave occurs when its amplitude becomes larger than its wavelength.

2.6.3. SRS as a plasma heating mechanism.

The damping mechanisms described above both have the ultimate effect of converting the ordered energy of the plasma wave into thermal energy of the plasma. Hence, SRS will always result in an increase in the electron and (eventually) the ion temperature. Some electrons accelerated by Landau damping will gain enough energy to escape the plasma. These electrons will either be dragged back at some later time by the electrostatic potential of the positively charged plasma they leave behind or they will drag some ions from the plasma which can be detected by a charged particle detector such as a Faraday cup (see section 5.5.1). However, most of the energy will be distributed among the electrons in the plasma at the laser focus, and so we can make an estimate of the level of heating by

recording the energy backscattered by SRS (the SRS reflectivity) by the following method:

1. The number of plasmons created in the plasma by SRS is equal to the number of scattered photons.
2. The energy of each plasmon can be determined from the frequency shift of the backscattered light, using the relation $\hbar\omega_k = \hbar\omega_0 - \hbar\omega_s$, where ω_k is the plasmon frequency, ω_0 is the laser frequency, and ω_s is the frequency of the scattered light. The wavevector of the plasmon can be determined, in a similar way, from the wavevector relation $k = k_0 + k_s$.
3. To determine the net heating of the bulk plasma we assume that this energy is thermalised in the plasma in the focal region. This region can be represented by a cylinder with a diameter of the FWHM of the laser spot and a length of twice the confocal parameter.
4. To determine the temperatures of 'hot' electrons created by Landau damping we can calculate the phase velocity of the plasma wave, $v_p = \omega_k/k$.

The increase in electron temperature due to SRS is given by

$$T_e(eV) = \frac{1}{e} \frac{ER}{NV} \frac{\omega_k}{\omega_s} \quad (2.6.7)$$

where R is the reflectivity, V is the focal volume, and E is the laser energy on target..

It is not a simple task to estimate the levels of SRS we expect to see in the experiments described in this thesis. This is because the growth of the instability is exponential, and

so it is extremely sensitive to temporal and spatial variations in the laser and the plasma. We do know that the SRS growth time is short compared to the laser pulse and that our laser intensity will be well above the threshold for SRS growth. The peak power Thomson scattered from thermal fluctuations (see equation 2.5.7 and section 4.4) in the plasma is of the order of 15 MW for a laser intensity of $8 \times 10^{17} \text{ W/cm}^2$ and a plasma density of 10^{21} cm^{-3} in a focal region of approximately $10 \mu\text{m}$ radius by $100 \mu\text{m}$ length (see section 4.4). At this density the growth rate for SRS is of the order of $2.6 \times 10^{14} \text{ s}^{-1}$, and it would only take about 90 fs for the stimulated Raman scattered power to equal the incident laser power. Obviously, the instability will saturate before this happens, and so we must wait to see experimentally how high the level of SRS will actually reach. The simple analysis above indicates that the levels will be high and that, when it does occur, SRS is likely to be the dominant heating mechanism.

2.7. Ponderomotive expulsion and ponderomotive heating.

A focused laser beam has a radial profile to its electric field amplitude (and intensity) which usually peaks at the centre of the focus and decreases with radius. This gradient produces a radial force that will act on the plasma and will drive electrons away from the focus. This ponderomotive force, F_P , is defined in terms of the ponderomotive potential, U_P

$$F_P(r) = -\nabla U_P(r) \quad (2.7.1)$$

$$\text{where} \quad U_P(r) = -\frac{e^2 E(r)^2}{4m\omega_0^2} \quad (2.7.2)$$

ω_0 is the laser frequency, and $E(r)$ is the magnitude of the electric field of the laser which varies as a function of radius, r . If we now assume a Gaussian profile for the laser, the ponderomotive force becomes

$$F_P(r) = \frac{e^2 E_0^2}{2m\omega_0^2} \frac{r}{r_0^2} \exp\left(-\frac{r^2}{r_0^2}\right) \quad (2.7.3)$$

where r_0 is the $1/e$ radius of the laser profile. This force will obviously result in a net acceleration of the electrons away from the centre of the focus. This can produce very high electron velocities. For our experiments, the laser pulse length is short enough (380 fs, 10 μm FWHM focal spot size) that significant electron motion does not occur during the time of the laser pulse, but it can result in significant motion at later time. In experiments conducted at 10 ps (and $\sim 3 \mu\text{m}$ focal spot size) the ponderomotive force results in a large depletion of electrons at the centre of the laser focus during the pulse [29,45].

As these ponderomotively accelerated electrons move away from the focus they leave behind them a positively charged plasma which creates a space charge that exerts a force which pulls the electrons back towards the focus and pulls the ions out of the focus. This space charge can be calculated by assuming that the ions remain stationary and that the electrons form a cylindrical sheath about a central ion core, shown in figure 2.7.1. The space-charge force, F_s , this creates can be calculated by applying Gauss's law to give

$$F_s = -\frac{Ne^2}{2\epsilon_0} r \quad (2.7.4)$$

The result of this is that some ions and electrons may leave the focus, but most of the electrons will return at some later time after the laser pulse has ended and the

ponderomotive force has vanished. This can set up oscillations of the plasma which will have a characteristic resonant frequency. If the laser pulse length is matched to this resonant frequency then plasma oscillations can be very large. The plasma waves created by these oscillations can cause significant heating.

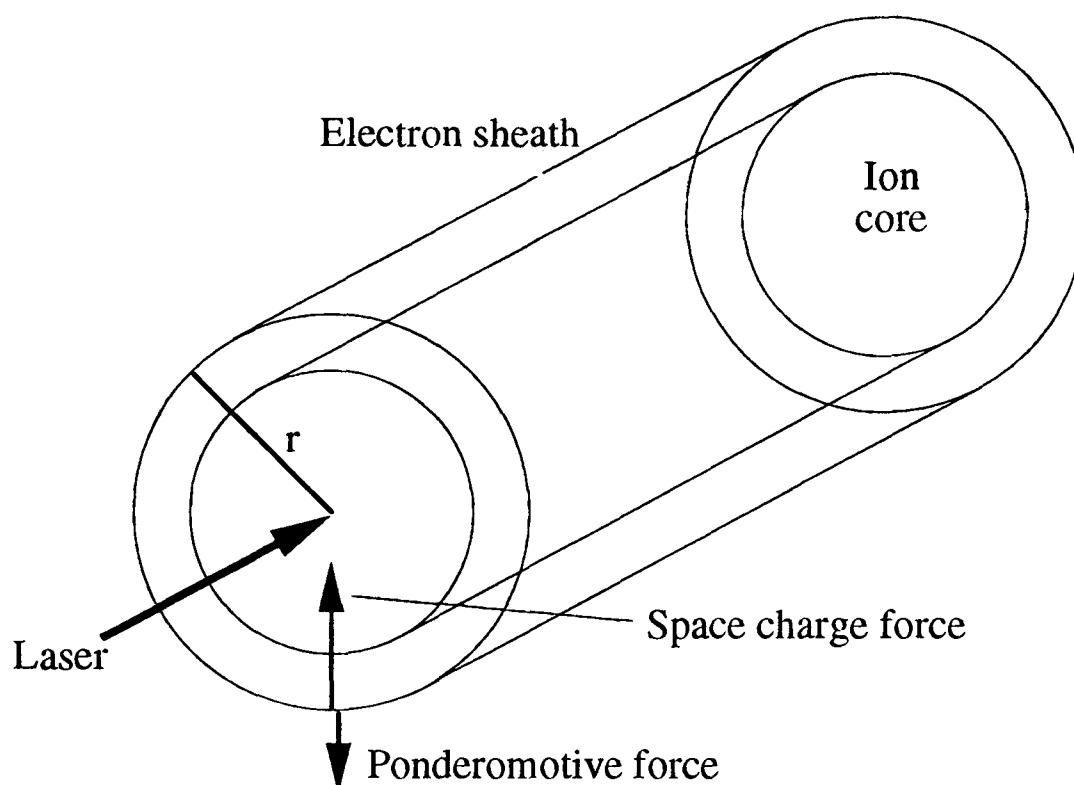


Figure 2.7.1. *Ponderomotive expulsion is limited by the space charge created between the cylindrical electron 'sheath' and the central ion core.*

Penetrante and Bardsley [46] discuss the effect of ponderomotive forces on the ATI energy. They show that if we include a restoring force in the equation of motion of an electron, i.e.

$$\ddot{x} + \omega_p x = -\frac{eE_0}{m} \sin \omega_0 t \quad (2.7.5)$$

where ω_p is the resonant frequency of oscillation of the plasma determined by F_s (not the electron plasma frequency, although the simple treatment above gives $\omega_p = \omega_{pe}/\sqrt{2}$), then the velocity of an electron created at rest at a time t_0 is given by

$$\dot{x} = -\frac{eE_0}{m(\omega_p^2 - \omega_0^2)} \left[\omega_0 \cos \omega_0 t + \cos \omega_p t \left(\omega_0 \cos \omega_0 t_0 \cos \omega_p t_0 + \omega_p \sin \omega_0 t_0 \sin \omega_p t_0 \right) \right. \\ \left. - \sin \omega_p t \left(\omega_0 \cos \omega_0 t_0 \sin \omega_p t_0 - \omega_p \sin \omega_0 t_0 \cos \omega_p t_0 \right) \right] \quad (2.7.6)$$

which is somewhat more complicated than equation (2.3.5) where ponderomotive effects are ignored. It can be shown that a minimum in the ATI energy occurs when $\omega_p \tau = \pi/2$, where τ is the laser pulse length [46].

2.8. Beam propagation.

When a short laser pulse propagates through a plasma its characteristics may be greatly altered from those it would exhibit when travelling through a vacuum. Some of the effects the plasma can have on the propagation of the beam include: refraction, filamentation, self focusing, frequency shifting, and pulse lengthening.

2.8.1. Refraction.

Refraction is caused by light passing from a region with one refractive index to one with a different refractive index. Let us first consider the simple case of a laser beam being focused into a slab of plasma of constant density. The refractive index of a plasma is given by $n = (1 - N/N_{cr})^{1/2}$. The phase change for light travelling through this plasma compared to a vacuum over a length, l , is approximately $\Delta\phi = (\pi l/\lambda)(N/N_{cr})$. A phase change of $\pi/2$ corresponds to doubling the diffraction limited beam divergence [47]. The corresponding 'defocusing length' is given by $l_D = (\lambda/2)(N_{cr}/N)$. When the defocusing length is shorter than the confocal parameter, $l_D < b$, we expect defocusing to be dominant. (The confocal parameter is defined as $b = \pi w_0^2/\lambda$ where w_0 is the 1/e width of a Gaussian laser beam at focus.)

In a homogeneous slab of plasma in which no ionisation is occurring there will be no adverse focusing effects except that at the boundary between the plasma and vacuum the complex radius of curvature of the Gaussian beam will be altered. However, for a real laser focus the radial intensity of a focused Gaussian beam will follow a Gaussian profile and the gas at the laser focus will typically have several ionisation stages, the critical intensity for which that can be reached in various regions of the laser focus. This means that the electron density profile of the plasma will roughly follow the laser profile, although at early times in the laser pulse the electron density profile may exhibit a step-like structure where the intensity rises above the threshold intensity for ionisation to a particular state and before electron motion has made the profile uniform. A MEDUSA simulation of the electron density profile from Neon with a neutral gas density of $1.25 \times 10^{19} \text{ cm}^{-3}$ at the peak of a 380 fs, 248 nm laser pulse with a focal spot size of $10 \mu\text{m}$ and a maximum intensity of $3 \times 10^{17} \text{ W/cm}^2$ is shown in figure 2.8.1. Note how the electron density is uniform to a radius of about $4 \mu\text{m}$, after which the fall-off in density is roughly constant except for a step which starts at about $6.5 \mu\text{m}$. Plasma simulations will be discussed in detail in chapter five.

The change in angle due to the radial electron density profile of a ray originally travelling parallel to the laser axis can be shown by a simple argument to be

$$\frac{d\theta}{dz} = -\frac{1}{2nN_{cr}} \frac{dN}{dr} \quad (2.8.1)$$

where $d\theta$ is the change in angle of propagation, z is the direction of propagation, n is the refractive index of the plasma, N is the electron density, and N_{cr} is the critical density. The effect of this is to cause the laser to focus in the plasma sooner and with a larger focal spot than it would in vacuum. This obviously reduces the focused laser intensity. This effect is shown schematically in figure 2.8.2.

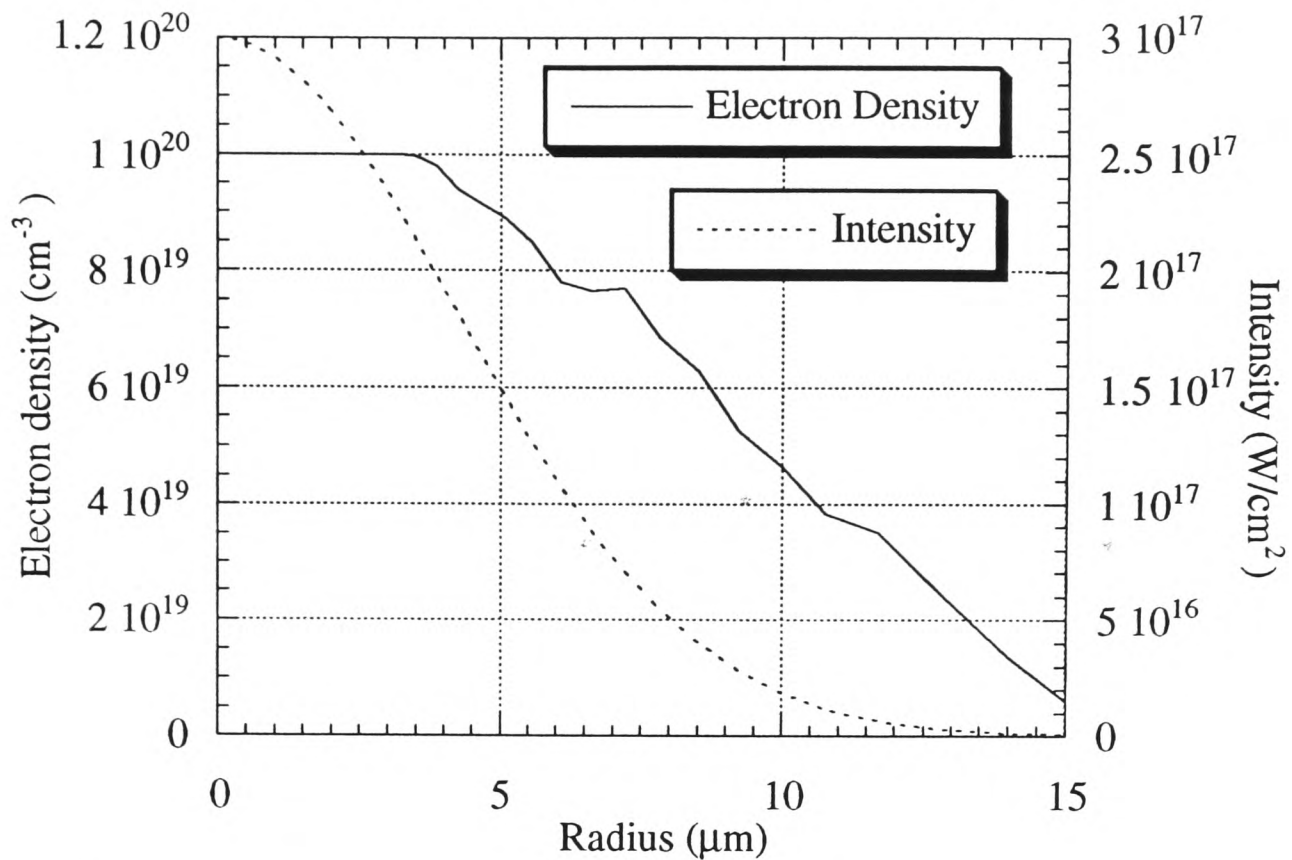


Figure 2.8.1. The radial variation of the electron density produced by optical ionisation in a Gaussian focus.

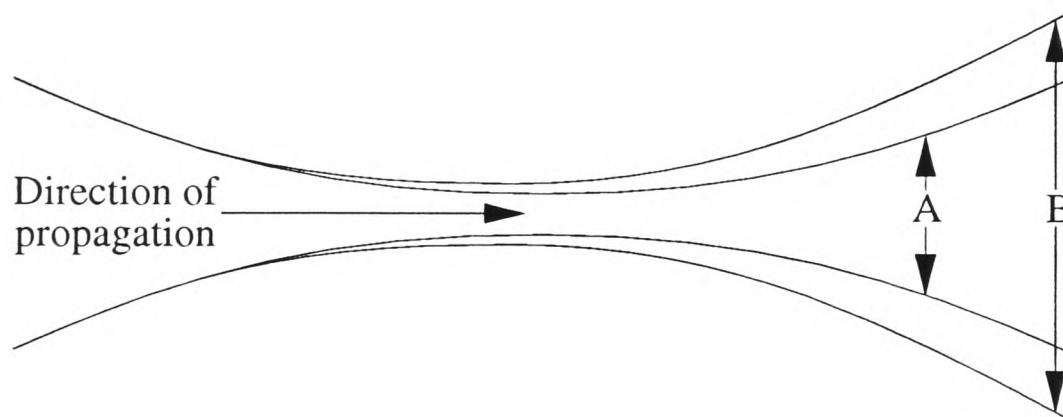


Figure 2.8.2. Refraction of a laser beam in a homogeneous plasma. The rays trace the beam profile in a vacuum (A), and in a homogeneous plasma (B).

2.8.2. Self-focusing.

If the laser profile is not a Gaussian, but exhibits some modulation in its intensity profile that is greater than the difference in the critical intensity for two ionisation stages, it is possible that the electron profile will follow the laser profile and the 'hot spots' in the

laser profile will be smeared out to make the beam more uniform. However, the process of self-focusing competes with this effect.

Self-focusing has been suggested as a method of creating a long channel in a plasma for applications such as the optically-ionised X-ray laser [38] and Wakefield electron acceleration [48,49]. The effect has been modelled extensively [48,50-53]. It has already been established that a radial density profile created by a Gaussian beam can cause defocusing. However, we have also shown in section 2.7 that a Gaussian beam can cause the ponderomotive expulsion of electrons from the laser focus. We can create an electron density minimum on the axis of the laser, either using a pre-pulse to the main laser or by using a laser pulse which is long enough that the tail of the pulse travels through the density minimum created by the leading edge of the pulse. The electron refractive index is given by $n = (1 - N/N_{cr})^{1/2}$, so the refractive index will be greatest on the laser axis where the electron density is smallest and rays travelling a distance, r , away from the axis will be guided towards $r = 0$. In this way it may be possible to keep the laser in a tight channel for much longer than the confocal parameter in vacuum [38,54].

The same effect can be achieved by relativistic self-focusing [38,51]. If the electron quiver velocity reaches relativistic speeds, the mass of the electron will start to increase. The critical density in the definition of the refractive index above is given by $N_{cr} = \omega_0^2 \epsilon_0 m / e^2$. We can see that, as the quiver velocity and mass of the electron increases, the electron refractive index will also increase. Since the laser intensity is greatest on-axis, the refractive index will be higher here and so self-focusing can again occur. This effect can cause the laser to focus down to a channel of the order of one wavelength in diameter. Borisov et al [55] claim to have seen experimental evidence of this effect and they identify a critical power for its onset given by

$$P_{cr} = 1.62 \times 10^{10} \frac{N_{cr}}{N} \text{ (Watts)}. \quad (2.8.2)$$

2.8.3. Frequency shifts of the ionising laser.

As an intense laser pulse passes through a gas target the electron density of the forming plasma rapidly increases as optical ionisation occurs. Since the refractive index of a plasma is given by $n = (1 - N/N_{cr})^{1/2}$, the rate of change of the refractive index is determined by the rate of change of electron density, i.e.

$$\frac{dn}{dt} = -\frac{1}{2nN_{cr}} \frac{dN}{dt}. \quad (2.8.3)$$

The change in phase of a monochromatic laser wavelength, λ_0 , over an interaction length, L , associated with this changing refractive index is just given by

$$\frac{d\phi}{dt} = \frac{2\pi L}{\lambda_0} \frac{dn}{dt} \quad (2.8.4)$$

where $d\phi/dt$ is a change in frequency of the propagating wave. If we now substitute equation 2.8.3 into equation 2.8.4 and put $N_{cr} = (\epsilon_0 m \omega_0^2 / e^2)$, we find that the changing refractive index induces a wavelength shift [34,56,57] given by

$$\Delta\lambda = -\frac{e^2 L \lambda_0^3}{8\pi^2 \epsilon_0 m n c^3} \frac{dN}{dt} \quad (2.8.5)$$

As optical-ionisation is a sequential process, the rate of change of electron density is given by the rates defined in equation 2.2.3 and shown graphically for Helium and Neon in figure 2.2.2. The result of this process is that the leading edge of the laser pulse which causes the optical ionisation is blue-shifted. If the pulse were long enough that significant recombination could occur at the trailing edge of the pulse, then this would lead to a red-shift. For the 380 fs pulse length considered in this thesis, this is not the case and only blue-shifting occurs. In the experiment presented in chapter five blue shifts

of up to 7 nm for a 248 nm pulse were measured. This blue shifting of the fundamental laser frequency will obviously cause a blue shift in any harmonics that are generated in the plasma (see section 3.3). The blue shift of the q th harmonic will be $1/q$ of that of the fundamental. This is an effect that results in an increase of the bandwidth of both the fundamental laser and its harmonics.

2.9. Summary.

In this chapter we have discussed how a plasma is formed by optical ionisation and how energy from the laser beam is absorbed to produce heating of the plasma. Optical ionisation is a method by which a plasma with a very low electron temperature may be formed, but the process does produce some additional heating by above threshold ionisation. ATI will dominate as a heating mechanism at low electron densities ($\sim 10^{17} \text{ cm}^{-3}$), when IB heating is small. Inverse bremsstrahlung heating will quickly come to dominate over ATI heating as the electron density increases. Recent calculations of the inverse bremsstrahlung rate [39] indicate that heating by this process will be higher than previously considered. This will lead to much higher electron temperatures in our experiments than were initially hoped. The SBS instability will not have time to grow to any significant level during a 380 fs laser pulse. However, SRS has a much faster growth rate and could become the dominant heating mechanism at high intensities ($\sim 10^{17} \text{ W/cm}^2$) and high electron densities ($\sim 10^{20} \text{ cm}^{-3}$).

We must also consider ponderomotive effects which were very large in previous experiments using a 10 ps pulse length [29]. However, we conclude that, for our experimental conditions, there will be no significant expulsion of electrons from the laser focus for at least 2 ps after the peak of the laser pulse.

Finally, we see that the formation of a plasma by the laser can significantly modify the propagation of that laser. This can lead to filamentation or self-focusing which could possibly be used to produce guiding of the laser beam and extend the laser focus well beyond the Rayleigh length in vacuum [54]. We also see that significant spectral shifts of the laser wavelength can occur. This will be detrimental to the production of harmonics (chapter three), as it will lead to a much wider (time dependent) bandwidth of the harmonic.

Chapter Three

Methods of Producing Coherent XUV Radiation

In this chapter we shall review techniques for generating coherent XUV radiation from the interaction of a laser with a plasma. These essentially consist of the X-ray laser in its various forms, and harmonic generation. The review of X-ray lasers will be brief, with most space being devoted to discussion of the optically-ionised recombination scheme in anticipation of the experimental measurements presented in chapter five. The discussion of harmonic generation will be in more depth to provide a background to the results presented in chapter six. First of all, however, we must consider why we want to produce coherent X-rays.

3.1. The need for coherent X-rays.

The application that is most often discussed with reference to X-ray lasers is that of high resolution microscopy. Consider the electron microscopy system currently employed [58]. A sample must be killed, sliced into wafers, and freeze-dried to remove all traces of water before placing it in a vacuum, it must also be gold-coated to provide good contrast for the structures of interest to be imaged. If this process is applied to the delicate structure of a cell or virus, it will inevitably distort or destroy much detail. Optical microscope systems have their imaging resolution ultimately restricted by the diffraction limit for the particular wavelength of light that they use. Since the diffraction limit is proportional to wavelength, higher resolution may be achieved by using shorter and

shorter wavelengths [59-62]. This eventually brings us to a region in the spectrum of great importance to biological microscopy: the water window.

The water window is a region of the electromagnetic spectrum between 43.7 Å and 23.2 Å, it is a window above the Oxygen K-edge, but below the Carbon K-edge where the radiation is transmitted by water, but is strongly absorbed by the Carbon-rich structures of a cell. This is shown in figure 3.1.1 which plots the wavelength dependence of the transmission of Carbon and Oxygen for a common areal density of 90 µg/cm². A transmission micrograph of a cell would then show good contrast between the cell structures and the aqueous solution in which it is bathed. Such radiation sources are available in the form of synchrotron radiation [63]. With the levels of brightness currently available from synchrotrons, however, a typical exposure can last for several hours [15,64]. This requires that the cell should not move over such a time period, i.e. we need the cell to be dead.

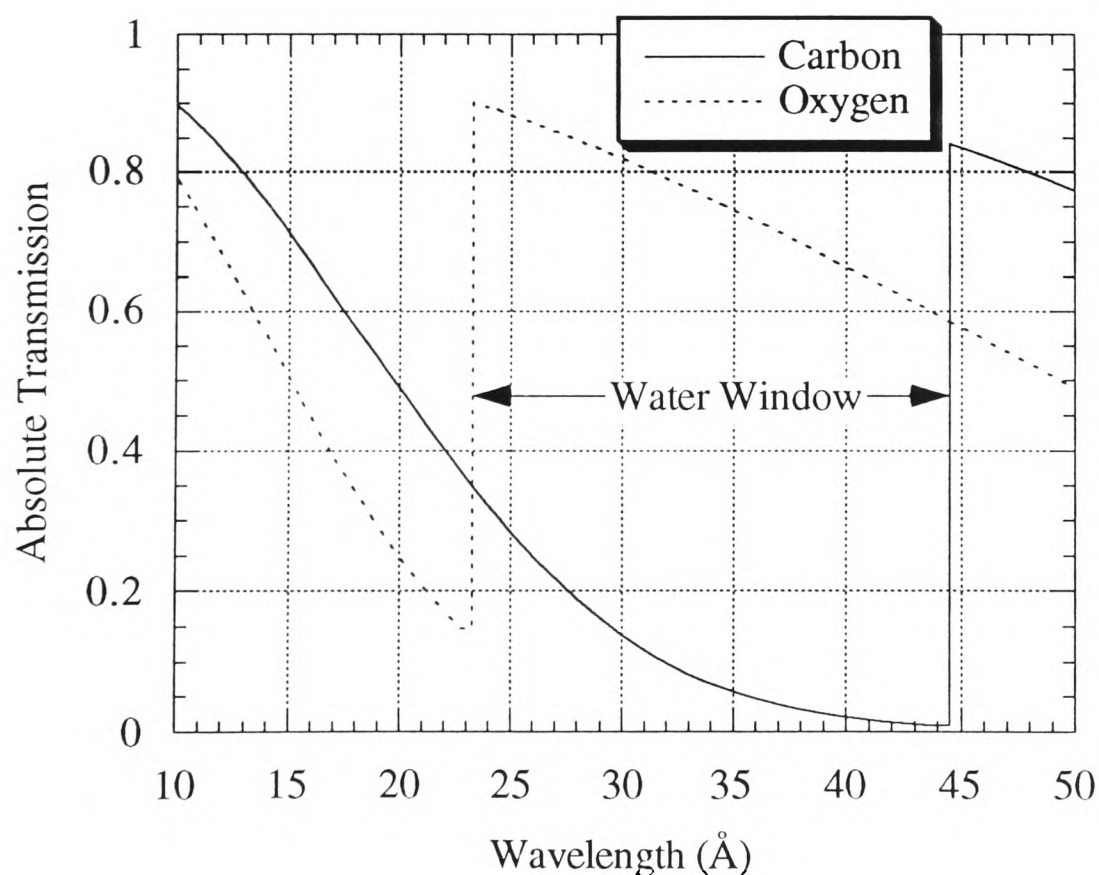


Figure 3.1.1. The water window.

Another use for coherent X-rays is as a probe where optical light cannot propagate. In a plasma, for example, electromagnetic radiation cannot propagate if the electron plasma frequency is greater than the frequency of the electromagnetic wave. Since the electron plasma frequency is proportional to the electron density, there is a critical density above which we cannot optically probe (see equation 2.1.3). By increasing the frequency of our radiation we can probe into denser regions of the plasma. In underdense plasmas a valuable diagnostic to determine the electron density is interferometry. By creating a coherent XUV source we may apply this technique to dense plasmas and produce interferograms of plasmas very near to solid density [18]. We may also use X-ray lasers to probe very small perturbations in optically thick systems. For example, illuminating a dense optical foil with an X-ray laser while it is being ablated by an optical laser will highlight density perturbations caused by the onset of the Rayleigh-Taylor instability [65].

3.2. The X-ray laser.

For high contrast microscopy of a living biological sample, we require a monochromatic radiation source with a short wavelength, a high brightness, and a short pulse length. In other words we need an X-ray laser. The short pulse length is required because, at the high brightness needed for a single shot exposure, the cell will be killed almost instantly by radiation exposure and will then start to move hydrodynamically as it explodes. It has been calculated that to image the cell before this happens, and to produce a resolution of 300 Å, we need a pulse length no longer than about 17 ps [64]. The added advantage of an X-ray laser source is that it exhibits both longitudinal and transverse coherence, making it possible to produce a hologram by taking a reflection from the cell and interfering it with a reference beam [16,17]. The optimum wavelength required for imaging depends upon whether we require a transmission image or a reflection image of our sample. The optimum wavelength for a transmission image is at the shorter

wavelength end of the water window, where the Oxygen atoms in the water have their lowest absorption. For a reflection image, the optimum wavelength is at the long wavelength end of the water window. In fact, for maximum reflectivity we should be just to the long wavelength side of the Carbon K-edge at about 44 Å [64].

X-ray laser action has been demonstrated in the water window [66], but the system has an extremely low efficiency, and is not very powerful. Many systems have now been developed at longer wavelengths which exhibit much higher efficiencies and powers, and have been used for X-ray holography of man-made objects [16,17] and as interferometric probes of dense plasmas in which optical radiation cannot propagate [18]. A shorter wavelength X-ray laser has been used to image rat sperm nuclei with a resolution of 500 Å [67].

The first X-ray laser scheme was demonstrated in 1985 at the Lawrence Livermore National Laboratory in the U.S.A., and was based on a collisionally-pumped scheme [68]. Since then recombination schemes have also been developed. Both these systems are described in this chapter, along with the newer optically-ionised laser schemes and a potential competitor to the plasma-based X-ray laser: harmonic generation.

3.2.1. The collisional X-ray laser.

To produce laser action we must create a population inversion in the laser medium. To do this we need some way of pumping and filling an upper laser level in preference to a lower level. Whether this population inversion is possible depends on the relative strengths of the recombination and collisional rates populating the laser levels. The collisional rate between an upper and lower level is given by

$$C_{ul} \propto \frac{Nf_{ul}}{\Delta U_{ul} T_e^{1/2}} \text{ s}^{-1} \quad (3.2.1)$$

whereas the spontaneous emission rate coefficient goes as

$$A_{ul} \propto f_{ul} \Delta U_{ul}^2 \text{ s}^{-1} \quad (3.2.2)$$

where f_{ul} is the oscillator strength of the transition, ΔU_{ul} is the energy difference between the two energy levels. High lying levels separated by small energy gaps will be dominated by collisional population and depopulation, will always be in local-thermodynamic equilibrium (LTE), and so will never achieve a population inversion. At some lower level in the atom, radiative transitions will dominate and it becomes possible to create a non-LTE population inversion. Hence, the lower laser level must be below this 'thermal limit' for population inversion to occur.

The collisional X-ray laser scheme relies on excitation by electron impact to pump an ion from the ground state to higher bound levels. A population inversion can then be built up between two levels of the same principal quantum number if the lower level has a fast ($\Delta l = \pm 1$) radiative decay route to the ground level and if the upper level has a much slower ($\Delta l = 0$) decay route. Such schemes are mostly based on a Neon-like electronic configuration (a ground state configuration of $2p^6$) with laser transitions between the $2p^5 3p$ and $2p^5 3s$ levels. Figure 3.2.1 shows a Grotrian diagram of the levels involved where the laser transitions are shown as bold arrows. The strongest transitions are those from the $(1/2, 1/2)_0$ and the $(1/2, 3/2)_0$ levels of the $2p^5 3p$ configuration to the $2p^5 3s$ configuration.

Iso-electronic scaling to shorter wavelengths on the Neon-like scheme is limited because the $3p$ - $3s$ (laser) splitting increases more slowly with ionic charge than the $2p$ - $3s$ excitation energy. For Nickel-like ions, however, the $4d$ - $4p$ splitting is a greater fraction of the excitation energy of the $3d$ - $4p$ transition. This leads to a much greater quantum efficiency of the laser scheme and so scaling to shorter wavelengths is much easier. A

Grotrian diagram of the Nickel-like laser scheme is shown in figure 3.2.2. The $J = 0 - 1$ transitions have the highest gain. A Nickel-like scheme, lasing in the water window, has been demonstrated at the Lawrence Livermore National Laboratory [66], but the driver laser energy required to pump the scheme is very large.

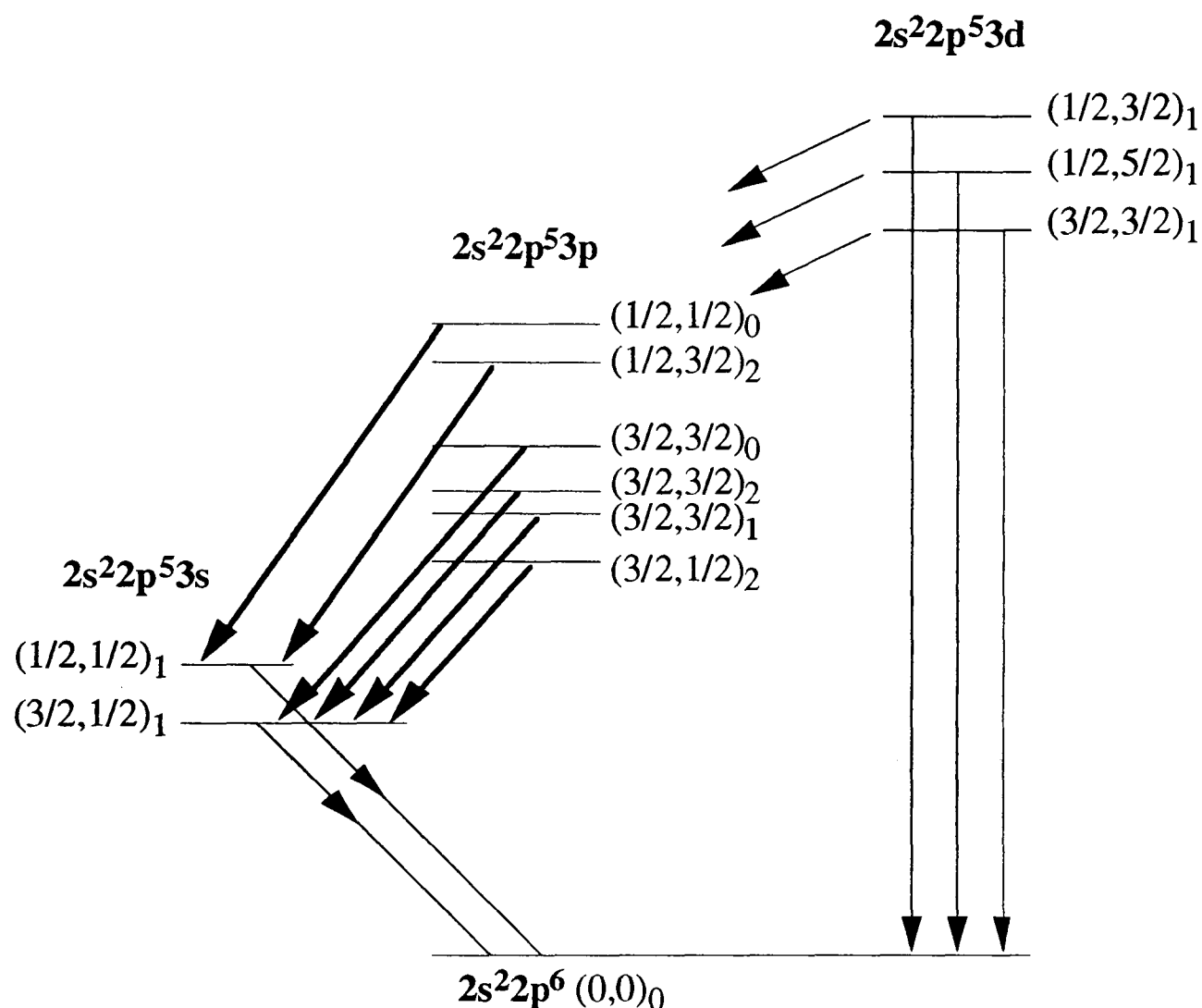


Figure 3.2.1. Grotrian diagram of the Neon-like collisionally-pumped laser scheme. The laser transitions are shown as bold arrows.

Various pumping geometries and target configurations have been developed for the collisional X-ray laser. Initial experiments were based on exploding foil systems [69,70], which then moved to slab geometries [70]. More recent developments have included bending slab targets [71] and double slab targets [72]. The development of multi-layer mirrors with reflectivities of up to 70% has enabled injector/amplifier systems and multi-pass geometries [73] to be used. These innovations have produced great improvements in the laser beam quality and the laser output power.

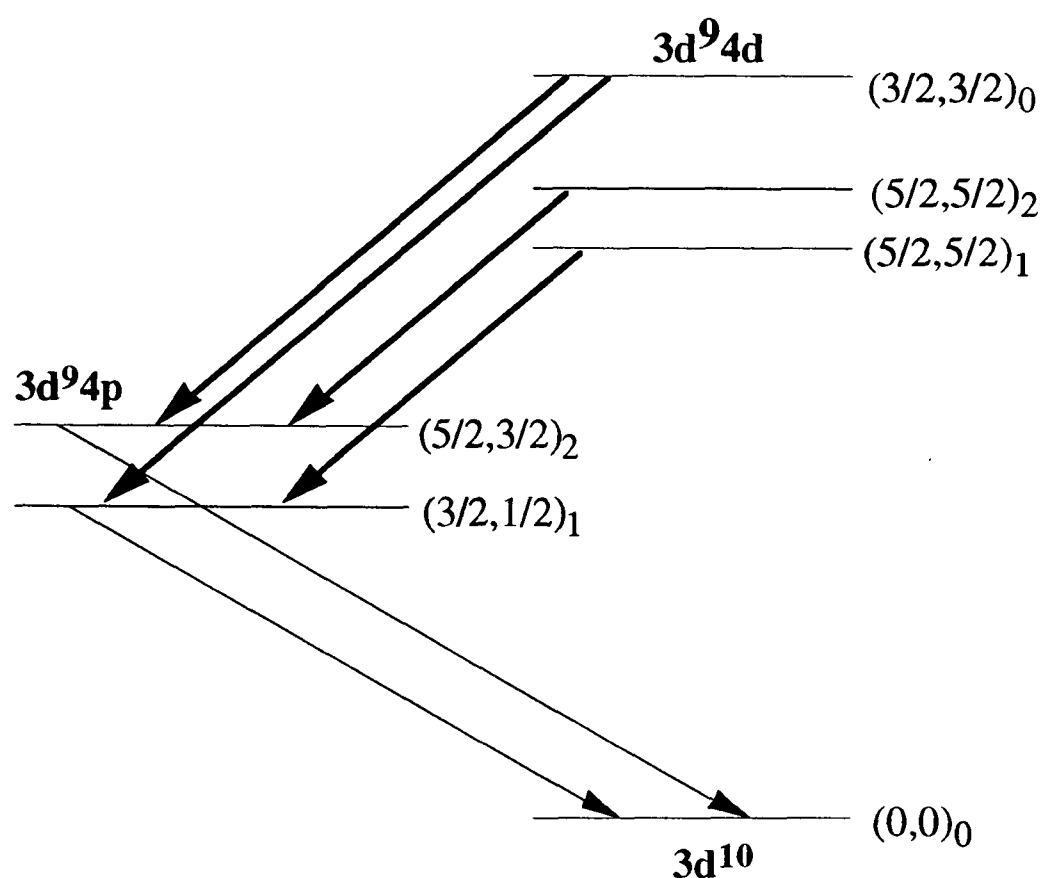


Figure 3.2.2. Grotrian diagram of the Nickel-like collisionally-pumped laser scheme. The laser transitions are shown as bold arrows.

3.2.2. The recombination X-ray laser.

The recombination pumping scheme relies on plasma production by laser ablation in the same way as the collisional scheme. Unlike the collisional laser, pumping of the upper laser level is via a fast radiative decay. Once formed, the plasma is rapidly cooled: either by adiabatic expansion; or by radiative cooling. The three body recombination rate scales inversely to the electron temperature and so, if a low enough plasma temperature is attained, electrons rapidly recombine into the upper levels of the ions. Above the thermal limit, transitions between levels are governed by collisions. Below this, electrons rapidly cascade down to lower bound levels via radiative transitions. With a fast enough radiative cascade, and if the lower laser level is a resonance transition with a fast decay route to the ground state, it is possible to build up a population inversion between two levels of differing principal quantum number (the $n = 3$ and $n = 2$ levels of a Hydrogen-

like ion, for example). Figure 3.2.3 schematically compares the recombination laser scheme to the collisional scheme.

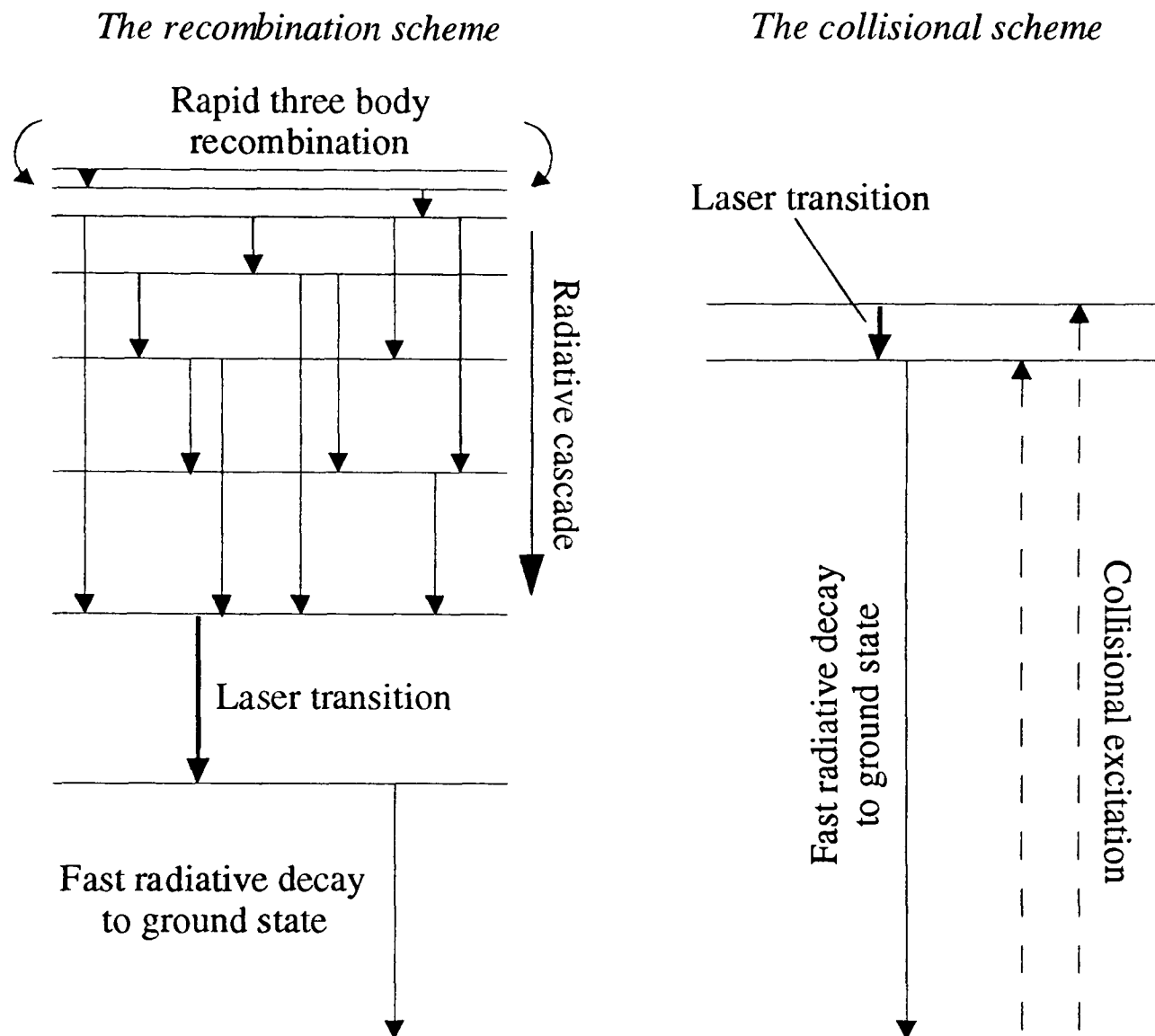


Figure 3.2.3. Schematic diagram comparing the recombination and collisional laser schemes.

Recombination laser schemes have the obvious advantage over collisional schemes in that they have a much higher quantum efficiency. The disadvantage is the need to quickly cool the plasma which can make it difficult to achieve a large population inversion. Some of the problems associated with this laser scheme are being overcome by the use of CPA laser systems to produce a driver laser pulse that is short enough that it doesn't significantly heat the plasma as it is supposed to be expanding adiabatically [74,75]. If heating continues for too long, the plasma will be quite tenuous by the time it has

expanded and cooled. This will prevent the rapid three body recombination that is needed to create a population inversion.

Recombination schemes are generally based on exploding fibre geometries. This causes considerable engineering problems with accurately aligning a fibre that is typically 1 cm long and 7 μm in diameter. The highest gain reported to date is a time and space integrated gain of 12.5 cm^{-1} at 18.2 nm in a C VI scheme [76].

3.2.3. The optically-ionised recombination X-ray laser.

An optically ionised XUV laser can be either recombination-pumped or collisionally-pumped. They are set apart from the schemes described in sections 3.2.1 and 3.2.2, not by the way in which pumping of the upper laser level occurs, but by how the plasma is produced.

Optically-ionised schemes make use of very short pulse-length pump lasers (typically $\sim 100 \text{ fs}$) to produce very high powers ($\sim 1 \text{ TW}$) and focused intensities ($> 10^{16} \text{ W/cm}^2$), but with very little energy ($\sim 100 \text{ mJ}$) to produce a plasma by optical-ionisation of a gas target (rather than by ablation of a solid target). To keep the pump energy requirements to a minimum the XUV laser is longitudinally pumped, i.e. the XUV laser action will occur along the same axis as the pump laser. The intensity required for optical ionisation to the charge state of interest can be calculated from the Ammosov equation given in section 2.2.2.

The optically-ionised, or optical-field ionised (OFI), recombination XUV laser scheme makes use of a linearly polarised pump laser to create a plasma with an extremely low electron temperature that is determined essentially by the ATI temperature of the electrons. If the pulse length of the pump laser is much shorter than the electron-electron

and the electron-ion collision times, then inverse bremsstrahlung heating should be negligible. This approach differs from the 'traditional' recombination laser scheme in that we are trying to create a cold electron distribution rather than letting the electrons heat up in the laser pulse and then cool by adiabatic expansion of the plasma. This is obviously a more energy efficient approach, but the task of generating such a cold plasma is far from trivial.

Extensive theoretical studies have been made of these schemes [44,77-80], the favoured system being a Lithium-like scheme where, at the appropriate temperatures and densities, a population inversion is predicted between the $3d_{5/2}$ excited level and $2p_{3/2}$ ground level. Two possible schemes are based on Ne VIII and N V. Hydrogen-like schemes are also possible, but the higher pump laser intensity required to ionise the 1s-shell is expected to cause problems with increased IB and SRS heating which will raise the electron temperature to too high a level.

Let us consider the Ne VIII scheme in more detail, with particular reference to simulations performed by Eder et al. [44,78-80]. They model a plasma which is optically-ionised by a laser with a wavelength of 248 nm, and a pulse length of 50 or 100 fs containing 25 - 100 mJ focused to an intensity of 3×10^{17} W/cm². A maximum gain of over 100 cm⁻¹ is predicted to occur about 300 fs after the peak of the ionising laser pulse on the 98 Å ($3d_{5/2}$ - $2p_{3/2}$) transition at an electron density of 5×10^{20} cm⁻³ and an electron temperature of 40 eV. If the temperature increases to 100 eV, the gain reduces to 13 cm⁻¹. Table 3.2 shows the variation of some important laser parameters as powers of T_e and N . The variation of G_{max} and $G_{max}L_{sat}$ with electron temperature at an electron density of 5×10^{20} cm⁻³ is shown graphically in figure 3.2.4. (L_{sat} is the length over which the laser will reach saturation).

<i>Scaling</i>	G_{max}	$G_{max}L_{sat}$	L_{sat}	I_{sat}	Δt	η
T_e	-2.6	+0.5	+3.1	+0.6	-0.1	-2.6
N	+1.5	-0.1	-1.6	+1.7	-0.6	+2.7

Table 3.2. *Scaling of XUV laser parameters as powers of T_e and N . A plasma radius of $5 \mu m$ was used to calculate L_{sat} and η . For the T_e scaling, we take $N = 5 \times 10^{20} cm^{-3}$ and for the N scaling, we take $T_e = 40 eV$. Δt is the gain duration of the laser action.*

Recent revised calculations [81] suggest that heating by three body recombination, which was left out of Eder et al.'s original simulations, is an important factor in determining the electron temperature during laser action and may reduce the gain to levels similar to those seen in traditional recombination XUV lasers, i.e. $4 - 5 cm^{-1}$. Experiment has also shown that, in a non-thermal plasma generated by OFI, the three-body recombination rate can be considerably reduced [82]. Since laser action commences so soon after ionisation, an upper limit is imposed on the length of the laser by the different speeds of propagation of the ionising laser and the XUV laser radiation through the plasma. The plasma refractive index takes the form $n = (1 - N/N_{cr})^{1/2}$, where N_{cr} is the critical plasma electron density. This means that the XUV radiation, travelling essentially at the speed of light, catches up with the 248 nm ionising laser, which has a speed less than the speed of light. The relative speed between the two pulses is $u = cN/2N_{cr}$, so for an electron density of $5 \times 10^{20} cm^{-3}$ an XUV pulse starting 300 fs after the peak of the ionising laser pulse will have caught up with it after a distance of just over 6 mm. During this time the XUV pulse will be moving ahead of the region of peak gain, so the total gain in the system will be somewhat reduced from the maximum values quoted above.

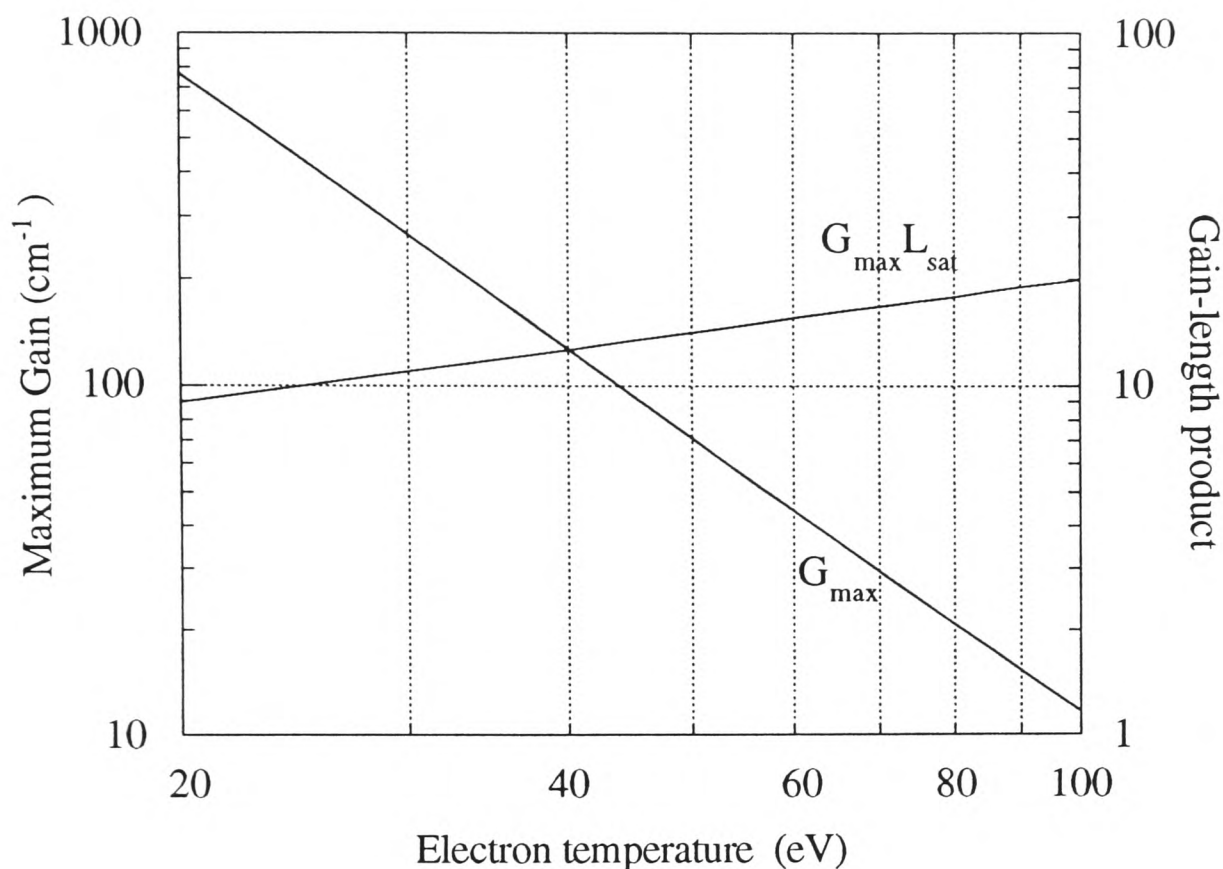


Figure 3.2.4. Variation of G_{max} and $G_{max}L_{sat}$ with the electron temperature at an electron density of $5 \times 10^{20} cm^{-3}$.

Claims have already been made of observation of gain in the OFI recombination scheme. For example, claims have been made of gain in Hydrogen-like Lithium [83,84], but there is some dispute as to the validity of the interpretation of the data. The main criticism of the papers is that the authors choose to compare the intensity ratios of two lines from different species of Lithium (Helium-like and Hydrogen-like). Another claim of possible gain has been made [22, 23], but this time in the 5f-3d transition of Lithium-like Nitrogen (at 512 Å) and in the 2s2p²-2s2p3d transition in N²⁺ (at 472 Å). These are obviously much easier transitions in which to create a population inversion, as they do not involve a transition to the ground state. However, this considerably reduces the quantum efficiency of the laser.

It is the possibility of this type of OFI recombination scheme that has stimulated much of the research in this thesis. Measurements of the density dependence of the electron

temperature in an OFI plasma produced using the 248 nm, 380 fs Sprite CPA system and their implications to the scaling of the laser system to shorter wavelengths are discussed in detail in chapter five.

3.2.4. The optically-ionised collisional X-ray laser.

Unlike the OFI recombination scheme, the OFI collisional laser uses circularly polarised light to generate a very hot electron temperature. The intensity of the pump laser is chosen to produce the required ionisation stage, and then the hot electrons excite an upper laser level by monopole electron excitation from the ground state. The population inversion occurs between two levels with the same principal quantum number (between which the electric dipole transition probability is relatively low). Such a scheme has been modelled [85] and demonstrated experimentally [86] by Lemoff et al. at Stanford University, California. Simulations predict gain in Ar IX (Neon-like), Kr IX (Nickel-like), and Xe IX (Palladium-like) schemes at wavelengths of 478, 319, and 410 Å respectively. Energy level diagrams, indicating the laser transitions, are shown in figure 3.2.5.

The Xe IX system has been shown to work experimentally using a 10 Hz, 800 nm Ti:Sapphire laser pulse with an energy of ~70 mJ and a pulse length of ~40 fs [86]. This is focused into a gas cell containing 5 to 12 torr of Xe. Amplification of the 41 nm line was seen with an estimated gain coefficient of 13 cm^{-1} over a length of 8.5 mm. Because the laser pulse is so short compared to the electron-ion collision time, the plasma is optically thick at 16.5 nm and so the lower laser level cannot empty into the ground state. This causes the laser to self-terminate after about one lifetime of the upper laser level (~30 ps for pressures <1 torr and ~3.5 ps at 12 torr) although this cannot be verified, because the laser output is time integrated on the detector. Interestingly, an attempt to reproduce this system using a 1 ps laser pulse failed to generate any significant gain [87].

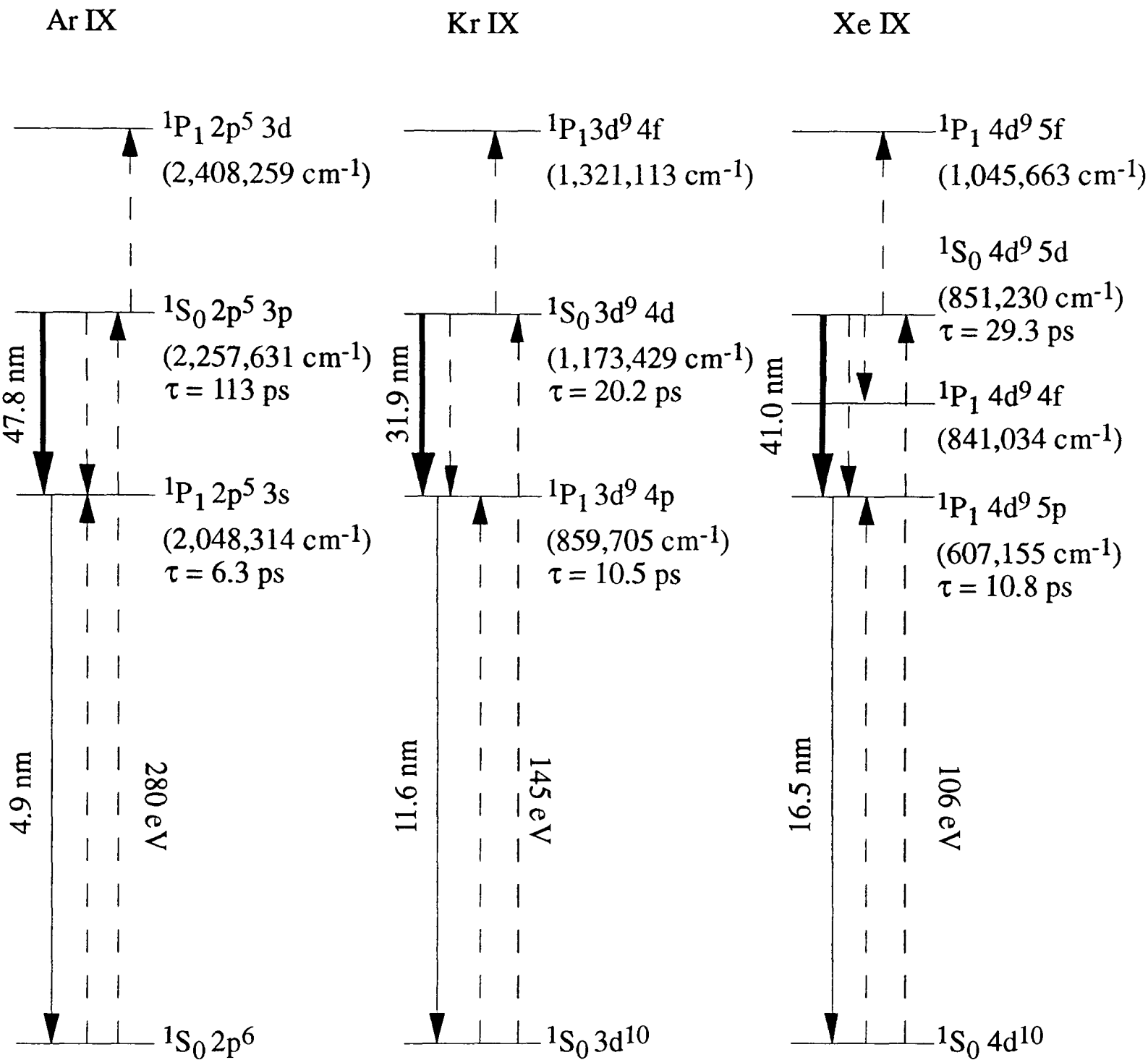


Figure 3.2.5. Grotrian diagrams of the Neon-, Nickel-, and Palladium- like OFI collisional laser schemes showing the laser transitions as bold arrows. Taken from Lemoff et al. [85].

3.3. Harmonic generation.

A relatively new phenomenon that is rapidly developing along with the progress in short pulse laser technology is the process of high harmonic generation. The first demonstration of the generation of a harmonic of a laser beam was performed in 1961 by

Franken et al. [88]. This experiment focused a 694 nm ruby laser into a quartz crystal. The light transmitted through the crystal was found to contain a second harmonic component. The conversion efficiency of this was of the order of 10^{-8} . Following over three decades of development, second harmonic conversion efficiencies can now exceed 50% and the technique is used in many laser systems.

In this section we shall discuss the process of high harmonic generation, both in the weak and strong field regimes. We will also discuss the effect of phase matching between the harmonic and fundamental. By weak or strong field we refer to the electric field amplitude that can be produced by a focused laser beam (the laser field). Non-linear effects become significant at laser intensities above about $10^{10} - 10^{11}$ W/cm². Below this intensity the interaction of light with matter is essentially a linear process. Above this, the laser field becomes strong enough to perturb the atomic potential. The weak field regime is that where perturbation theory can be used. The upper intensity limit of this regime is dependent on the structure of the atom under consideration. For example, the electric field at one Bohr radius from a Hydrogen nucleus is 5.14×10^{11} V/m. This is equivalent to the electric field amplitude produced at a laser intensity of 3.5×10^{16} W/cm². The critical intensity for the optical-ionisation of Hydrogen is about 1.4×10^{14} W/cm², i.e. optical ionisation starts to occur when the external field is greater than about 6% of the atomic field. We will define the weak field limit to be well below this level and the strong field limit to be above this. Obviously, this limit will be different for different atomic species depending on their ionisation potential.

3.3.1. Harmonic generation in the weak field regime.

When an atom is exposed to an electric field, the centre of charge of its electrons is displaced from its equilibrium position about the nucleus. This effect is shown in figure 3.3.1 where we depict schematically the electron distribution (with a total charge of $-Ze$)

of an atom in a laser field which has been shifted away from its equilibrium position about the nucleus of charge Ze .

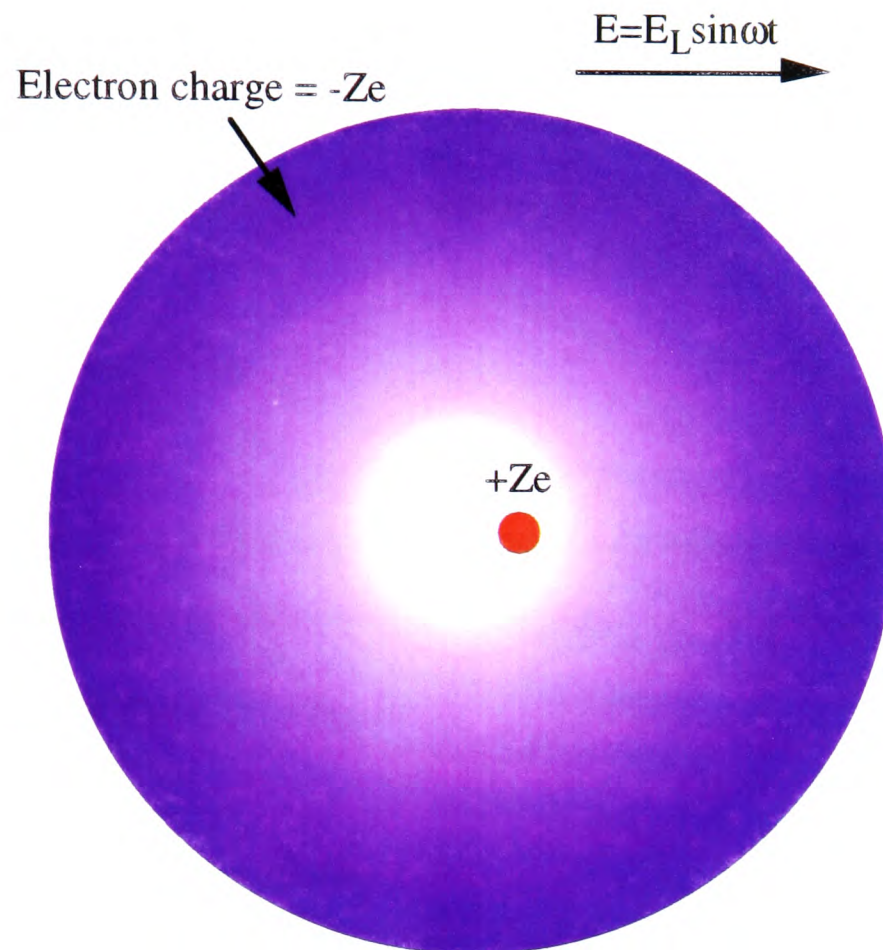


Figure 3.3.1. Schematic diagram showing the movement of the electron distribution of an atom away from its equilibrium position in the presence of an electric field.

The restoring force for small displacements (produced by small electric fields) can be considered to be purely linear, and we represent the Coulomb force on the electrons as that of a simple harmonic oscillator potential. This displacement induces a net polarisation of the medium in which the atom is sited. For small electric fields the polarisation is given by the linear relationship

$$\mathbf{P}^L = \epsilon_0 \chi^{(1)} \mathbf{E} \quad (3.3.1)$$

where $\chi^{(1)}$ is the complex linear susceptibility, given by

$$\chi^{(1)} = \bar{\chi}^{(1)} + i\tilde{\chi}^{(1)} = \frac{N_0}{\epsilon_0 \hbar} \sum_a \frac{|\mu_{ag}|^2}{(\Omega_{ag} - \omega)} \quad (3.3.2)$$

where N_0 is the number density of atoms or molecules in the medium, μ_{ag} is the dipole matrix element for the transition between states $|g\rangle$ and $|a\rangle$, and where

$$\Omega_{ag} = \omega_{ag} - i\Gamma_{ag} \quad (3.3.3)$$

is the complex transition frequency containing the damping frequency Γ_{ag} . The linear susceptibility defines the refractive index n according to $1 + \chi^{(1)} = \epsilon_r = n'^2$ where n' is the complex refractive index, $n' = n + i\tilde{n}$

3.3.1.1. The non-linear susceptibility.

As we increase the laser field, this linear relation starts to break down. This can be explained if we consider the modification of the Coulomb field by the laser field. Figure 3.3.2 shows how the centre of charge of the electron distribution is moved by the addition of a laser field to the Coulomb potential. If we assume that the photon energy of the laser is much less than the energy of the ground state of the atom, the potential, U , at $-a_0$ is lowered by approximately $E_L a_0$ (where E_L is the external electric field applied to the atom, and a_0 is the Bohr radius) and the potential at $+a_0$ is raised by the same amount, ΔU . The displacement the electron from its equilibrium position by this perturbation is given by

$$r = (r)_{r=a_0} + \Delta U \left(\frac{dr}{dU} \right)_{r=a_0} + \frac{(\Delta U)^2}{2!} \left(\frac{d^2 r}{dU^2} \right)_{r=a_0} + \frac{(\Delta U)^3}{3!} \left(\frac{d^3 r}{dU^3} \right)_{r=a_0} + \dots + \frac{(\Delta U)^n}{n!} \left(\frac{d^n r}{dU^n} \right)_{r=a_0} \quad (3.3.4)$$

if we substitute a hydrogenic potential into this we derive

$$r(\Delta U) = a_0 \left(-1 + \frac{\Delta U}{ea_0 E_{at}} - \left(\frac{\Delta U}{ea_0 E_{at}} \right)^2 + \left(\frac{\Delta U}{ea_0 E_{at}} \right)^3 + \dots \right. \\ \left. \dots + (-1)^{n+1} \left(\frac{\Delta U}{ea_0 E_{at}} \right)^n \right) \quad (3.3.5)$$

where $E_{at} = e/4\pi\epsilon_0 a_0^2$ and $\Delta U = \pm a_0 E_L$.

The displacement of the centre of charge of the electron from its central equilibrium position is given by $x = r(a_0 E_L) - r(-a_0 E_L)$ which gives

$$x = 2a_0 \left(\left(\frac{E_L}{eE_{at}} \right) + \left(\frac{E_L}{eE_{at}} \right)^3 + \left(\frac{E_L}{eE_{at}} \right)^5 + \dots + \left(\frac{E_L}{eE_{at}} \right)^{n(=odd)} \right) \quad (3.3.6)$$

A result which is only valid in the weak field regime, i.e. when the laser field is much smaller than the atomic Coulomb field. Since the susceptibility, $\chi = Nex/\epsilon_0 E_L$, we can define a series of non-linear susceptibilities which are given approximately by $\chi_n = (2Nea_0/\epsilon_0 E_{at})(E_L/E_{at})^{n-1}$. Hence we see that the non-linear susceptibilities scale as E_L^{q-1} , where q is the harmonic order, and the ratio of one susceptibility to the next varies as $\chi^{n+2}/\chi^n = (E_L/eE_{at})^2$.

It should be noted that the disappearance of even order susceptibilities is a consequence of the atomic potential being symmetrical with respect to reflection about the origin. In the case of a non-centrosymmetric potential (as can be experienced in a crystal lattice), the even order susceptibilities will not disappear. This results in the generation of even as well as odd harmonics.

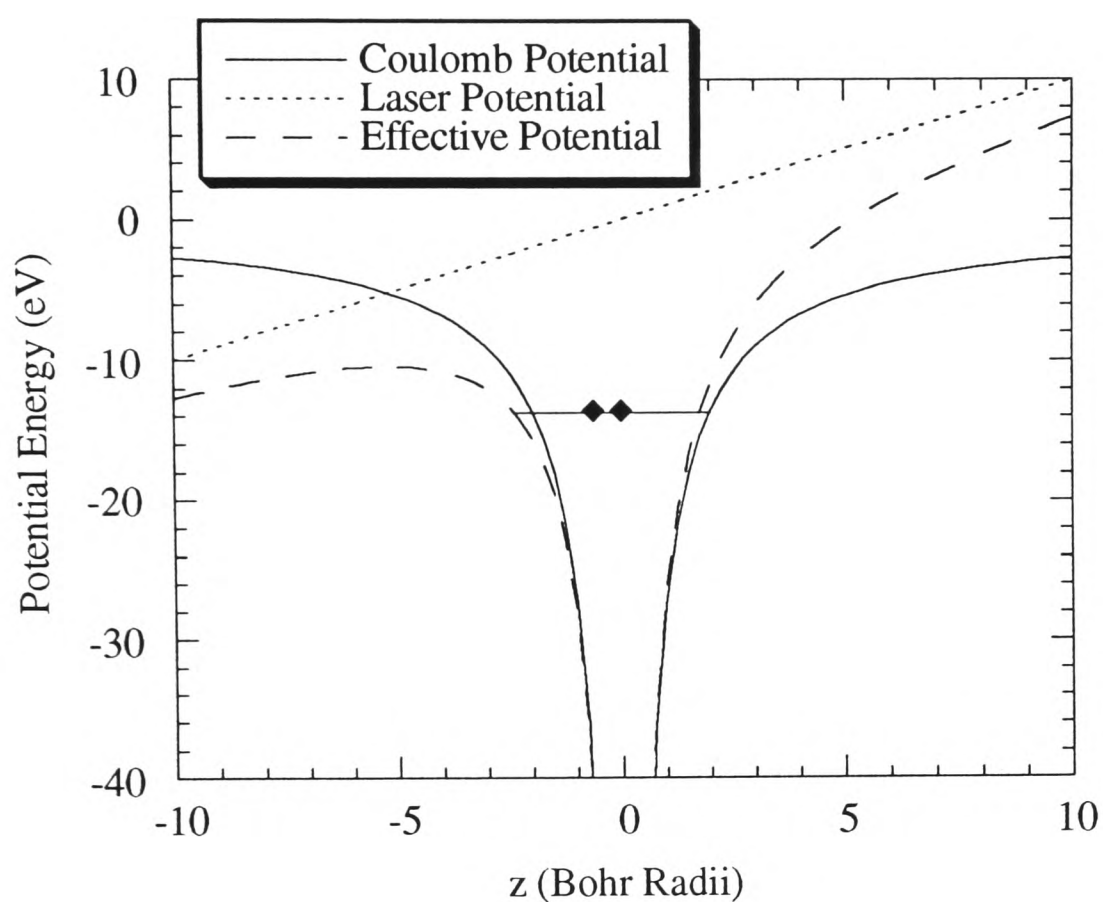


Figure 3.3.2. The movement of the centre of charge of the electron distribution in a Hydrogen potential modified by the electric field of a laser.

These non-linear susceptibilities obviously lead to a non-linear polarisation of the medium which can be introduced by including a non-linear polarisation term

$$\mathbf{P}^{NL} = \sum_{n=2}^{\infty} \mathbf{P}^{(n)} \quad (3.3.7)$$

A more rigorous definition of the nth order non-linear susceptibility is given by

$$\mathbf{P}_{\alpha_s}^{(n)}(r, \omega_s) = \frac{n!}{2^{n-1}} \sum_{\alpha_1 \dots \alpha_n} \chi_{\alpha_s \alpha_1 \dots \alpha_n}^{(n)}(-\omega_s; \omega_1 \dots \omega_n) \times \mathbf{E}_{\alpha_1}(r, \omega_s) \dots \mathbf{E}_{\alpha_n}(r, \omega_n) \quad (3.3.8)$$

where $\alpha_1, \alpha_2 \dots \alpha_s$ are Cartesian subscripts (x, y or z). This shows that the non-linear susceptibility $\chi^{(n)}$ is a $(n+1)$ -rank tensor which in almost all cases of practical interest reduces to just a few significant numbers.

As we are considering high harmonic generation we must use a gas, as solids will absorb radiation below about 200 nm (which corresponds to approximately the 5th harmonic of a 1053 nm Nd:YAG laser, the 4th harmonic of an 800 nm Ti:Sapphire laser, or any harmonic of a 248 nm KrF laser. Since a gas is isotropic, its even order terms $\chi^{(2)}, \chi^{(4)} \dots$ will generally vanish because the polarisation must be unaffected by reflection of the co-ordinate system at its origin. This can be seen for the case of a Hydrogen potential in equation 3.3.6.

3.3.1.2. The third order non-linear susceptibility.

To consider the non-linear susceptibilities involved in harmonic generation we shall specifically introduce the third order susceptibilities. These have been chosen to simplify the equations, but the conclusions can be applied to any non-linear order. We are looking at the specific case of four wave mixing in which a sum frequency signal at

$$\omega_s = \omega_1 + \omega_2 + \omega_3 \quad (3.3.9)$$

is generated. This process is shown schematically in Figure 3.3.3. We are considering only the special case of $\omega_1 = \omega_2 = \omega_3 = \omega_s/3$ i.e. third harmonic generation. If we expose a non-linear medium to an incident wave of angular frequency ω we have to consider the following three non-linear susceptibilities (only the dominant terms of interest are shown):

$$\begin{aligned}
\chi^{(3)}(-3\omega; \omega\omega\omega) &= \chi_T^{(3)}(3\omega) \\
&= \frac{N}{\epsilon_0 \hbar^3} \sum_{abc} \frac{\langle g|e_s\mu|a\rangle\langle a|e_1\mu|b\rangle\langle b|e_2\mu|c\rangle\langle c|e_3\mu|g\rangle}{(\Omega_{ag} - \omega)(\Omega_{bg} - 2\omega)(\Omega_{cg} - 3\omega)}
\end{aligned} \tag{3.3.10}$$

$\chi_T(3\omega)$ specifies a polarisation at 3ω which gives rise to the desired third harmonic signal.

$$\begin{aligned}
\chi^{(3)}(-\omega; \omega - \omega\omega) &= \chi_{SA}^{(3)}(\omega) \\
&= \frac{N}{\epsilon_0 \hbar^3} \sum_{abc} \frac{\langle g|e_s\mu|a\rangle\langle a|e_1\mu|b\rangle\langle b|e_2\mu|c\rangle\langle c|e_3\mu|g\rangle}{(\Omega_{ag} - \omega)(\Omega_{bg} - 2\omega)(\Omega_{cg} - \omega)}
\end{aligned} \tag{3.3.11}$$

$\chi_{SA}(\omega)$ causes a polarisation at ω and can be viewed as a field dependent correction to the refractive index. Its real part is responsible for a field dependent change in the refractive index, whereas the imaginary part entails a two photon absorption.

$$\begin{aligned}
\chi^{(3)}(-\omega; \omega 3\omega - 3\omega) &= \chi_{SB}^{(3)}(\omega, 3\omega) \\
&= \frac{N}{\epsilon_0 \hbar^3} \sum_{abc} \frac{\langle g|e_s\mu|a\rangle\langle a|e_1\mu|b\rangle\langle b|e_2\mu|c\rangle\langle c|e_3\mu|g\rangle}{(\Omega_{ag} - 3\omega)(\Omega_{bg} - 2\omega)(\Omega_{cg} - \omega)}
\end{aligned} \tag{3.3.12}$$

$\chi_{SB}(\omega, 3\omega)$ causes a polarisation at the fundamental frequency which originates from a simultaneous interaction of the fundamental and its third harmonic wave with the non-linear medium. Its real part is a change of refractive index at the fundamental frequency due to the harmonic wave, the imaginary part characterises a Raman-type gain or loss of the fundamental wave under the influence of the harmonic wave.

All three of these expressions are similar and differ only in their denominators, i.e. in the frequencies at which the susceptibilities become resonant.

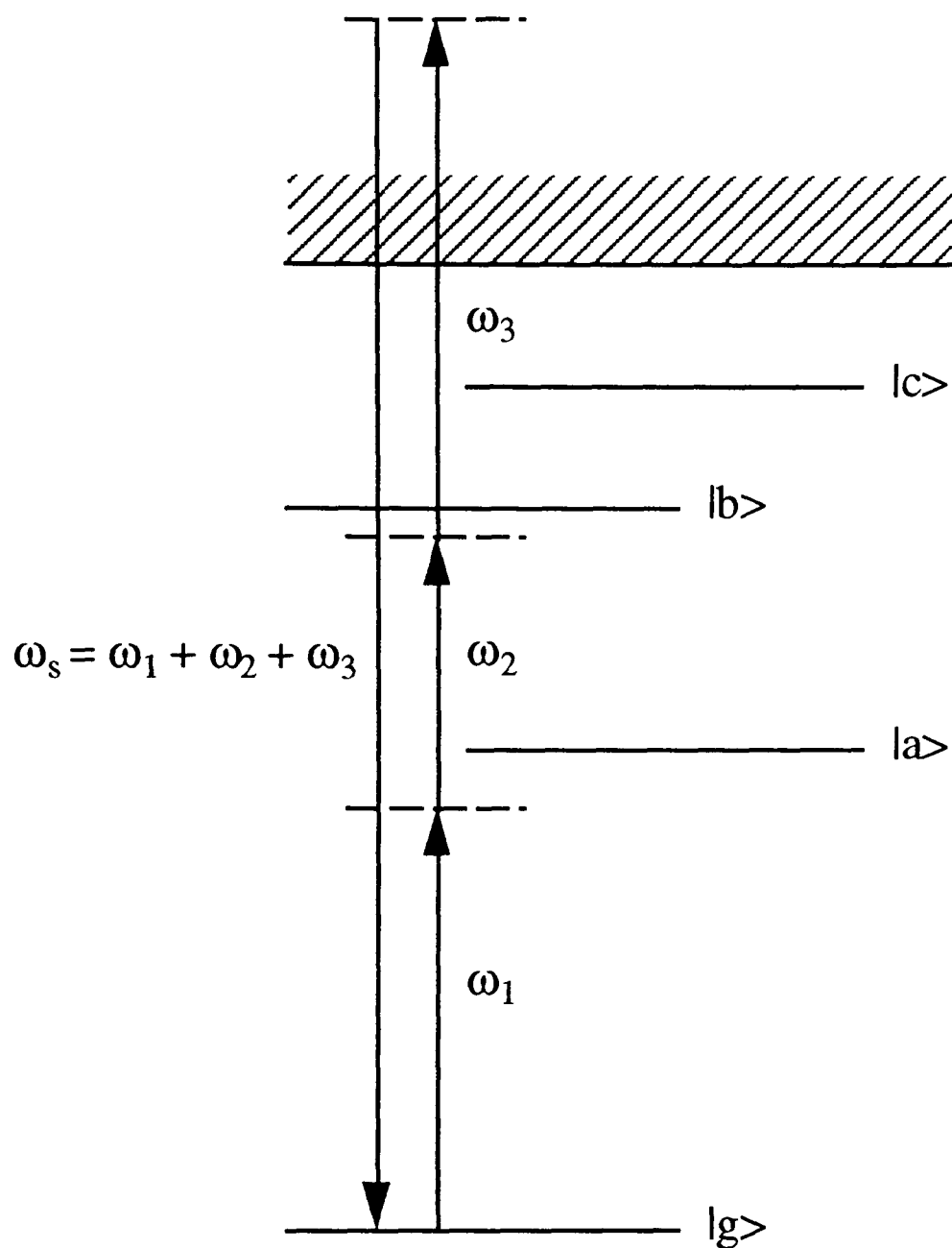


Figure 3.3.3. Energy level representation of the general four wave mixing process. We are interested in the case $\omega_1 = \omega_2 = \omega_3 = \omega_s/3$.

3.3.1.3. Selection rules for harmonic generation.

Using the Wigner-Eckart theorem the individual matrix elements in the above numerators can be written as

$$\langle \alpha J_a m_a \| \beta J_b m_b \rangle = (-1)^{J_a - m_a} \begin{pmatrix} J_a & 1 & J_b \\ -m_a & \Delta m_k & m_b \end{pmatrix} \langle \alpha J_a \| \mu \| \beta J_b \rangle \quad (3.3.13)$$

where $\langle \alpha J_a \| \mu \| \beta J_b \rangle$ is the reduced matrix element. Δm_k inside the 3j-symbol is equal to +1(-1) for right-(left-) circularly polarised radiation and equal to zero for linear polarisation. Using the selection rule $-m_s + \Delta m_k + m_b = 0$ we immediately obtain

$$\Delta m_s + \Delta m_1 + \Delta m_2 + \Delta m_3 = 0 \quad (3.3.14)$$

This relation implies conservation of angular momentum. Note that two left- and one right-hand waves result in a right-hand wave, but three waves of the same handedness do not result in any sum frequency wave at all. Since the numerators contain electric dipole matrix elements, we see that states $|a\rangle$ and $|c\rangle$ have opposite parity with respect to states $|g\rangle$ and $|b\rangle$.

For enhancing χ_T we must minimise the resonant denominators in equation 3.3.10. However we must be careful, as minimising the one photon resonance may lead to strong absorption of the fundamental, and minimising the three photon resonance may strongly absorb the third harmonic. The third harmonic generation efficiency is ultimately limited by competition between χ_T and χ_{SA} .

3.3.2. Harmonic generation in the strong field regime.

In the weak field regime the amplitude of the q th harmonic scales with the electric field as E^q (see equation 3.3.6). However, the q th order non-linear polarisation diminishes rapidly with increasing q . As the laser field is increased the harmonic intensities are seen to saturate and increase in magnitude much more slowly. As the laser field continues to increase, higher and higher harmonics become saturated and form a plateau. The plateau harmonics will scale similarly with intensity and seem to exhibit an E^m dependence, where m is somewhere between 6 or 8. We are now seeing a different mechanism of

harmonic generation. Figure 3.3.4 shows schematically the progression of the harmonic spectrum from the weak field to the high field regime.

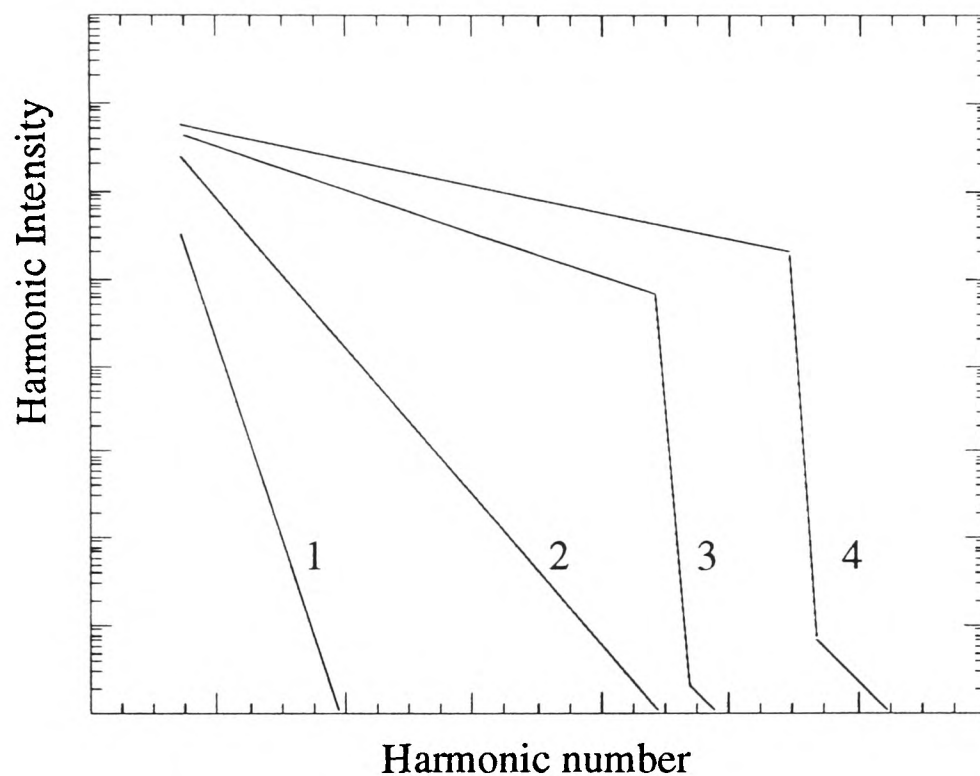


Figure 3.3.4. *The development of a harmonic spectrum with increasing fundamental laser intensity, 1 - 4. 1 and 2 are in the weak field regime, whereas 3 and 4 show the response in the strong-field case. Note the perturbative tail at the root of the cut-off in 3 and 4.*

3.3.2.1. The classical equations of motion and the cut-off energy.

In the strong-field regime, i.e. at laser intensities when the laser electric field is of the same order as the atomic electric field, we may consider harmonic generation using a semi-classical approach [89]. In this approximation, the electron is optically-ionised (the rate of which we may determine by the Ammosov equation [33] discussed in section 2.2.2). This ionisation may occur at any time during the laser cycle, but is most probable at the cycle peak. If the laser field varies as $E = E_0 \cos \omega_0 t$ then the peak laser intensity will occur at $\omega_0 t = p\pi$ where p is an integer. The electron is then driven away from the atom by the electric field of the laser. At a later time the laser field reverses and the electron is accelerated back towards the atomic core. If the electron is re-incident on the

atomic core, it may recombine into the ground state. When this recombination occurs the electron emits a photon which carries away its excess kinetic energy and momentum. The equations of motion of the electron displacement from the nucleus, z , are

$$\frac{d^2z}{dt^2} = \frac{qE_0 \cos \omega_0 t}{m} \quad (3.3.15)$$

$$\frac{dz}{dt} = v = \frac{qE_0 \sin \omega_0 t}{m\omega_0} + v_i \quad (3.3.16)$$

$$z = -\frac{qE_0 \cos \omega_0 t}{m\omega_0^2} + v_i t + z_i \quad (3.3.17)$$

At $t = t_0$ (the time of ionisation) let $z = 0$, $v = 0$ then

$$v = \frac{qE_0}{m\omega_0} \{\sin \omega_0 t - \sin \omega_0 t_0\} \quad (3.3.18)$$

$$z = -\frac{qE_0}{m\omega_0^2} \{\cos \omega_0 t + \omega_0 t \sin \omega_0 t_0 - \cos \omega_0 t_0 - \omega_0 t_0 \sin \omega_0 t_0\} \quad (3.3.19)$$

We must then solve for v when the electron returns to the core ($z = 0$). The energy at impact will be $mv^2/2$. The solution of these equations is shown in figure 3.3.5 and gives us a local maximum in the impact energy of the electron of

$$U = U_I + 3.2U_P. \quad (3.3.20)$$

which corresponds to $\omega_0 t = 17^\circ$ or 197° . The emitted photon will be at a frequency that is an odd-integer multiple of the laser frequency: a harmonic. Figure 3.3.5 shows the dependence of the probability of tunnel ionisation with time and the corresponding energy of the electron when it is re-incident on the atom. The calculation was performed for a Hydrogen potential at an intensity of 10^{14} W/cm².

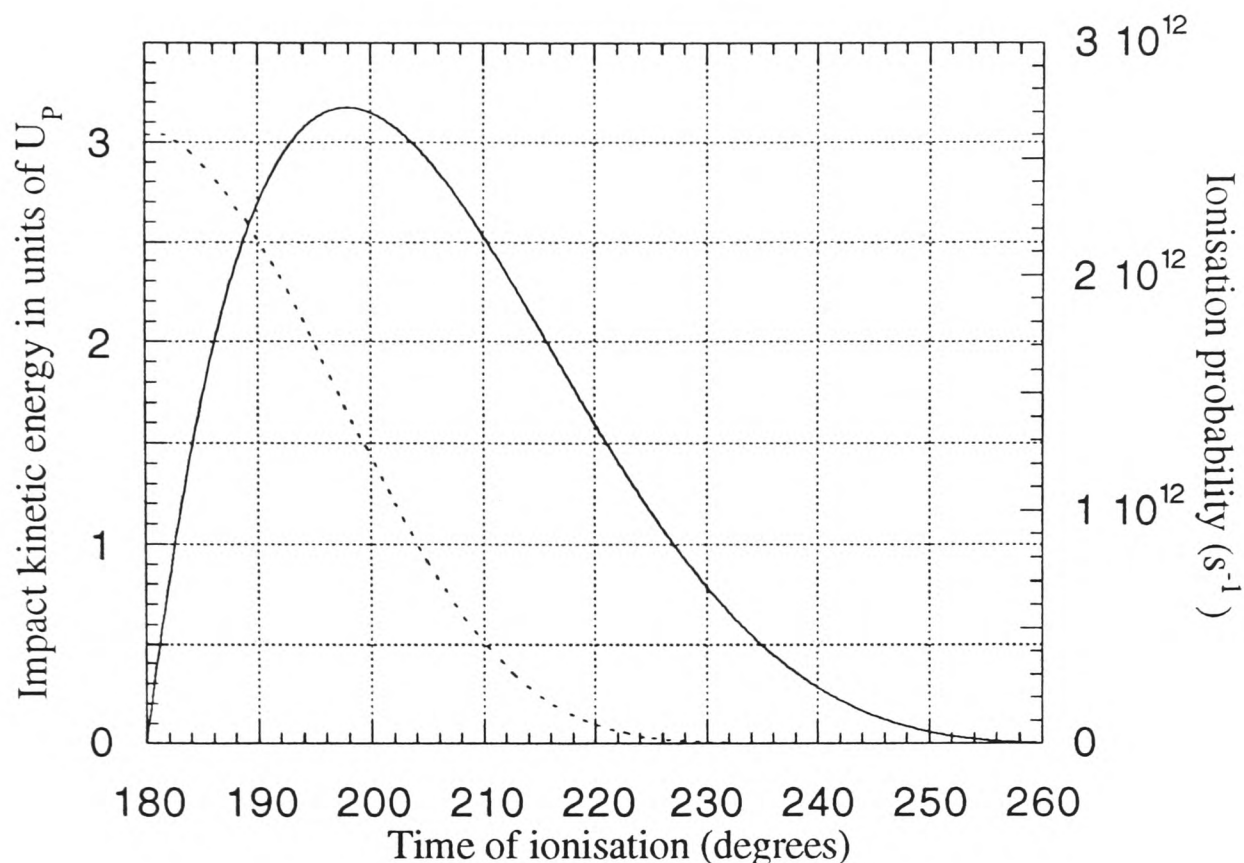


Figure 3.3.5. *Energies of returning electron trajectories at the nucleus (solid line, left axis) and the approximate Hydrogen tunnelling rate (dotted line, right axis) as a function of the phase of the oscillating laser field (in degrees) at a laser intensity of 10^{14} W/cm². The electron energies are expressed in units of U_p , the ponderomotive energy.*

At this point it should be noted that high harmonic generation in the high-field regime is still governed by electric dipole transitions, but in this case most of the dipole operators will be between continuum states, i.e. the excitation transitions shown in figure 3.3.3 will continue well beyond the ionisation limit of the atom. The final recombination transition is then between a state in which the electron is essentially free from the atomic Coulomb potential (quivering in the laser field with its ponderomotive energy, U_p) and the atomic ground state. It is for this reason that the semi-classical approach may be used. In a real situation (as will be seen in chapter six) the harmonic plateau is not completely flat. The individual harmonic intensities may vary by up to an order of magnitude from one harmonic to the next. This is an indication that resonances in the

electric dipole transitions as the atom is pumped to a continuum state can greatly enhance the conversion of q photons at frequency ω to one photon at frequency $q\omega$. The electric dipole recombination transition will be discussed further when we consider the harmonic simulation code used to model our experimental data in section 6.1.

3.3.2.2. *The saturation intensity.*

The plateau cut-off energy derived above (equation 3.3.20) matches the experimentally observed cut-off of the harmonic plateau very well [90]. Experiments conducted at varying intensities see the harmonic response reach a maximum and then start to fall as the intensity is increased further. This is due to depletion, by optical ionisation, of the atomic population that is generating the harmonics. Krause et al. [90] defined the saturation intensity which is the intensity at which 20% of the atoms survive the laser pulse without being ionised. This saturation intensity is an effective upper limit on the intensity that can be used for efficient harmonic generation and sets a limit on the scaling of the maximum harmonic energy with U_P . Harmonic response beyond this cut-off imposed by the saturation intensity was seen in early experiments [91,92]. Considering this data without taking the time dependence of the laser pulse into account leads us to the incorrect conclusion that this could be evidence of harmonic response from ions. It is in fact possible to generate harmonics beyond this limit [93]. The few atoms that remain un-ionised for a short period above the saturation intensity can generate harmonics quite efficiently. In this regime, however, the total harmonic response rapidly diminishes as the atomic population is depleted by optical ionisation.

3.3.2.3. *The advantage of short wavelengths.*

While the electron is free from the atom and in the influence of the laser field, its wavefunction will spread transversely to its direction of motion. This is because the

electron will appear outside the atomic core with a small (random) velocity which will have a finite component perpendicular to the laser field [94]. Such a situation is depicted in figure 3.3.6. This has the effect of reducing the overlap between the free electron wavefunction and the ground state wavefunction, reducing the probability of recombination. Obviously, the time during which the wavefunction can spread is determined by the wavelength of the laser. The area of the wavefunction as it returns to the atom is then proportional to the square of the laser wavelength which means that the probability of recombination is inversely proportional to the square of the fundamental laser wavelength. This simple result indicates that a short wavelength fundamental laser should produce a much higher harmonic response than a longer wavelength laser.

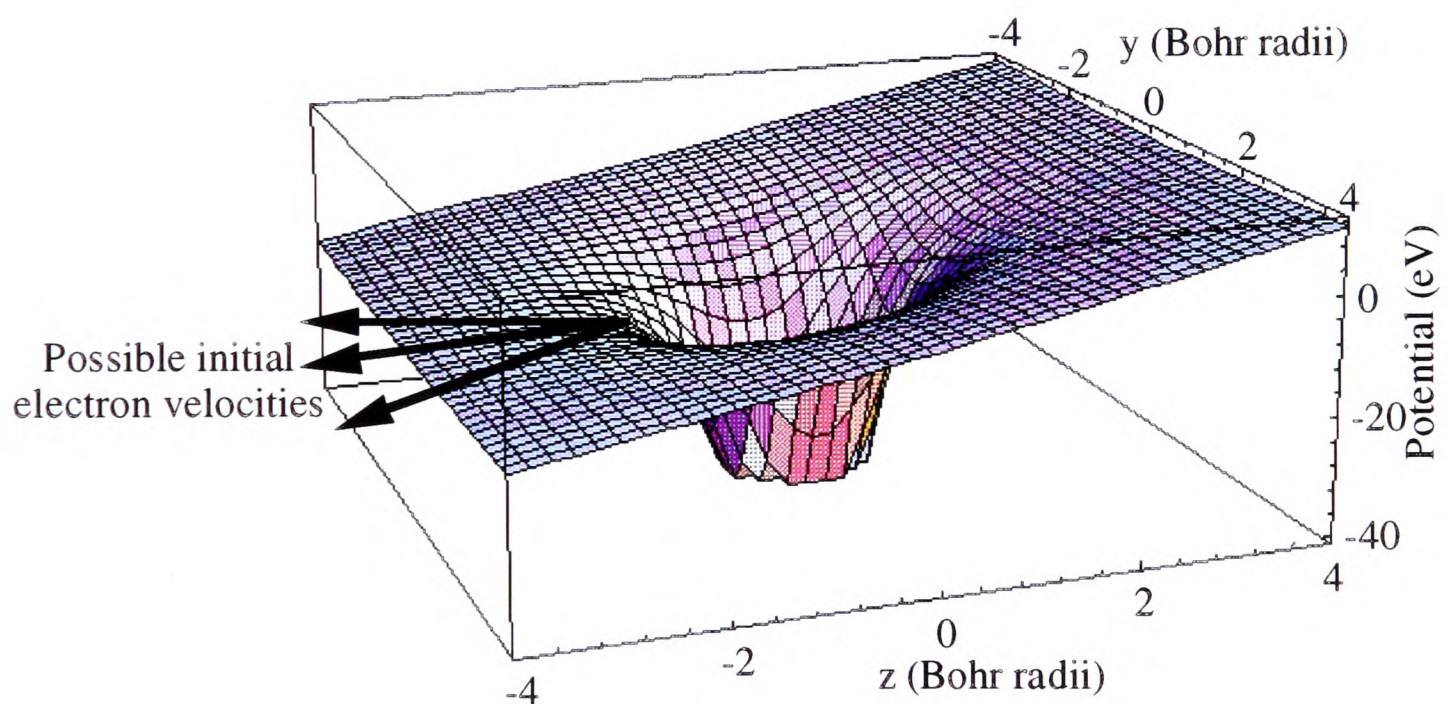


Figure 3.3.6. Three dimensional plot of the Hydrogen potential modified by an external electric field. The spread of the arrows indicate the diffusion of the electron wavefunction transversely to the electric field (y-axis).

Most harmonic generation research to date has been performed using 1053 nm Nd:YAG or 800 nm Ti:Sapphire lasers [19,20,95-104]. This is partly because these are the most commonly available laser systems, but also because it was considered that the best way to

generate short wavelength harmonics was to maximise the ponderomotive term in the plateau cut-off equation which is proportional to the square of the laser wavelength [19,20,101].

Recent work using a 248 nm KrF laser by Sarukura et al. [92] and Kondo et al. [105] indicates that shorter wavelength drivers can produce short wavelength harmonics not just from the ponderomotive energy term in the cut-off equation, but by using the higher ionisation potentials of ions rather than neutrals. He⁺, for example, has an ionisation potential of 54.4 eV compared to 24.6 eV for neutral Helium. This, of course, leads to a much higher ionisation (and saturation) intensity for the ion compared to that for the neutral atom. Harmonic generation efficiencies from ions using long wavelengths are, at best, very weak in comparison to those from neutrals [100]. For 248 nm, however, ionic efficiencies can be of the same order as those from atoms. The results of Sarukura et al. and Kondo et al. are indeed startling in that they demonstrate unambiguously that high harmonics can be generated from ions with short wavelength driver lasers. However, the number of harmonic photons produced and the highest harmonic energy achieved were no better than those produced with long wavelength lasers.

It should be noted that harmonic generation depends on good phase matching as well as a high single atom/ion response. In the case of generating harmonics near the saturation intensity, or generating harmonics from ions, phase matching will usually be dominated by the free electron refractive index. This will set a coherence length that is smaller than in an un-ionised gas. Hence harmonic generation is always a trade off between producing high photon energies and high efficiencies.

With harmonic generation from ions already demonstrated, it is timely to investigate whether higher harmonic photon energies can be produced from ions using short wavelength lasers, than from neutrals with longer wavelength lasers. We will present

data and simulations in chapter six that go beyond those of Sarukura et al. and show that 248 nm light can produce much shorter harmonic wavelengths than 800 nm or 1053 nm light, and that generation efficiencies are much higher at the longer harmonic wavelengths.

3.3.3. Phase matching between the harmonic and the fundamental.

Phase matching is an important mechanism for optimising the generation of high harmonics. If the fundamental and the harmonic can be made to travel together and in phase through the non-linear medium, then constructive interference can occur between the amplitudes of the harmonic wave generated in different parts of the medium. In this section we shall derive some of the fundamental equations of non-linear optics and we shall consider the phase matching factor for several cases of interest.

3.3.3.1. The equations of non-linear optics.

The derivation of the equations of non-linear optics in this section follows that of C.R.Vidal [106]. Using Maxwell's Equations (assuming $M = 0$; $\mu = 1$; $j = 0$; $\rho = 0$)

$$\nabla \cdot \mathbf{E} = 0 \quad (3.3.21)$$

$$\nabla \cdot \mathbf{B} = 0 \quad (3.3.22)$$

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \quad (3.3.23)$$

$$\nabla \times \mathbf{B} = \frac{1}{c^2} \frac{\partial \mathbf{E}}{\partial t} \quad (3.3.24)$$

We obtain the well known wave equation :

$$\nabla^2 \mathbf{E} - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{P}}{\partial t^2} \quad (3.3.25)$$

Putting $\mathbf{P} = \mathbf{P}^L + \mathbf{P}^{NL}$ in equation 3.3.25 we obtain

$$\nabla^2 \mathbf{E} - \frac{n^2}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} - i \frac{n\tilde{n}}{c\omega} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{P}^{NL}}{\partial t^2} \quad (3.3.26)$$

the driven wave equation.

At this stage we must specify the Fourier components of the electric field amplitudes. For simplification we will make the plane wave approximation $\hat{\mathbf{E}}(r, \omega) = \hat{\mathbf{E}}(z, \omega)$ and assume that the electric field amplitudes change slowly within times of the order of ω^{-1} and over a length of typically k^{-1} ($\frac{\partial \hat{E}_j}{\partial t} \ll \omega \hat{E}_j$; $\frac{\partial \hat{E}_j}{\partial z} \ll k \hat{E}_j$). Note that we denote here the Fourier component of the electric field by $\hat{E}(r, \omega_j) = \hat{E}_j(r)$.

We then arrive at the fundamental equations of non-linear optics

$$\frac{d\hat{E}_j}{dz} = i \frac{j\omega}{2\epsilon_0 c n_j} P_j^{NL} \exp(i\Delta k z) - \frac{\tilde{n}_j}{2} \hat{E}_j \quad (3.3.27)$$

where $\frac{d\hat{E}_j}{dz} = \frac{\partial \hat{E}_j}{\partial z} + \frac{n_j}{c} \frac{\partial \hat{E}_j}{\partial t}$ and $\Delta k = jk_0 - k_j$

So in the plane wave approximation, the spatial dependence of the electric field amplitude in a non-linear medium is given by

$$E_j = \hat{E}_j \exp(ik_j z) \quad (3.3.28)$$

3.3.3.2. Phase matching for a plane wave.

In the small signal limit it can be shown that the intensity of the q th harmonic (Φ_s) in the generated beam is given by the equation :

$$\frac{\Phi_s}{n_s} = \left[\frac{q\omega}{cn_s} L \chi^{(q)}(-q\omega; \omega\omega\omega\dots) \right]^2 \frac{\Phi_0^q}{\epsilon_0 n^3} F(\Delta kL, \tau_i, \tau_s) \quad (3.3.29)$$

where n is the refractive index at the fundamental frequency, n_s is the refractive index at the q th harmonic frequency, Φ_0 is the initial intensity of the pump beams and F is the phase matching factor given by

$$F(\Delta kL, \tau_\omega, \tau_{q\omega}) = \frac{\exp(-\tau_\omega) + \exp(-\tau_{q\omega}) - 2 \exp[-(\tau_\omega + \tau_{q\omega})/2] \cos(\Delta kL)}{\left[(\tau_{q\omega} - \tau_\omega)/2 \right]^2 + (\Delta kL)^2} \quad (3.3.30)$$

where $\Delta k = k_{q\omega} - qk_\omega$ and $\tau_i = \tau_1 + \tau_2 + \tau_3 + \dots$ is the total optical depth for all the incident waves. τ_ω and $\tau_{q\omega}$ are the optical depths for the fundamental and harmonic waves respectively ($\tau = \tilde{n}\omega L/c = \zeta L$, where ζ is the $1/e$ length for the electromagnetic wave).

F depends on three variables: ΔkL , τ_ω , and $\tau_{q\omega}$ which we must try to minimise in order for F to approach its maximum value of unity. Let us consider two limiting cases:

(i) For negligible absorption cross sections, the phase matching factor has the form

$$F(\Delta kL, \tau_\omega = 0, \tau_{q\omega} = 0) = \left(\frac{\sin(\Delta kL/2)}{\Delta kL/2} \right)^2 \quad (3.3.31)$$

(ii) For an optically thick system with $\tau_\omega, \tau_{q\omega} \gg 1$, the minima of the Sinc function fill up and we move towards a Lorentzian type profile for the phase matching factor.

$$F(\Delta kL, \tau_\omega \gg 1, \tau_{q\omega} \ll 1) = F(\Delta kL, \tau_\omega \ll 1, \tau_{q\omega} \gg 1) \\ = \frac{1}{\left[(\tau_{q\omega} - \tau_\omega)/2 \right]^2 + (\Delta kL)^2} \quad (3.3.32)$$

Figure 3.3.7 shows the variation of the phase matching factor, F , with the optical depth of the generated wave. For practical applications, it is generally not very difficult to keep the incident wave away from resonances which will significantly increase the optical depth. This is not necessarily the case for the high harmonics that are generated. Figure 3.3.8 shows the variation of the transmission with wavelength for 1 mm of neutral Helium and Neon at an atomic density of 10^{18} atoms/cm³. Certainly the lower harmonics will frequently cause photo-ionisation or auto-ionisation and so will often have a relatively large optical depth. This can be seen for the case of Helium, where the transmission tails off towards zero at 1200 Å. The transmission curve for Neon is more complicated, because of its higher atomic number. A transmission of 0.4 is equivalent to an optical depth of about 0.9. All harmonics of KrF (248 nm) have an optical depth (in 1 mm of gas) of less than two in both gases.

The reduction of the peak phase matching factor with increasing optical depth can eventually limit the effectiveness of enhancing harmonic generation by increasing the length of the non-linear medium, L , over which the harmonic is generated. For example, if we take $\tau_\omega = 0$, and if we assume optimum phase matching ($\Delta kL = 0$), we find that (for large $\tau_{q\omega}$) the factor F in equation 3.3.29 (and defined by equation 3.3.30) is proportional to $\tau_{q\omega}^{-2}$ (and so is proportional to L^{-2}). Equation 3.3.29 also contains a factor of L^2 , and so the two factors will cancel each other out resulting in no net enhancement of the harmonic intensity with increasing length. For small optical depths,

$\tau_{q\omega} < 1$, increasing the interaction length will lead to enhancement of harmonic generation.

Of course, for a given optical depth we must ensure that phase matching is optimised (ΔkL close to zero). This can sometimes be achieved by using two gases in the generation cell: one to convert the pump beam to the higher frequency signal; the other to satisfy the phase matching condition. Efficient phase matching requires very good homogeneity in the generating medium.

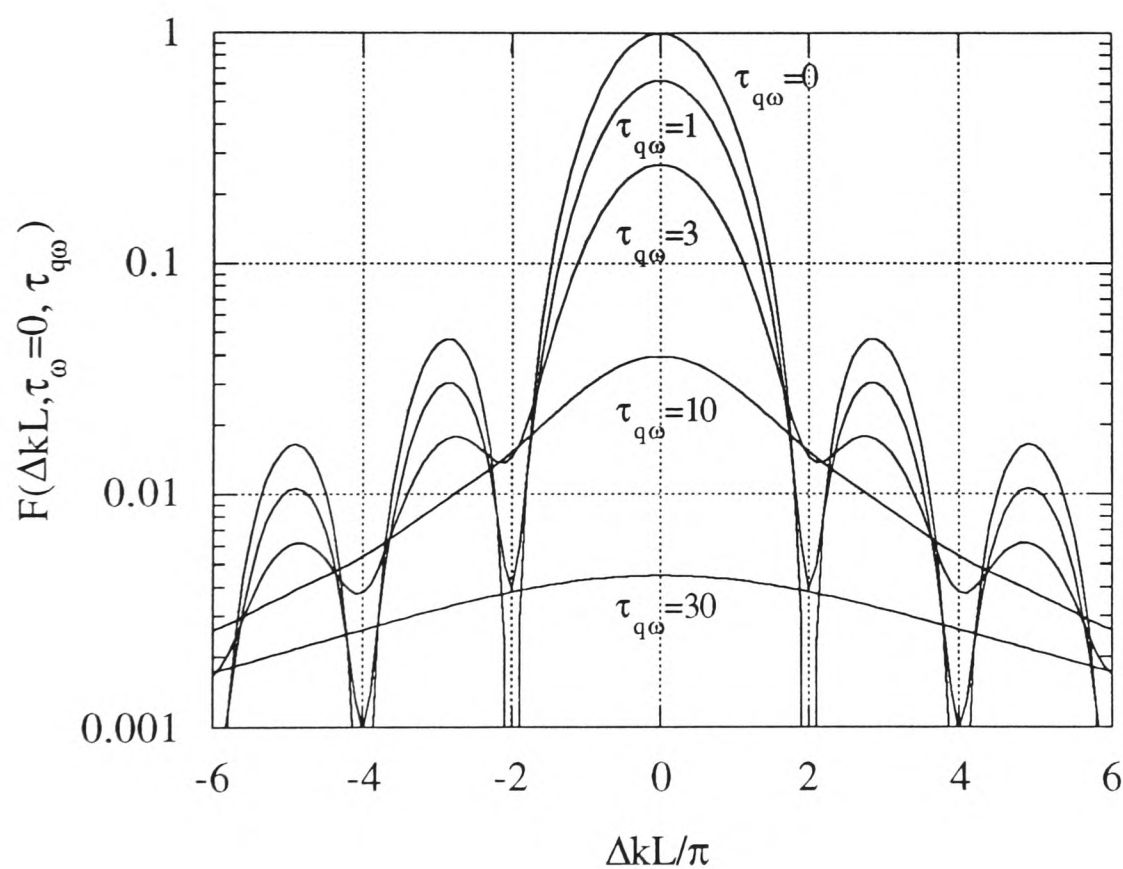


Figure 3.3.7. Variation of the phase matching function, F , with $\tau_{q\omega}$. Note the transition from a Sinc function to a Lorentzian.

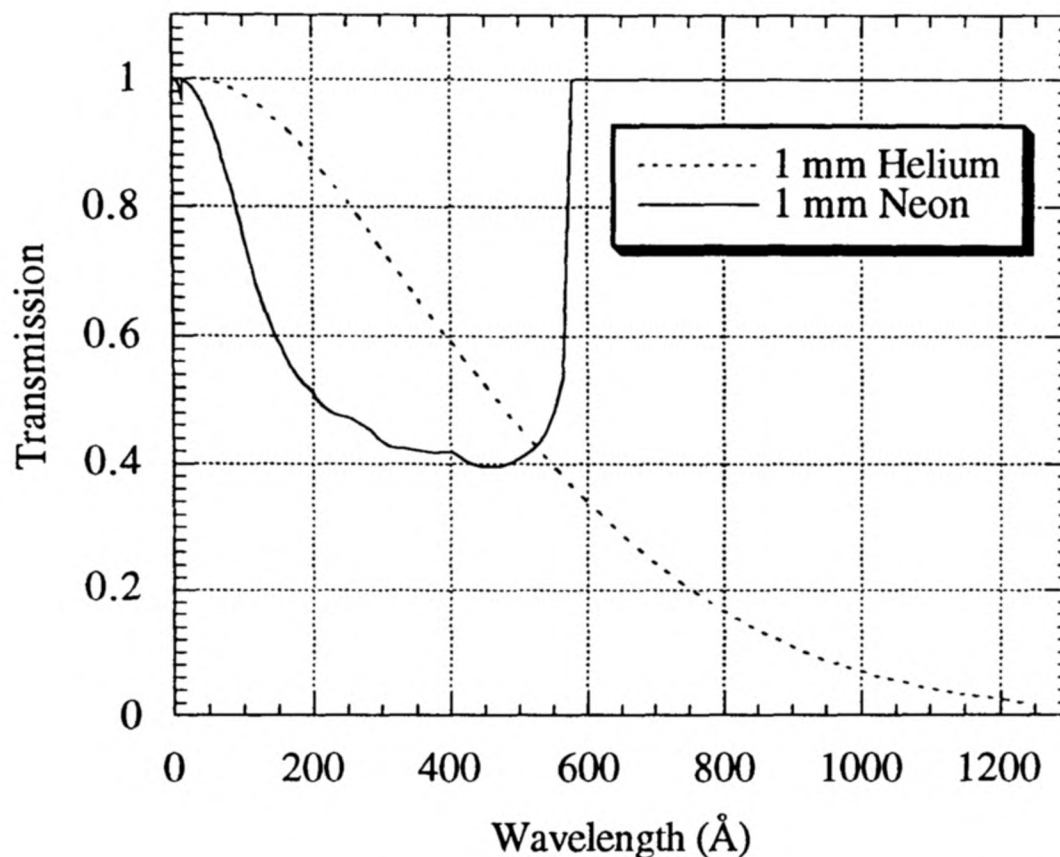


Figure 3.3.8. The variation of transmission with wavelength for 1 mm thicknesses of Helium and Neon. The neutral gas density is 10^{18} atoms/cm³ in both cases.

3.3.3.3. Phase matching for a focused Gaussian beam.

The above treatment is for the special case of a plane wave. If we assume that the medium is optically thin for both the fundamental and the harmonic and that the medium is dispersionless, we immediately see that (for the case of negligible absorption) for any harmonic, q , $\Delta k = k_q - qk_0 = 0$. This implies that the two waves will remain in phase for the entire length of the non-linear medium and that phase matching is not a problem. In practice, however, the fundamental light wave will be a focused Gaussian laser beam. The amplitude of a Gaussian (TEM₀₀) beam is given by

$$E(r, z) = \frac{E_0}{z - ib} \exp \left[-i \left(\tan^{-1} \left(\frac{z}{b} \right) - \frac{kr^2 z}{2(z^2 + b^2)} \right) \right] \quad (3.3.33)$$

where the exponential term is a phase factor, E_0 contains the factors determining beam propagation, and b is the confocal parameter.

The q th harmonic generated at some point in the plasma will propagate with a different phase factor, which is the q th power of the fundamental phase factor. Again assuming a dispersionless medium, the phase mismatch between harmonic and fundamental in a Gaussian beam is given by $\Delta\phi(z) = (1 - q)\tan^{-1}(z/b)$ [98]. The coherence length is defined as the distance the two waves must propagate before they become dephased by π and interfere destructively. Hence we arrive at the expression for the coherence length, L_{coh} , in a dispersionless medium: $L_{coh} = b \tan(\pi/(q - 1))$. For large q this becomes

$$L_{coh} = b \pi / (q - 1) \quad (3.3.34)$$

In the weak field regime, the coherence length is a good measure of the length over which harmonic generation can occur. In a homogeneous, dispersionless medium, the amplitude of the harmonic wave will grow over half a coherence length and then will reduce again over the next half coherence length. If the laser intensity is constant over this region, then there will be no net harmonic generation, as the harmonic polarisation generated in the second half of the coherence length will exactly cancel that in the first half of the focus. This situation is depicted schematically below in figure 3.3.9. The cancellation occurs because the non-linear polarisation contributing to harmonic generation in the second half of the coherence length will be of the same magnitude, but opposite to that in the first half (i.e. $\mathbf{P}^{NL}(L) = -\mathbf{P}^{NL}(L + L_{coh}/2)$). Hence for a non-linear medium of arbitrary length, we would expect the amplitude of the harmonic wave exiting the medium to be somewhere between zero and the maximum reached after travelling a distance $L_{coh}/2$.

A real non-linear medium will not usually be homogeneous, and the laser intensity will vary across the focus. In this situation, the harmonic wave will not exactly cancel after a coherence length. Indeed, at high laser intensities we would expect to see several fringes of non-linear polarisation in the medium, the fringe separation being L_{coh} . The harmonic field will not exactly cancel after each fringe, and the net harmonic amplitude exiting the medium will saturate at close to the maximum amplitude generated in a single coherence length. It has been shown by L'Huillier et al. that scanning the laser focus through a non-linear medium produces a fringe structure in the recorded harmonic intensity that emerges from the medium [96-98]. This structure is caused by the scanning of the polarisation fringes through the medium. A maximum in the recorded intensity corresponds to a fringe maximum moving to the output edge of the medium.

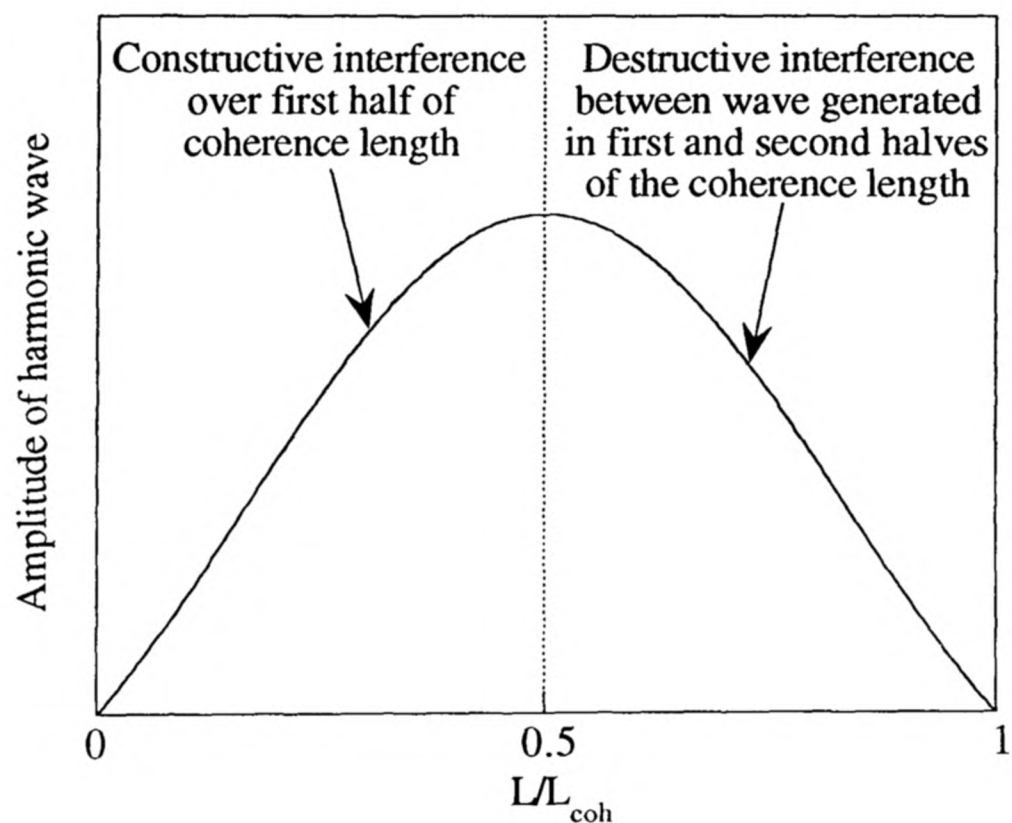


Figure 3.3.9. Schematic diagram of the growth and decay of a harmonic wave in a homogeneous non-linear medium at a constant laser intensity.

3.3.3.4. Phase matching in a plasma.

In the strong field limit we reach a stage when significant ionisation occurs during the laser pulse and very few atoms survive the entire pulse. In this situation, or when generating harmonics from ions in a plasma, the coherence length is usually governed not by the Gaussian beam mismatch or the confocal parameter, but by the refractive index due to free electrons in the plasma. The refractive index of a plasma is given by $n = (1 - N/N_{cr})^{1/2}$, where N_{cr} is the critical density of the plasma (which is wavelength dependent). Hence the phase mismatch is $\Delta\varphi = q\pi L/\lambda (N/N_{cr})$, where the difference of the refractive index from unity is taken to be negligible for the harmonic, giving a coherence length, for high harmonics, of

$$L_{coh} = (\lambda/q)(N_{cr}/N). \quad (3.3.35)$$

To summarise, for efficient harmonic generation we require :

1. χ_T to be large. This may be enhanced by exploiting a multiphoton resonance.
2. Good phase matching - to exploit long column densities and produce high efficiencies.
3. Small optical depths - for both the fundamental and harmonic frequencies.

Conditions (1) and (2) can sometimes be met for lower harmonics by using a two component gas system. It remains to be seen whether the same system can be used for high harmonics. Condition (3) means we have to use gaseous media for harmonic generation.

3.4. Summary.

Two methods of producing coherent XUV radiation are the X-ray laser and high harmonic generation. Various X-ray laser schemes have been developed over a period of years, the most successful being the Neon-like collisional X-ray laser. Target geometries and injector/amplifier configurations for this system have been developed that have improved the power, efficiency and coherence of these lasers. Increases in gains in recombination laser schemes are now being seen, but the technology is not as advanced as for collisional schemes. The advent of CPA technology is now making OFI collisional and recombination schemes possible. These have the advantage that they require much lower pump energies and potentially lend themselves to a table top X-ray laser system.

Research into high harmonic generation, on the other hand, is a relatively new field. Harmonic generation requires a high laser power (rather than energy) and so is ideally suited to the use of the new CPA technology that is now appearing. Most harmonic generation research has been performed using long wavelength fundamental lasers, but there are now indications that better results may be achieved using shorter wavelengths. Generation efficiencies can be greatly enhanced by phase matching the harmonic and the fundamental as they travel through the non-linear medium. The power and efficiency of XUV radiation production from this phenomenon is still relatively low [21,107], but the field is advancing rapidly. It should be remembered that second harmonic generation in crystals originally had conversion efficiencies as low as 10^{-8} [88] whereas now these efficiencies can be greater than 50%. Harmonic generation has an advantage over traditional X-ray laser systems because it uses a gas target rather than a solid. This leads to a potentially higher repetition rate and eliminates problems with the production of debris from the laser ablation of a solid material, it is also potentially a tuneable source. Applications for high harmonic generation are still very limited, the only currently

published work being a study of the inner shell structure of semiconductors by photo-emission [108], and a study of the radiative lifetime of the $1s2p^1P$ state of Helium [109].

As well as developing these two applications separately, it is possible that they could be combined to exploit some of the advantages of each. For example, one or more low energy, wide bandwidth, but very short harmonic pulse(s) could be passed through an X-ray laser with a long gain lifetime and narrow bandwidth to produce a well collimated, high power, narrow bandwidth, short pulse XUV source. This has been demonstrated as a proof of principle experiment [110], but remains to be developed into a serious application. The future of both X-ray lasers and harmonic generation systems depends upon the development of useful applications that utilise their unique properties.

Chapter Four

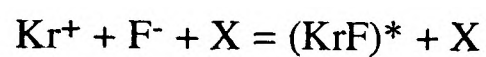
Experiments and Diagnostics

In the following two chapters (chapter five - Electron Temperature Measurements, and chapter six - Harmonic Generation) we shall present data from experiments performed using the Sprite KrF laser which is a part of the Central Laser Facility of the Rutherford Appleton Laboratory, Chilton, Didcot [26]. Sprite is a unique ultraviolet laser facility which can produce multiple pulses with pulse lengths from several nanoseconds down to a fraction of a picosecond. An experiment on Sprite typically lasts for six weeks. This usually consists of two weeks to set up diagnostics and four weeks to perform the experiment. Since there is much overlap between the diagnostics used in obtaining the data presented in both chapters five and six, and since obviously the same laser system is used, we shall describe in this chapter the Sprite laser and its diagnostics along with the equipment used as plasma diagnostics. We shall also briefly review the use of Thomson scattering as a plasma diagnostic. This will enable the data in chapters five and six to be presented more succinctly.

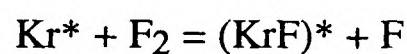
4.1. The Sprite laser system.

The main amplifiers of the Sprite laser are two electron-beam (e-beam) pumped KrF modules, Goblin (7 cm aperture) and Sprite (27 cm aperture). Population inversion in KrF can be achieved because the molecule only exists in excited electronic states. Once it has decayed to the ground level, it quickly dissociates into its constituent Krypton and Fluorine atoms within one vibrational period of the molecule. Hence, if a population of excited molecules can be produced (i.e. the pump rate must be faster than the decay rate)

then laser action will occur. A simplified schematic energy level diagram of the KrF excimer molecule is shown in figure 4.1.1. This shows the general principle of how population inversion can be achieved, but does not include the full electronic structure of the molecule, in which some levels compete with the upper laser level and can reduce the laser gain. The interaction of electrons from the e-beam with the Krypton and Fluorine atoms either produces excited states of the atoms or causes ionisation. The dominant method of production of KrF molecules is the three body recombination reaction



(where X is a third body, such as Argon), although reactions such as



also play a part in the excitation process [111].

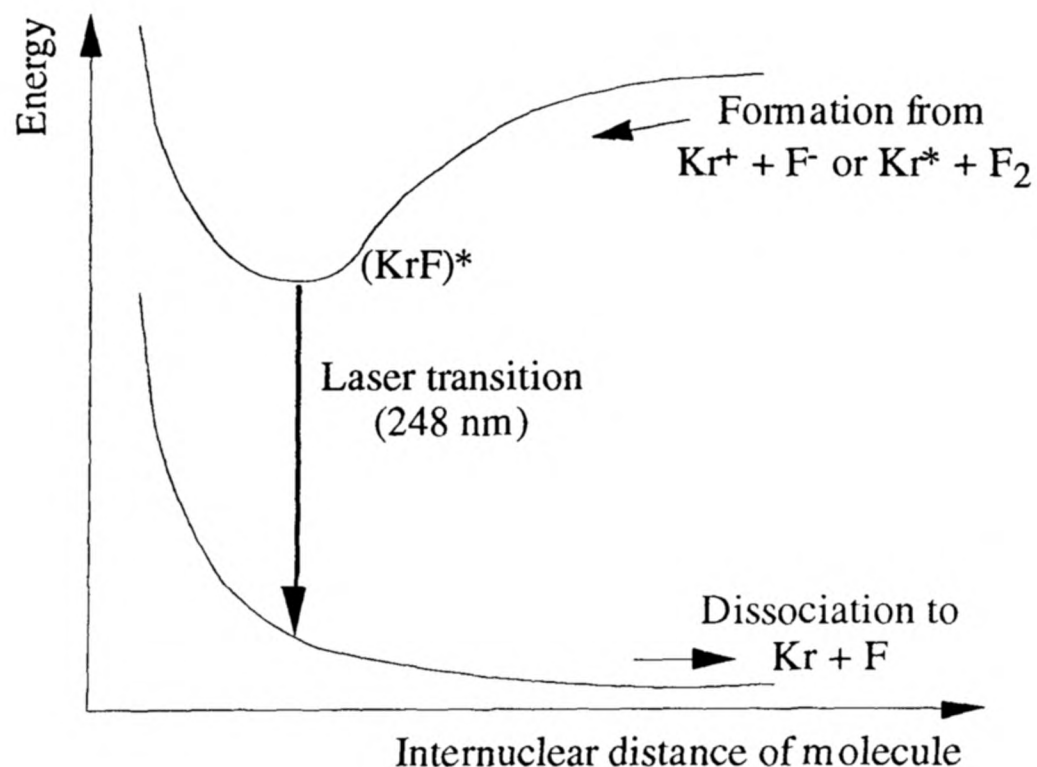


Figure 4.1.1. Simplified schematic energy level diagram of the KrF molecule.

4.1.1. The laser parameters.

Sprite produces pulsed output of ultraviolet radiation with a full shot repetition rate of about two minutes. Two modes of operation are available: Raman amplified mode [112] which utilises stimulated Raman effects in Methane to produce an output at 268 nm; and chirped pulse amplification (CPA) mode [26,113,114] which uses pulse expansion and compression techniques to produce a very short pulse at 248 nm. The parameters of these two systems are outlined below in table 4.1.

	<i>Raman Beam</i>	<i>CPA Beam</i>
<i>Wavelength</i>	268 nm	248 nm
<i>Pulse duration</i>	12 ps	300 - 400 fs
<i>Pulse energy</i>	3 - 5 J	150 - 250 mJ
<i>Focused spot diameter</i>	~3 μ m	~8 μ m

Table 4.1. Performance characteristics of the Sprite laser system. The focused spot radius is that produced with a 33 cm off-axis parabolic mirror.

Experiments performed using the Raman amplified configuration of Sprite are described in the thesis of W.J. Blyth [29] which contains a description of the Raman amplifier system. The experiments described in this thesis are all performed in CPA mode, so we will not discuss the Raman mode further. The wide bandwidth (5 nm) of the laser transition in the KrF molecule gives it the potential to amplify pulses shorter than 100 fs to multi-joule energies [115]. The main limits on the pulse length are placed by two photon absorption and self phase modulation of the high power beam in the laser and in the air-path to the vacuum chamber. One method of avoiding this is chirped pulse amplification and compression. The basic technique is to create a very short (transform limited) pulse and then to expand it temporally before passing it through the main laser amplifiers. The pulse can then be recompressed before focusing it on target. It should

also be noted that an absorption dip has been identified in the KrF laser transition which limits the bandwidth that can be amplified in KrF. This has been attributed to internal absorption to an excited molecular state which dissociates to $\text{Kr}^*(^3\text{P}_1)$ and $\text{F}(^2\text{P}_{3/2})$ [116].

4.1.2. The Sprite CPA system.

The Sprite CPA system makes use of such a pulse stretching and compression system [26]. A schematic of the Sprite CPA amplifier chain is shown in figure 4.1.2. Fourier transform limited 110 fs pulses at 746 nm are generated by an Argon ion pumped mode-locked Ti:Sapphire oscillator and are amplified to 400 μJ in two Rhodamine 700 dye amplifiers and a double passed Ti:Sapphire amplifier. (These amplifiers are all pumped by a Q-switched frequency doubled Nd:YAG system). The pulses are then frequency tripled to 248 nm by passing them through two 0.3 mm long beta-Barium Borate (BBO) crystals. At the output of the frequency tripling stage the laser bandwidth is 95 cm^{-1} , sufficient for a 150 fs pulse duration. The laser pulse is then chirped in a single-pass, folded geometry, diffraction grating stretcher (figure 4.1.3).

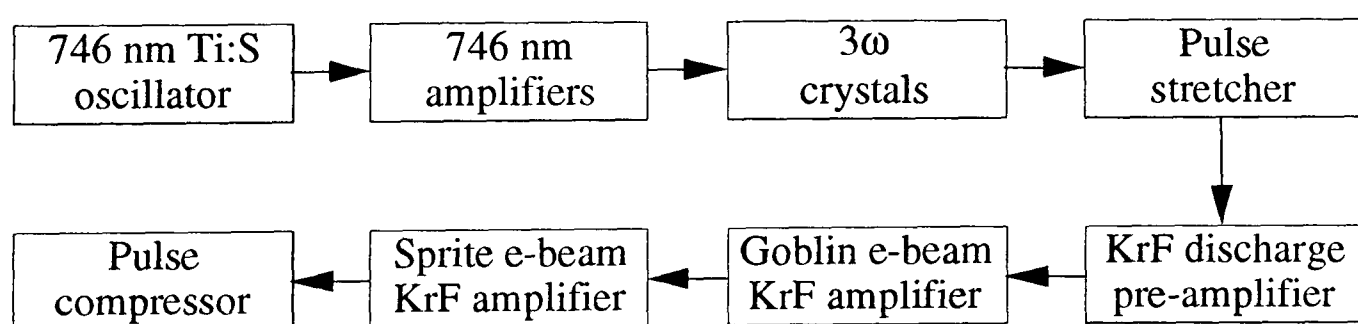


Figure 4.1.2. Schematic diagram of the Sprite CPA KrF laser amplifier chain.

The stretcher, shown in figure 4.1.3, uses a 3600 g/mm holographic diffraction grating. The dispersed beam is focused by a 99 cm focal length lens onto a dielectric UV mirror,

which reflects the beam back to the grating. The 2° biprism ensures that the input and output beams are separated horizontally. This configuration produces a longer path length through the stretcher for higher frequencies than for lower frequencies. We have now produced a pulse in which the low frequency components lead the high frequency components. The duration of the stretched pulse is given analytically by

$$\tau = \frac{2(f-l)(ma)^2 \lambda \cdot \Delta\lambda}{c \cos^2 \theta_d} \quad (4.1)$$

where f is the focal length of the lens, l is the lens-grating spacing, m is the grating diffraction order, a is grating line density, $\Delta\lambda$ is the input pulse bandwidth, θ_d is the angle of diffraction of the input beam at the grating, and c is the speed of light. The stretcher parameters are adjusted to produce an output pulse length of around 10 - 14 ps which is then pre-amplified to an energy of about 300 μJ in a Lambda-Physik EMG103-MSK KrF discharge laser which is pulsed at a frequency of 8 Hz. The beam is then expanded and can be passed through the Goblin and Sprite e-beam pumped amplifiers without problems from high intensity effects. After the Sprite amplifier the pulse bandwidth has reduced to around 70 cm^{-1} .

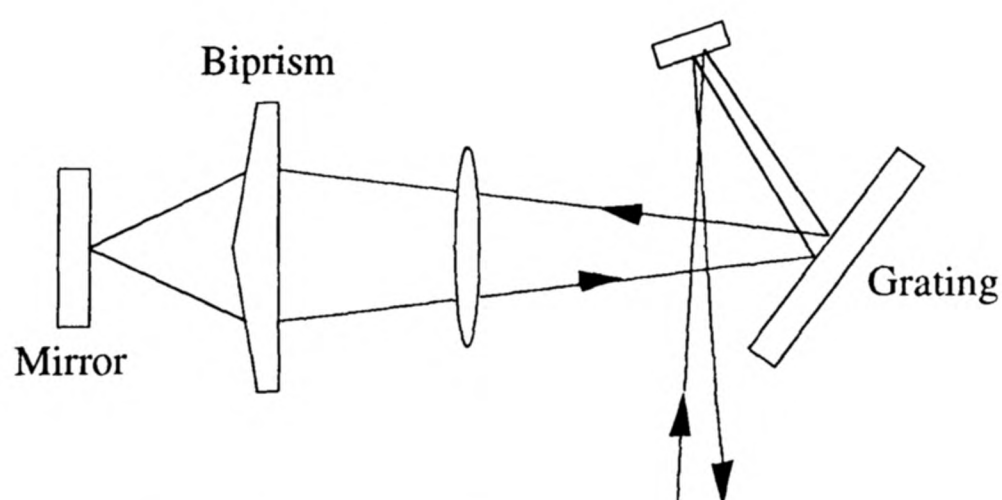


Figure 4.1.3. The configuration of the pulse stretcher.

The output beam of the Sprite laser is compressed by passing it through two parallel gratings, shown in figure 4.1.4. The gratings have 3600 g/mm and are blazed at first

order for 248 nm. One grating is placed in an extension tube linked to the target chamber, while the other is mounted in the chamber itself. This eliminates problems associated with transmitting the compressed pulse through a chamber window or through air.

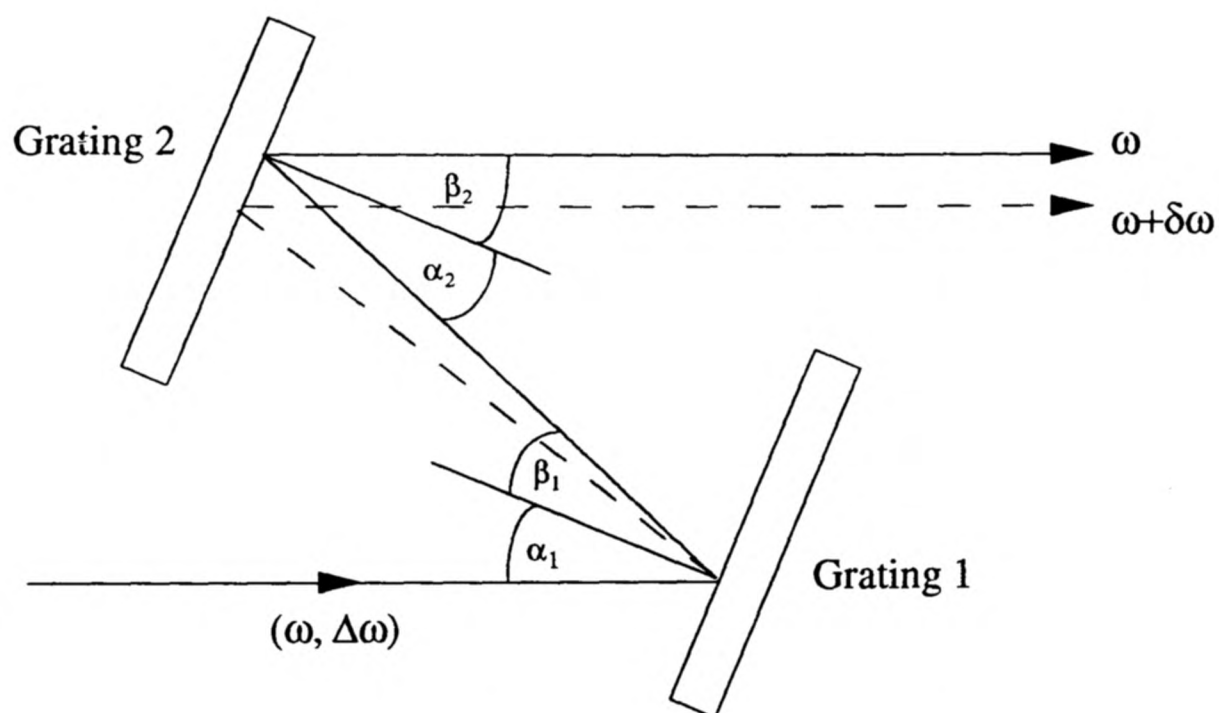


Figure 4.1.4. Pulse compression using parallel gratings showing the different paths travelled by differing frequencies in the input beam.

The gratings are at a working angle of 26° , and must be parallel to within 100 microradians for the residual dispersion to have a negligible effect on the pulse length. Figure 4.1.4 illustrates that higher frequencies travel a shorter distance through the compressor and so will catch up with the front end of the pulse. That the output beam emerges parallel to the input beam can be shown using the grating equation,

$$d(\sin \alpha + \sin \beta) = \lambda \quad (4.2)$$

From geometry we see that $\alpha_2 = \beta_1$, hence

$$\sin \beta_2 = \frac{\lambda}{d} - \sin \alpha_2 = \sin \alpha_1 \quad (4.3)$$

Using the Sprite laser in CPA mode with grating pairs to stretch and compress the source pulse it is possible to produce pulses on target as short as 280 fs, but more typically at 350 - 380 fs. The final compressor gratings are installed in the target chamber to limit non-linear losses in the propagating beam. Using a 33 cm off-axis parabolic mirror (OAP), energies up to 250 mJ may be focused on target with an effective focal spot area of $64 \mu\text{m}^2$ and intensities up to 10^{18} W/cm^2 at a wavelength of 248 nm.

Amplified spontaneous emission (ASE) levels are determined by the excited lifetime of the EMG103-MSK KrF amplifier cell. Typically the ASE pulse has a duration of 20 ns, and a total energy of 5 mJ (reduced to 2.5 mJ after the compressor gratings). This is focused in the target chamber to an intensity 10^{10} W/cm^2 , which gives a contrast of 10^8 between the ASE and the main pulse intensities [117].

The laser energy delivered on target is limited by the incident laser energy that the Aluminium coated compression gratings can withstand without damage. Energies greater than 600 mJ can be used for a short period without immediate grating damage, but in the interests of longevity this has been set as an upper limit. The grating pair have a combined transmission of approximately 50%

4.1.3. Monitoring the laser far-field.

Once the laser pulse has passed the compression gratings, it is focused onto target by an OAP. The focal spot size is determined by the f-number of the OAP and by the beam divergence. The CPA beam divergence is considerably greater than the diffraction limit. This is due to the generally poor quality multimode structure of the output from Sprite which is further degraded by non-flatness of the compression gratings. The beam quality before compression is monitored for every shot using an equivalent plane monitor to give an indication of the divergence. This is done by taking a 4% reflection from a quartz

pellicle and focusing it (after suitable attenuation) onto a UV sensitive CCD camera using a 20 m focal length lens combination. A schematic of this is shown in figure 4.1.5.

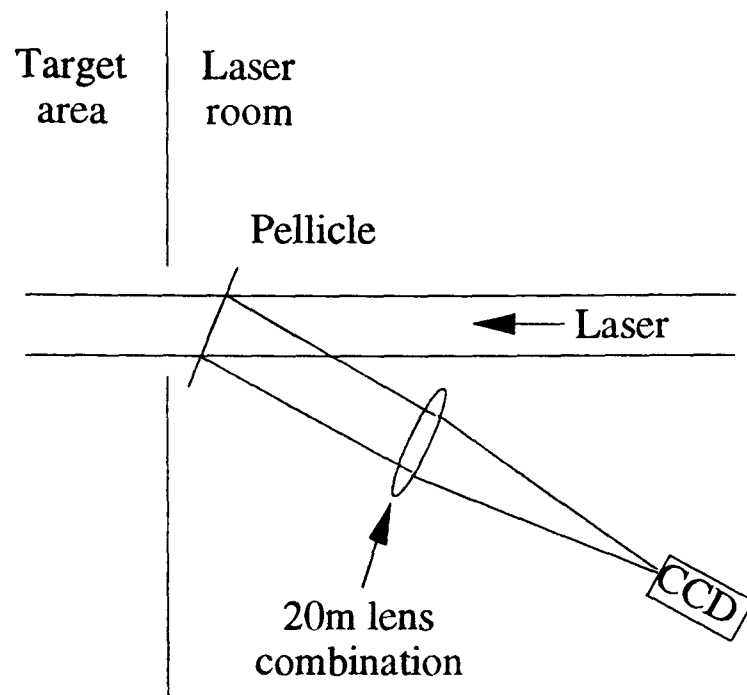


Figure 4.1.5. *The Sprite far-field monitor.*

The air path between the far field monitor and the vacuum chamber is 6 m and there are two 12.5 mm thick CaF_2 windows downstream of the diagnostic point. The stretched Sprite pulse length is 8 ps and the equivalent beam area for estimating peak intensity is 50 cm^2 . This gives a B-integral (i.e. the integral over the beam path of the intensity dependent second order non-linear refractive index [118]) between the monitor and the chamber of 0.9 times the energy in joules from the laser. Since the laser energy is never more than about 0.5 J, the B-integral is always less than 0.5 [117]. This tells us that beam break-up downstream of the far-field monitor is negligible under normal operating conditions. Hence, any degradation of the focal spot at the chamber centre with respect to the far-field monitor is due to non-uniformities in the compression gratings caused during manufacture or by grating damage which inevitably occurs over the grating lifetime.

Measuring the focal spot after the gratings is difficult, as a full energy shot can easily destroy any lens used to image it. An alternative is to image the 8 Hz oscillator. This

gives a measure of the beam quality, but does not allow for gain saturation effects in the Sprite amplifier. A typical oscillator focal spot is shown in figure 4.1.6. It is plain to see that the beam exhibits much structure. Some of this will be removed in a full shot when gain saturation occurs, but data from the far-field monitor (figure 4.1.7) shows that much structure still remains. The FWHM seen at the far-field monitor will increase when the beam has passed through the compression gratings. Note that the grey scales used in figures 4.1.6 and 4.1.7 are different and have been chosen to highlight the structure in each image.

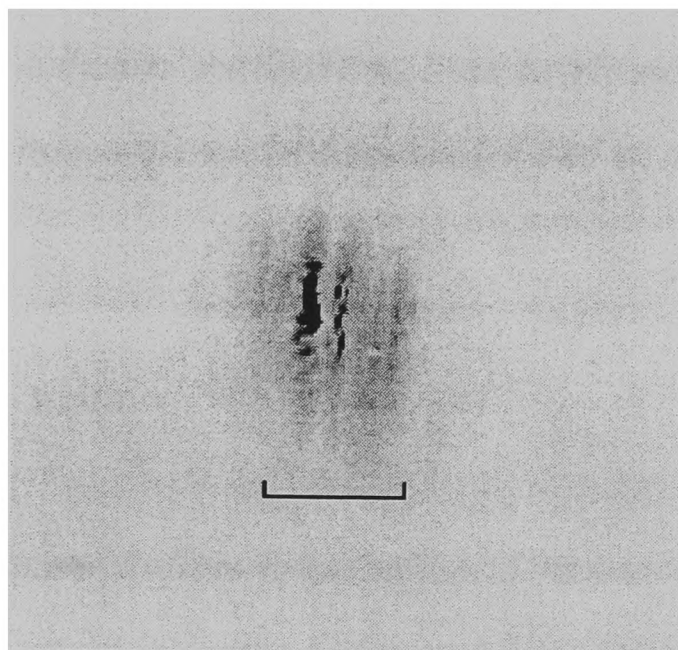


Figure 4.1.6. Image of the Sprite oscillator focused by the 33 cm OAP. The solid line indicates the FWHM of the image which is $10\ \mu\text{m}$ or 30 microradians. This is about 10 times the diffraction limit.

The CPA oscillator focal spot size has been measured for the 33 cm and 100 cm OAPs used in our experiments. The 33 cm (f/3) OAP produces an approximately 10 times diffraction limit spot, the 100 cm (f/10) OAP produces a 6 times diffraction limited spot. All of these contain fine structure down to the diffraction limit. This beam structure is undesirable as it leads to increased ponderomotive heating; it can seed SRS growth; and it can reduce the coherence length for harmonic generation.

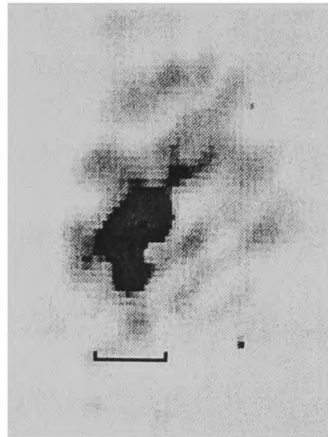


Figure 4.1.7. Image of full shot (SN16144) on the Sprite far field monitor. The horizontal line indicates the FWHM of the bright central lobe. Its length corresponds to 10.2 microradians (or $3.4\ \mu\text{m}$ if focused by an $f/3$ OAP).

4.2. The Target area.

The Sprite target area houses a 45 cm radius spherical vacuum chamber. A schematic of a typical experimental set-up is shown in figure 4.2.1. The laser enters the chamber through a Calcium Fluoride anti-reflection coated window and passes through the compression gratings. These gratings must be aligned in air and an offset included to allow for the slight movement of the extension tube holding the second grating when the chamber is pumped down. The beam path through the gratings is aligned using near- and far- field monitors and a small sliding mirror housed at the interface between the chamber and the extension tube. This mirror is aligned to be normal to the beam and can be moved into the beam path to reflect the beam before and after it passes through the gratings. The gratings are moved until the far-fields from both positions are coincident, which indicates that they are parallel.

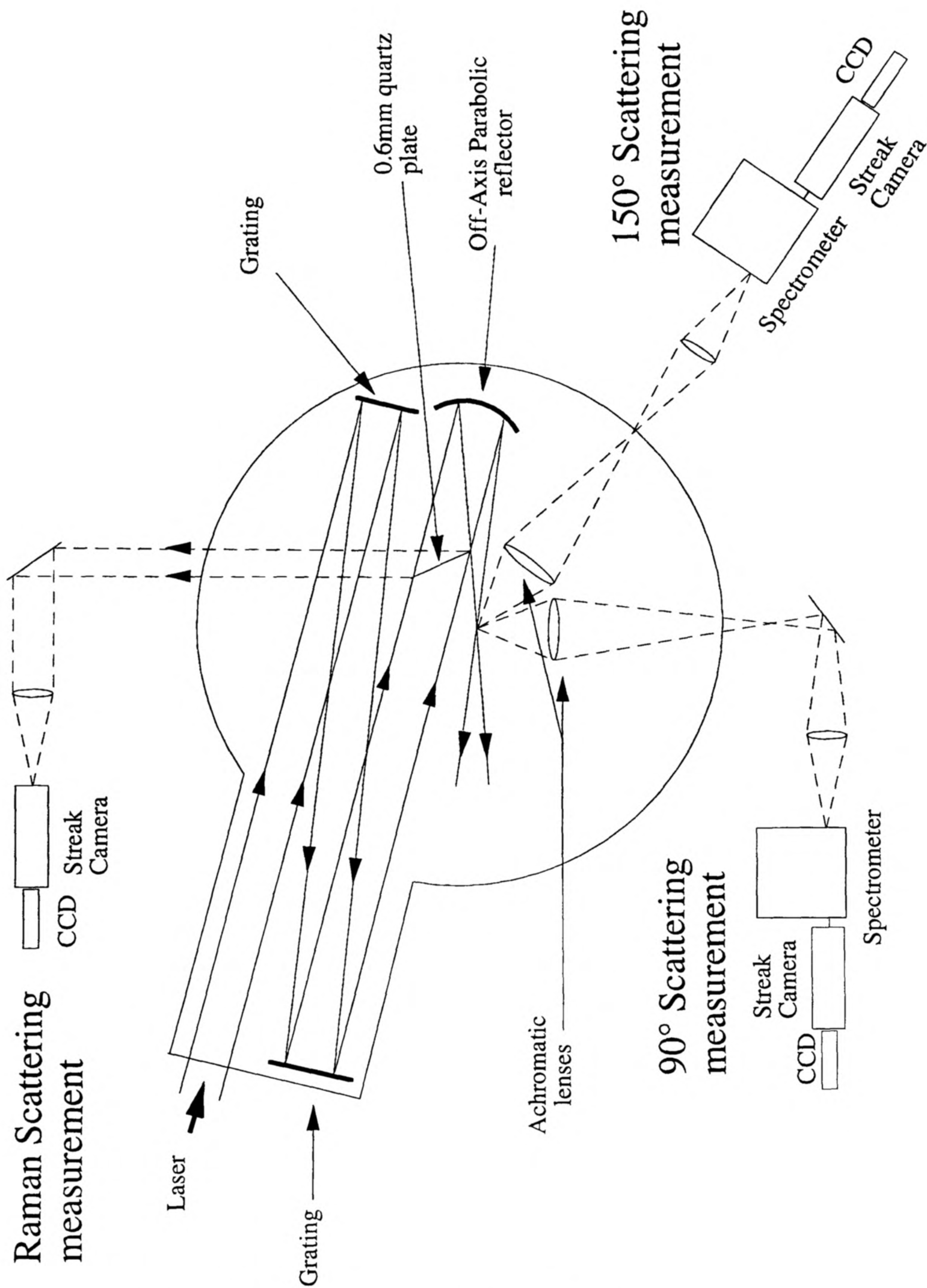


Figure 4.2.1. Schematic diagram of a typical experimental set-up.

From the second grating, the beam is directed onto an off-axis parabolic mirror. The 33 cm OAP fits easily into the chamber and is designed so that the angle subtended at the OAP between the second grating and the focus matches the off-axis angle of the parabolic mirror. This angle can be fine adjusted manually in air, and the position of the focus can be adjusted under vacuum by driving the OAP on x, y, z translation motors. The 100 cm OAP must be housed in an extension tube to the chamber, which means that the beam must be directed onto the OAP by a turning mirror. To overcome problems with movement of the OAP extension tube when pumping down, the OAP is mounted on a vacuum isolation stand which allows the tube to bend by means of a bellows arrangement, but holds the OAP in place. The OAP is adjusted to produce best focus at the centre of the chamber. The plasma diagnostics shown in the figure will be described in detail in the next section.

4.3. Plasma diagnostics.

The experiments described in this thesis have been concerned with determining the electron temperature of optically-ionised plasmas by Thomson scattering, or with measuring harmonic generation. The division between these experiments is somewhat blurred as Thomson scattering is also used as a density diagnostic in harmonic generation experiments. We describe below the experimental configurations used, but there is much overlap and some diagnostics have been omitted from the diagrams for clarity.

4.3.1. Thomson scattering experiment.

This experiment used the 33 cm OAP and was primarily concerned with determining electron temperatures and densities from Thomson and stimulated Raman scattering measurements. A schematic of the experimental set up has already been shown in figure 4.2.1.

Scattered light was collected at 90° and 150° by chromatically corrected f/5 10 cm focal length lenses and focused (with approximately 10x magnification) onto the entrance slits of two Czerny-Turner monochromators (a Spex 270 mm for the 90° channel and a Bentham 300 mm for the 150° channel). Two Aluminium coated mirrors were used in the 90° channel to rotate the direction of the laser beam to lie along the spectrometer slits. The 90° monochromator used two interchangeable gratings: one with 300 g/mm blazed at 250 nm; the other with 600 g/mm blazed at 300 nm. The spectrally dispersed output was then collected by a Hadland 500 series streak camera and passed through a Kentech 50/40 intensifier. The output of this was fibre optically coupled to a Photonic Science intensified CCD camera which was generally operated at its lowest gain setting. The streak direction was oriented perpendicular to the dispersion direction. This allowed spectral measurements which were time integrated at the slow streak speeds used (typically 100 - 200 ps/mm). The streak camera acted as a temporal filter to separate the weak Thomson scattered signal from the strong signal scattered from the chamber walls that arrived later in time.

The 150° monochromator used a 600 g/mm grating blazed at 250 nm. This was coupled to another streak camera and intensified CCD camera system (the same as for the 90° channel). Again the streak direction was perpendicular to the dispersion direction. The spectrometer slits were aligned either parallel or perpendicular to the axial direction to permit viewing of the whole or partial scattering region. The spectral resolution of both the 90° and 150° channels was 0.8 nm.

Light reflected at 180° was collected via a 0.6 mm thick quartz pellicle placed between the OAP and the second compressor grating. This light was then focused onto another streak camera and intensified CCD camera combination where the light scattered at all wavelengths was integrated. The streak camera was used to distinguish light scattered from the laser focus from stray light scattered from in and around the vacuum chamber.

Light at the laser wavelength could be removed using a filter (KG5) which blocked all wavelengths below 300 nm. The KG5 filter is made from a 2mm glass substrate and has a transmission of less than 0.001% below 280nm. Using this filter it is possible to collect only light produced from stimulated Raman scattering. Absolute reflectivity was determined by normalising scattered light levels to a Fresnel reflection at normal incidence from a quartz window. As this diagnostic used a filter to remove wavelengths below 300 nm, it could only be used with relatively high electron densities which could produce stimulated Raman scattering with wavelength shifts of greater than 50 nm.

Other diagnostics used in this experiment were an X-ray mini-spectrometer and two Faraday cups. The X-ray spectrometer contained a Thallium Acid Phthalate (TlAP) crystal and was used to look at X-ray emission in the region of the Helium- and Hydrogen- like Neon recombination series (5 - 15 Å). The spectrometer was placed about 6 cm from the laser focus in the horizontal plane just to the side of the 90° Thomson channel (i.e. at an angle of slightly less than 90°). A 10 µm Beryllium filter was used to eliminate stray laser radiation.

The Faraday cups produced a trace of ion current against time and could therefore be used to determine the energy distribution of the ions produced in, and ejected from, the plasma. These were simple devices which did not have a biasing mesh, but the whole cup could be biased (typically to 6 V) to prevent the escape of secondary electrons produced by ion impact. One Faraday cup was placed in the horizontal plane between the 90° and 150° channels at a distance of 11 cm from the laser focus, and the other was placed above the X-ray mini-spectrometer 22 cm from the laser focus.

Helium and Neon gas targets were provided by both static fill and pulsed gas jet (a high pressure solenoid valve with backing pressures up to 100 bar [119]). The gas jet, with a 1 mm diameter circular orifice, was positioned using a stepper motor driven translation

mount and oriented with the gas flow perpendicular to the laser beam axis and to the Thomson scattering channels. Gas densities up to 10^{20} atoms/cm³ could be achieved with the laser focused into the gas jet at a height of 100 μ m above the orifice. The gas jet pulse length was set to about 12 ms.

4.3.2. Harmonic generation experiment.

This was primarily a harmonic generation experiment, but Thomson scattering was used as a density diagnostic for the gas jet. In this experiment, the focusing was varied by changing between the 33 cm and 100 cm OAPs. A turning mirror was used to direct the beam down an extension tube. The turning mirror was also used with the 33 cm OAP so that when the OAPs were changed, the other diagnostics could remain in place. A schematic of this experiment is shown in figure 4.3.1. The Thomson channels are omitted for clarity.

The Thomson scattering diagnostic consisted of a channel at 90° to the laser axis. This was essentially the same as in the Thomson scattering experiment, with an instrument width (with the spectrometer slits set to 100 μ m) of about 1.4 nm and 0.7 nm for the 300 g/mm and 600 g/mm gratings respectively.

The primary diagnostic for the harmonic generation experiment was an XUV spectrometer based on a Hitachi variable line-spaced 1200 g/mm grating (ref. no. 001-0266) blazed at 3° [120]. A schematic diagram of this spectrometer is shown in figure 4.3.2. The spectrometer was used in a slitless configuration with the grating at an angle of 3.77° to image the source (at the chamber centre) onto the detector plane. The detector was a Galileo double micro-channel plate (MCP) detector (Galileo Model Number 1390-0020). The output from the MCP phosphor was fibre optically coupled onto a Photonic

Science Darkstar intensified CCD camera incorporating a 4:1 fibre-optic reducer. The CCD output was then passed to a computer controlled image acquisition system.

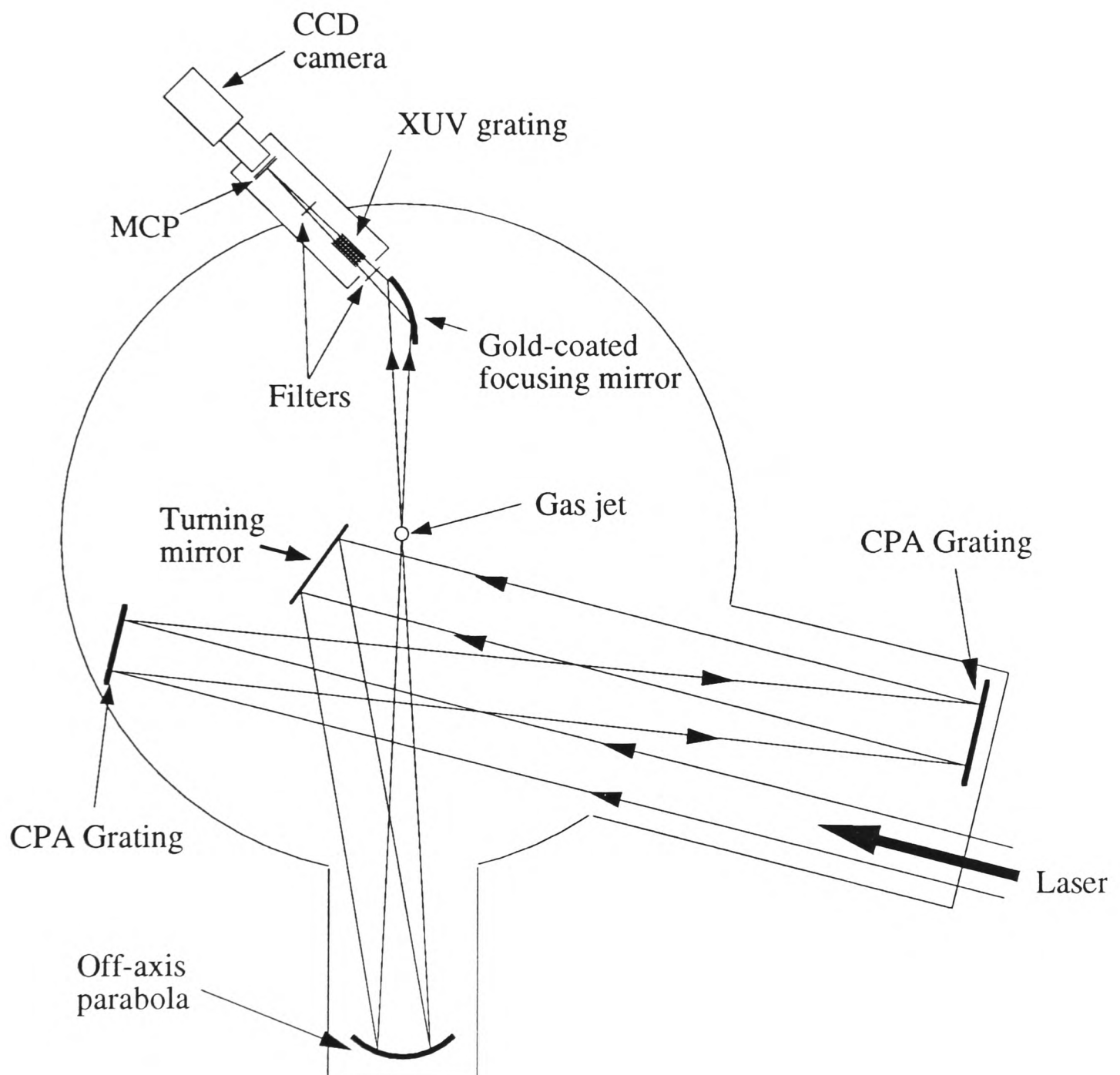


Figure 4.3.1. Schematic diagram of the harmonic generation experiment. The Thomson channels are omitted for clarity.

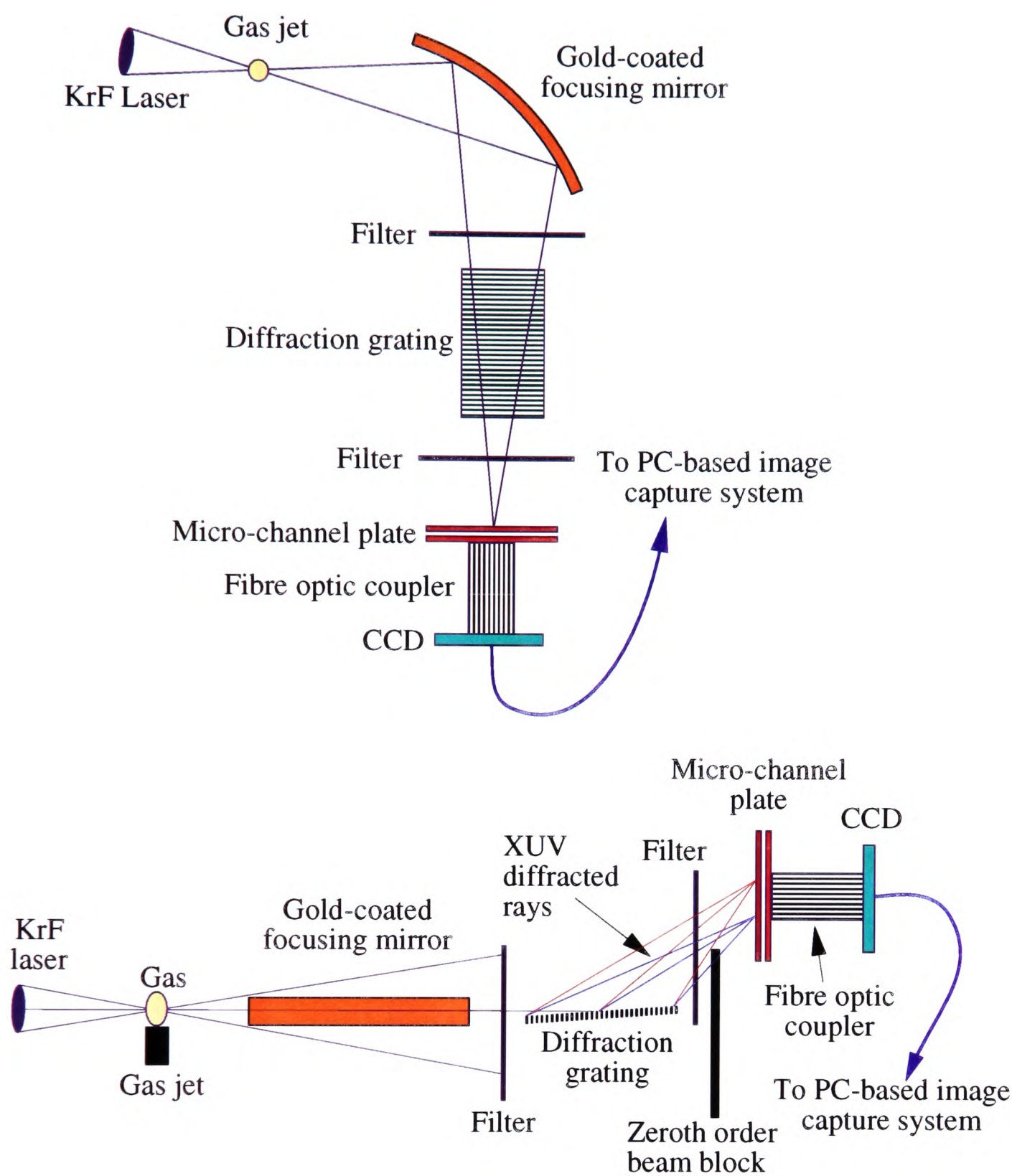


Figure 4.3.2. Top and side schematic views of the focusing XUV spectrometer. The angles and distances are not to scale.

A single toroidal gold-coated mirror was positioned before the grating to focus the chamber centre, and nominal laser focus, onto the detector. The mirror and grating were oriented to produce focusing in perpendicular directions resulting in a series of spectrally dispersed images of the focus at the detector. The total acceptance solid angle of the

spectrometer and focusing mirror was 1.5×10^{-5} steradians, i.e. 1/40th of the f/10 laser beam or 1/460th of the f/3 beam.

Filters were placed before and after the grating to protect both the grating and the detector from focused stray light. 20 - 200 $\mu\text{g}/\text{cm}^2$ Carbon filters were used when studying short wavelength harmonics ($<170 \text{ \AA}$), and 27 - 108 $\mu\text{g}/\text{cm}^2$ Aluminium filters were used for longer wavelength harmonics. The wavelength dependence of both the Aluminium and Carbon filter transmission [121] is shown in figure 4.3.3. The dispersion of the XUV spectrometer was approximately $0.4 \text{ \AA}/\text{pixel}$ and the instrument width varied between 10 pixels FWHM at $350 \text{ \AA}$ and 4 pixels FWHM at $70 \text{ \AA}$. The variation in instrumental width was due to the wide wavelength range covered by the spectrometer being too large for the Hitachi grating to produce a focus at the detector for all wavelengths.

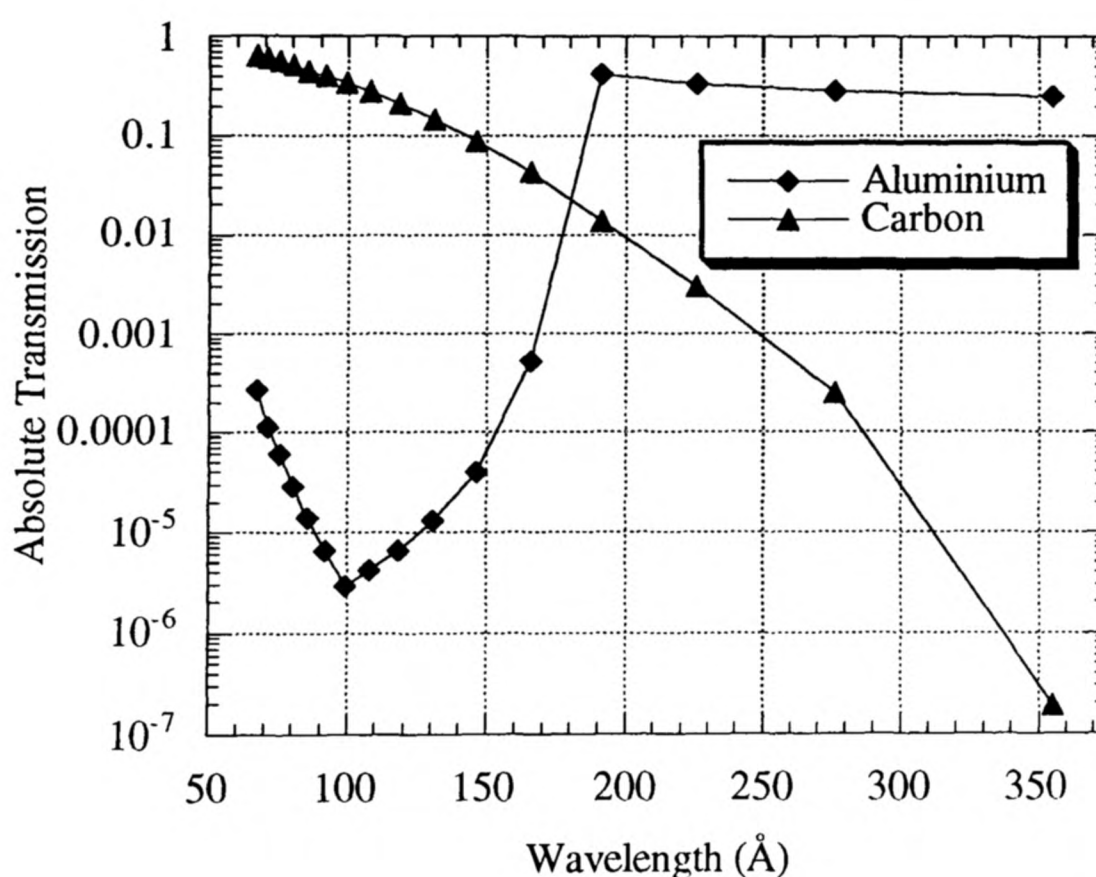


Figure 4.3.3. Transmission of $70 \mu\text{g}/\text{cm}^2$ Carbon and $108 \mu\text{g}/\text{cm}^2$ Aluminium. The data points indicate harmonic wavelengths of 248 nm starting with 7th harmonic at $355 \text{ \AA}$.

The MCP detector was typically operated with a voltage of 3 kV between its phosphor and the rear microchannel plate. The voltage between the front and rear plates was varied between 0.9 and 1.8 kV. The combined response of the MCP and DarkStar CCD camera was determined in a separate experiment [122] to be 0.014 photons per pixel count at a voltage of 1.5 kV which was essentially independent of wavelength. The gain of the MCP increases by a factor of two for an increase of 50 eV in the voltage between the two microchannel plates. The wavelength dependent reflectivity of the grating in first order was taken from data collected by Edelstein et al. [123] and was combined with a calculation of the focusing mirror reflectivity [124]. The focusing mirror was made from a 300 nm gold coating on a fused silica substrate. It had a surface roughness of 1 nm and the glancing angle at its centre was 4° . No allowance was made for either the grating being positioned at 3.77° rather than the 3.2° angle for which the reflectivities were determined, or for possible contaminants on the grating surface. The combined reflectivity of these two components is shown in figure 4.3.4.

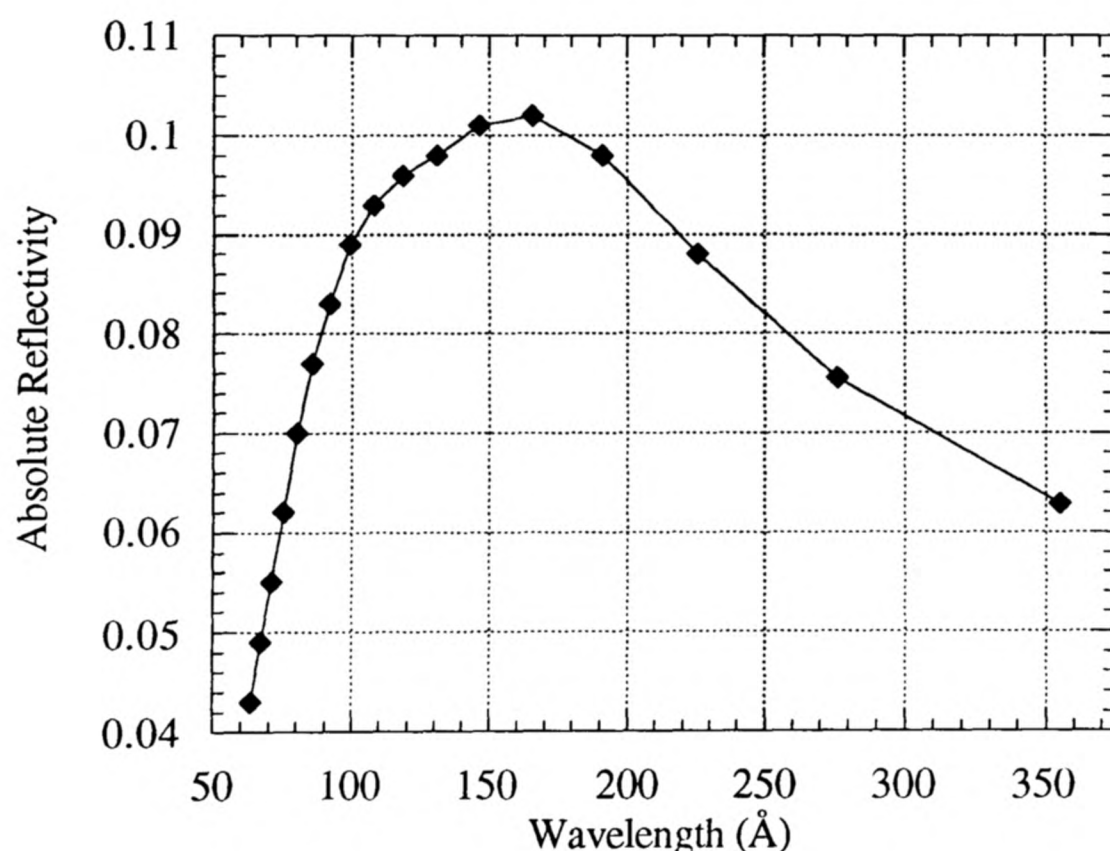


Figure 4.3.4. The combined absolute reflectivity of the Hitachi grating and gold-coated, toroidal focusing mirror. Each data point represents a harmonic wavelength of 248 nm starting with 7th harmonic at 355 Å.

4.4. Thomson scattering as a plasma diagnostic.

A powerful technique for studying the structure and dynamics of matter is the scattering of electromagnetic radiation. If we use such a frequency that the medium under study is essentially transparent and has a refractive index close to unity, the scattered radiation results primarily from interactions between the radiation and the electrons in the medium. Study of the angular distribution of the scattered radiation provides an indication of the spatial distribution of charge density in the medium, as for Bragg scattering of X-rays from crystals. Wavelength shifts, caused by Doppler shifting of the light scattered from moving electrons, give us a good diagnostic of the electron velocity distribution.

These methods can be used as a plasma diagnostic. This was first shown by studies of the incoherent scattering of radio waves from the ionosphere where the large physical scale of the medium counters the extremely low electron densities [125]. Since a plasma will, in general, be homogeneous we are interested in studying the frequency shift of the scattered light in order to determine densities and temperatures in the plasma. This scattering of light from the electrons in a plasma is known as Thomson scattering.

The requirement that the plasma be virtually transparent to the probing radiation and display a refractive index close to unity means that the radiation frequency must be much greater than the electron plasma frequency, ω_{pe} . In the experiments discussed in this thesis, the maximum electron density is of the order of 10^{21} cm^{-3} (i.e. $\omega_{pe} = 1.8 \times 10^{15} \text{ s}^{-1}$), which is approximately the critical density for an Nd:Glass laser (1053 nm). The critical density for a KrF laser (248 nm) is about sixteen times higher than this. Ideally, the laser beam which is being Thomson scattered should not perturb the plasma it is probing. In our experiments, however, we are looking at Thomson scattering from the beam which is creating the plasma. This can lead to problems with SRS growing from thermal noise created in the plasma by the Thomson scattering which

will then mask the Thomson scattered signal. We will also experience problems in interpreting the scattered spectrum, because the plasma is still forming and equilibrating while scattering is taking place.

There are two main regimes which characterise Thomson scattering. The first is the scattering of light from a single electron or from a group of uncorrelated electrons. In this case the electrons can be considered to be oscillating in the electric field and re-radiating the energy at the same frequency, ω_0 . The cross section for this process is given by the Thomson scattering cross section $\sigma_T = (8\pi/3)(e^2/4\pi\epsilon_0 mc^2)^2$ which is equal to $6.65 \times 10^{-25} \text{ cm}^2$. The spectrum of the light which arrives at the detector is Doppler shifted due to the motion of the electron in the direction of the scattering vector, $\mathbf{k}$, defined in figure 4.4.1 below. This process can also be thought of as the decay of the incident photon into a photon and a plasmon. For scattering from a collection of uncorrelated electrons, the measured spectrum will be centred on ω_0 and have a width which is characteristic of the Doppler shift due to the thermal velocity of the electrons.

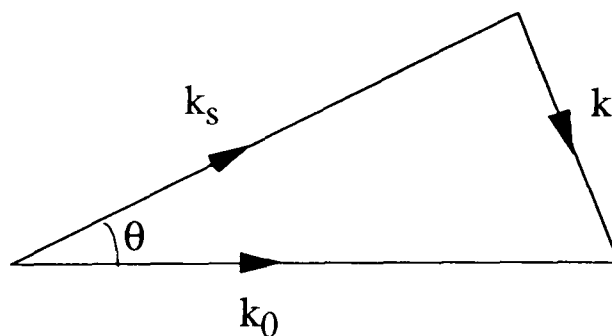


Figure 4.4.1. The change in wavevector, $\mathbf{k}$, resulting from the scattering of an incident wavevector, $\mathbf{k}_0$, to a scattered wavevector, $\mathbf{k}_s$, by an electron. The magnitude of the Doppler shift is determined by the component of the electron velocity along $\mathbf{k}$.

The condition that determines whether the electrons can be considered to be uncorrelated is the Debye length, $\lambda_D = (\kappa T_e \epsilon_0 / Ne^2)^{1/2}$. The Debye length represents the distance

over which electron shielding will hide a deviation from neutrality in a plasma. Therefore, if density fluctuations have a scale length less than the Debye length, they will be shielded by an electron sheath and will not contribute to the scattering. In this case the scattering can be considered to be from uncorrelated electrons.

Alternatively, scattering is dominated by collective effects of the electrons in the plasma (coherent scattering). This occurs when there are plasma density fluctuations which are large compared to the Debye length. The scattered photon can now be considered to be scattering off plasma waves rather than off individual electrons. In this case the spectrum of the scattered light is affected by the spectrum of the density fluctuations. In general there will be a maximum in the spectrum at a frequency shifted from ω_0 by the Bohm-Gross frequency.

Thomson scattering is a very useful plasma diagnostic [29,126-132], because the spectrum is determined by the plasma conditions. In the incoherent scattering case, the width of the spectrum gives a direct measure of the thermal velocity of the electrons and hence the temperature. In the collective, or coherent, scattering case the position of the maximum in the spectrum is given by the Bohm-Gross dispersion relation

$$\omega^2 = \omega_{pe}^2 + 3 \left(\frac{\kappa T_e}{m_e} \right) k^2 \quad (4.4.1)$$

Thus the spectrum is determined by both the temperature and the density.

We will not derive the Thomson scattering spectrum here, as this has been done in great detail in numerous publications [29,133-135]. The intensity of light which is Thomson scattered into a frequency range $d\omega$ and a solid angle $d\Omega$ is given by

$$I_s(\mathbf{k}, \omega) d\omega d\Omega = NI_0 \sigma_T S(\mathbf{k}, \omega) d\omega d\Omega \quad (4.4.2)$$

where N is the number of electrons, I_0 is the incident laser intensity, and σ_T is the Thomson scattering cross section, given by

$$\sigma_T = \left(\frac{8\pi}{3} \right) \left(\frac{e^2}{4\pi\epsilon_0 mc^2} \right)^2 \sin^2 \psi \quad (4.4.3)$$

where ψ is the angle between the plane of scattering and the laser polarisation. The form of the scattered spectrum is given by the form factor $S(\mathbf{k}, \omega)$ which for an arbitrary one-dimensional electron (ion) velocity distribution function f_{e0} (f_{i0}) is given by

$$S(\mathbf{k}, \omega) = \frac{2\pi}{k} \left| \frac{1 - G_i}{\epsilon} \right|^2 f_{e0} \left(\frac{\omega}{k} \right) + \frac{2\pi Z}{k} \left| \frac{G_e}{\epsilon} \right|^2 f_{i0} \left(\frac{\omega}{k} \right) \quad (4.4.4)$$

where G_e and G_i are the 'screening integrals' which make up the longitudinal dielectric function

$$\epsilon = 1 - G_e - G_i \quad (4.4.5)$$

The first term in equation 4.4.4 containing f_{e0} is called the 'electron term', and the second term containing f_{i0} is called the 'ion term'. The form of the Thomson scattered spectrum is then given by the electron and ion velocity distribution functions and the plasma dielectric function.

4.4.1. The form of the Thomson scattered spectrum.

In order to determine the exact form of the Thomson scattered spectrum we must specify the electron and ion velocity distributions, f_{e0} and f_{i0} . However, in the case of a plasma

which is optically-ionised by a short pulse laser (where Thomson scattering is from the ionising laser beam) we do not necessarily know the form of these distributions. When the plasma is created we expect the initial electron distribution to be characteristic of the ATI distribution for the charge states that have been ionised. This is a series of Gaussian distributions centred at a different temperature for each ionisation stage. Inverse bremsstrahlung heating and collisions in the plasma will convert this to a Maxwellian distribution with a single temperature, but the onset of SRS will tend to create a 'hot' electron wing in the velocity distribution. We expect the ion distribution to evolve from that of the gas from which the plasma was created to a higher temperature distribution as electron-ion and ion-ion collisions occur. Since we do not know the exact velocity distributions we will assume them to be Maxwellian for simplicity. Now let us introduce the dimensionless frequency parameters

$$x_e = \frac{\Delta\omega}{\sqrt{2}kv_{te}} \text{ and } x_i = \frac{\Delta\omega}{\sqrt{2}kv_{ti}} \quad (4.4.6)$$

(where $\Delta\omega$ is the frequency shift from the incident laser frequency) and the scattering parameter

$$\alpha = \frac{1}{k\lambda_D} = \frac{1}{k} \left(\frac{e}{\epsilon_0} \right)^{1/2} \left(\frac{N}{T_e(\text{eV})} \right)^{1/2} \quad (4.4.7)$$

The Thomson scattering form factor for a Maxwellian distribution functions is

$$S(\mathbf{k}, \omega) = \frac{2\pi^{1/2}}{k} \left\{ \frac{A_e}{|\epsilon|^2} + \frac{A_i}{|\epsilon|^2} \right\} \quad (4.4.8)$$

where A_e is the electron term and A_i is the ion term

$$A_e = \exp(-x_e^2) \left[\left(1 + \alpha^2 Z \frac{T_e}{T_i} \operatorname{Re}\{W(x_i)\} \right)^2 + \left(\alpha^2 Z \frac{T_e}{T_i} \operatorname{Im}\{W(x_i)\} \right)^2 \right] \quad (4.4.9)$$

$$A_i = Z \left(\frac{m_i T_e}{m T_i} \right)^{1/2} \exp(-x_i^2) \left[\left(\alpha^2 \operatorname{Re}\{W(x_e)\} \right)^2 + \left(\alpha^2 Z \operatorname{Im}\{W(x_e)\} \right)^2 \right] \quad (4.4.10)$$

where

$$|\varepsilon|^2 = \left[1 + \alpha^2 \left(\operatorname{Re}\{W(x_e)\} + Z \frac{T_e}{T_i} \operatorname{Re}\{W(x_i)\} \right) \right]^2 + \left[\alpha^2 \operatorname{Im}\{W(x_e)\} + \alpha^2 Z \frac{T_e}{T_i} \operatorname{Im}\{W(x_i)\} \right]^2 \quad (4.4.11)$$

The plasma dispersion function, $W(x)$, is given by

$$W(x) = \left(1 - 2x e^{-x^2} \int_0^x e^{-p^2} dp \right) - i \left(\pi^{1/2} x e^{-x^2} \right) \quad (4.4.12)$$

The imaginary part of this gives rise to Landau damping. The real part can be approximated by [136]

$$\operatorname{Re}\{W(x)\} = 1 - 2x^2 \left(1 - \frac{2x^2}{3} + \dots \right) \text{ for } x \ll 1 \quad (4.4.13)$$

and

$$\operatorname{Re}\{W(x)\} = -\frac{1}{2x^2} \left(1 - \frac{3}{2x^2} + \dots \right) \text{ for } x \gg 1 \quad (4.4.14)$$

Salpeter [135] first demonstrated that, when the electron temperature is approximately equal to the ion temperature, the spectrum takes a relatively simple form, allowing the

electron and ion features to be treated separately. In the Salpeter approximation the ratio of energy in the central 'ion' feature to the rest of the spectrum is given by

$$R_I = \frac{Z\alpha^4}{1 + \alpha^2(1 + ZT_e/T_i)}$$

In the experiments presented in this thesis, the Salpeter approximation does not hold, as the electron temperature is much higher than the ion temperature. In this case the fraction of energy scattered into the ion feature compared to the energy in the total Thomson scattering spectrum is given by $R_I = \alpha^2 / (1 + \alpha^2)$, and the ratio of energy scattered into the ion feature compared to the electron feature is $R = \alpha^2$, which can be used to estimate the value of α from a Thomson scattered spectrum. Figure 4.4.2. shows R_I as a function of T_e/T_i for both the full Thomson scattering form and the Salpeter approximation for $\alpha = 1$ in Helium. The fraction for the full solution approaches the expected value of 0.5 at high T_e/T_i , whereas the Salpeter approximation approaches zero.

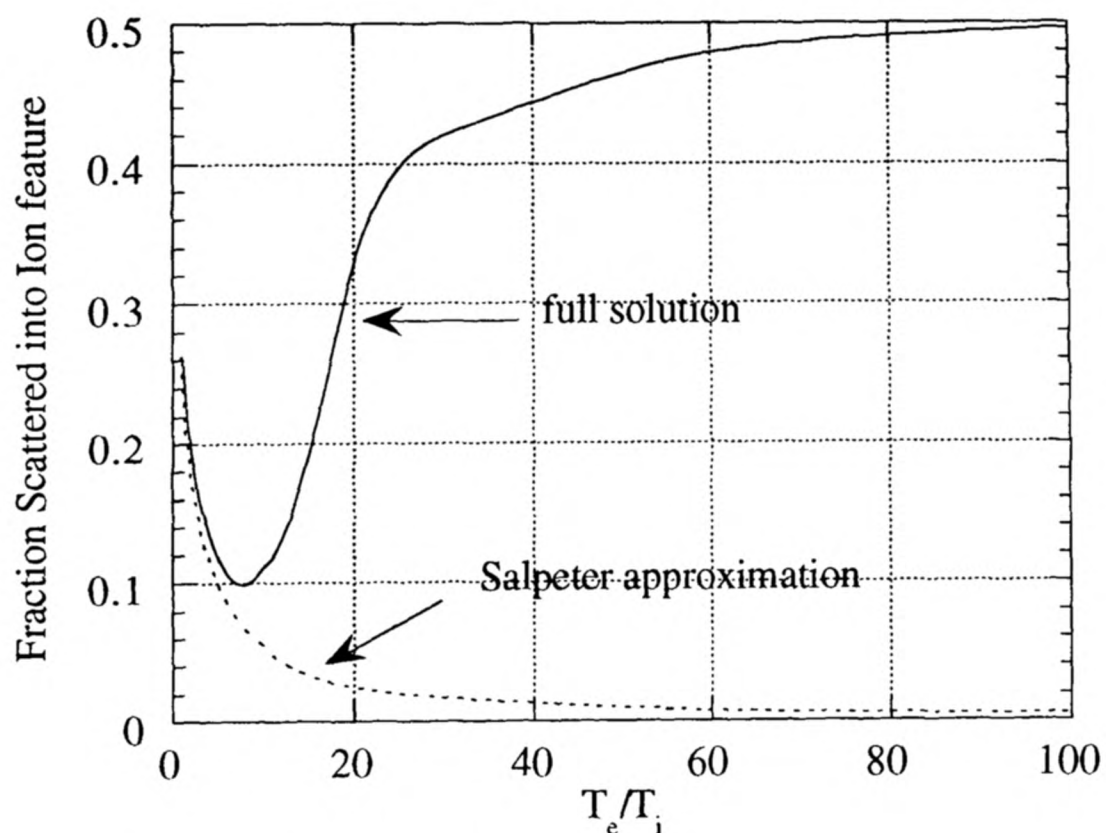


Figure 4.4.2. The fraction of energy Thomson scattered into the ion feature as a function of T_e/T_i for a fully ionised Helium plasma with $\alpha = 1$.

There are two regimes of Thomson scattering corresponding to uncorrelated and correlated electron behaviour (as mentioned previously), which we will now describe.

4.4.2. Incoherent Scattering ($\alpha \ll 1$).

Since both G_e and G_i are proportional to α^2 , in the limit that α goes to zero the expression for $S(\mathbf{k}, \omega)$ becomes

$$S(\mathbf{k}, \omega) = \left(\frac{2\pi}{k} \right) f_{e0}(\omega/k) \quad \text{for } \alpha \ll 1 \quad (4.4.15)$$

Thus the expression is reduced to a measure of the velocity distribution of the electrons. This corresponds to the incoherent limit where the spectral width of the scattered light comes from the Doppler shift due to the electron motion. Figure 4.4.3 shows a Thomson scattered spectrum for $\alpha = 0$, generated by a program written by J.S. Wark [137] and modified by W.J. Blyth [29] which computes the Thomson scattering form factor using the full plasma dispersion function described in section 4.4.1. The data has been arbitrarily normalised to a peak value of 10.

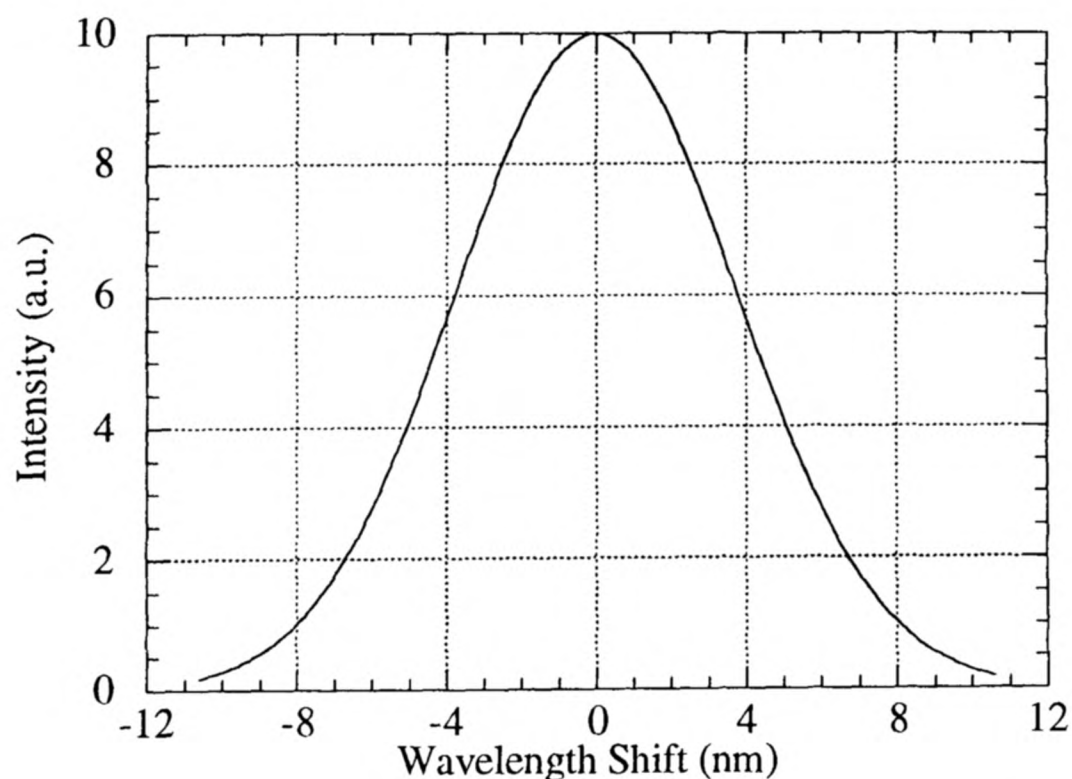


Figure 4.4.3. Simulated 90° Thomson scattering spectrum for He^{2+} with $\alpha = 0$, $T_e = 20 \text{ eV}$, $T_e/T_i = 100$.

4.4.3. Coherent Scattering ($\alpha > 1$).

The spectrum of scattered light can be significantly affected if the dielectric function, ϵ , becomes small. This occurs particularly at the natural resonances of the plasma: the electron plasma frequency, ω_{pe} ; and the ion-acoustic wave frequency, ω_{pi} . We will now study the form of $S(\mathbf{k}, \omega)$ in different parts of the spectrum.

At high frequencies ($\omega \gg \omega_{pi}$ and $x_i \gg 1$), the second term in equation 4.4.8 (the ion feature) is negligible. In this case, $S(\mathbf{k}, \omega)$ is dominated by the electron term which contains the electron distribution function, f_{e0} .

At low frequencies ($\omega \sim \omega_{pi}$), the relative contribution of the ion and electron terms depend on the value of T_e/T_i . Now we have $x_i \sim 1$ and $x_e \ll 1$, so $\text{Re}\{W(x_i)\} \sim 1$ and $\text{Re}\{W(x_e)\}$ is non-vanishing. Also $\exp(-x_e^2) \approx 1$ and so the competing terms are

$$\left[1 + \alpha^2 \frac{ZT_e}{T_i} \text{Re}\{W(x_{pi})\}\right]^2 \quad (\text{electron term}) \quad (4.4.16)$$

$$\text{and} \quad Z \left(\frac{m_i T_e}{m T_i}\right)^{1/2} \alpha^4 \exp(-x_{pi}^2) \quad (\text{ion term}) \quad (4.4.17)$$

with x_{pi} evaluated at the ion-acoustic frequency ω_{pi} . The condition for the electron term to dominate the ion term can be approximately written as

$$Z \left(\frac{T_e}{T_i}\right)^{3/2} > \left(\frac{m_i}{m}\right)^{1/2}. \quad (4.4.18)$$

which will be satisfied for Hydrogen with $T_e/T_i > 10$, and for Ne^{8+} with $T_e/T_i > 7$. If this condition is satisfied, there can be a strong resonance signal at low frequencies due to

the electron response to an ion-acoustic resonance. This is typically true for values of T_e/T_i greater than 10 to 50 depending the value of α . This condition corresponds physically to an ion-acoustic mode in the plasma which is only weakly attenuated by Landau damping. A typical Thomson scattering spectrum from Helium for $\alpha = 2$ and $T_e/T_i = 100$ is shown in figure 4.4.4. Note the very intense, narrow, central feature caused by the electron response which peaks at a value of 10. In our experiments this energy is broadened out by the instrument function and so the form of the ion-acoustic mode cannot be determined.

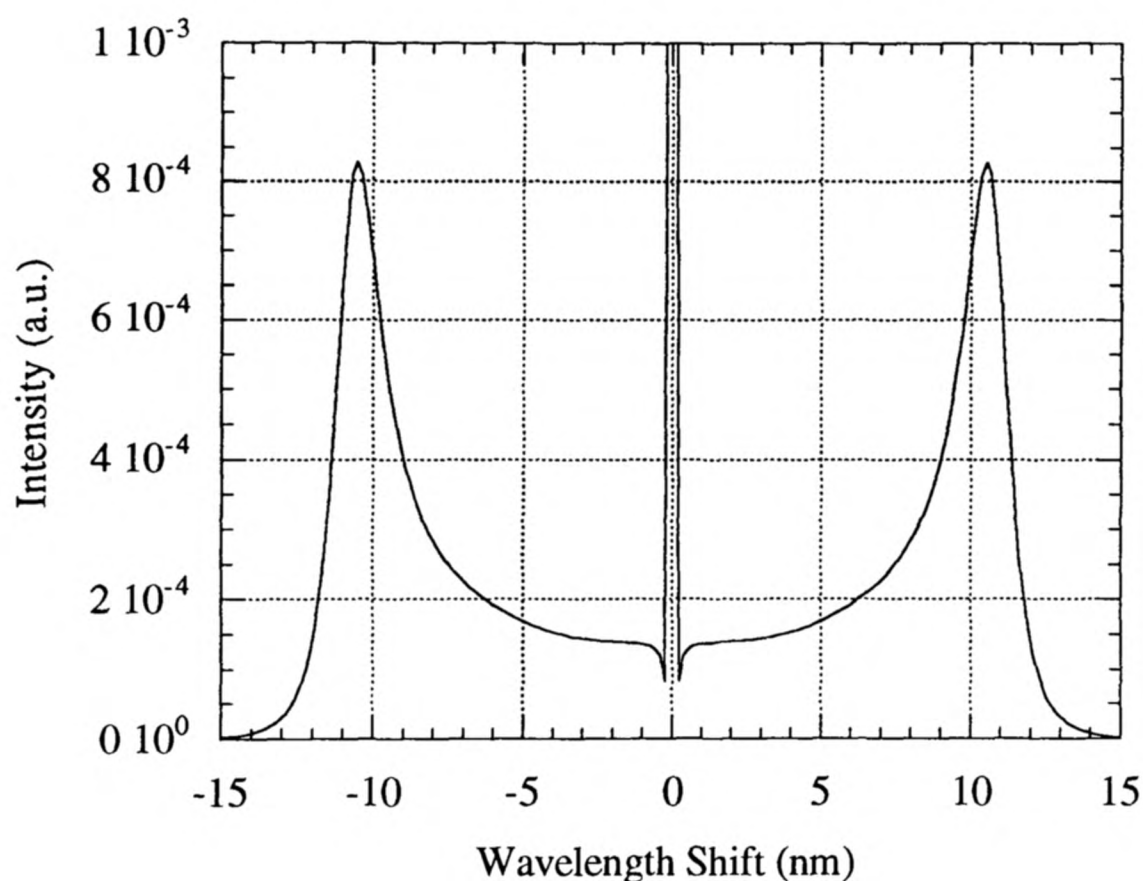


Figure 4.4.4. Simulated 90° Thomson scattering spectrum for He^{2+} with $\alpha = 2$, $T_e = 50 \text{ eV}$, $T_e/T_i = 100$.

4.5. Summary.

In this chapter we have reviewed the Sprite laser system, paying particular attention to its CPA mode of operation. We have also described in detail the experimental diagnostics used for electron temperature and for harmonic generation measurements, and we have

reviewed the theory of Thomson scattering. This is in anticipation of the next two chapters (chapters five and six), where we will describe the measurements taken using the diagnostics we have described here.

Chapter Five

Electron Temperature Measurements

5.1. Plasma simulations.

In this chapter we shall review the data obtained from our plasma diagnostics described in chapter four. These will include analysis of Thomson and stimulated Raman scattered optical signals, and an analysis of Faraday cup and X-ray spectrometer data. The justification for this investigation of electron temperatures of OFI plasmas is to determine whether the conditions required for the OFI recombination laser scheme described in section 3.2.3 are attainable. These conditions were set out by Eder [44] assuming a 100 fs laser pulse at a wavelength of 248 nm. The short pulse length is required to minimise the total inverse bremsstrahlung heating. The short wavelength is required to minimise SRS which has a growth rate which is proportional to $\omega_0^{-1/2}$ (equation 2.5.6) and the inverse bremsstrahlung heating rate which is proportional to ω_0^{-2} in the standard high frequency, low intensity regime (equation 2.4.3).

We do not have a 100 fs, 248 nm laser available for our experiments, the Sprite laser in CPA mode has a pulse length of around 380 fs. However, it is possible to determine the viability of the recombination XUV laser scheme by taking measurements at 380 fs and scaling to a shorter pulse length. Before looking at the experimental data we shall attempt to simulate our experimental conditions using the MEDUSA one dimensional plasma simulation code [41].

5.1.1. The MEDUSA code.

The MEDUSA code was originally developed to simulate conditions for inertial confinement fusion [6]. It has now been developed over many years to model many aspects of laser-plasma interactions, including optically-ionised plasmas [35,138]. The simulations presented in this chapter are modelled using MEDUSA version 104 [139] which includes ATI heating and the modified high intensity inverse bremsstrahlung rate [39].

MEDUSA is a one dimensional Lagrangian code, which means that the planar region of the laser-plasma interaction is divided up into cells of equal mass using either cylindrical or spherical geometry. In our simulations we will use cylindrical geometry with the laser incident axially down the cylinder. The cells therefore characterise the radial profile of the laser and plasma. A schematic of this geometry is shown in figure 5.1.1. The initial cell configuration for these simulations is a 10 μm core divided into 30 cells, followed by a 100 μm ring of 20 cells, and then a 1000 μm outer ring of 10 cells, but these cells will expand or contract during the laser pulse as they follow the movement of plasma mass in the system. The code includes IB and ATI heating, but does not include SRS. At high electron densities ($N > 10^{19} \text{ cm}^{-3}$), Landau damping of plasma waves generated by SRS is a major cause of heating. This means that the simulations presented here will only set a lower bound on the electron temperature produced by a given set of parameters. Ponderomotive effects can also be included approximately in MEDUSA with ponderomotive expulsion of electrons and ions from the laser focus causing a significant reduction in density along the axis of the laser for certain conditions [29,45]. However, for the sub-picosecond laser pulses used in these experiments ponderomotive expulsion does not significantly alter the plasma density during the laser pulse. MEDUSA simulations can only model ponderomotive expulsion qualitatively, because the code demands charge neutrality within each cell. This approximation is good for the longer

time scales associated with the 10 ps Raman amplified mode of the Sprite laser, but is not necessarily valid for the shorter 380 fs CPA mode.

Much debate has been generated recently over the correct form of heat conduction to use in simulations of sub-picosecond lasers. The usual form is the Spitzer and Härm heat diffusion [36,140], $Q_f = fNkT_e v_{th}$, where f is the flux-limiter to prevent heat flow reaching non-physical values. The flux-limiter is typically set to a value of around 0.1. This formulation assumes that the electron mean free path is much smaller than the scale length of the temperature gradient and that the electron temperature distribution is Maxwellian. However, recent comparisons to experiment [35] indicate that a more realistic heat conduction is achieved if we use a flux-limiter of close to unity, or if we use a convolution integral formula [141-143]. Such a formulation is computationally simpler than solving the Boltzmann equation and also accounts for non-local effects. In the simulations performed in this thesis we shall use non-local heat conduction of the same form as in a recent paper by Djaoui and Offenberger [35]. This convolution (or non-local) form of the heat conduction is more suited to situations where the electron mean free path is larger than the scale length of the temperature gradient. It gives flux inhibition under sharp temperature gradients and allows for preheat ahead of the main heat front. In the diffusive limit it leads to the classical Spitzer and Härm result.

To summarise, the MEDUSA parameters used in simulations presented in this chapter are:

Geometry :	Cylindrical, laser on axis.
Cell configuration :	30 cells to a radius of 10 μm , 20 cells to a radius of 100 μm , 10 cells to a radius of 1000 μm .
Heat Flow :	Non-local.

Ponderomotive effects :	Not used.
IB heating :	Form of Polishchuk et al. [39].
Laser intensity :	Variable (10^{15} - 10^{19} W/cm ²), default = 3×10^{17} W/cm ² .
Laser pulse profile :	Gaussian; FWHM variable (30 fs - 10 ps), default = 380 fs.
Laser focal spot profile :	Gaussian; FWHM variable (1.8 - 10 μ m), default = 10 μ m.
Gas at laser focus:	Neon (and Helium).
Initial electron temperature :	0.1 eV.
Initial ion temperature :	0.1 eV.
Time step :	10 fs for a 380 fs pulse.
Simulation start time :	4 pulse lengths before peak of pulse.

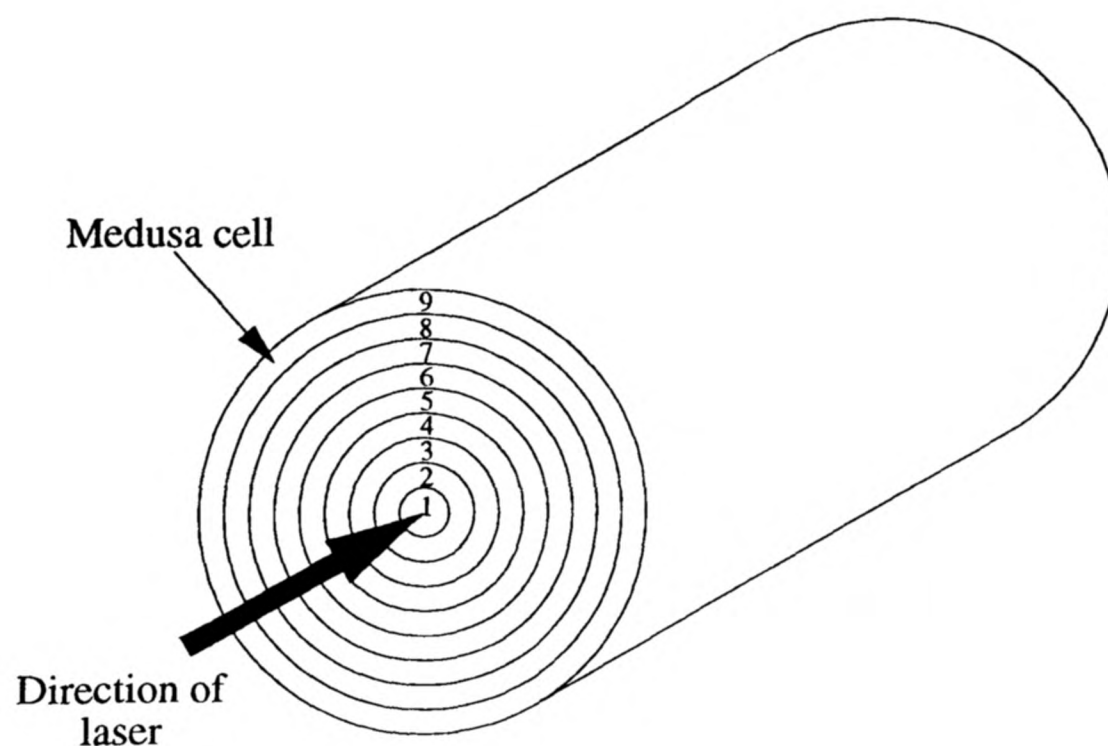


Figure 5.1.1. Schematic diagram of the MEDUSA code simulation geometry.

5.1.2. Predictions of inverse bremsstrahlung and ATI heating.

The simulations of Eder et al. [44,78,80] predict that gains of over 100 cm^{-1} can be produced on the $98 \text{ \AA}$ $3d_{5/2}-2d_{3/2}$ transition during the recombination of Neon from its Helium-like ion to its Lithium like ion at an electron density of $5 \times 10^{20} \text{ cm}^{-3}$ and an electron temperature of 40 eV. If the temperature increases to 100 eV then the gain will

drop to about 10 cm^{-1} . We must determine whether such conditions can possibly be met by the Sprite laser CPA system, or by any other laser system. In figure 5.1.2 we show a simulation for Neon at a gas density of $6.25 \times 10^{19} \text{ atoms/cm}^3$ (producing an electron density of $5 \times 10^{20} \text{ cm}^{-3}$ when eight times ionised) at the centre of the focus of a 248 nm laser with a Gaussian temporal profile with a FWHM of 380 fs and a Gaussian radial focal profile with a FWHM of $10 \mu\text{m}$. The peak laser intensity is $3 \times 10^{17} \text{ W/cm}^2$. In the simulations described in this section, the laser parameters are the same as these unless stated otherwise. The electron density for all simulations except this one should be taken to be 10^{20} cm^{-3} unless stated otherwise. 'Time' refers to the time relative to the peak of the laser pulse in all plots of the variation of a plasma parameter during the laser pulse.

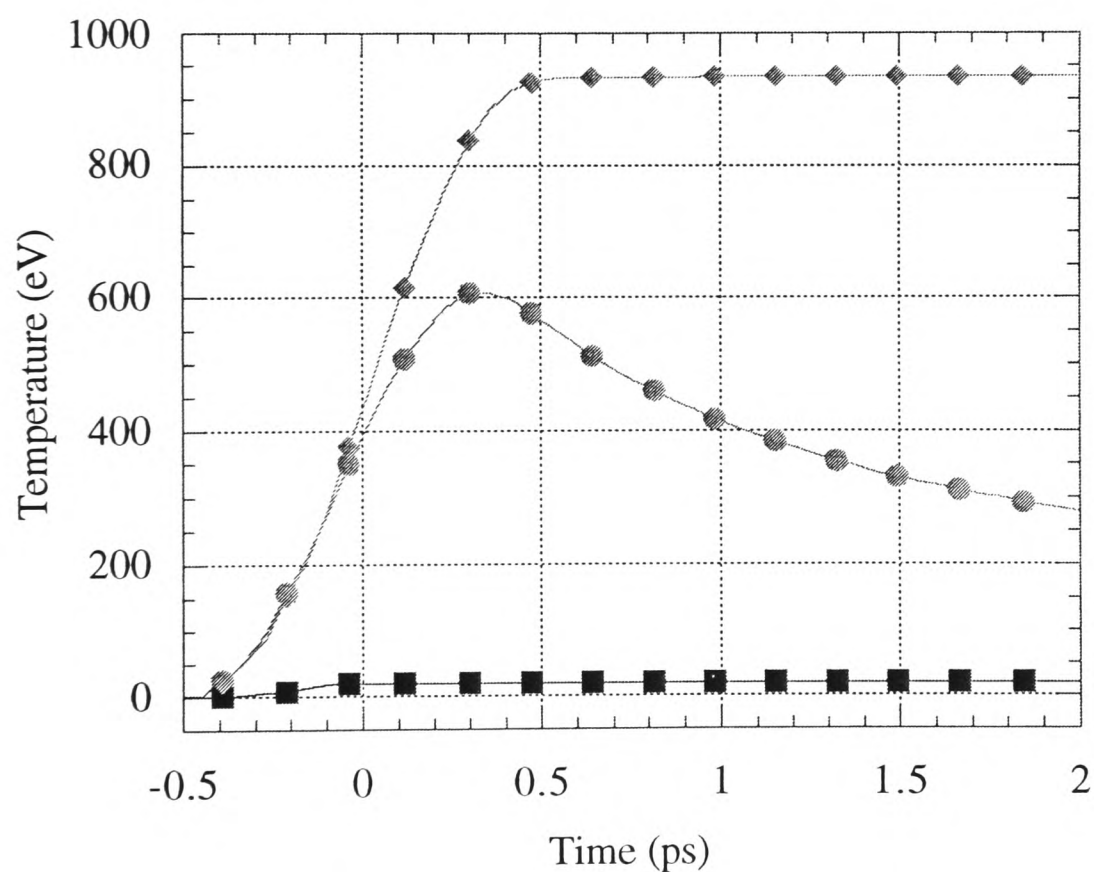


Figure 5.1.2. Variation of electron temperature throughout a 380 fs laser pulse. Components of the temperature shown are ATI (squares), inverse bremsstrahlung (diamonds), and the total temperature including heat diffusion (circles).

The total electron temperature shown is the sum of ATI and IB heating allowing for heat diffusion out from the centre of the focus. The ATI and IB components in the absence of

diffusion are also shown. The electron temperature peaks at over 600 eV about 200 - 300 fs after the peak of the laser pulse, the time when peak gain is predicted to occur. Obviously the electron temperature is much too large for the Neon recombination scheme at these densities. To lower this temperature we may vary one or more of the density, the laser intensity, the laser spot size, or the laser pulse length. Reducing the density will reduce the electron temperature (by reducing IB), but will lead to reduced gain. The time dependence of the total electron temperature is plotted in figure 5.1.3 for a range of densities from 10^{17} cm^{-3} to 10^{21} cm^{-3} . Note that the electron density quoted is the maximum density that is produced by the laser pulse and corresponds to eight times ionisation of Neon at these laser intensities. Figure 5.1.3 indicates that, to achieve an electron temperature of the order of 40 eV with a 380 fs laser pulse, we must reduce the electron density to about 10^{19} cm^{-3} . This will reduce gain in the Neon system to a negligible level.

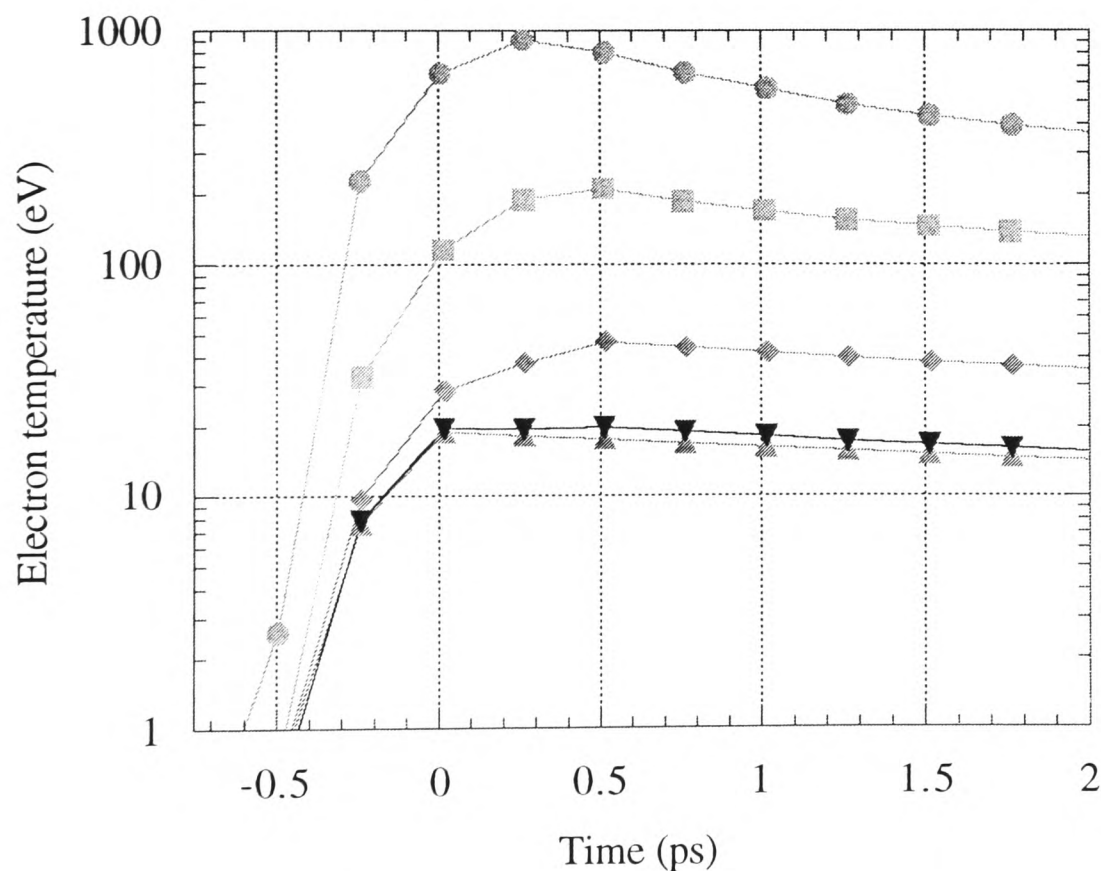


Figure 5.1.3. Variation of the total electron temperature on axis with time for differing electron densities. The data correspond to electron densities of 10^{17} cm^{-3} (triangles), 10^{18} cm^{-3} (inverted triangles), 10^{19} cm^{-3} (diamonds), 10^{20} cm^{-3} (squares), 10^{21} cm^{-3} (circles). The peak laser intensity is $3 \times 10^{17} \text{ W/cm}^2$.

Another way of reducing the electron temperature is to lower the laser intensity. Figure 5.1.4 shows the variation of the total electron temperature throughout the laser pulse for a range of intensities from 10^{15} W/cm² to 10^{19} W/cm². The figure implies that we can achieve any electron temperature we require just by lowering the laser intensity.

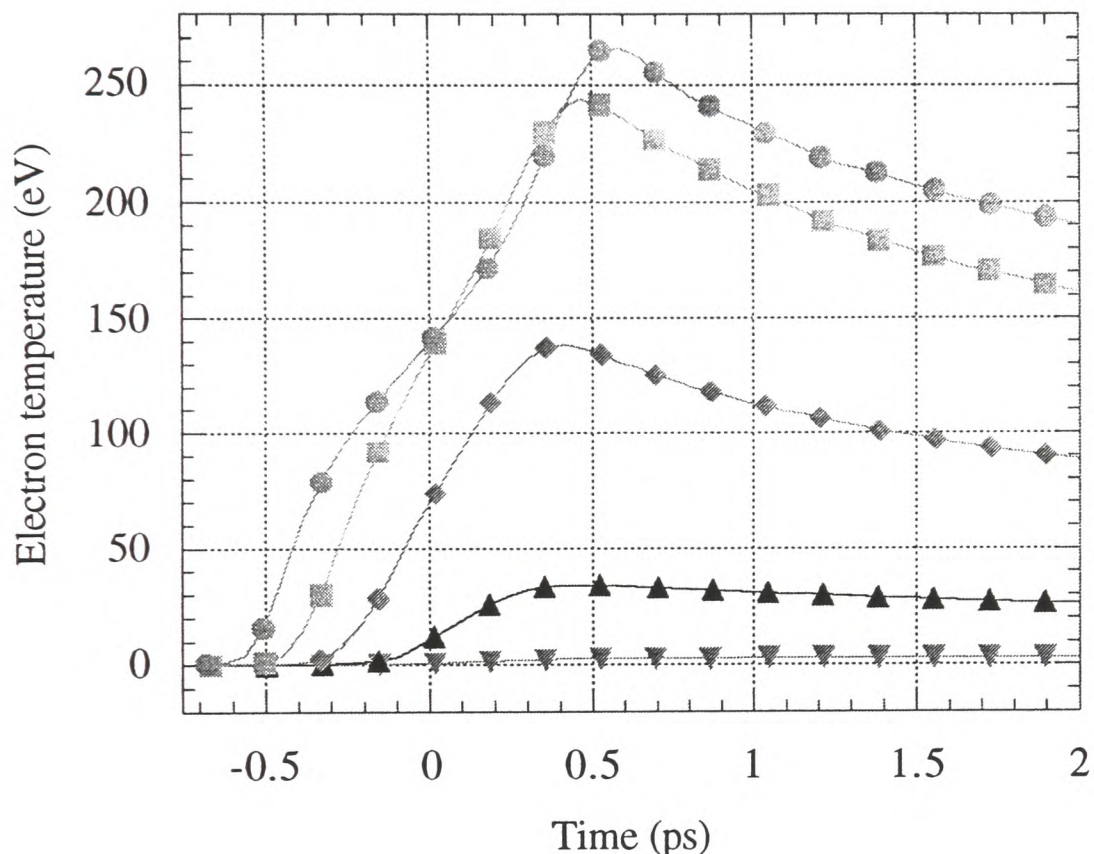


Figure 5.1.4. Variation of total heating rate with time for differing laser intensities. The data correspond to intensities of 10^{15} (inverted triangles), 10^{16} (triangles), 10^{17} (diamonds), 10^{18} (squares), 10^{19} (circles) W/cm². The Neon gas density is fixed at 1.25×10^{19} atoms/cm³.

Once we go below a certain threshold intensity, however, we will no longer be able to optically-ionise to our required ionisation stage. Figure 5.1.5 shows the effective ionisation state of the plasma at the same intensities shown in figure 5.1.4. This confirms the conclusions in section 2.2 which indicates a threshold intensity of about 3×10^{17} W/cm² for the production of Helium-like Neon (Ne VIII). However, this intensity scaling may work if we iso-electronically scale to a longer XUV laser wavelength by moving to a lower atomic number. Nitrogen, for example, has a

threshold intensity of just over 10^{16} W/cm² for optical-ionisation to a Helium-like ion (N V). The same transition as for Neon would then produce radiation at 247.6 Å. Figure 5.1.4 shows that the electron temperature produced in Neon at the appropriate intensity is about 40 eV. In fact, the Nitrogen scheme would not require such a high electron density, and is predicted to show a gain of around 30 cm⁻¹ at an electron density of about 10^{19} cm⁻³. This lower density would lessen the IB heating and hopefully reduce the electron temperature to the required level of approximately 10 eV.

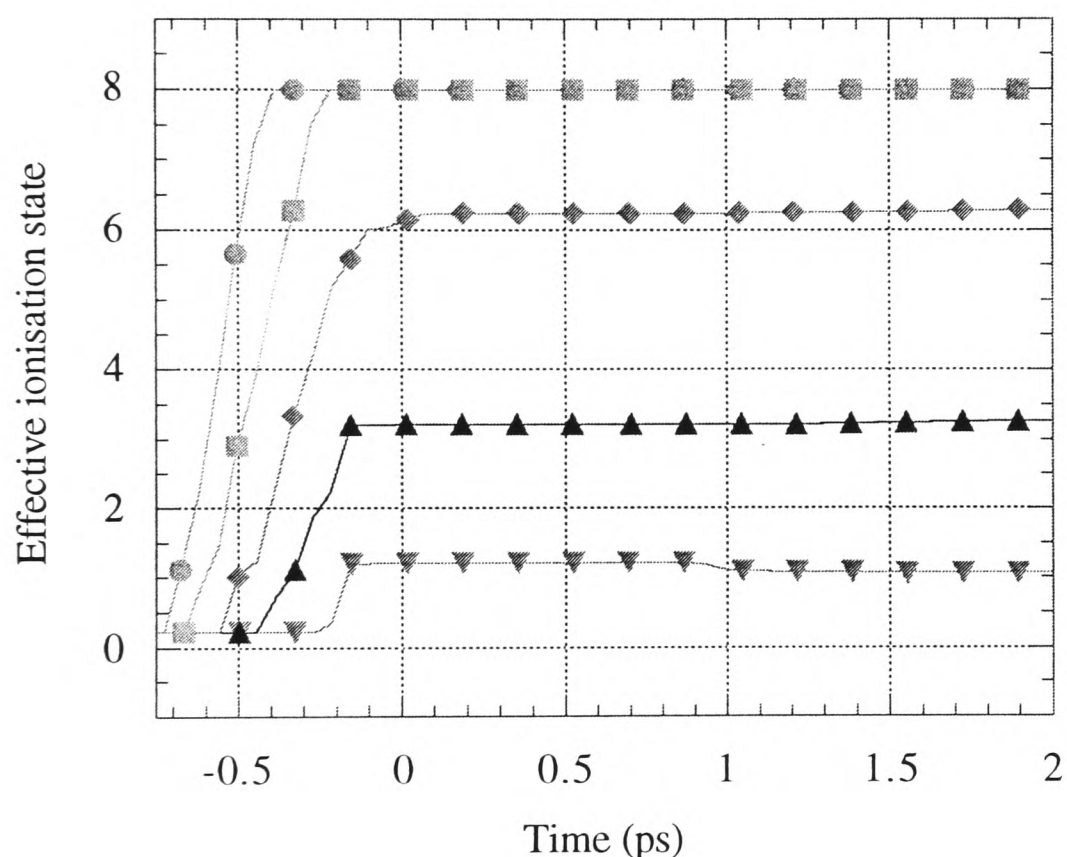


Figure 5.1.5. Variation of the effective ionisation state, Z^* , with time for different laser intensities. The data correspond to intensities of 10^{15} (inverted triangles), 10^{16} (triangles), 10^{17} (diamonds), 10^{18} (squares), 10^{19} (circles) W/cm².

The simulation conditions presented so far indicate that the dominant heating mechanism is inverse bremsstrahlung. This is not surprising as many of the lower ionisation stages of Neon are reached very quickly during the rise of the pulse. The electrons then oscillate in the laser field during the remainder of the pulse and will undergo small angle collisions

with the Neon ions. To reduce the amount of IB heating we must reduce the time these free electrons have to scatter and convert their quiver energy into thermal energy. This can be achieved by shortening the laser pulse length so that Neon is optically-ionised to its Helium-like stage as quickly as possible and then ending the laser pulse before significant electron-ion collisions can occur. Figure 5.1.6 shows how the ATI temperature, the IB temperature, and the total electron temperature vary with the laser pulse length from 30 fs to 10 ps.

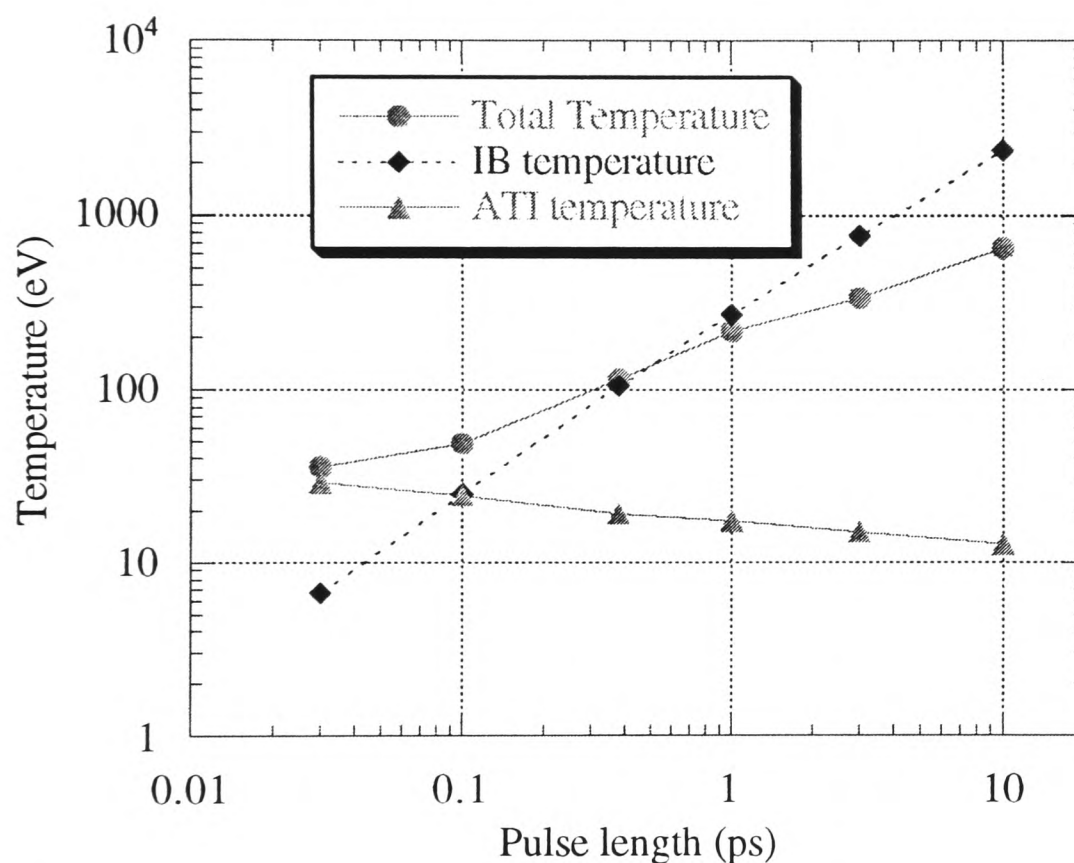


Figure 5.1.6. The dependence of the ATI, IB and total temperatures on the laser pulse length. The temperatures shown are those at the peak of the laser pulse. The neutral Neon density is 1.25×10^{19} atoms/cm³ and the peak laser intensity is 3×10^{17} W/cm².

As expected, the inverse bremsstrahlung heating is greatly reduced if we shorten the laser pulse. We also see that the effect of cooling on the total temperature reduces to a negligible level as we approach 100 fs and below. This is simply because the electrons do not have time to move through the plasma. Unfortunately, some of the reduction in

temperature achieved by lessening the IB heating is offset by the increase in ATI heating. The ATI energy increases from just below 13 eV for 10 ps to almost 30 eV at 30 fs. The increase in the ATI energy is because of the higher intensity required to completely ionise an atom or ion in the shorter laser pulse. This can be seen in the plots of the Ammosov equations (figure 2.2.2). To completely ionise Neon to Ne^+ in a 10 ps laser pulse requires an ionisation rate of about 10^{11} s^{-1} and an intensity of approximately $2 \times 10^{14} \text{ W/cm}^2$. However, complete ionisation of the same atom in 30 fs requires an ionisation rate of over $3 \times 10^{13} \text{ s}^{-1}$ and an intensity of about $6 \times 10^{14} \text{ W/cm}^2$. This creates a fundamental problem for producing gain in the Neon-like scheme, because once the IB heating is brought down to a low enough level then ATI starts to dominate the total electron temperature. However, it does seem that, at pulse lengths of about 100 fs or below, it will be possible to generate conditions that are favourable for the Neon OFI recombination laser scheme to work.

This requirement for such a short pulse length causes an additional problem for our laser pumping scheme. The bandwidth of the KrF laser transition is approximately 5 nm, which imposes minimum pulse length of 80 fs [115]. Therefore, we will have to operate very close to the theoretical bandwidth of the laser to produce a pulse of the appropriate length. This is a difficult task, as the higher the gain in KrF, the more gain narrowing occurs. This effect is exacerbated by the presence of an absorption dip in the KrF gain spectrum which is attributed to internal absorption to an excited molecular state which dissociates to $\text{Kr}^*(^3\text{P}_1)$ and $\text{F}(^2\text{P}_{3/2})$ [116]. The result of this gain narrowing and absorption is a reduction in the bandwidth of the amplified pulse and a consequent increase in the laser pulse length after recompression. These effects can be overcome by introducing appropriate spectral filters into the laser amplifier train, but this leads to a reduction in overall efficiency of the laser.

A final laser parameter that can be altered in an attempt to lower the electron temperature is the focal spot diameter. As shown in the previous figures the electron temperatures can be significantly reduced from the sum of the ATI and IB temperatures by cooling via heat conduction. Obviously a smaller focal spot will create a much larger radial temperature gradient and so a much higher cooling rate. The simulations presented so far have used a spot size that models as closely as possible that of our experimentally measured focus. The FWHM of our focus is $10\text{ }\mu\text{m}$, but the diffraction limit for $f/3$ focusing of 248 nm light produces a width (first minimum to first minimum) of $1.8\text{ }\mu\text{m}$. In figure 5.1.7 we set our FWHM to this value to compare with the non-diffraction limited simulation shown in figure 5.1.2.

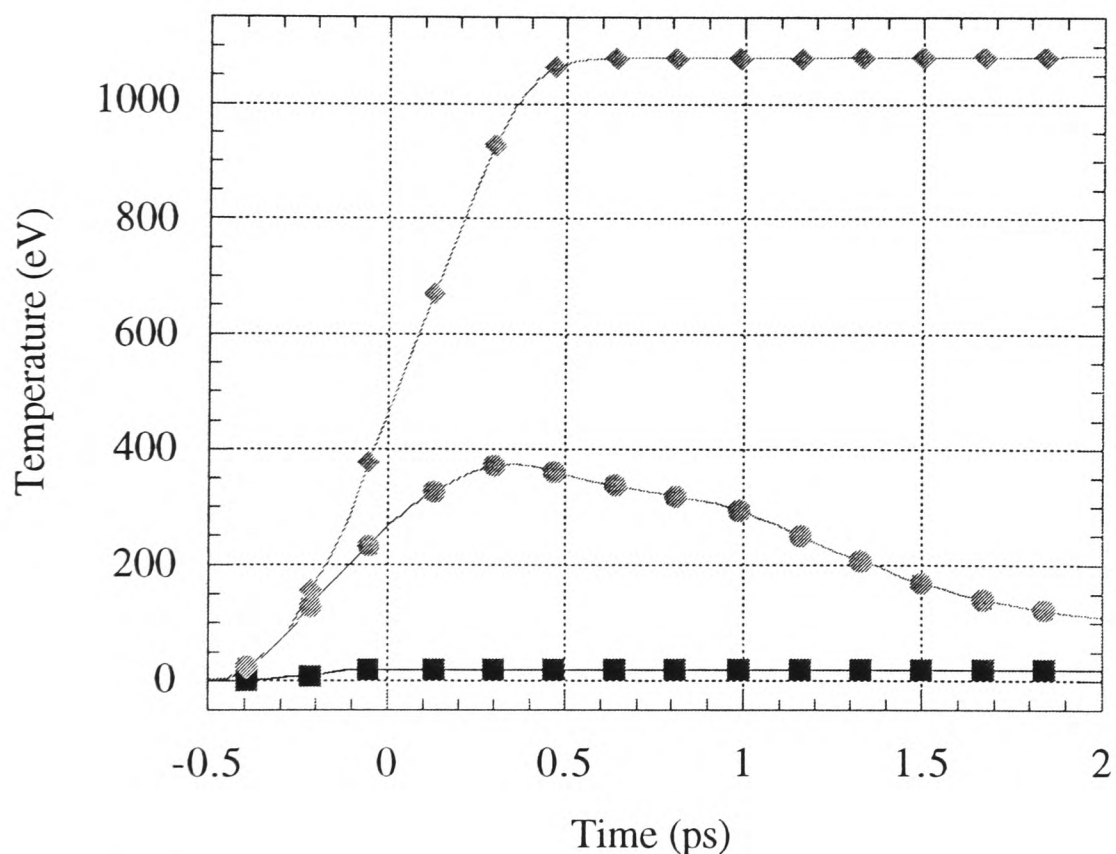


Figure 5.1.7. The variation of the total electron temperature over the laser pulse for a focal spot with a FWHM of $1.8\text{ }\mu\text{m}$ and an electron density of $5 \times 10^{20}\text{ cm}^{-3}$.

A comparison of figure 5.1.2 and 5.1.7 shows that, because of the reduced focal spot size, the electron temperature 500 fs after the peak of the laser pulse is reduced from 550 eV to 350 eV. This increased cooling, although significant, is not high enough to

provide the low temperatures we require without also reducing the intensity, density, or pulse length..

5.2. Fitting Thomson spectra to theory.

Having determined some of the expected levels of electron heating in the above section and having briefly discussed the physics of Thomson scattering in section 4.4, we must now consider how accurate Thomson scattering will be at actually determining the electron temperature and density in the laser focus.

5.2.1. Validity of the Thomson scattering data.

Thomson scattering can be a very powerful plasma diagnostic. However, there are some problems in using this technique with very short laser pulses when the thermalisation of the plasma occurs on a time scale longer than the laser pulse. The theory of Thomson scattering assumes a Maxwellian distribution of electron and ion temperatures, but our data comes from light scattered from a rapidly evolving plasma which has not yet reached thermal equilibrium. When imaging the laser focal spot we have no choice but to integrate light scattered at all times during the 380 fs laser pulse. We will also have to image a finite region of the focus, the size of which is determined by the width of our spectrometer slits. For our experiments the 90° Thomson scattering channel had a typical magnification of 10x and the slits were typically 100 μm . This means that we will be imaging a radial section of the plasma that is at least 10 μm wide and integrated along the line of sight of the lens, with a reduction in light levels collected away from the centre of the focus governed by the depth of field of the lens (i.e. the light is defocused on the spectrometer slits). Figure 5.2.1 shows a simplified schematic diagram of the integration of Thomson scattered light from the laser focus onto the spectrometer slits.

Let us now take the worst case scenario and assume that our spectrometer samples Thomson scattered light from the whole focal volume. Figure 5.2.2 shows a MEDUSA simulation of the variation of the α parameter across the laser focus at the peak of the laser pulse. The $1/e$ width of the focus is $6\ \mu\text{m}$ (FWHM = $10\ \mu\text{m}$), the laser pulse length is 380 fs, and the gas is Neon at an atomic density of $1.25 \times 10^{19}\ \text{cm}^{-3}$. Also shown as a dotted line in the same graph is the product of the laser intensity, the electron density and the cell area (given by $2\pi r\Delta r$ where r is the radial position of the cell and Δr is the cell width). This gives us a product of the number of scatterers and intensity in a given areal element (the N.I product) expressed as W/cm^3 . Multiplying this by the Thomson scattering cross section and by the length over which the laser interacts gives us the total power Thomson scattered into 4π steradians, $P = \sigma NVI$ where V is the volume of the laser focus.

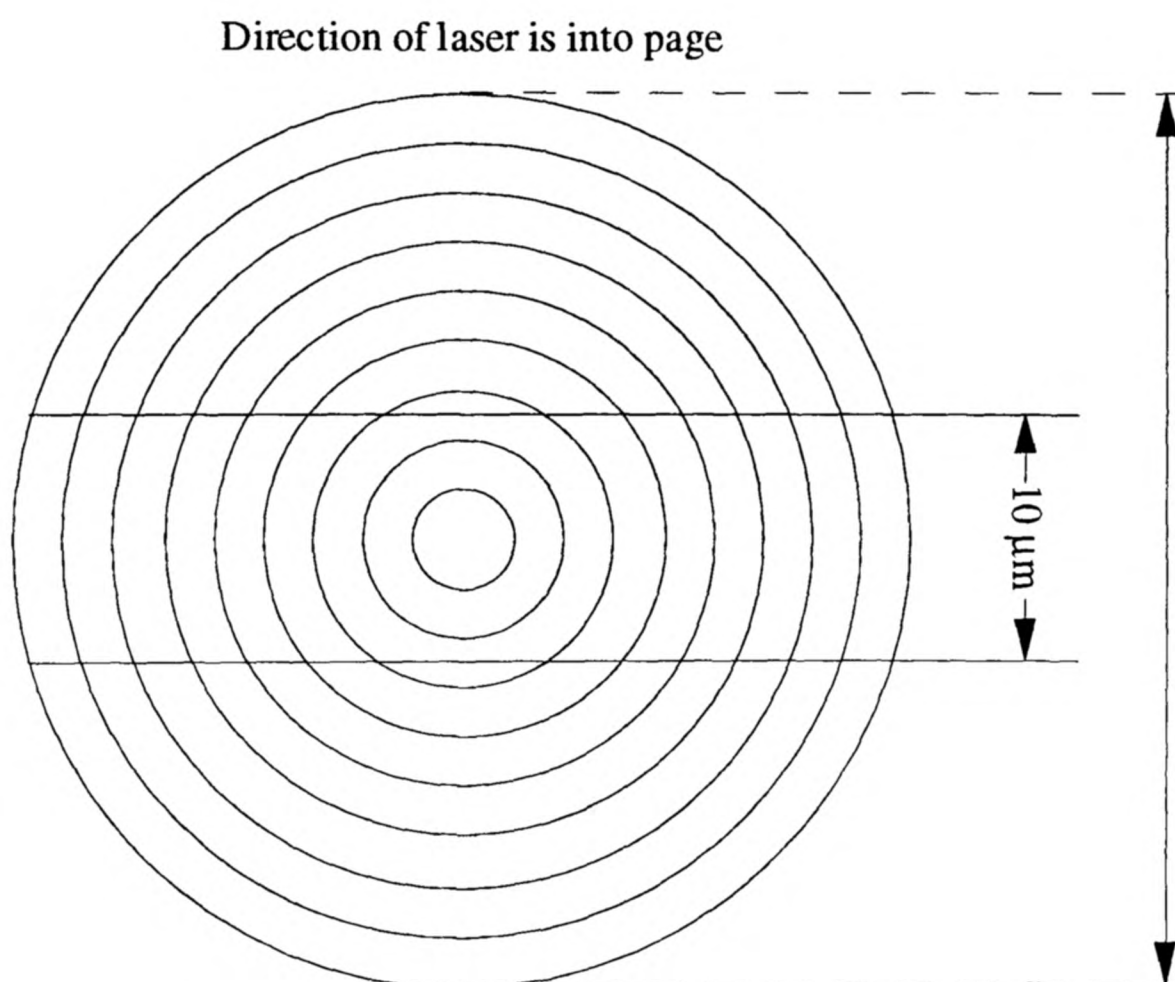


Figure 5.2.1. Spatial integration of scattered light from the laser focus. The concentric rings represent the cells of the MEDUSA simulation. The solid lines indicate the best case imaging when the slits cover $10\ \mu\text{m}$ of the focus, the dotted lines indicate the worst case when the whole focus is imaged.

Most of the power scattered instantaneously at the peak of the laser pulse comes from a radius of 5 - 6 μm from the laser centre. Even with finite slits our experiment would have preferentially measured this region, because the effective slit width at the source was 10 μm . Since the electron density and temperature profiles are relatively flat from the axis to this radius, the mean α parameter we would deduce from our Thomson scattered signal is close to that on axis. In this case $\alpha = 3.5$ on axis and 3.84 at a radius of 5 μm , a difference of only 10% which is small compared the error in fitting a theoretical Thomson scattered spectrum to the experimental data.

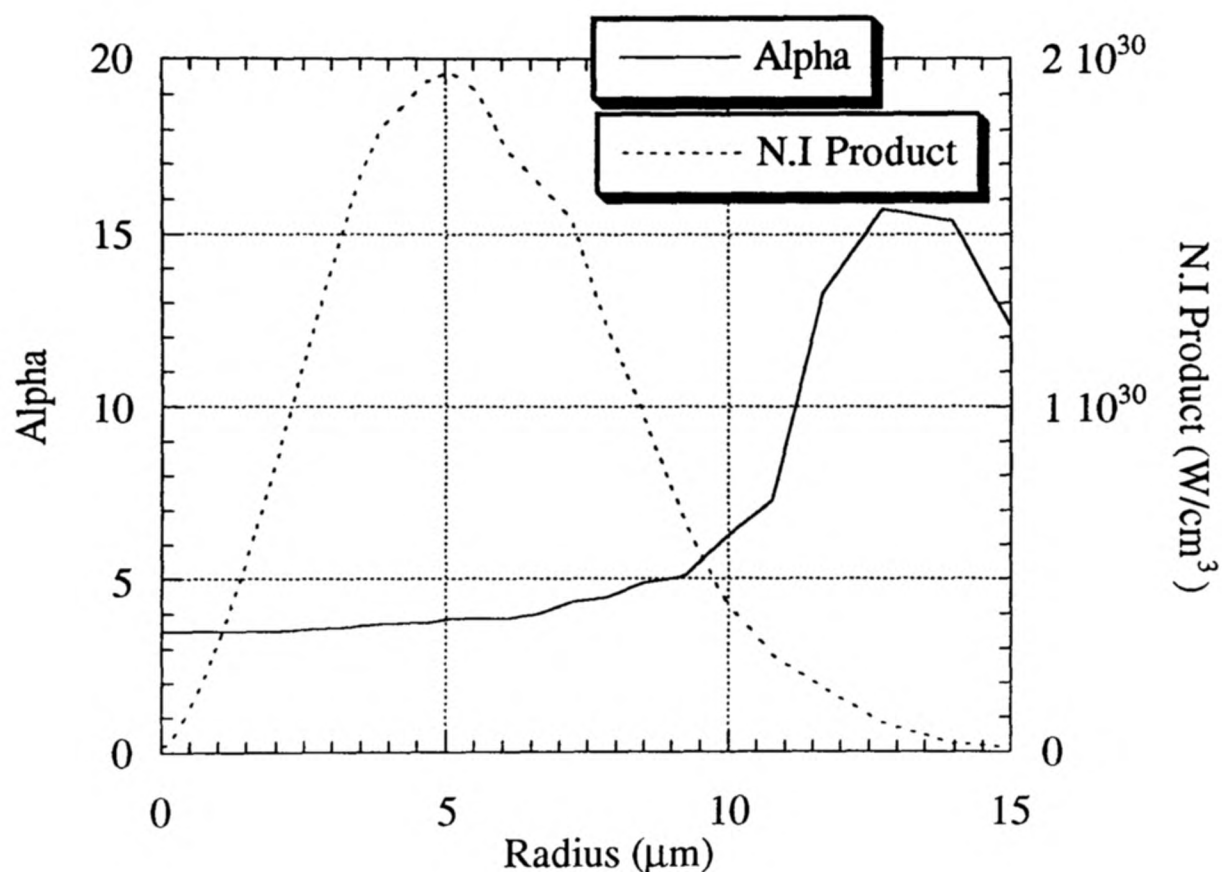


Figure 5.2.2. Plot of the variation of α with radius (solid line). Also shown is the product of the electron number and the laser intensity (dotted line). The simulation is for a peak laser intensity of $3 \times 10^{17} \text{ W}/\text{cm}^2$ with a $1/e$ width of 6 μm and an atomic density of $1.25 \times 10^{19} \text{ cm}^{-3}$ of Neon.

Now let us consider the effect of the time integration of the Thomson scattered signal. Figure 5.2.3 shows how α varies on axis with time during the laser pulse. We see that this time the N.I product is largest at the laser peak, but in a 500 fs time about the peak α

varies from 5.8 to 2.7 with a value of 3.5 at the peak. This is a variation of 66% above and 23% below the most favoured value.

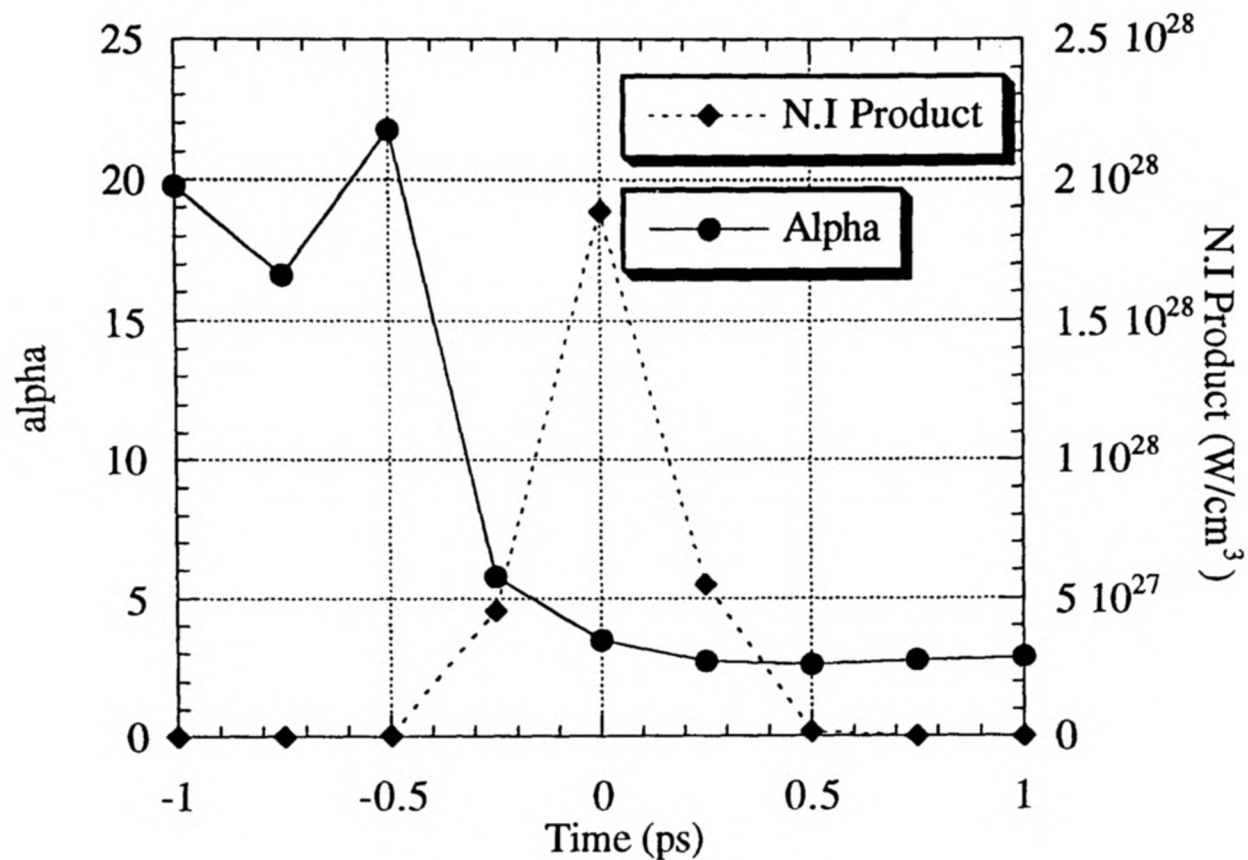


Figure 5.2.3. Variation of α with time (solid line) through the laser pulse. The $N.I$ product is shown as a dotted line. Data points are depicted by symbols.

We now combine the radial and temporal variations in α by taking a weighted mean defined as

$$\bar{\alpha} = \frac{\sum_{r,t} \alpha N I \Delta A}{\sum_{r,t} N I \Delta A} \quad (5.2.1)$$

where N is the electron density, I is the laser intensity, and ΔA is the area of the cell at radius r and time t . The value of α integrated over all time and space in the focus is 4.4 compared to a value of 3.5 at the peak of the pulse and in the centre of the focus, a difference of 26%. Since α is proportional to $(N/T_e)^{1/2}$ the error in determining the

electron density and temperature at the centre of the laser focus will be twice the error on α . However, interpretation of the experimental results will still give us a rough measure of the electron temperature and will certainly give an indication of how the electron temperature scales with electron density and laser intensity.

5.2.2. Determination of the electron temperature and density from Thomson scattering data.

The equations governing the form and scale of Thomson scattered light from a plasma were discussed in section 4.4. The important conclusions were that the form of the Thomson scattered spectrum at a given angle is determined solely by the α parameter and the scaling of the signal with frequency is determined by the electron temperature (and, to a lesser extent, the ion temperature).

To model the spectra observed in the experiments we have developed a simple program that computes the Thomson scattering form factor as a function of α , T_e and T_i , and convolves it with a known instrument function to simulate the experimental spectra [137]. In the case of Thomson scattering from static fills of gas, we know the atomic density exactly. Given that we also know the laser intensity and the ionisation rates of the atom, we can easily calculate the electron density of the plasma. In our experiments performed to date the observed frequency spread of the 'ion response' close to the laser wavelength is dominated by the instrument width of our spectrometer and imaging system and so we cannot resolve the ion resonance. The 'electron response' shifted from the laser frequency by the Bohm-Gross frequency is also instrument broadened, but this resonance is over a large enough frequency range that it can be resolved. The ion and electron resonance terms are shown graphically in figure 4.4.4. As a first measure we can determine α from the ratio of the energy scattered into the central ion feature compared to the energy scattered into the whole Thomson spectrum. In the limit of

$T_e \gg T_i$ this ratio is $\alpha^2/(1+\alpha^2)$ and so the ratio of scattering into the ion feature to the scattering into the electron feature goes as α^2 . Hence as we move to higher α the ion response dominates over the electron response.

From this initial estimate of α we may obtain a good fit to the form of the spectrum by varying α about this value in the simulation program. Once this has been done the frequency scale of the spectrum can be found by altering T_e until the simulated spectrum matches experiment. Once α and T_e have been fitted we may calculate N which we can check against the known electron density in the case of a static gas fill. When we use a gas jet we must trust that our inferred electron density is correct. If we take Thomson spectra at two different scattering angles we may compare the inferred electron density from each to check for consistency. A difference in electron temperature may be inferred from the two scattering directions. This does not necessarily mean that the fitting of the Thomson spectrum is inaccurate, it could point to an anisotropic electron temperature distribution caused by the optical-ionisation process or by Landau damping of plasma waves produced by SRS.

The method of analysing the Thomson scattered spectra described above is good for a simple gas, such as Helium, which is fully ionised at relatively low intensities. For a gas such as Neon, however, which has multiple ionisation stages that exist over a wide range of intensities and which produce electrons which greatly different ATI temperatures, the Thomson scattered spectrum becomes more complicated. The observed spectrum consists of a central ion feature like the simple Thomson spectrum, but the electron wings are replaced by shoulders created from a superposition of several Thomson spectra with several different values of α [29]. For a spectrum like this we can determine an upper bound on the electron temperature by fitting an $\alpha = 0$ (Gaussian) spectrum to the electron wings.

We have already shown in section 5.2.1 that, if we integrate Thomson scattered light from the entire focus, the electron temperature and density we infer will have an error of the order of 50%. This uncertainty is compounded by the form of the Thomson scattered spectrum that we see experimentally. Figure 5.2.4 shows lineouts of Thomson scattering data taken at 150° from static fills of Helium at 5 and 100 torr. In both spectra the red shifted wing is enhanced relative to the blue shifted wing. (The effect is more pronounced in the 100 torr data). There are several possible reasons for this phenomenon, including:

1. The detector response may fall off on the short wavelength side of the laser wavelength. This is unlikely, as the enhancement of the 5 torr spectrum occurs within two or three nanometres of the 248 nm laser wavelength and the enhancement of the 100 torr signal is only at a 6 nm shift.
2. Thomson scattering can be viewed as the decay of a photon into a photon and a plasmon to cause red shifting (loss of energy from the radiation field), and the absorption of a plasmon by a photon to cause blue shifting (gain in energy by the radiation field). We have already mentioned that the plasma is still equilibrating during the entire laser pulse, so there is perhaps a net increase in the number of plasmons during this time, i.e. there is more red shifted scattering. This would be a modification to the Thomson scattering form factor from that we derived in section 4.4.
3. This could be a sign of the onset of SRS.

This asymmetry to the Thomson spectra causes a problem when we try to fit a theoretical spectrum (which is symmetric) to our experimental data. Do we fit to the red wing or the blue wing? If the red wing is enhanced by SRS then the value of α we infer will be much too large, which will lead to a consequent error in the electron density and temperature.

If the blue wing is suppressed due to lack of plasmons then this will lead us to a value of α that is too low.

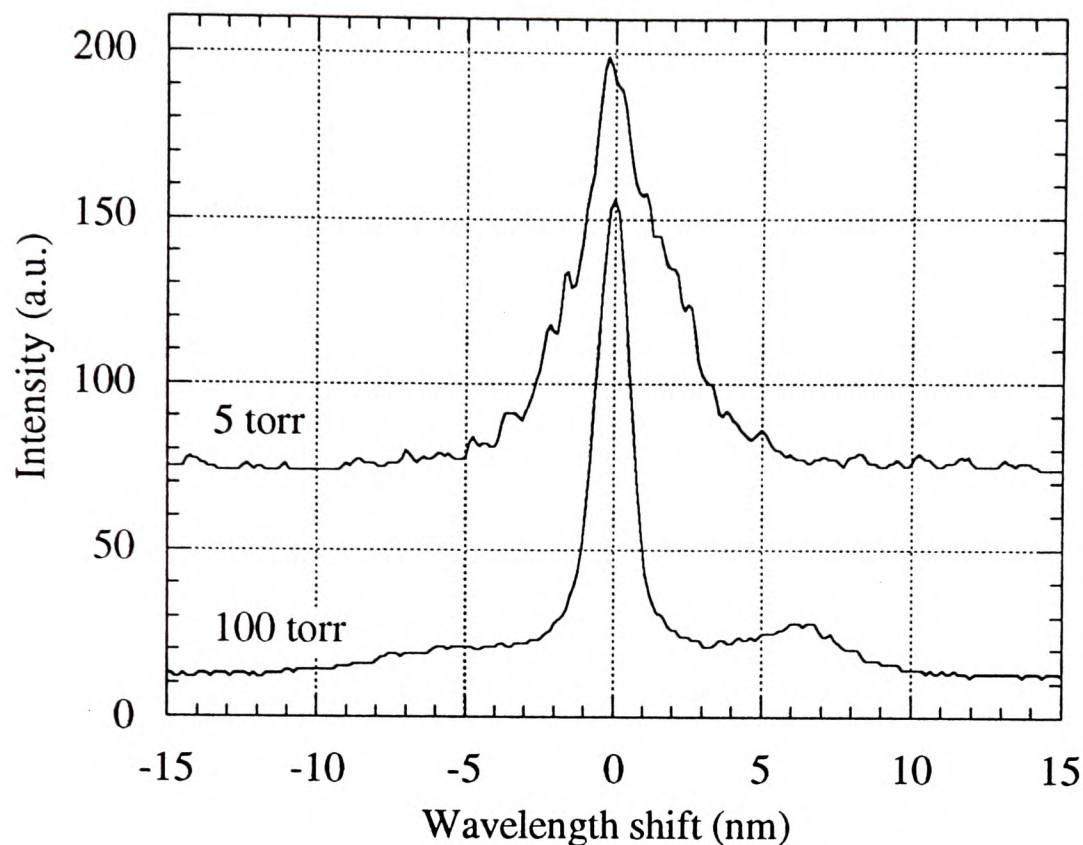


Figure 5.2.4. Comparison of Thomson scattering at 150° from Helium at 5 torr and 100 torr. Note the enhanced red shifted wings in the 100 torr spectrum.

As Helium only has two ionisation stages, and since ponderomotive expulsion of electrons from the focus is negligible in our experiments, we can assume that the electron density of the plasma is twice the neutral density of the static gas fill in the vacuum chamber. If our Thomson scattering data gives us an electron density that differs greatly from this density then we know that it is not valid. We will analyse the static fill data in the following ways. We will fit a theoretical spectrum to the red shifted and to the blue shifted electron wings which will give us values for α , N , and T_e . We will also subtract the blue shifted wing from the red shifted wing, which will reveal the peak of the enhancement. If we compare this shift to the Bohm-Gross frequency with an electron plasma frequency, ω_{pe} , calculated from the known static fill density, we can deduce an electron temperature. This latter method was used in a preliminary analysis of this data

by W.J. Blyth in his thesis [29] and some of that data will be reproduced in this chapter. We will then use the three methods to set limits on the possible temperature scaling with electron density.

5.2.3. Determination of the electron temperature and density in the absence of Thomson scattering data.

At low electron densities, analysis of the Thomson scattered spectrum gives us a value for the α parameter for the plasma from which we can determine N and T_e , or, alternatively, we may deduce a temperature from the Bohm-Gross frequency of a plasma with a known density. At higher electron densities produced from the solenoid-valve high pressure gas jet we find that the Thomson scattered signal is swamped by plasma emission. At the lower end of this density range there is no scattered signal visible above the plasma emission. However, a very strong SRS signal is seen at both 90° and 150° at higher densities.

In this situation we do not know the absolute electron density from which the laser scatters, because the gas profile created by the gas jet is non-uniform and we do not know where in this profile the SRS will be strongest. We cannot model the SRS spectrum in the same manner as the Thomson scattered signal, as its form and magnitude are very sensitive to the history of the laser propagation through the plasma. This sensitivity comes from the fact that the SRS signal grows exponentially in time and/or space. However, the dispersion relation for the plasmons produced by SRS is the same as for Thomson scattering and is given by the Bohm-Gross formula

$$\omega^2 = \omega_{pe}^2 + 3k^2 \left(\frac{\kappa T_e}{m} \right) \quad (5.2.2)$$

where ω_{pe} is the plasma frequency, ω and k are the frequency and wavevector for the electrostatic plasma wave. If we assume that stimulated scattering in both directions comes from the same region of the plasma, we can calculate the temperature from the frequency shifts as follows :

$$90^\circ: \quad \omega_1^2 = \omega_{pe}^2 + 3\left(\frac{\kappa T_e}{m}\right)(k_{in}^2 + k_{90}^2) \quad (5.2.3)$$

$$150^\circ: \quad \omega_2^2 = \omega_{pe}^2 + 3\left(\frac{\kappa T_e}{m}\right)(k_{in}^2 + k_{150}^2 + 2k_{in}k_{150}\cos 30^\circ) \quad (5.2.4)$$

where k_{in} is the incident wavevector and k_{90} , k_{150} are the scattered light wavevectors. The geometric relations that give these are shown in figure 5.2.5. Subtracting equation 5.2.4 from equation 5.2.3 gives us the relation

$$\omega_1^2 - \omega_2^2 = 3\left(\frac{\kappa T_e}{m}\right)(k_{90}^2 - k_{150}^2 - \sqrt{3}k_{in}k_{150}) \quad (5.2.5)$$

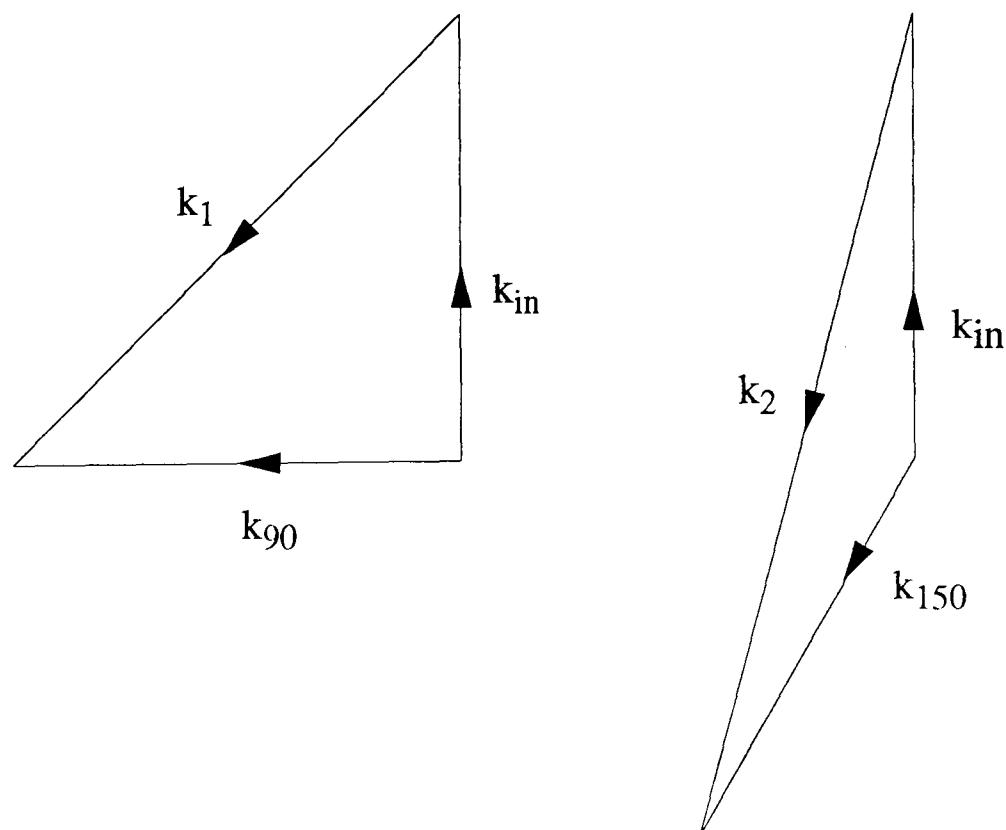


Figure 5.2.5. Addition of wavevectors to show conservation of momentum in the decay of a light wave into a light wave and an electron plasma wave.

This gives us an expression for the thermal energy of the scattering electrons

$$\kappa T_e = \frac{m(\omega_1^2 - \omega_2^2)}{3(k_{90}^2 - k_{150}^2 - \sqrt{3}k_{in}k_{150})} \quad (5.2.6)$$

We can then substitute the thermal energy back into equations 5.2.3 or 5.2.4 to derive the plasma frequency and hence the electron density in the plasma.

$$N = \frac{m\epsilon_0}{e^2} \left(\omega_2 - 3 \left(\frac{\kappa T_e}{m} \right) k^2 \right) \quad (5.2.7)$$

This technique will give us a measure of the electron temperature and density of the plasma, provided that our assumption that both scattering signals come from the same region (and time) in the plasma is correct. A consideration of the SRS spectra at both 90° and 150°, discussed in section 5.4.1, indicates that the 150° SRS signal has a very large bandwidth which is probably caused by filamentation of the laser beam and its subsequent propagation through different parts of the plasma. This introduces large errors in determining values for the electron density and temperature determined by this method. However, we can verify the magnitudes of these electron temperatures by considering levels of SRS scattering (section 5.4.2), Faraday cup measurements and X-ray spectra recorded on an X-ray mini-spectrometer (section 5.5).

5.3. Thomson scattering measurements.

Thomson scattering was collected at both 90° and 150° to the direction of propagation of the laser. A description of the lens set up and the detector configuration used for this is given in chapter four. Scattering was detected from static fills down to 5 torr of Helium on the 150° channel. Levels of scattering on the 90° channel were below the threshold of the streak camera detector until pressures reached about 200 torr (of Helium). For this

reason, we will not review the Thomson scattered spectrum at 90° . However, we will consider scattering in this direction when we consider SRS in section 5.4.

5.3.1. Helium.

Figure 5.3.1 shows images of Thomson scattering at 150° from various densities of Helium. We can clearly see the progression from an incoherent scattering spectrum ($\alpha \ll 1$) at 5 torr to a coherent scattering ($\alpha > 1$) spectrum above 100 torr. In all the images the red shifted wing is to the right and is obviously enhanced over the blue shifted wing. The contrast of the 50 and 100 torr images has been enhanced to make the electron wings visible, which causes the central 'ion' feature to appear saturated.

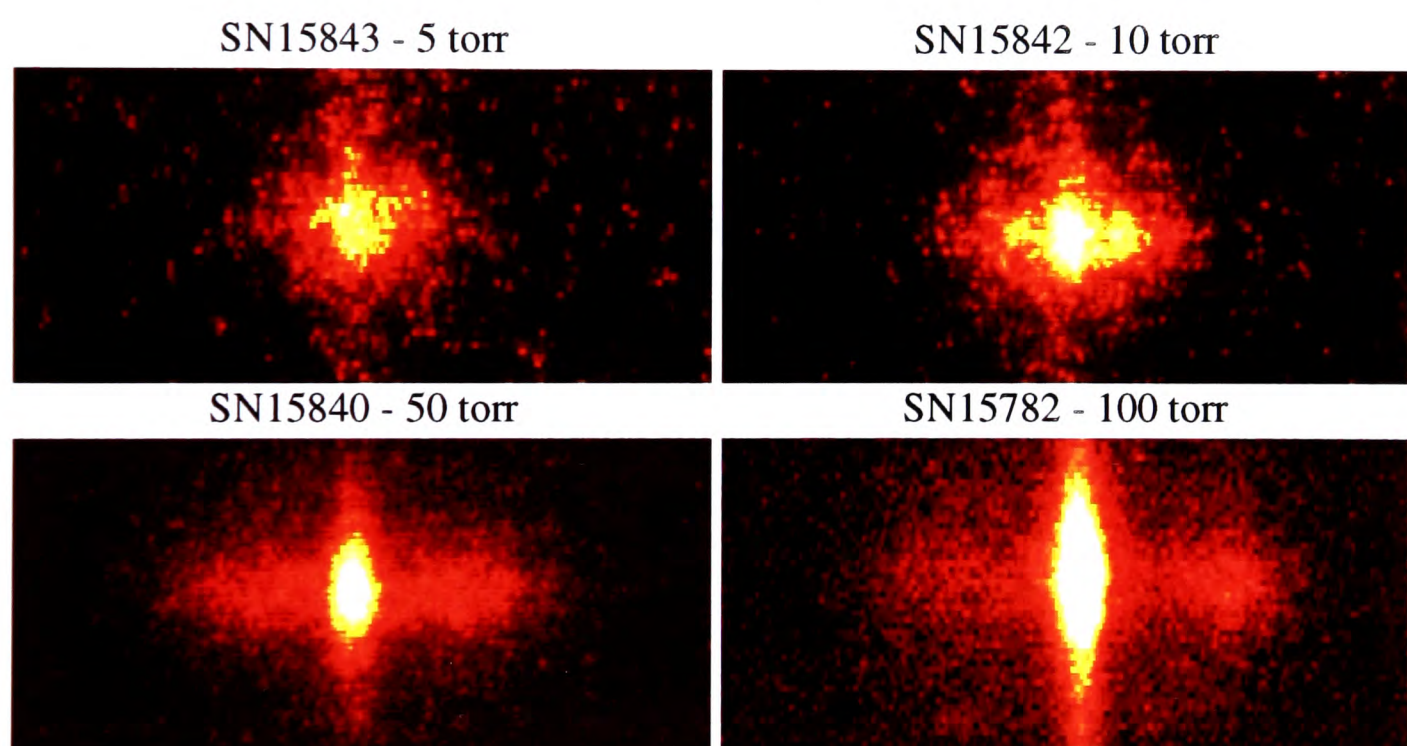


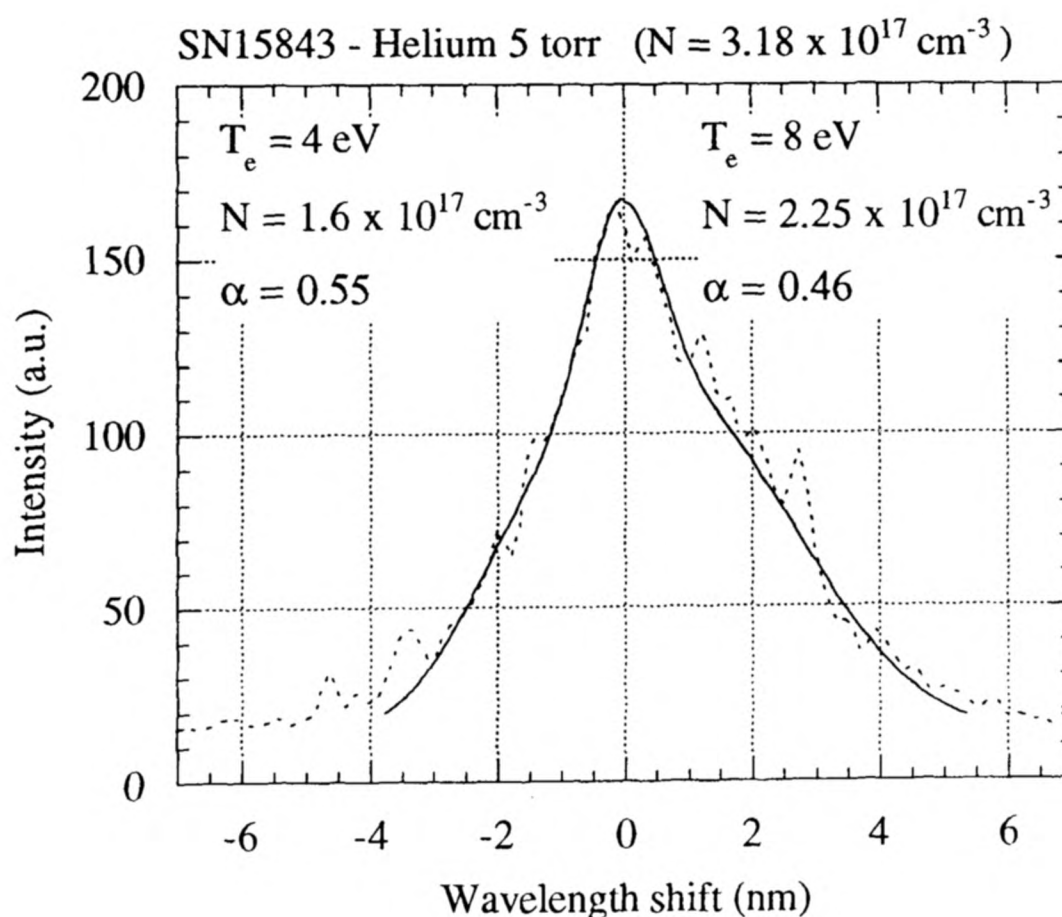
Figure 5.3.1. Thomson scattering at 150° from Helium at static fill pressures of 5, 10, 50 and 100 torr. Note the progression from a Gaussian distribution at low pressure to a coherent scattering spectrum at higher pressures.

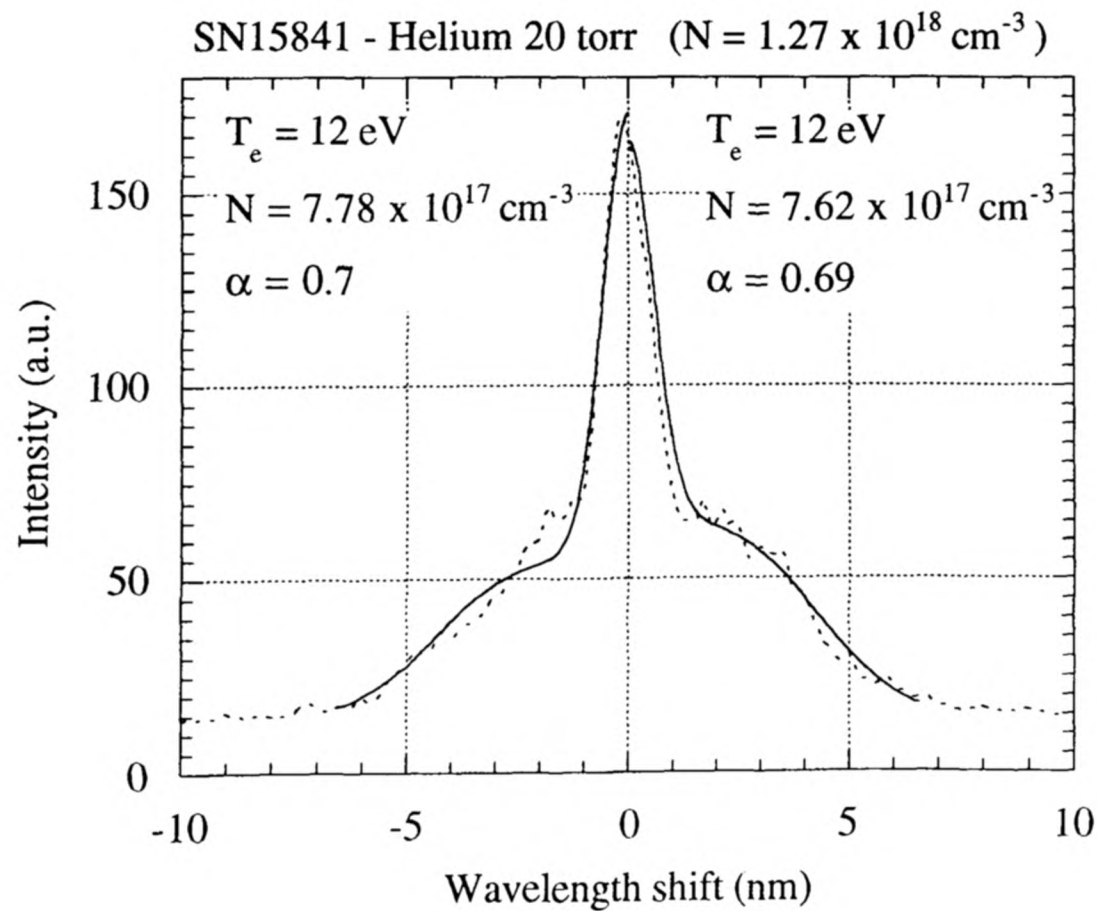
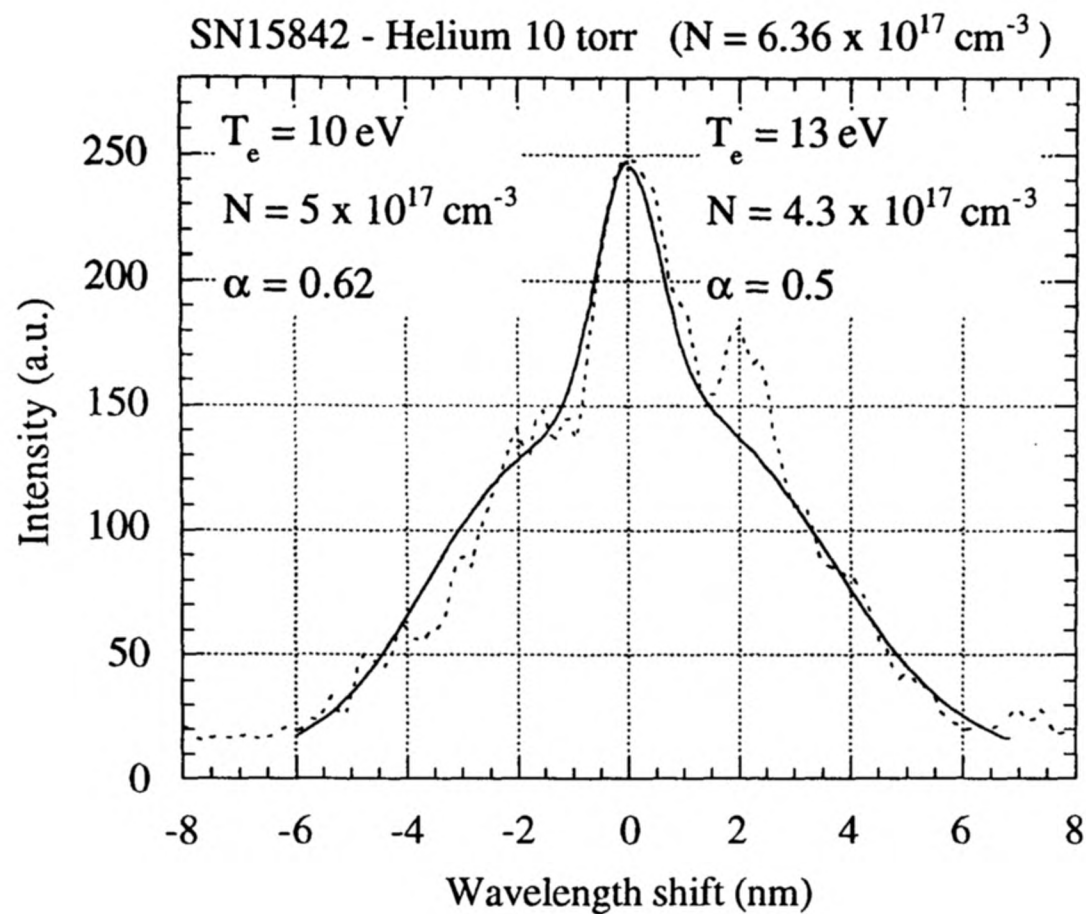
5.3.1.1. Fitting the blue and red shifted electron wings.

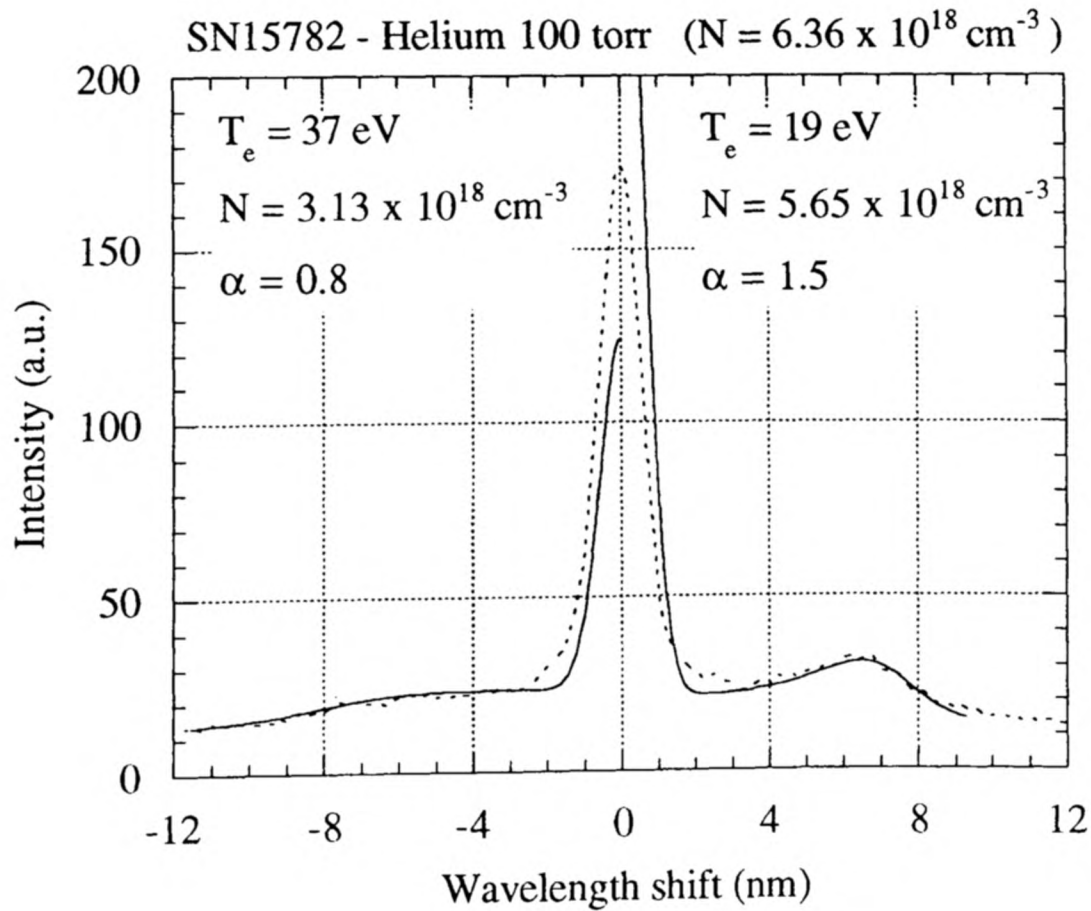
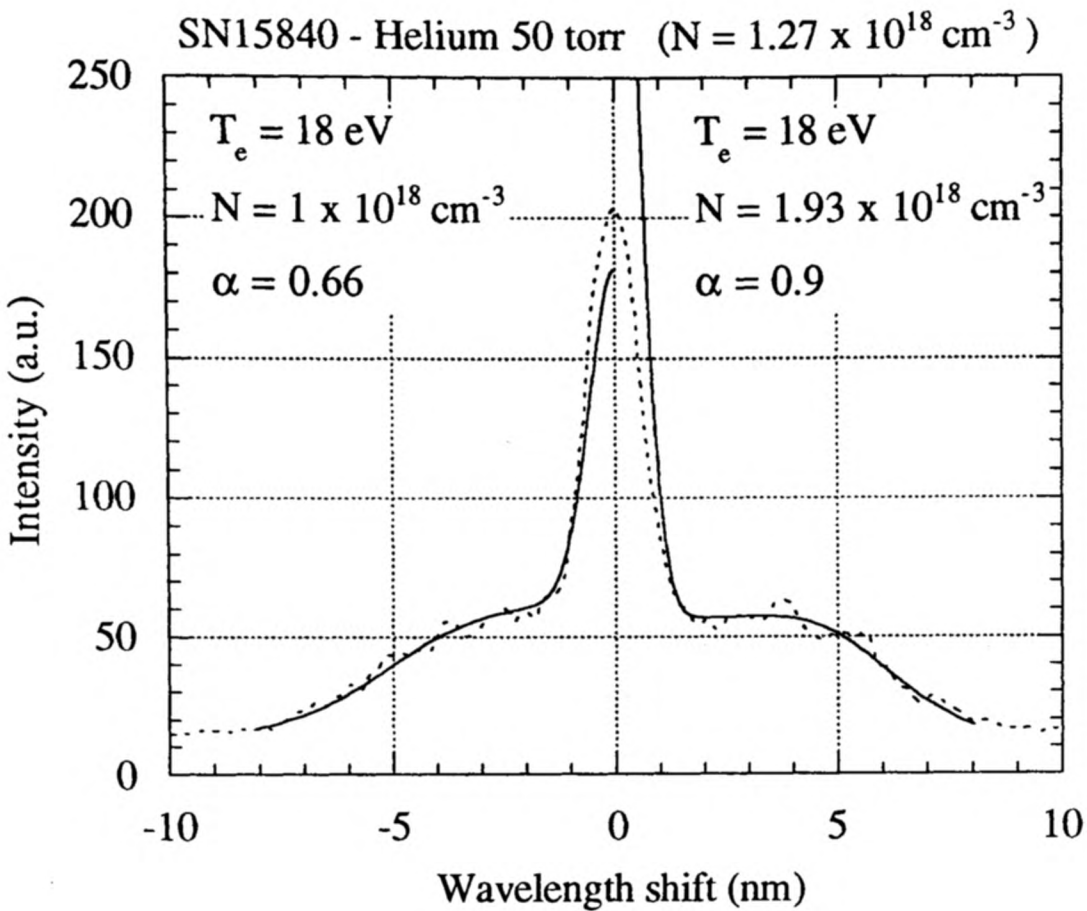
Let us first of all attempt to fit the form of the Thomson scattering spectrum on the blue and red side of the spectrum. In figure 5.3.2 we show lineouts of Thomson scattered

spectra from Helium at densities of 5, 10, 20, 50, 100, 200 and 400 torr. The red and blue side of each spectrum is fitted independently. The parameters of each fit are shown in the figures, with the parameters for the blue wing on the left and those for the red wing on the right. The actual density produced by doubly-ionised Helium at the known static fill pressures is given in the title of each graph.

It should be noted that, due to the structure of the Sprite focal spot, it is possible that some regions of the focus may experience incomplete ionisation for some gas targets (such as Neon, for example). However, the intensity required for complete ionisation of Helium is 100 times less than the peak intensity of our laser focus. This, combined with the fact that the levels of Thomson scattering are proportional to the laser intensity, leads us to ignore such wing effects when determining the electron density from which scattering occurs.







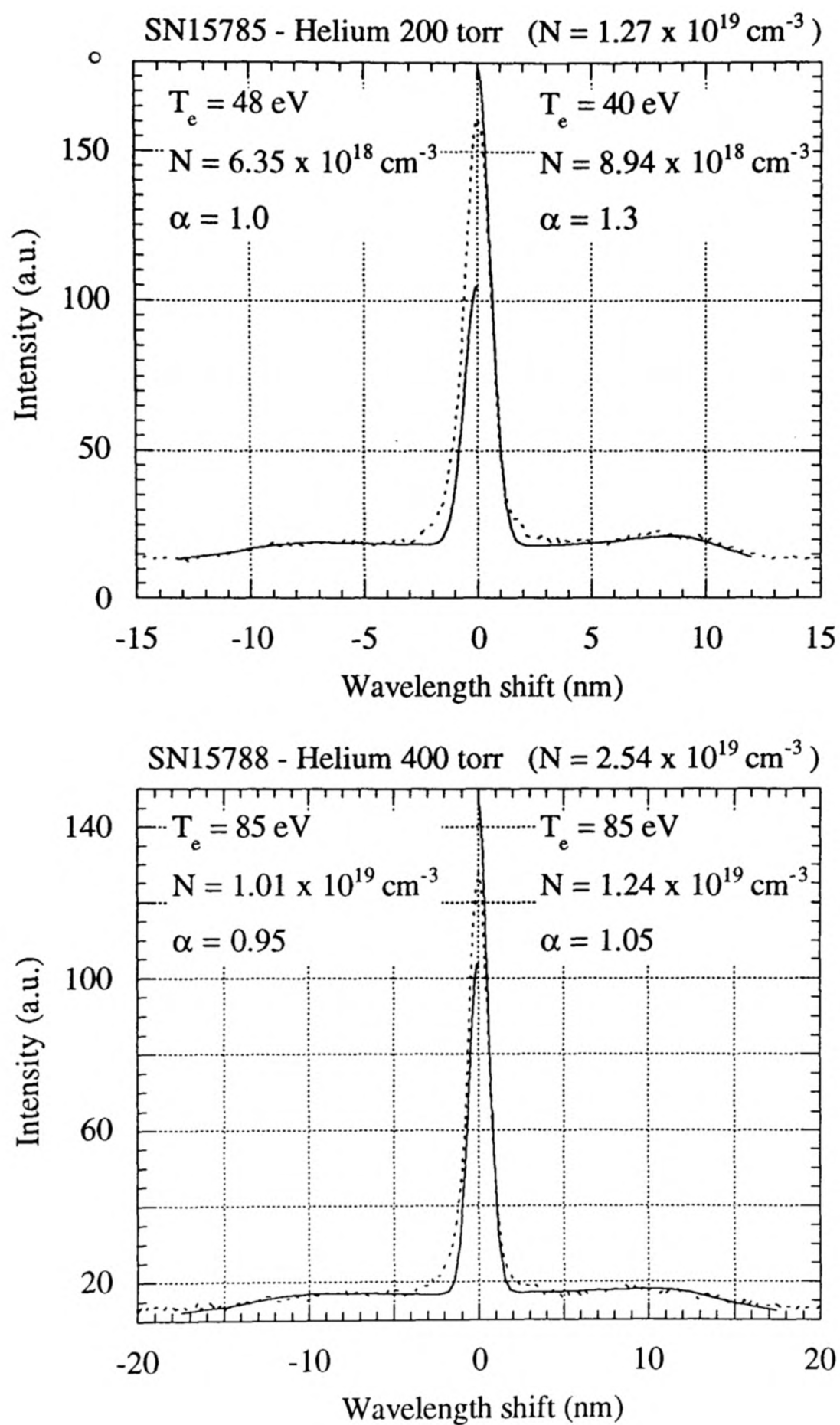
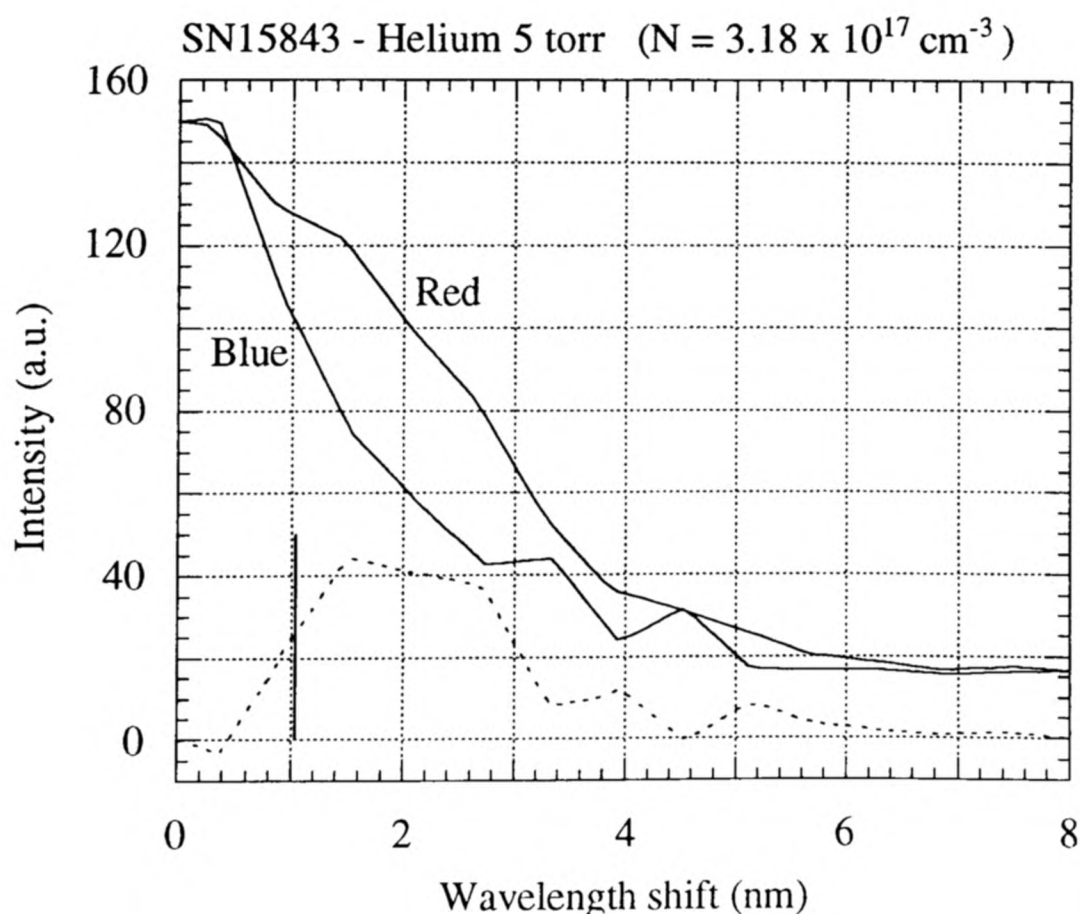


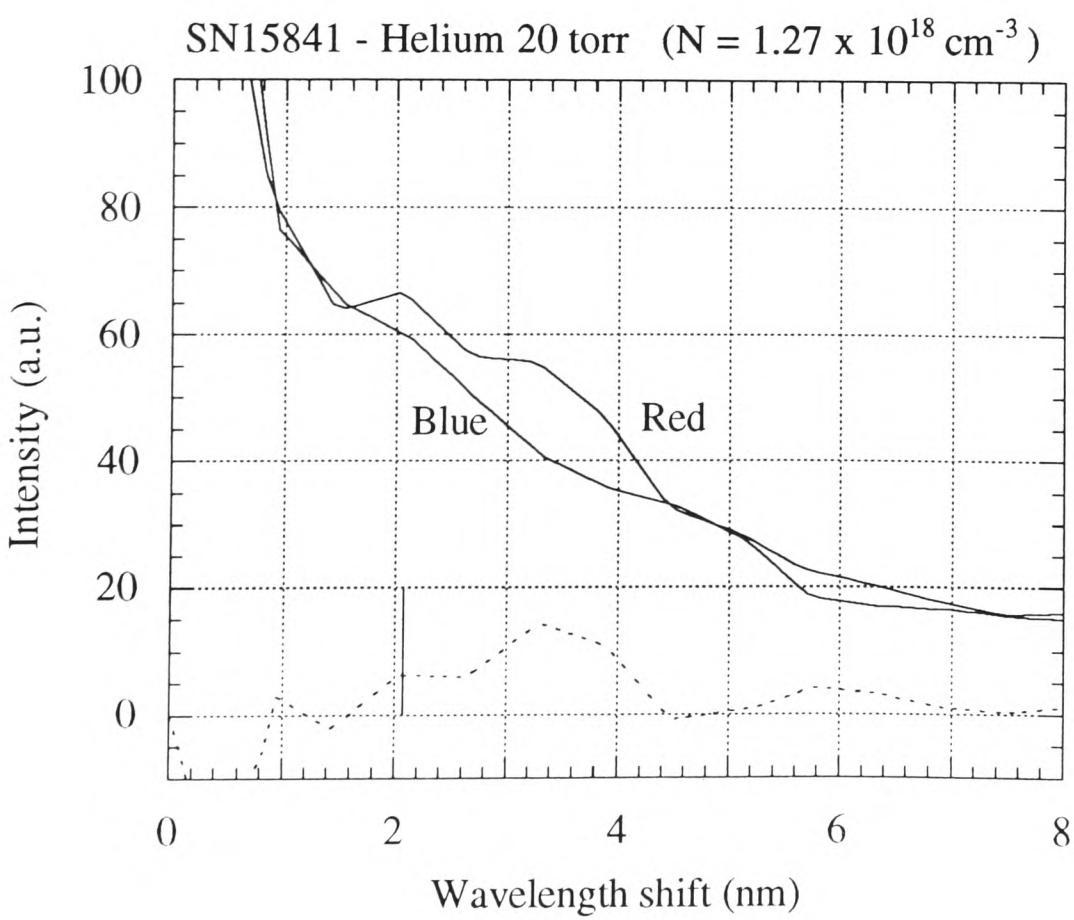
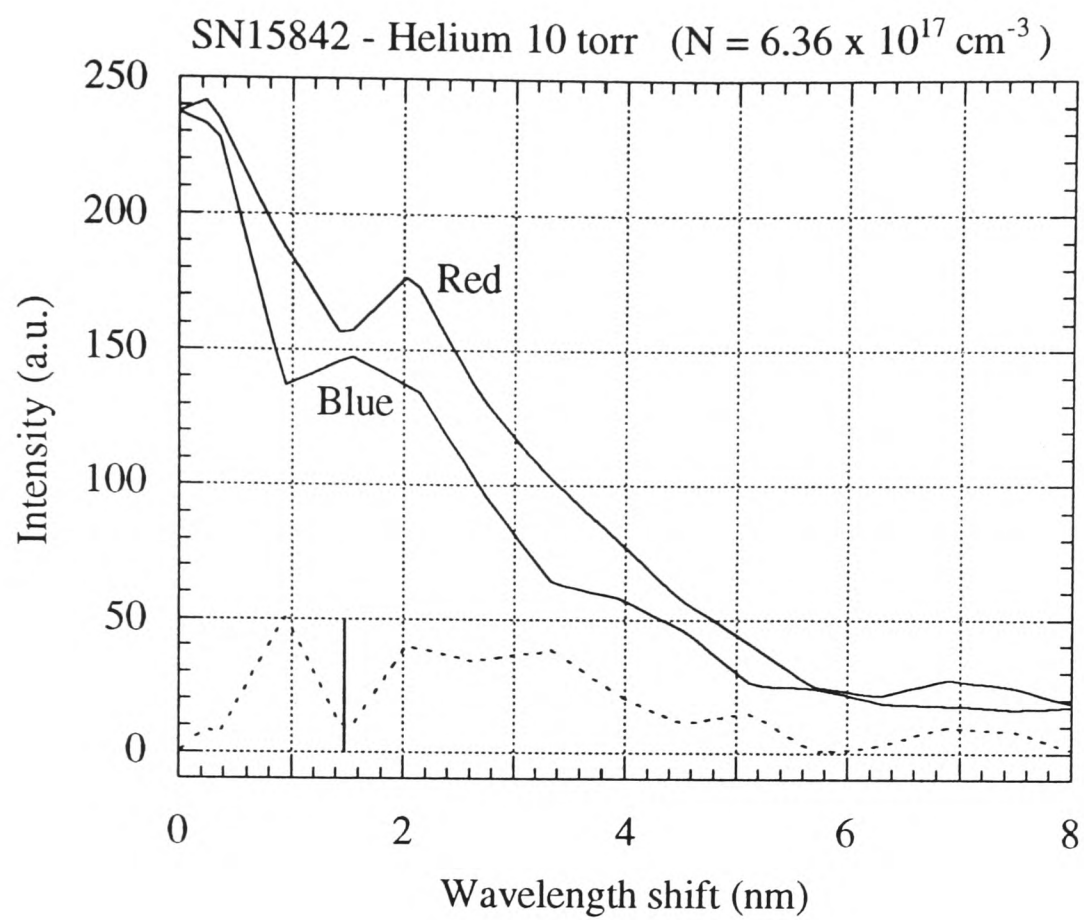
Figure 5.3.2. Lineouts of 150° Thomson scattering from static fill Helium at pressures of 5, 10, 20, 50, 100, 200 and 400 torr. Independent fits to the red and blue side of each spectrum are shown and their parameters listed. The density of doubly-ionised Helium at these pressures is also shown.

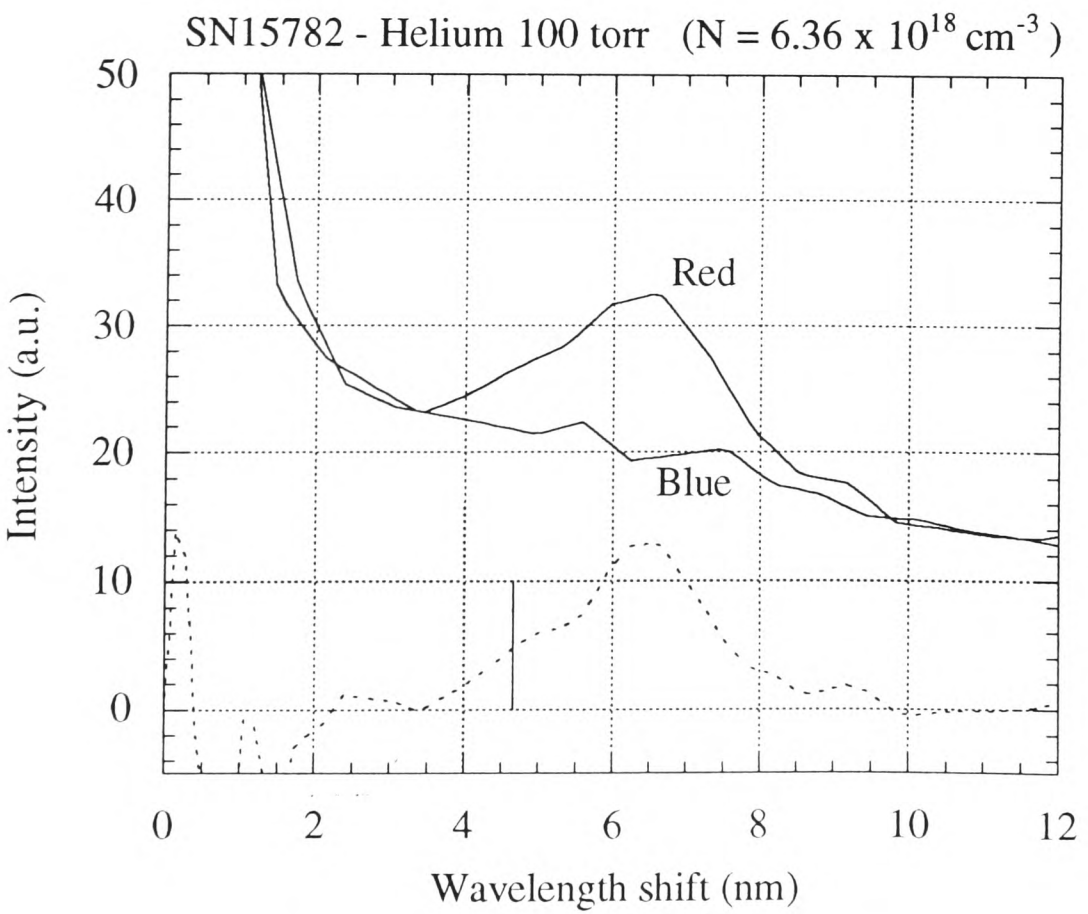
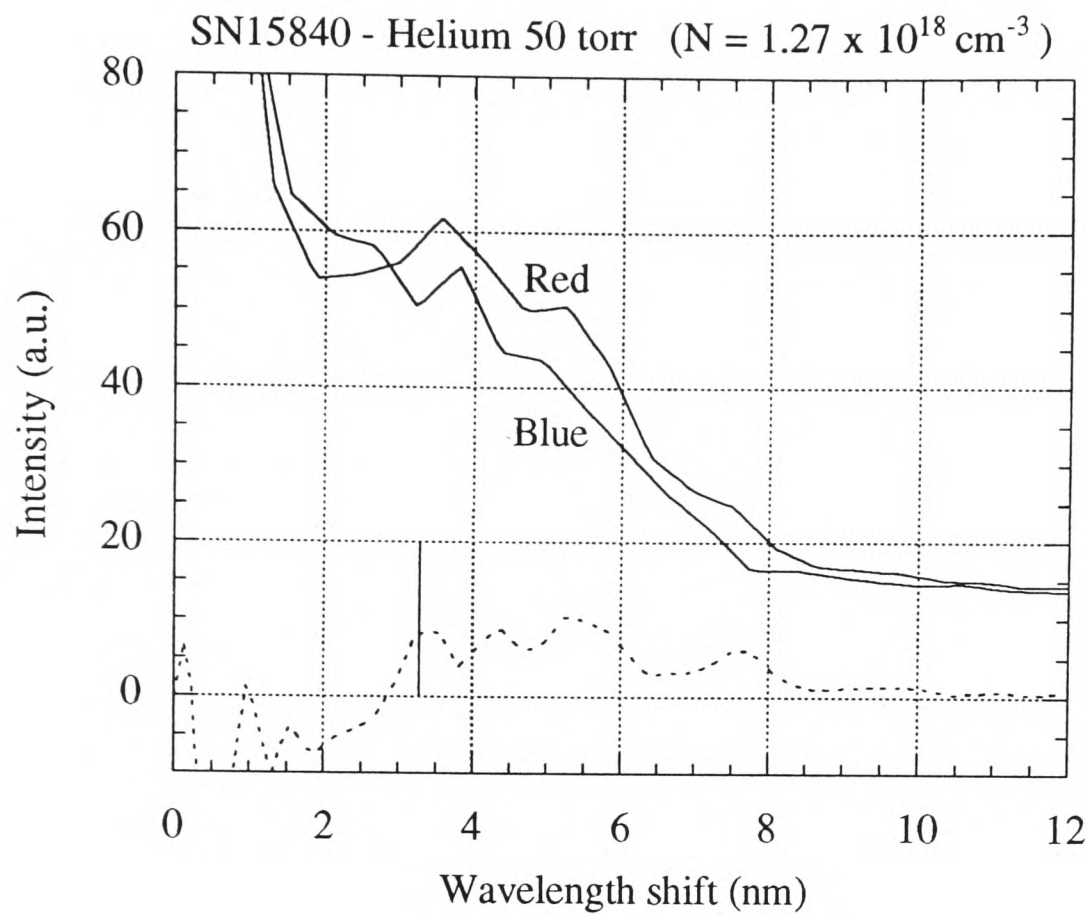
5.3.1.2. Fitting the Bohm-Gross frequency.

The densities inferred from both the red shifted and blue shifted wings of each of the Thomson scattered spectra in the figure above gives an acceptable match to the known electron density, given the errors caused by time and space integration. It is not surprising that the temperature from red and blue fits are similar, since the wavelength scale of any Thomson scattered spectrum is determined by the electron temperature. Enhancement of the red wing (by SRS or otherwise) would then lead to an increase in the fitted α , and so to an increase in the inferred electron density.

We will now subtract the blue shifted side of each of the above spectra from the red shifted side to determine the frequency at which the enhancement in the red shifted wing occurs. This should tell us the Bohm-Gross frequency, from which we may infer an electron temperature. Figure 5.3.3 shows the results of this subtraction. The electron plasma frequency is labelled by a solid vertical line in each lineout. A cubic spline has been fitted to the experimental lineouts to smooth the noise in the data.







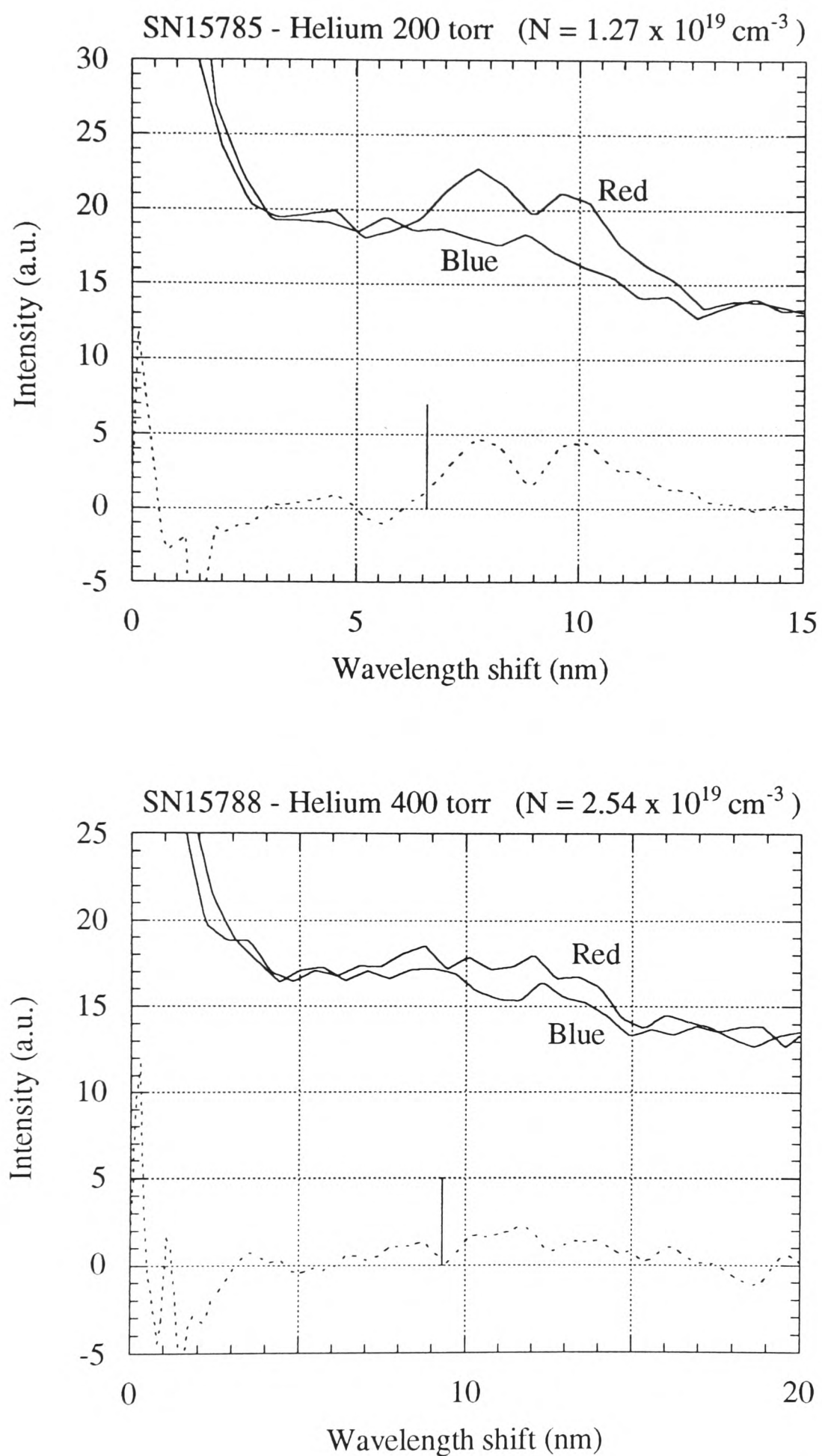


Figure 5.3.3. Overlaid red and blue shifted wings from 150° Thomson scattering in Helium. The difference between the two is shown as a dotted line. The electron plasma frequency is shown as a solid vertical line.

5.3.1.3. Electron temperatures in Helium.

We now collate the inferred temperatures from the three methods of determining the electron temperature described above, and present (in figure 5.3.4) the variation of electron temperature with the electron density for static fill Helium. In the figure we have assumed that the electron density is given by twice the static fill density. We can see the electron temperature rise from a level of a few eV (determined essentially by ATI) to almost 100 eV as IB (and SRS) becomes dominant.

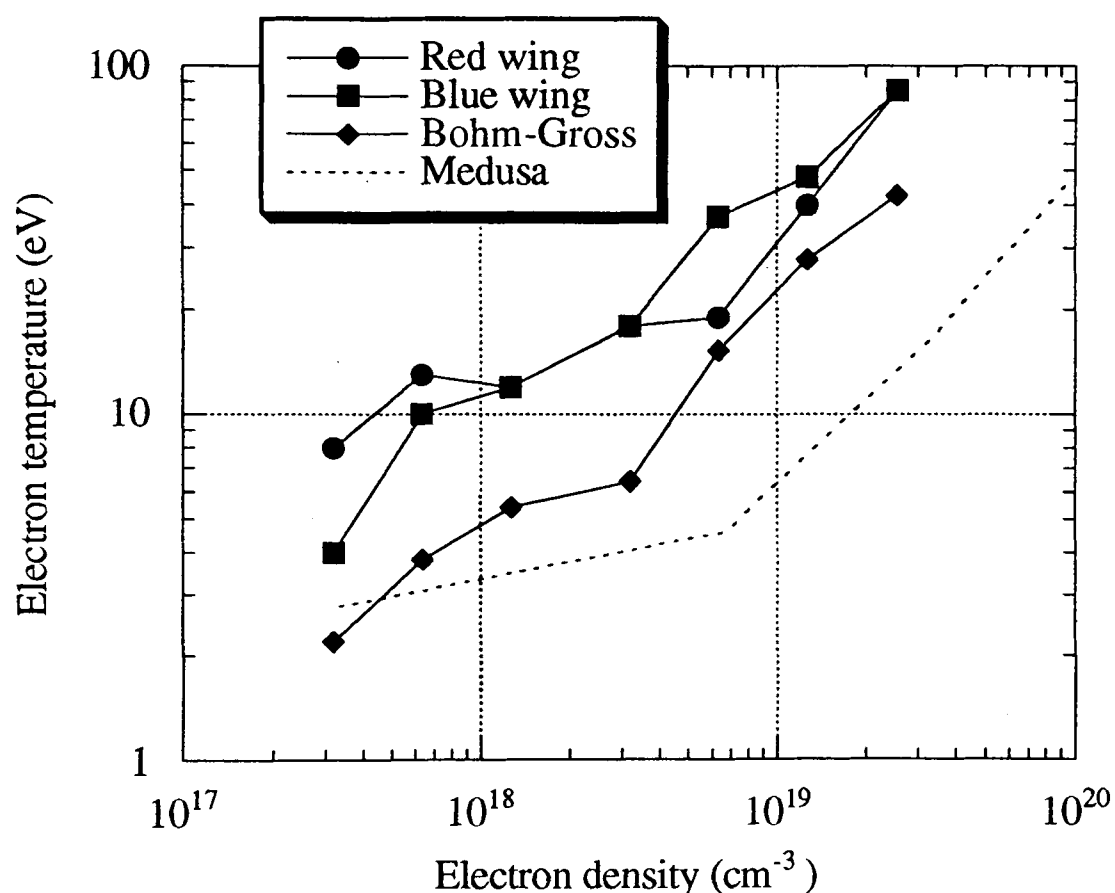


Figure 5.3.4. Variation of the electron temperature with electron density in Helium.

Taking the most conservative estimate of the electron temperature given by fitting the Bohm-Gross frequency to the difference of the red and blue shifted sides of the spectra we see that the electron temperature at $2 \times 10^{19} \text{ cm}^{-3}$ is already above 40 eV. However, the laser intensity used in the experiment is about $8 \times 10^{17} \text{ W/cm}^2$ which is higher than the optimum intensity for the Neon VIII recombination laser scheme ($3 \times 10^{17} \text{ W/cm}^2$). The measured electron temperatures are consistently higher than those predicted at the

peak of the pulse by the MEDUSA code (dotted line). This supports the theory that the enhancement of the red shifted wing is caused by SRS. An alternative explanation is that the poor quality of the Sprite focal spot leads to additional ponderomotive heating which is not modelled by the TEM₀₀ focus used in the MEDUSA simulations.

5.3.2. Neon.

As we have previously discussed, analysis of the Thomson scattering spectrum of Neon is complicated by its many ionisation stages. We cannot make the assumption, as we did for Helium, that the ionisation profile is uniform throughout the focus. Therefore we do not know the electron density, and so we cannot use the Bohm-Gross frequency to determine a plasma temperature. For the same reason we cannot easily fit a single Thomson scattering spectrum. Figure 5.3.5 shows the spectrum of Thomson scattering from Neon at static fill pressures of 10 torr, 60 torr, and 100 torr. Again we see enhancement of the red shifted wing.

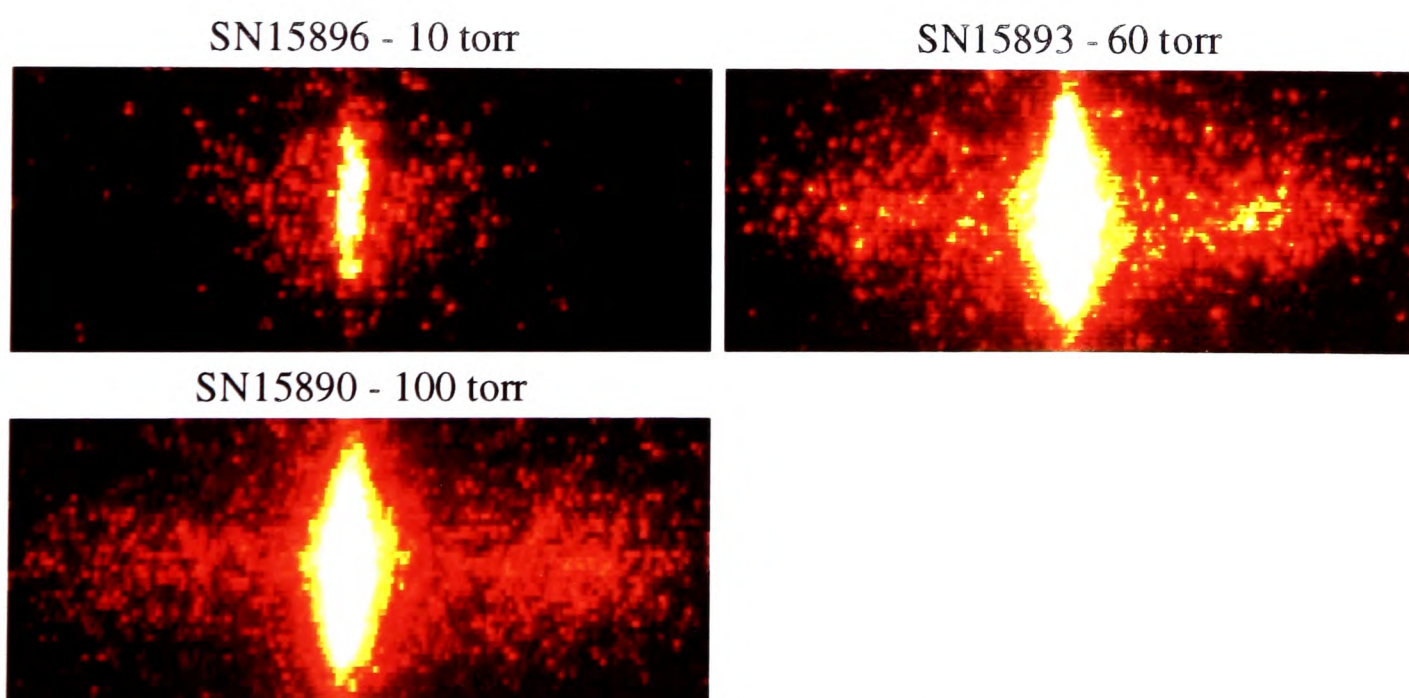
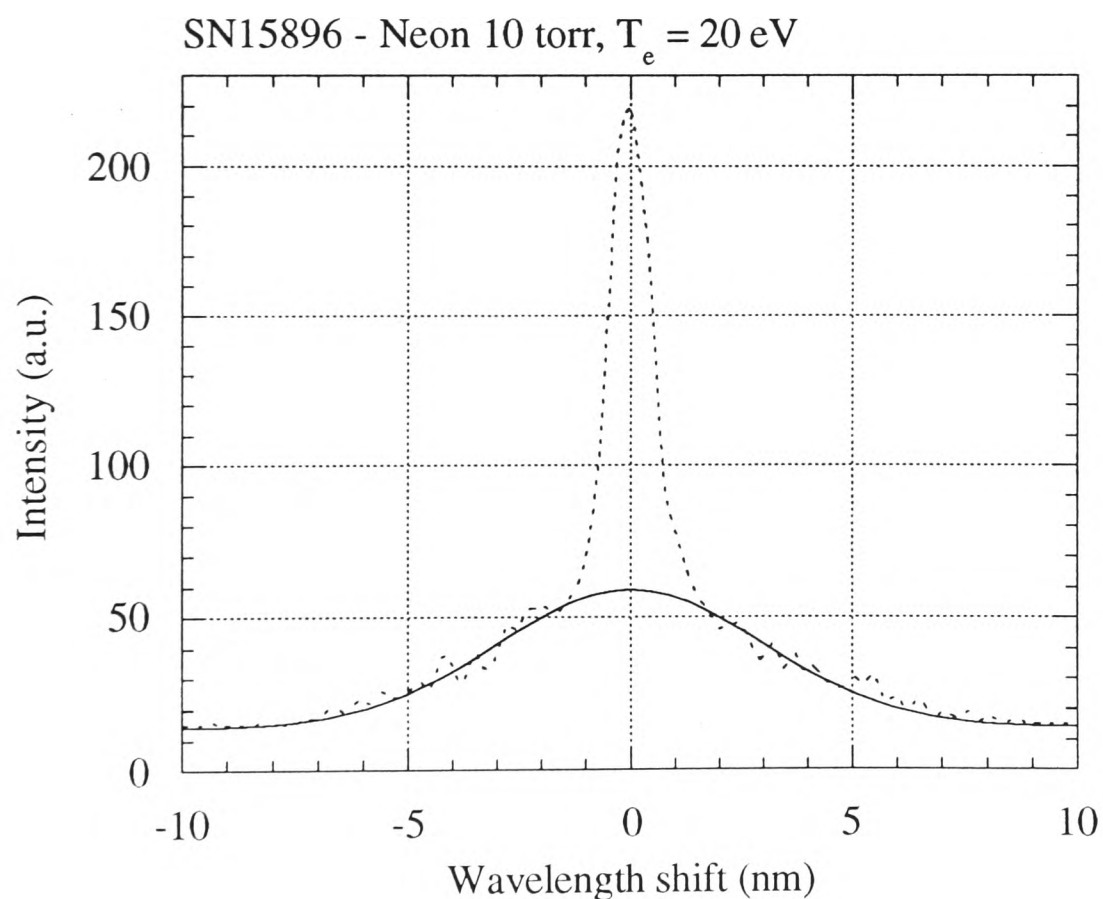


Figure 5.3.5. Images of Thomson scattering at 150° from Neon at static fill pressures of 10, 60 and 100 torr. The central 'ion' feature of both SN15890 and SN15893 is slightly saturated.

5.3.2.1. Fitting a Gaussian curve to the electron wings.

To estimate an upper bound to the electron temperature we assume that the scattering in the electron wings is purely incoherent. In this way we can fit an $\alpha = 0$ (Gaussian) spectrum to the blue shifted electron feature. We cannot infer an electron density using this method, but we do know that the maximum ionisation stage that can be reached at these laser wavelengths is Ne^{8+} . We will therefore assume that the electron density is eight times the static fill density. $\alpha = 0$ fits to Thomson scattering at 150° from Neon at static fill pressures of 10, 60, and 100 torr are shown in figure 5.3.6.



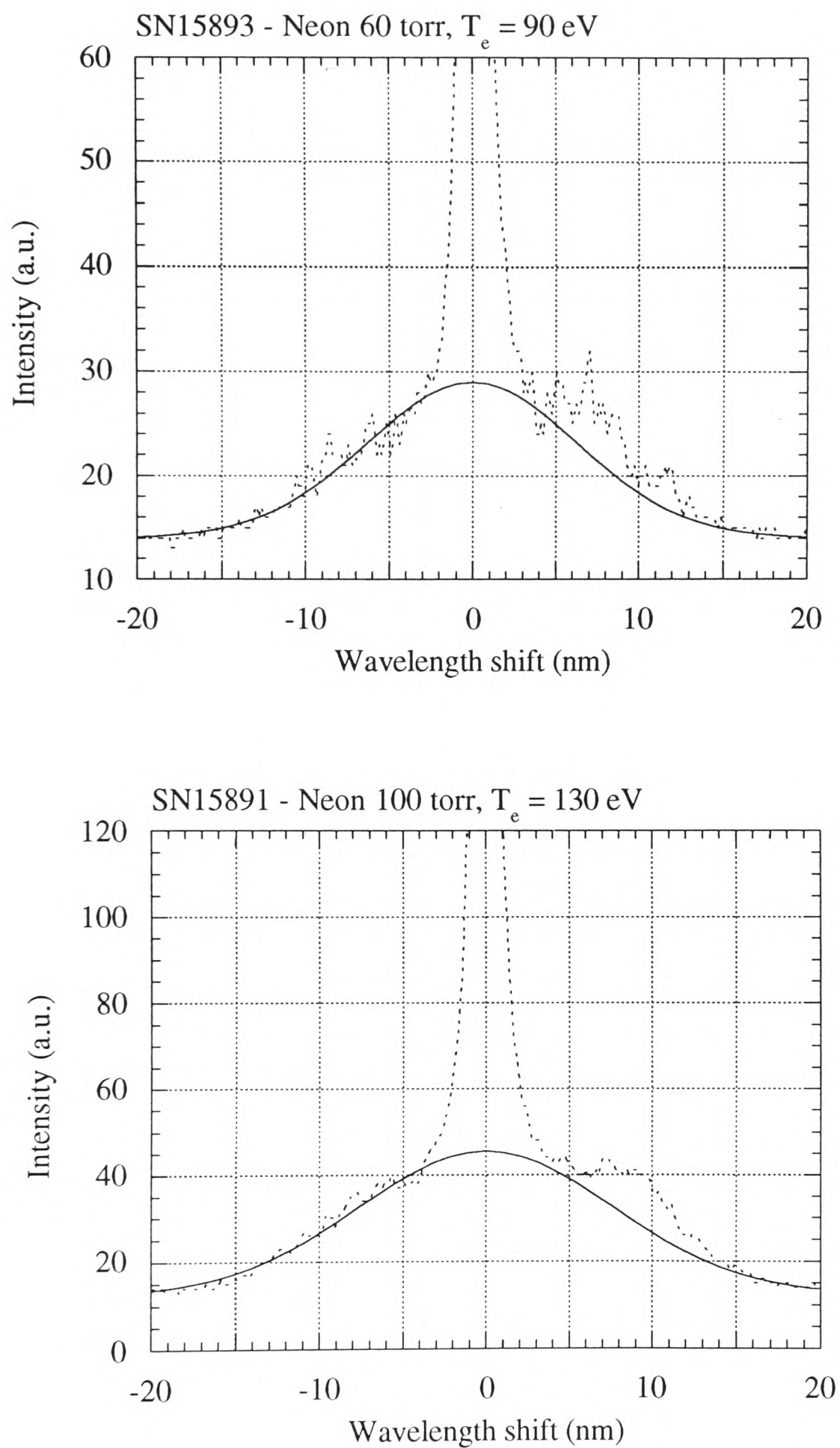


Figure 5.3.6. Lineouts of Thomson scattering at 150° from Neon at static fill pressures of 10, 60 and 100 torr. Temperatures have been estimated by fitting an $\alpha = 0$ spectrum to the blue shifted electron feature.

Figure 5.3.7 shows the variation in electron temperature with electron density (assuming eight times ionised Neon). Again we see that the measured temperatures rise more quickly with density than the MEDUSA simulation (solid line) predicts. This again can be attributed to either SRS or ponderomotive heating. Interestingly, the Neon electron temperatures at a given electron density are of the same order as the Helium temperatures of figure 5.3.4.

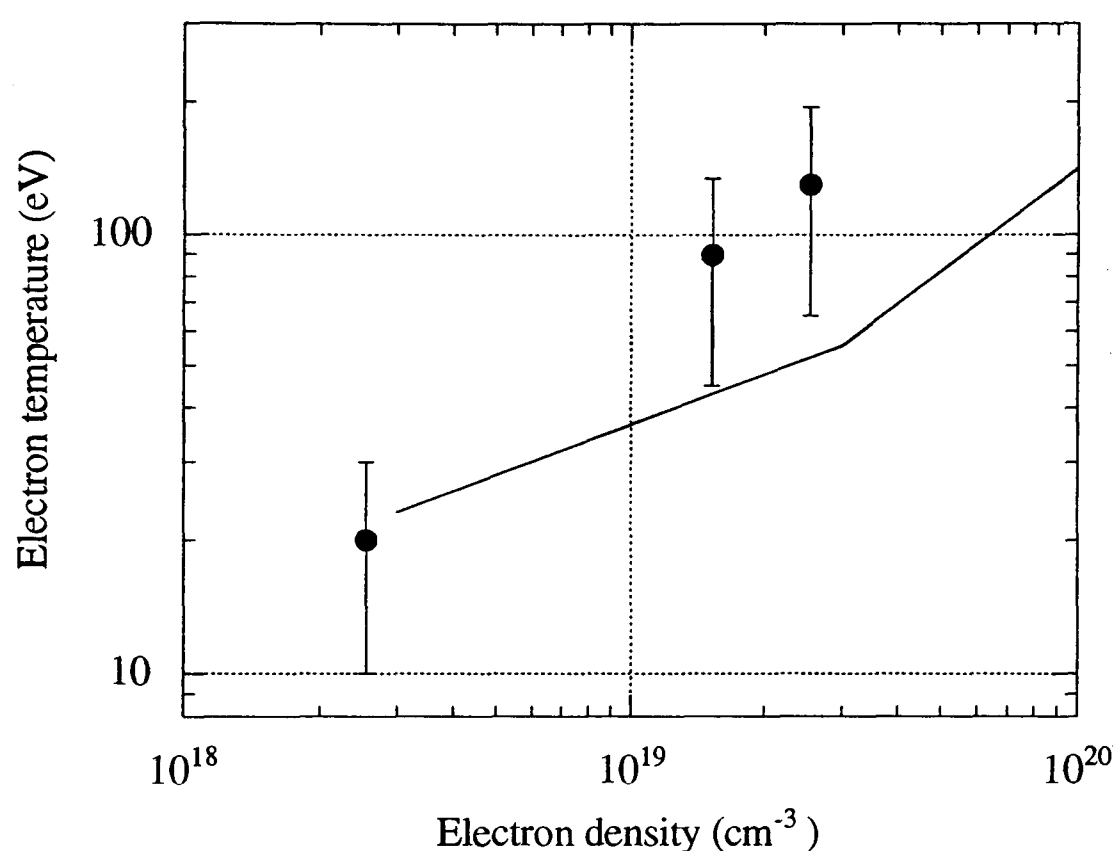


Figure 5.3.7. Variation of the electron temperature with electron density in Helium. The solid line shows the temperature predicted by MEDUSA.

5.4. SRS measurements.

Determining how the electron temperature varies with density in a plasma created from a gas at a known density is relatively simple. (Although we have seen that even this can present problems). However, to study higher densities we cannot continue to use a static gas fill, as the presence of the gas will severely affect both the focusing and the pulse length of the CPA laser pulse. To study plasmas with electron densities above about

10^{19} cm^{-3} we must use a gas jet to create a finite region of gas into which we focus the laser. This creates an additional problem for determining the variation of the electron temperature with density, as we do not know the initial gas density from which the plasma is created. Even if we characterise the profile of the gas jet by interferometry we cannot be certain where in this profile the laser will focus, especially at densities where the laser filaments [144].

At electron densities from about $5 \times 10^{19} \text{ cm}^{-3}$ to about $2 \times 10^{20} \text{ cm}^{-3}$ we cannot see Thomson scattering as it is masked by very strong plasma emission. This plasma emission is broadened out into a continuum by the Stark effect [145]. Therefore we cannot use scattering of the laser light to diagnose the plasma. Above these densities, however, SRS reaches such a level that it is brighter than the plasma emission. We can then use this SRS to probe the plasma once more.

In section 2.5 we discussed the physics of parametric instabilities in the laser-plasma system with particular reference to SRS and SBS. In this section we shall review experimental data, which consists of SRS spectra at 90° ; SRS spectra at 150° ; and the calibrated measurement of the amount of energy SRS backscattered through 180° (the SRS reflectivity). For the 380 fs Sprite laser pulse we do not expect significant SBS to develop, as the typical growth time for the instability is much longer than the laser pulse length. As we will see, this is not the case for SRS.

5.4.1. SRS spectra.

Let us consider the case of the KrF laser focused 100 μm above a 1 mm circular aperture nozzle of the high pressure solenoid-valve gas jet with a backing pressure of 1000 psig (67 bar gauge) Neon. Figure 5.4.1 shows the 150° scattered spectra from two shots with nominally the same conditions (SN16042 and SN16045). The only quantifiable

difference between the two is the laser energy on target which was 220 and 200 mJ respectively.

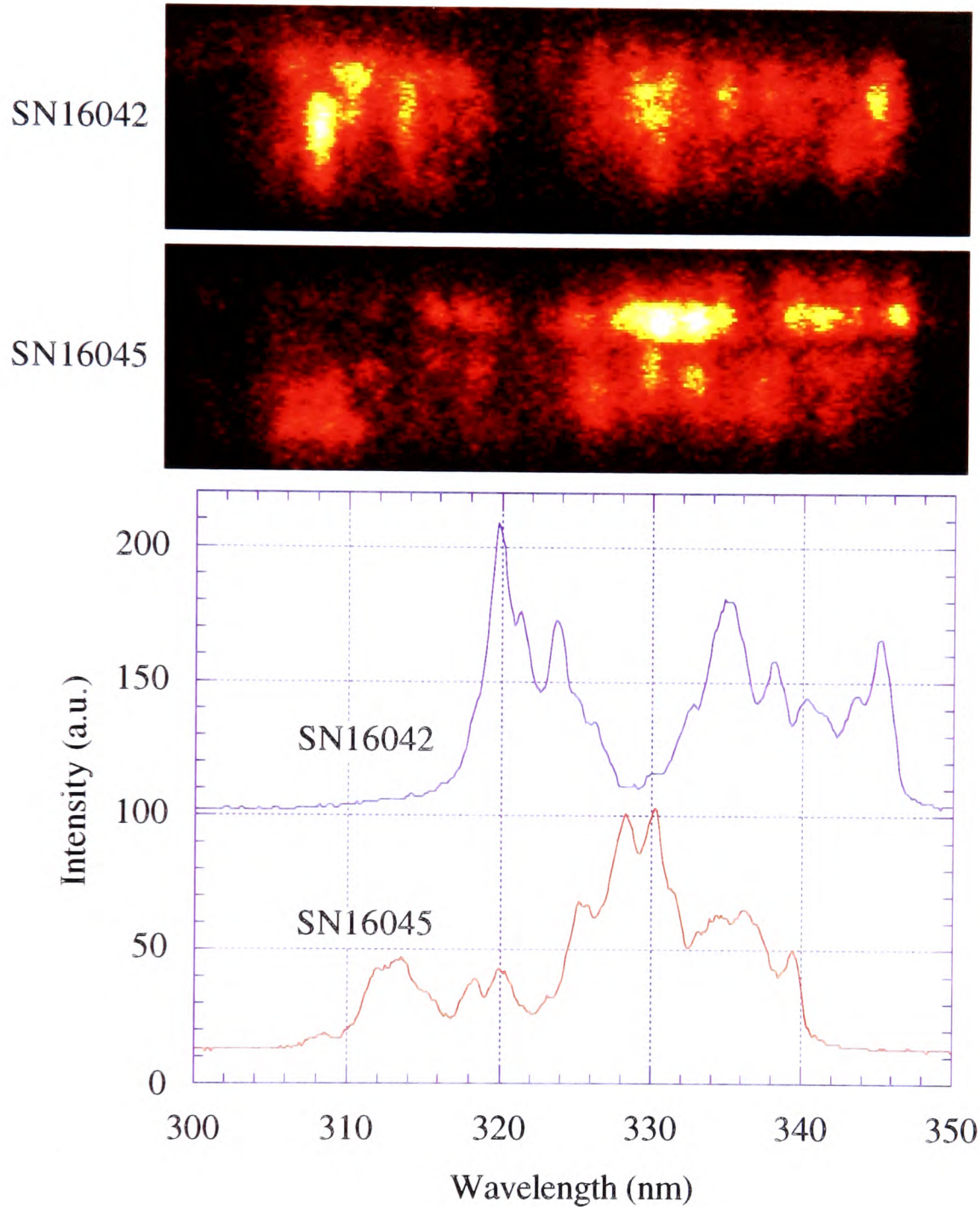


Figure 5.4.1. Images and lineouts of 150° stimulated Raman scattering spectra.

Note that the images are not aligned to the same wavelength position.

Both spectra exhibit considerable structure and are spread in wavelength over a range of over 30 nm. This structure is due to the filamentation of the laser beam as it passes through the plasma. Each filament travels through a region of plasma with a different density and temperature and so the criteria for conservation of energy and momentum result in a different frequency shift of the scattered light. This frequency shift is given by

the Bohm-Gross frequency which can include a large temperature correction. The Bohm-Gross frequency shift is in the range $1.71 - 2.06 \times 10^{15} \text{ s}^{-1}$ for SN16042 and $1.56 - 2.14 \times 10^{15} \text{ s}^{-1}$ for SN16045. The peak electron density at this gas jet backing pressure is of the order of 10^{21} cm^{-3} . At this density our MEDUSA simulation (figure 5.1.3) tells us that the electron temperature should be of the order of 1000 eV. This gives a thermal correction ($\sqrt{3}k\nu_{th}$) to the electron-plasma frequency (ω_{pe}) of $10 - 9.7 \times 10^{14} \text{ s}^{-1}$ for both SN16042 and SN16045. If we assume this temperature and consider the maximum and minimum wavelength shifts in figure 5.4.1 to be solely due to variation in the electron density then equation 5.2.4 gives us a density range of $0.6 - 1.2 \times 10^{21} \text{ cm}^{-3}$ for SN16042 and $0.4 - 1.0 \times 10^{21} \text{ cm}^{-3}$ for SN16045. Hence, the wavelength spread of the scattered signal can be explained by the laser beam scattering from electron densities that vary by a factor of only two if we assume that the electron temperature is constant. This variation will be less if the electron temperature is less in regions of lower electron density.

Figure 5.4.2 shows the scattering signal at 90° for the same two laser shots as in figure 5.4.1. Unfortunately the scattered signal is so strong that it has saturated our detector, but we can see the wavelength shift of the scattered light. SN16042 has limiting slits in the vertical (space and time) plane. The light to the left of the image is at the laser wavelength, the signal on the right is the SRS scattered light shifted by the Bohm-Gross frequency. Note that the SRS signal of SN16042 exhibits structure in a similar manner to the 150° spectrum. Any structure in SN16045 is blurred out by saturation and by the instrument response. The high background signal across the lower half of the images is caused by plasma emission. At electron densities (of the order of $10^{20} - 10^{21} \text{ cm}^{-3}$) present in these optically-ionised plasmas we would expect any spectral emission in the optical region to be Stark broadened into a continuum [145]. This was confirmed by scanning the 150° spectrometer over a wide wavelength range ($\sim 200 - 400 \text{ nm}$) over a series of laser shots where a continuous background was observed.

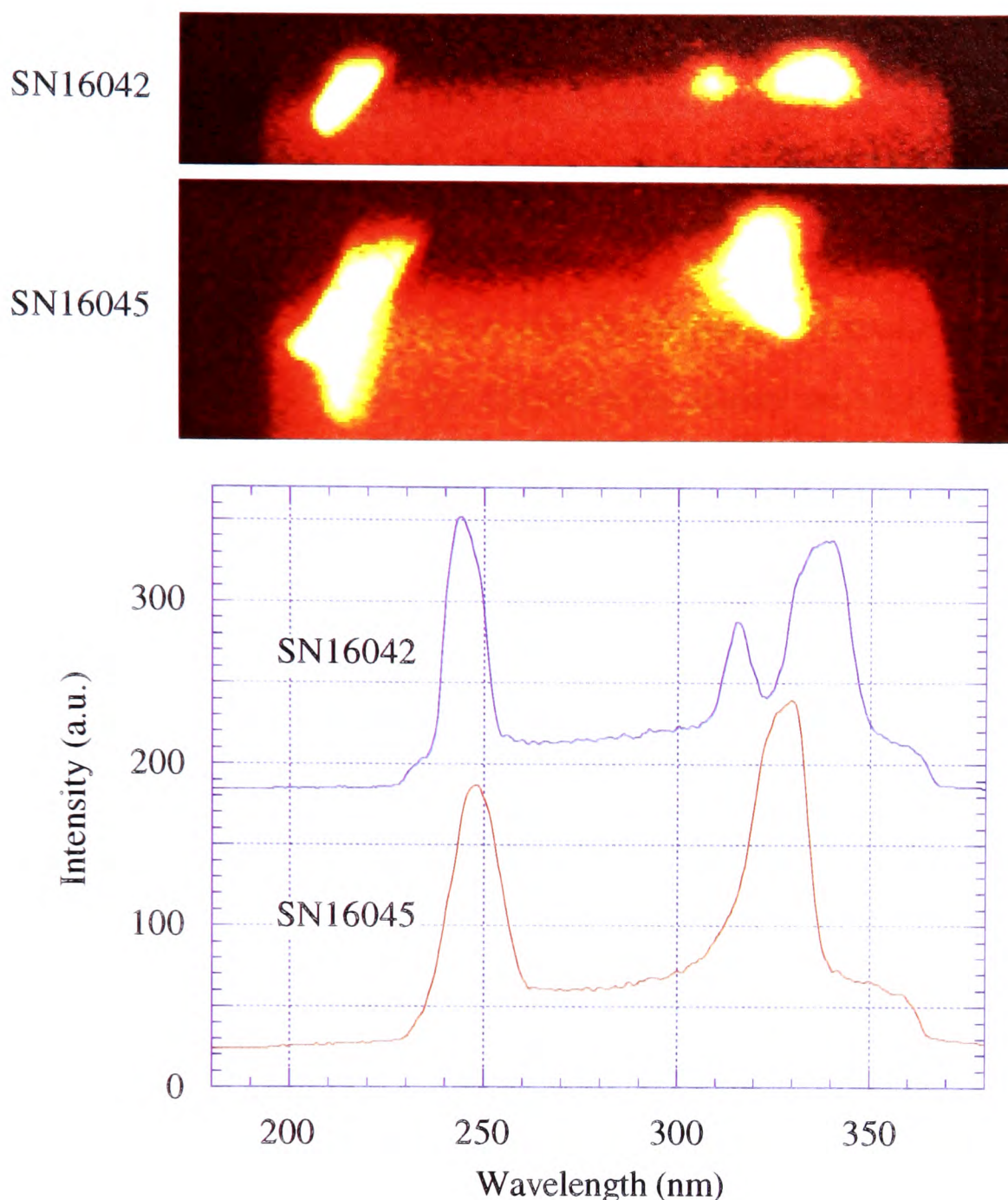


Figure 5.4.2. Images and lineouts of stimulated Raman scattering spectra at 90° .

If we assume that the 90° SRS comes from the same region of the plasma as the 150° SRS then we would (again) expect an electron density of the order of 10^{21} cm^{-3} and an electron temperature of about 1000 eV. This electron temperature then gives thermal corrections (to the Bohm-Gross frequencies corresponding to the peaks in the scattered signal of $1.63 \times 10^{15} \text{ s}^{-1}$ and $2.02 \times 10^{15} \text{ s}^{-1}$ for SN16042 and $1.84 \times 10^{15} \text{ s}^{-1}$ for SN16045) of $7.39 \times 10^{14} \text{ s}^{-1}$, $7.20 \times 10^{14} \text{ s}^{-1}$ and $7.29 \times 10^{14} \text{ s}^{-1}$ respectively. This gives us electron densities of $6.7 \times 10^{20} \text{ cm}^{-3}$, $1.1 \times 10^{21} \text{ cm}^{-3}$ and $9.0 \times 10^{20} \text{ cm}^{-3}$ which are consistent with those calculated from the 150° signals.

We shall now calculate both the electron temperature and density by the method described in section 5.2.3 to compare with the simple calculation presented above. If we compare the shorter wavelength 90° signal with the shorter wavelength 150° signal we derive an electron temperature of 540 eV at an electron density of $6.7 - 7.5 \times 10^{20} \text{ cm}^{-3}$. The longer wavelength signals combine to give an electron temperature of 1340 eV at a density of $0.9 - 1.1 \times 10^{21} \text{ cm}^{-3}$. For SN16045 the shorter wavelength end of the 150° signal gives unphysical negative temperatures when combined with the peak of the 90° signal. The longer wavelength end of the signal gives an electron temperature of 1830 eV at an electron density of $5.3 - 7.8 \times 10^{20} \text{ cm}^{-3}$.

Figure 5.4.3. shows a plot of the electron temperatures inferred from several laser shots using this method. In this case, all the peaks in the 90° and 150° spectra were used to calculate electron temperatures. The temperature was then used to calculate a density for the 90° and 150° peak. The data shown on the graph corresponds to the peaks that produce the smallest spread in density between the two channels for each shot. The data covers an electron density range from about $3 \times 10^{20} \text{ cm}^{-3}$ to 10^{21} cm^{-3} . The calculated temperature covers a range from about 500 eV to 2500 eV. This spread in temperature is partly due to the relative inaccuracy of this technique, giving an accuracy no better than about 50%. However, it is also because the variable focal spot structure of the beam combined with filamentation of the beam in the plasma can result in greatly differing conditions for each shot. This shot-to-shot variation is much greater than the error on any particular point. We can say with certainty that electron temperatures at these electron densities are of the order of 1000 eV.

Landau damping of the electron-plasma waves produced by SRS will cause a hot tail to the electron temperature distribution. If we assume that electrons drawing energy from the plasma wave are accelerated to the phase velocity of the plasma wave, then the hot electron temperature, T_H , is given by $T_H = (m/2)(\omega/k)^2$ [146]. The hot electron

temperatures at 150° range from 3000 to 5000 eV for SN16042 and from 2900 to 4600 eV for SN16045. The corresponding hot electron temperatures at 90° are 7300 to 11600 eV and 9700 eV respectively. These hot electrons can cause significant ionisation in the plasma resulting in production of 9 and 10 times ionised Neon: states which cannot be produced by optical-ionisation at our laser intensity. This will be shown by the X-ray spectrum discussed in section 5.5.2.

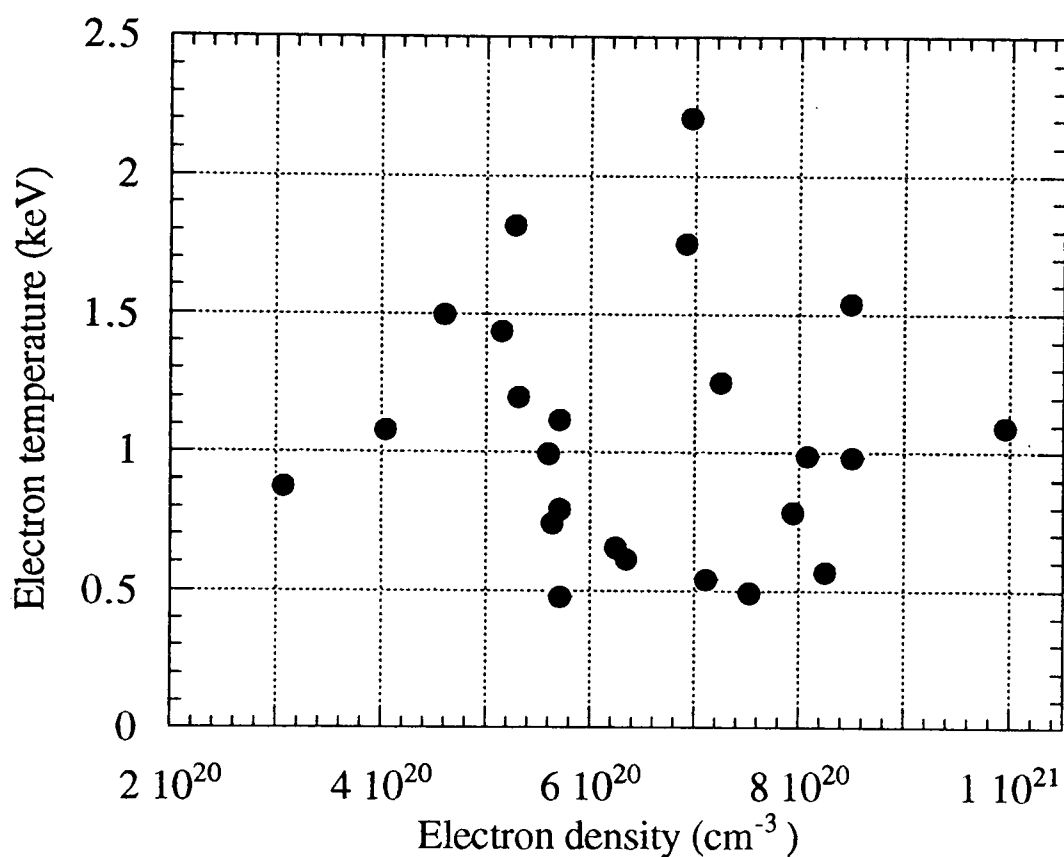


Figure 5.4.3. Electron temperatures and densities calculated from 90° and 150° SRS scattering.

We now combine the electron temperatures deduced from Thomson scattering at low densities (section 5.3) with electron temperatures deduced from SRS spectra at higher densities. Figure 5.4.4 shows the variation in electron temperature with electron density for both Helium and Neon at electron densities ranging from about $3 \times 10^{17} \text{ cm}^{-3}$ to just over 10^{21} cm^{-3} . Data for Helium is shown as filled symbols, and data for Neon is shown as unfilled symbols. Triangles represent temperatures inferred from Thomson

scattered data and diamonds represent temperatures inferred from SRS. The temperatures from the SRS data are all calculated by comparing SRS spectra at 90° and 150° . The spread in the data gives an indication of the error in fitting the electron temperature. For the Thomson scattered Neon data, temperatures are inferred from fitting a Gaussian to the blue shifted electron wing. The upper and lower temperatures plotted at each density for the Helium Thomson scattered data inferred from fitting the blue shifted wing and the Bohm-Gross frequency respectively. The solid lines represent MEDUSA calculations of the electron temperature at the centre of the laser focus and at the peak of the laser pulse. We expect the experimental data to be weighted towards the peak of the pulse, as this is when the laser intensity is highest and most scattering occurs.

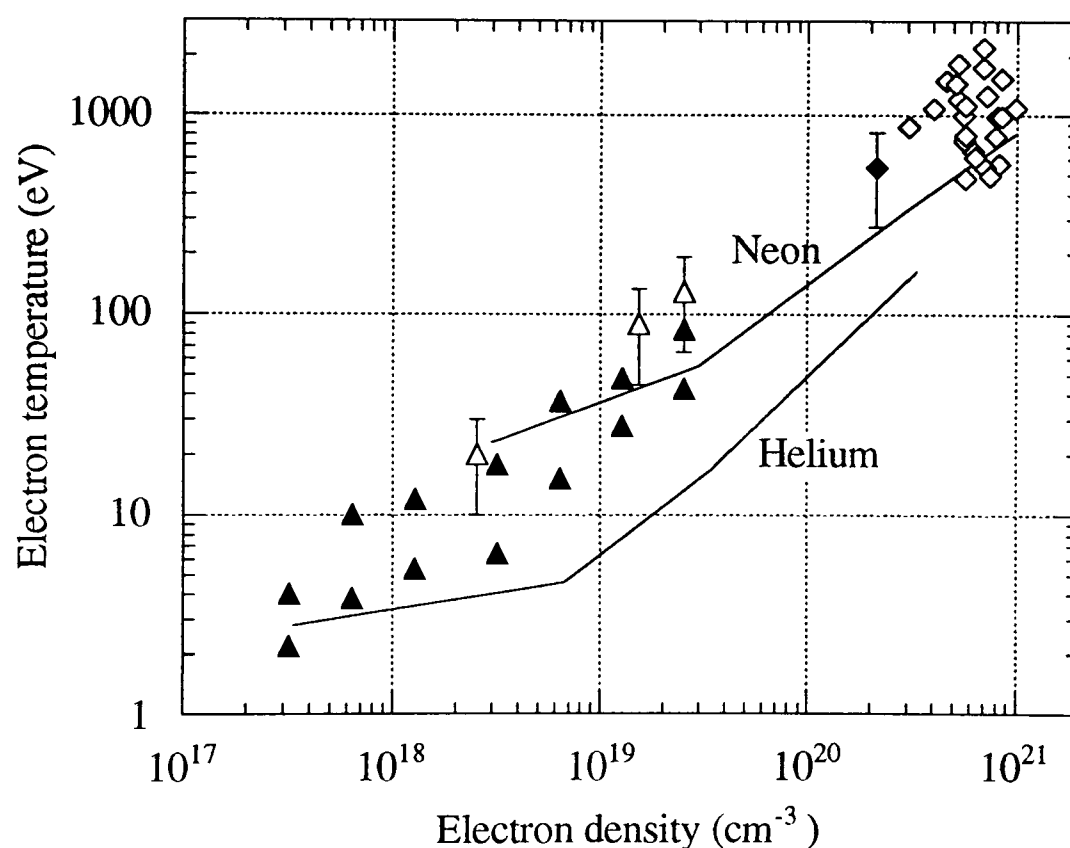


Figure 5.4.4. The variation of electron temperature with density in Helium (filled symbols) and Neon (unfilled symbols). Triangles represent temperatures inferred from Thomson scattered data and diamonds represent temperatures inferred from SRS. The solid lines depict temperatures calculated from MEDUSA simulations.

We see that the experimentally inferred temperatures, for both Helium and Neon, generally rise more quickly than predicted by MEDUSA. This is due to a combination of ponderomotive heating by the poor quality of the Sprite focal spot and heating by Landau damping of plasma waves created by SRS, with SRS causing the greatest additional heating. It will be shown in the next section that, at the higher electron densities ($>10^{20} \text{ cm}^{-3}$), SRS alone can cause heating of several keV. However, we showed in figure 5.1.6 that reducing the laser pulse length from 380 fs to 100 fs (at an intensity of $3 \times 10^{17} \text{ W/cm}^2$ and an electron density of $5 \times 10^{20} \text{ cm}^{-3}$) will reduce the contribution of IB to the total electron temperature from just over 100 eV to about 25 eV, i.e. IB scales linearly with the laser pulse length. We also showed in figure 5.1.4 that a reduction in laser intensity from that used in our experiment ($8 \times 10^{17} \text{ W/cm}^2$) to the threshold required for production of Ne^{8+} ($3 \times 10^{17} \text{ W/cm}^2$) will cause a further decrease in the electron temperature. We therefore predict that, if SRS can be eliminated, the conditions required by Eder et al. [44] can be met. Hence, we must now determine the scaling of SRS with laser intensity.

5.4.2. SRS reflectivities.

The form of the SRS spectrum gives us an indication of the plasma density and temperature at the peak of the laser pulse, but the energy in the electron-plasma waves created by this process will be converted to thermal energy by Landau damping at some later time. To determine the magnitude of this additional heating we must consider how much energy is absorbed from the laser by this process. We expect the strongest SRS to occur at 180° where the instability has the longest length over which to grow and the wavevector matching requirements are most easily achieved. A series of laser shots at a range of energies were taken with the laser focused 100 μm above the gas jet nozzle with a gas jet backing pressure of 500 psig (33 bar gauge). Such a backing pressure should

produce an electron density of around $5 \times 10^{20} \text{ cm}^{-3}$ at the focus. Figure 5.4.5 shows the variation of the SRS reflectivity with laser energy. The scatter in the data is due to the variable focal spot of Sprite which leads to differences in the focused intensity at the same energy.

The SRS reflectivity shows a remarkable fall off as we reduce the laser energy. Assuming a Gaussian intensity distribution at the laser focus with a $10 \mu\text{m}$ FWHM, we calculate that 250 mJ laser energy would correspond to a peak intensity of $6 \times 10^{17} \text{ W/cm}^2$ (in the absence of defocusing effects) at which intensity the SRS reflectivity reaches 3%. If we assume a similar spectral shift as that seen at 150° (figure 5.4.1) then the frequency of the plasmon created from each scattered photon is of the order of $1.6 - 2.1 \times 10^{15} \text{ s}^{-1}$ and $1.2 - 1.3 \times 10^{16}$ photons are scattered. If all this energy is uniformly absorbed within a cylinder of radius $5 \mu\text{m}$ (the width of our laser focus) and length $100 \mu\text{m}$ (approximately twice the confocal parameter of our laser focus) with an electron density of $5 \times 10^{20} \text{ cm}^{-3}$, then the electron temperature due to SRS will be 2100 - 3100 eV. If we reduce the laser intensity by just a factor of two (to $3 \times 10^{17} \text{ W/cm}^2$), the SRS reflectivity plummets to about 0.002%. The electron temperature due to SRS is now just 1.5 - 2.0 eV.

Before considering the implications of this SRS heating to the OFI recombination laser scheme we should consider a few factors that may affect the levels of SRS heating. Firstly, we should note that the Sprite CPA focal profile exhibits considerable structure (see figure 4.1.6) and is a long way from that of a Gaussian (TEM_{00}) beam. This non-uniformity can act as a seed for SRS growth and can greatly enhance the scattering levels. Secondly, we should consider that, at such high electron densities, a uniform beam would be greatly defocused [34,47] with a consequent reduction in laser intensity. Since our beam exhibits considerable structure it is likely to split into many filaments, each of which creates a channel through the plasma [144]. These filaments slightly counter the

defocusing effects by creating regions of high and low intensity in the laser profile. We have assumed that the peak intensity in these filaments is at the same level as that calculated for a clean Gaussian beam in a vacuum.

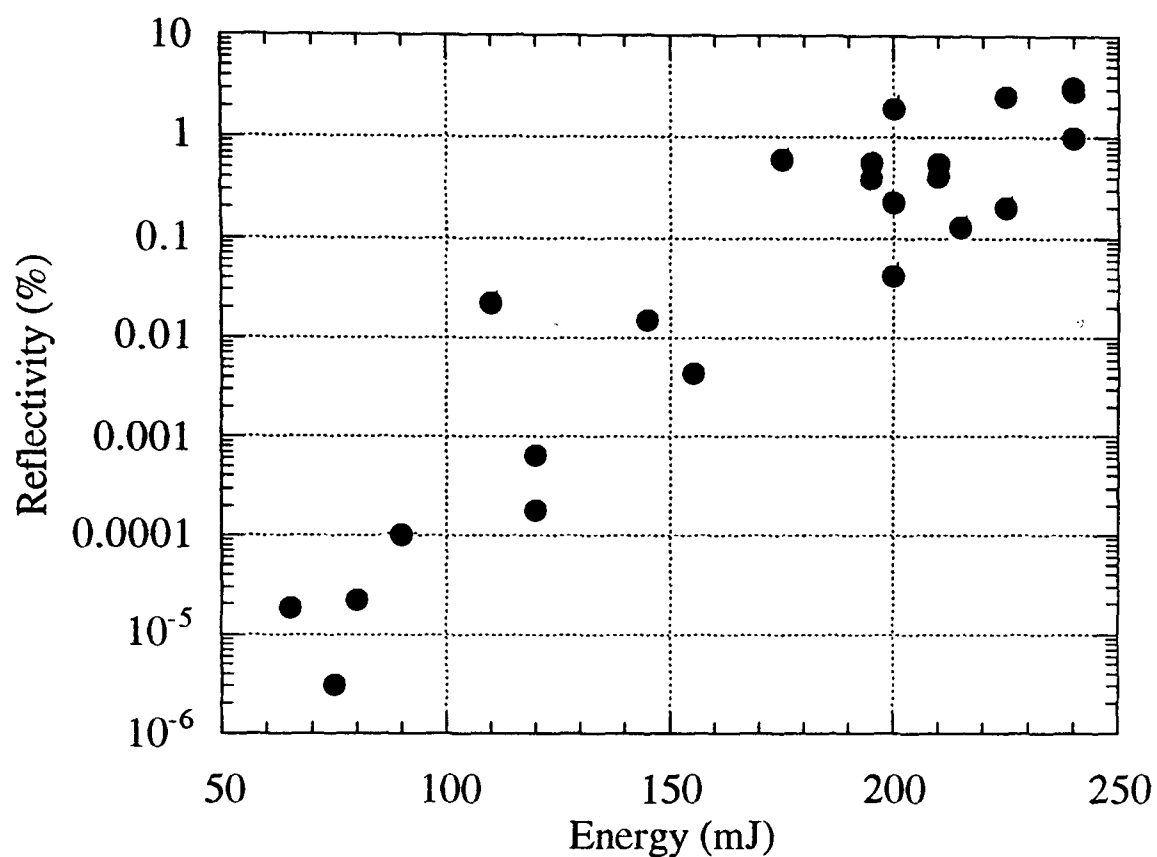


Figure 5.4.5. SRS reflectivity as a function of laser energy on target. The laser was focused 100 μm above a 1 mm circular aperture gas jet nozzle with a backing pressure of 500 psig Neon.

The calculations of Eder et al [44] were performed for a pulse length of 100 fs and an intensity of $3 \times 10^{17} \text{ W/cm}^2$. We have already shown in the discussion of figure 5.4.5 that reducing the vacuum focused intensity to this level brings SRS heating down to a negligible level. Since the SRS gain scales as $I^{1/2}t$, where t is the time over which SRS is allowed to grow, a reduction in pulse length will further decrease the SRS levels. Hence, we can say with some certainty that for the conditions of Eder et al. the instability will be suppressed.

It also follows that increasing the atomic number, Z , will also require an increase in laser intensity. Equation 2.2.2 shows that the critical intensity, I_c , required to optically ionise

a given energy level, U_I , is proportional to U_I^4/Z^2 . Since the binding energy, U_I , in a hydrogenic potential scales roughly as Z^2 , it is trivial to see that scaling to shorter wavelengths by moving to higher atomic numbers will cause a scaling of the laser intensity approximately proportional to Z^6 . This intensity scaling, together with an increase in the required electron density for fast three body recombination, implies that the SRS instability will rapidly recur for higher Z . The pulse length cannot be lowered significantly for a KrF system because of the practical limitation of about 80 fs set by the bandwidth of the KrF laser transition. OFI recombination lasers based on Lithium-like schemes will therefore be restricted to atomic numbers less than 12, and so to wavelengths longer than about 60 Å.

5.5. Verification of SRS measurements by other diagnostics.

Having used SRS spectra as a diagnostic for determining the electron density at the peak of the laser pulse, and the SRS reflectivities for determining the levels of additional heating caused by Landau damping of the strong electron-plasma waves created by the instability, we now turn to other diagnostics to verify our interpretation of the data so far.

5.5.1. Faraday cup measurements.

Our first alternative diagnostic is the Faraday cup. This was placed 11 cm from the laser focus and in the horizontal plane between the 90° and 150° optical channels (i.e. somewhere near 120°). The signal from the Faraday cup recorded on a fast storage oscilloscope was extremely weak, in fact the signal on another Faraday cup positioned 22 cm away was too weak to measure. Figure 5.5.1 shows a typical trace which has been smoothed to remove some of the electrical noise from the firing of the laser. The Faraday cup measures the time dependence of the arrival of charged ions ejected from the

laser focus. These ions are dragged out of the laser focus by the electrostatic charge created when electrons heated by SRS escape the laser focus. By converting the time of flight signal to an ion energy distribution we can estimate the temperature of electrons that initially left the focus.

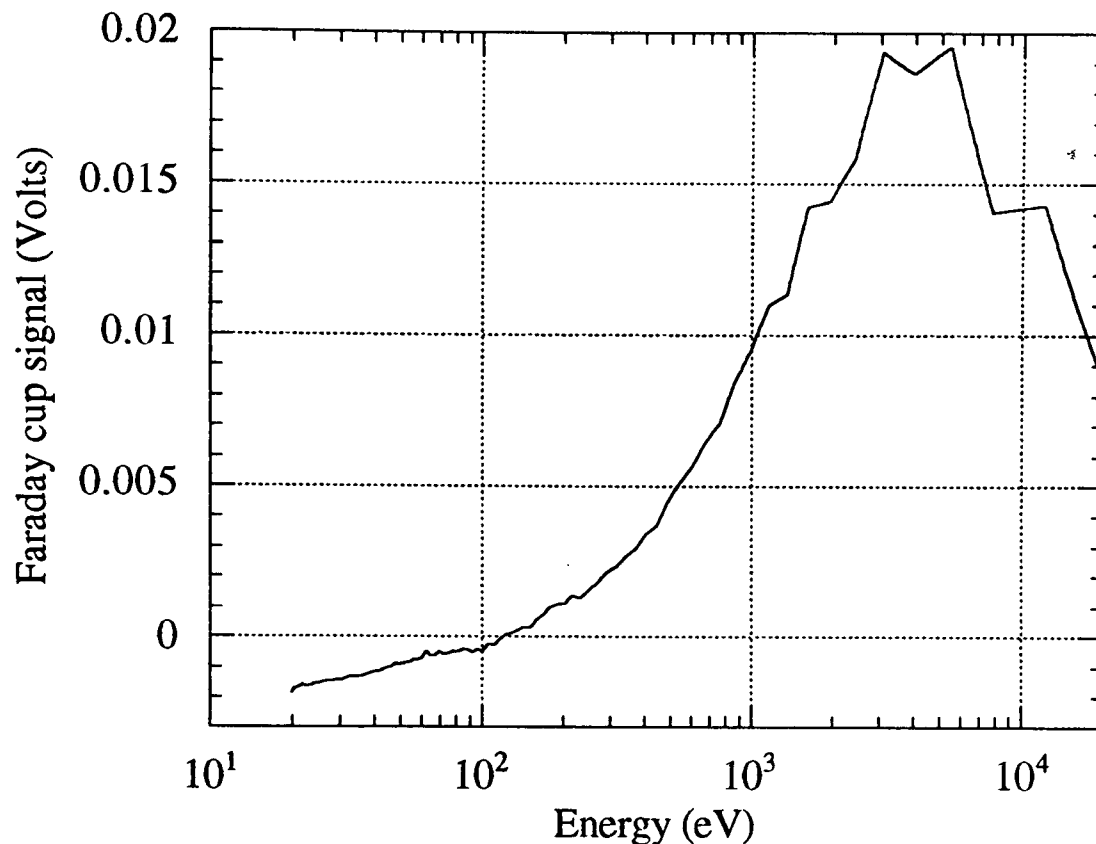


Figure 5.5.1. Faraday cup signal of ion emission from 400 psig Neon with a cup bias of 6V (SN16437). The laser was focused 100 μm above the gas jet nozzle.

We make the simple assumptions that eight electrons will drag one Ne^{8+} ion from the laser focus, and that, by the time the electrons reach the Faraday cup, the electrostatic force between electrons and ions will have equilibrated their velocities. Since the ion mass is so much greater than the electron mass it is simple to show that the kinetic energy of eight electrons at the laser focus is essentially converted to the kinetic energy of one ion at the Faraday cup. The distribution of the ion kinetic energy peaks somewhere between 3000 and 6000 eV. If we assume that the electron energy is directed motion from the electron-plasma waves (i.e. it is a one dimensional energy distribution) we have

the relation $4\kappa T_e \approx U_{ion}$, where U_{ion} is the ion energy measured by the Faraday cup. This implies an electron temperature of 750 - 1500 eV. This approximation is very simplistic, but the order of magnitude of the inferred electron temperature supports the conclusions of our analysis of the SRS spectra and reflectivities.

The signal from the Faraday cup showed some interesting variation over different laser shots. It was noticed that any laser shot that produced a strong SRS signal on the 90° optical channel also produced a Faraday cup signal similar to that in figure 5.5.1. However, some laser shots did not produce any SRS signal at all. The reason for this is not known, but it is probably due to variations in the laser focal spot. These 'blank' SRS shots produced no Faraday cup signal (except for the characteristic noise signal from the laser firing, which was also seen for vacuum shots). This phenomenon supports our belief that the ejection of ions is caused by the SRS heated electrons.

5.5.2. X-ray spectrometer measurements.

A TIAP crystal X-ray mini-spectrometer placed approximately 6 cm from the focus of the Sprite CPA laser was used to record the X-ray spectrum from a Neon gas jet target (with densities of the order of 10^{21} atoms/cm³) in the region of 5 - 15 Å emitted from the focus. The X-rays passed through a 10 µm Beryllium filter placed over the spectrometer entrance slit and were recorded on Kodak X-ray double emulsion film (DEF). Twenty laser shots were integrated onto the film.

The raw data (on film) was densitometered on a Joyce-Loebl densitometer using a x10 objective with a numerical aperture of 0.25. The slit was 50 µm in the spectral direction (1000 steps) by 100 µm in the other direction (40 steps). A C1232 (0.06 D per cm) wedge was used for the densitometry. Once the spectral lines in the spectrum were

identified the distance on film was converted into a wavelength scale. An image and lineout of the densitometered data, including identification of lines from Helium- and Hydrogen- like Neon, is shown in figure 5.5.2. The resonance lines and the recombination continuum of Helium-like Neon together with the resonance lines of Hydrogen-like Neon are clearly visible. The K- α line between 14 and 15 Å is from neutral Neon. No Hydrogen-like recombination continuum is present.

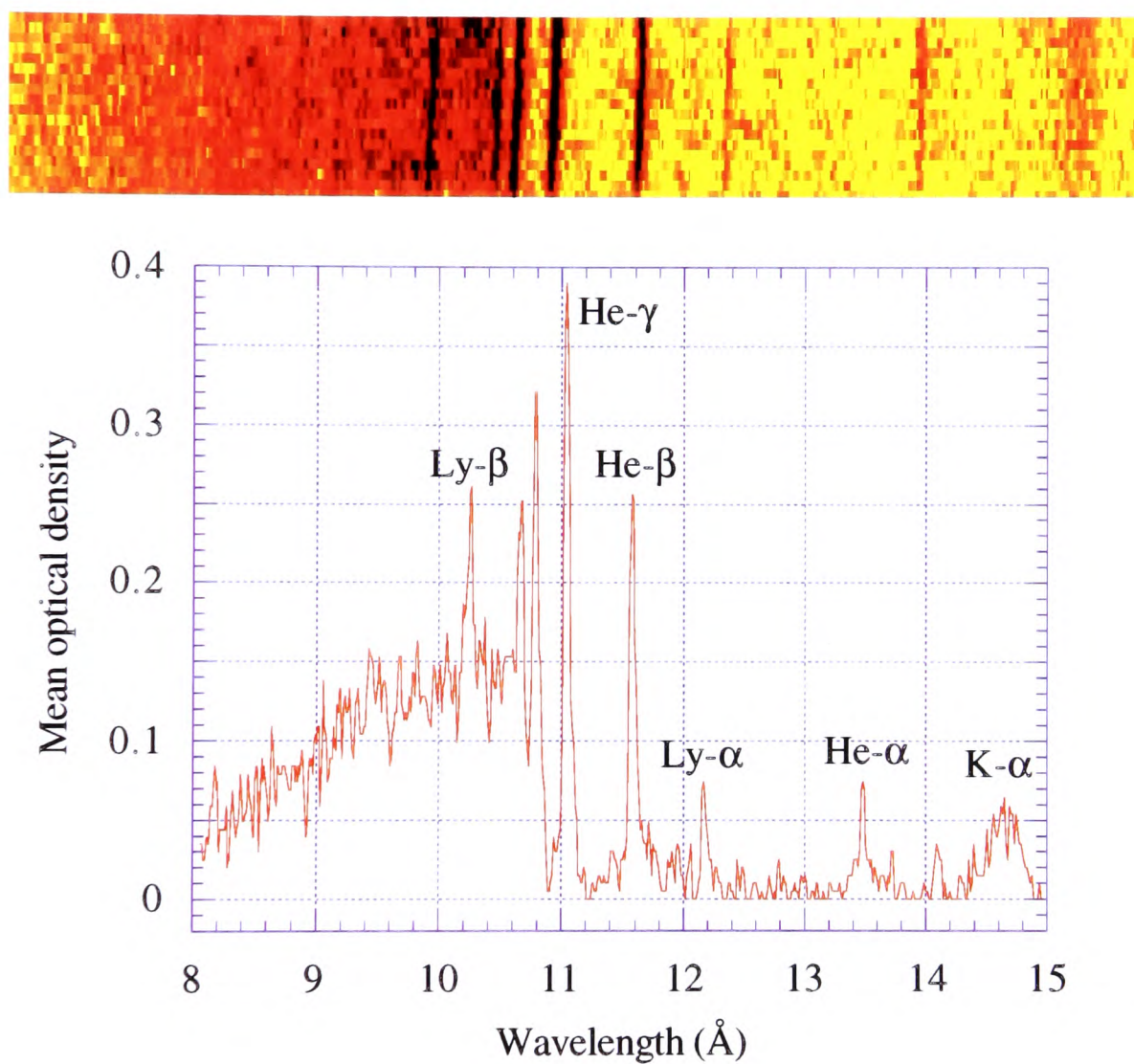


Figure 5.5.2. Densitometered image and lineout of the integrated X-ray spectrum from a 400 psig Neon gas jet with the laser focused 100 μm above the jet nozzle.

The intensity spectrum, I , was derived from the density above fog on film, D , using the formula for the low energy X-ray response of the DEF [121]

$$\alpha D = a \ln(1 + b\beta I) \quad (5.5.1)$$

$$\text{in which } \beta = [1 - \exp(-\mu_1 d)] \exp(-\mu_0 t / \sin \theta) \quad (5.5.2)$$

$$\text{and } \alpha = \mu' / (\sin \theta + \mu' d_0) \quad (5.5.3)$$

$$\text{where } \mu' = \mu_0 - (1/d) \ln(1 - V \{1 - \exp[-(\mu_1 - \mu_0)d]\}) \quad (5.5.4)$$

and θ is the angle of incidence of the photons on film, calculated from the Bragg condition, μ_m are the linear absorption coefficients of gelatin ($m = 0$), AgBr ($m = 1$), d is the effective AgBr grain thickness ($1.6 \mu\text{m}$), d_0 is the effective emulsion-surface-layer thickness, which we assume to be negligible, t is the thickness of the gelatin-like overcoat ($1.0 \mu\text{m}$), V is the volume fraction of AgBr, and a ($= 0.680 \mu\text{m}^{-1}$) and b ($= 1.69 \mu\text{m}^2$) are fitting parameters, experimentally determined for the DEF.

The values shown for the above parameters are taken from an experimental study of the Kodak DEF [147]. The background fogging level was removed by converting raw densities to intensities and plotting intensity against density. The offset from the origin gives the background fogging level which must be subtracted from the raw density on film. This gives us the intensity on film in terms of photons/ μm^2 . Next we remove the response of the spectrometer and crystal using the spectrometer dispersion and sensitivity functions (given by ref. [121]). To do this we must determine whether the resolution is crystal rocking curve or source limited.

We determine the distance, L , of the film from the source for all wavelengths, using

$$L = \frac{2d \cos \theta \sin \phi}{d\lambda/dz} \quad (5.5.5)$$

where θ is now the angle of incidence of X-rays on the crystal, $\phi = \theta + 22.5^\circ$ is the angle of incidence of X-rays on the film.

Taking $dz = 50 \mu\text{m}$, and performing a linear line fit to find a smoothly varying $d\lambda$, we find that L varies from 5.6 to 9 cm. If we assume that the source width is about $10 \mu\text{m}$, then the resolution of the spectrometer is crystal rocking curve limited for all lines except for the continuum recombination and perhaps the K- α line. This gives us a spectrometer sensitivity function, S_c , of

$$S_c = \frac{R_c \sin \phi}{L^2 \Delta\beta} \quad (5.5.6)$$

where R_c is the crystal reflectivity, and $\Delta\beta$ is the rocking curve FWHM. Dividing the intensity (in photons/ μm^2) by S_c gives us the emission of the source in terms of photons/steradian.

Finally we must correct for the X-rays having passed through $10 \mu\text{m}$ of Beryllium foil. This is done by multiplying by a factor of $\exp(-\mu_{Be} t_{Be})$, where μ_{Be} is the linear absorption coefficient, and t_{Be} is the thickness of the Beryllium foil. Multiplying the result of this by 4π gives us the total number of photons emitted by the source.

In the crystal limited case, the convolution of the spectral line with the rocking curve has already integrated the line. To obtain the number of photons in a particular line we must read off the value at its peak. The source emission for an integration of 20 shots is shown in figure 5.5.3. It is surprising that we see Helium- and Hydrogen- like X-ray emission from our Neon plasma. At the intensities we expect in the laser focus there is no chance of any optical ionisation of the 1s shell electrons of Neon. The Ammosov equation (section 2.2.2) shows that significant optical ionisation of these levels would

require an intensity of over $4 \times 10^{19} \text{ W/cm}^2$. This could only be achieved if the laser were to self-focus (relativistically or otherwise) into a channel of less than $1.5 \mu\text{m}$ diameter. MEDUSA modelling [148] predicts that, if this occurs, the X-ray photon yield would be less than 1% of that observed, even if the channel were 1 mm in length (the diameter of the gas jet).

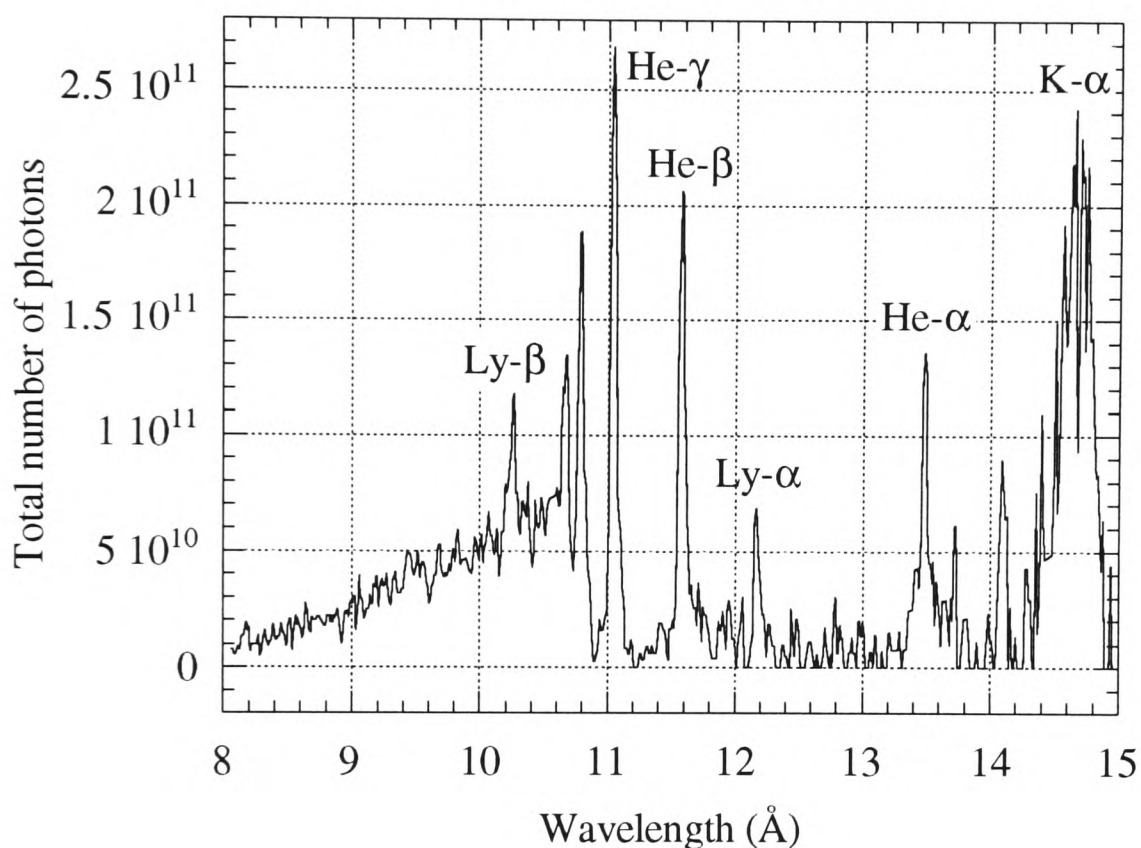


Figure 5.5.3. X-ray spectrum of 400 psig Neon corrected for film, spectrometer and filter response.

The alternative to optical ionisation as a means of production of these ionisation states is collisional ionisation. We have determined that electron temperatures of one or two keV are produced by a combination of SRS and IB. These electrons are energetic enough to collisionally excite and ionise Helium-like Neon, and to collisionally excite Hydrogen-like Neon. MEDUSA simulations [148] were performed to calculate the X-ray yield where we assumed deposition of energy by SRS and thermalisation of the electron energies. The X-ray yield was calculated with IB and SRS heating corresponding to 2% reflectivity and a focal volume of length $100 \mu\text{m}$ and radius $10 \mu\text{m}$, giving an electron

temperature of almost 2000 eV. These yields were compared to those with just IB heating. The SRS and IB heating is predicted to cause transient ionisation to Ne^{9+} and to produce X-ray photon yields that are more than five times higher than for just IB heating. These yields are consistent with those measured experimentally. We therefore attribute the presence of Ne^{9+} to collisional ionisation and associate the X-ray yield with the additional heating caused by SRS.

5.6. Summary.

In this chapter we have presented Thomson scattered and SRS spectra for Helium and Neon and discussed the electron temperatures and densities inferred from these. Thomson scattering is used to determine electron temperatures for electron densities up to about $2 \times 10^{19} \text{ cm}^{-3}$, above which plasma emission masks the signal until the onset of strong SRS. We support our analysis of SRS spectra and reflectivities with Faraday cup data and an X-ray spectrum showing line emission from Helium- and Hydrogen- like Neon. All three of these diagnostics indicate electron temperatures of the order of 1 keV in Neon at an electron density of $5 \times 10^{20} \text{ cm}^{-3}$ created by a laser with an intensity of $6 \times 10^{17} \text{ W/cm}^2$ and a pulse length of 380 fs. The conclusions of this investigation are that the plasma conditions required by the Ne VIII scheme proposed by Eder et al. [44] should be possible for a laser pulse length of 100 fs and a focused intensity of $3 \times 10^{17} \text{ W/cm}^2$ provided the laser beam quality is good. However, the onset of SRS and the increase in IB with electron density will limit the system to elements with $Z < 12$. This limits the shortest wavelength that can be produced by this scheme to about 60 Å, which will unfortunately prevent such a scheme from generating wavelengths in the water window. It also means that the OFI recombination scheme will be in direct competition with harmonic generation, which is discussed in the next chapter.

Chapter Six

Harmonic Generation

In this chapter we shall consider the production of coherent XUV radiation by the process of harmonic generation in Helium and Neon plasmas. We shall present experimental and theoretical results that show that, contrary to commonly held beliefs, a short wavelength fundamental laser can generate harmonics with both shorter wavelengths and with higher efficiencies than a long wavelength laser. (The advantages of using a short wavelength laser were discussed in section 3.3.2.3).

We shall introduce the work by presenting, in section 6.1, simulations of the expected form of the harmonic spectrum which predict that a short wavelength KrF laser is better suited for generating both higher efficiencies and shorter wavelengths than longer wavelength systems such as Nd:YAG or Ti:Sapphire. In section 6.2 we shall review experimental results and discuss the dependence of harmonic generation on laser focusing and gas densities for both Helium and Neon. We will then compare the Helium and Neon data with each other and with the simulations.

6.1. Computer simulations of harmonic spectra.

When making a full simulation of the harmonic experimental data one should consider not only the single atom response but also all the issues of beam propagation which may introduce effects such as: blue-shifting of the fundamental laser frequency; filamentation of the laser resulting in large intensity variations over a small radial scale in the focus; ponderomotive expulsion of electrons and ions from the focus which can significantly

change the plasma density; and phase matching effects which can greatly enhance the harmonic generation efficiencies, especially of the lower harmonics. The simulations presented here do not consider beam propagation, they model the single atom/ion response to the laser field. However, these numerical simulations of the individual atoms/ions can be made to match experimental data remarkably well by combining the response of the neutral single atom response with that of their ions for a full pulse [93].

In this section we will review the simulation code used to model the experimental data and try to predict the form of the harmonic spectrum for both Helium and Neon. We will compare simulations using one- and three- dimensional models of the atomic potential, and we will show that the single atom/ion harmonic response is higher for shorter wavelength driver lasers. It should be noted that the single atom/ion simulations were performed (at the request of the author) by Dr A. Sanpera of the Clarendon Laboratory.

6.1.1. The single atom computer simulation.

Our simulations are performed for a number of laser intensities in the range of 10^{15} - 10^{17} W/cm² at a fundamental wavelength of 248 nm for Helium and its ion and for the Neon atom and its charge states up to Ne³⁺. The simulated laser pulse length has a sine-squared temporal profile with a FWHM of 350 fs. These intensities range from just below to well above the saturation intensity of the neutral and it is therefore necessary to study the contributions both from neutral atoms and from ions.

Our approach is based on a step procedure where, as a first step, the time-dependent ionisation rate $P_{ion}(t)$ and harmonic generation from the atom in its ground state is calculated from the time-dependent Schrödinger equation (TDSE) in the single active electron (SAE) approximation [90,149]. The Hamiltonian in the TDSE is of the form

$$H(t) = -\frac{\hbar^2}{2m} \nabla^2 + V(r) + e\mathbf{E}(t) \cdot \mathbf{r} \sin \omega t \quad (6.1.1)$$

where all symbols have their usual meaning. $\mathbf{E}(t)$ contains the time dependence of the laser pulse.

The wavefunction $\Psi(r,t)$ is expanded as

$$\Psi(\mathbf{r},t) = \sum_0^{l_{\max}} (1/r) \chi_l(r,t) Y_l^0(\theta) \quad (6.1.2)$$

where for simplicity we consider only spherical harmonics, Y_l^m , with $m = 0$. $(1/r)\chi_l$ is the radial part of the wavefunction. This approximation is valid if the atom is initially in an s-state. Using this angular basis we obtain a set of $l_{\max}$ coupled one-dimensional partial differential equations. As the norm of $\Psi(r,t)$ reduces with time (because of ionisation) the ion population at time t is given by $P_{ion}(t) = 1 - \langle \Psi(\mathbf{r},t) | \Psi(\mathbf{r},t) \rangle$. The harmonic spectrum is calculated as

$$P(\omega) = \left| \int_0^T \ddot{d}(t) e^{-i\omega t} dt \right|^2 \quad (6.1.3)$$

where T is the duration of the laser pulse and $\ddot{d}(t)$ is the acceleration of the electric dipole moment

$$\ddot{d}(t) = \frac{d^2}{dt^2} \langle \Psi(\mathbf{r},t) | z | \Psi(\mathbf{r},t) \rangle \quad (6.1.4)$$

The acceleration is calculated using Ehrenfest's theorem

$$\ddot{d}(t) = -\langle \Psi(\mathbf{r}, t) | \frac{\partial}{\partial z} [V(r) + H_{\text{int}}] | \Psi(\mathbf{r}, t) \rangle. \quad (6.1.5)$$

Once the ionisation and harmonic generation for the neutral is calculated, we solve the TDSE for the ion, and then the ion's wavefunction is weighted by the time-dependent ion population function $P_{\text{ion}}(t)$ previously calculated. In this step the double ionisation function $P'_{\text{ion}}(t)$ and the harmonic generation from the ion is obtained. The procedure is repeated for each charge state of the atom we want to consider. In this way, the problem of solving the time propagation of an N-electron atom is reduced to solving N one-electron wavefunctions, where each wavefunction includes the ionisation of the previous charge states along the pulse [93].

The experimental Helium data was modelled using this approach with $V(r)$ set to an effective potential using the SAE approximation for the neutral and set to a hydrogenic potential for the ion. However, due to the extreme demands on computation, we have also used a 1D soft core model of the form $V(x) = Z_{\text{eff}} / (a^2 + x^2)^{1/2}$ (where a is chosen to correctly match the ionisation potential of the atom) to calculate the response of Neon and its ions. We have also used this potential to model Helium and its ion to compare the form of the one- and three- dimensional simulations and to compare the Helium response to short and long wavelength lasers.

The saturation intensities of Neon, Ne^+ , Ne^{2+} and Ne^{3+} are well below 10^{17} W/cm^2 and therefore they are mostly ionised before the experimental peak laser intensity. Although higher ionised states of the atom, i.e. Ne^{4+} and above, have higher saturation intensities and could therefore experience larger intensities before being completely ionised, their contribution to the harmonic generation is negligible [57]. Since they are so tightly bound they require a high intensity to ionise. At such intensities the classical velocity that

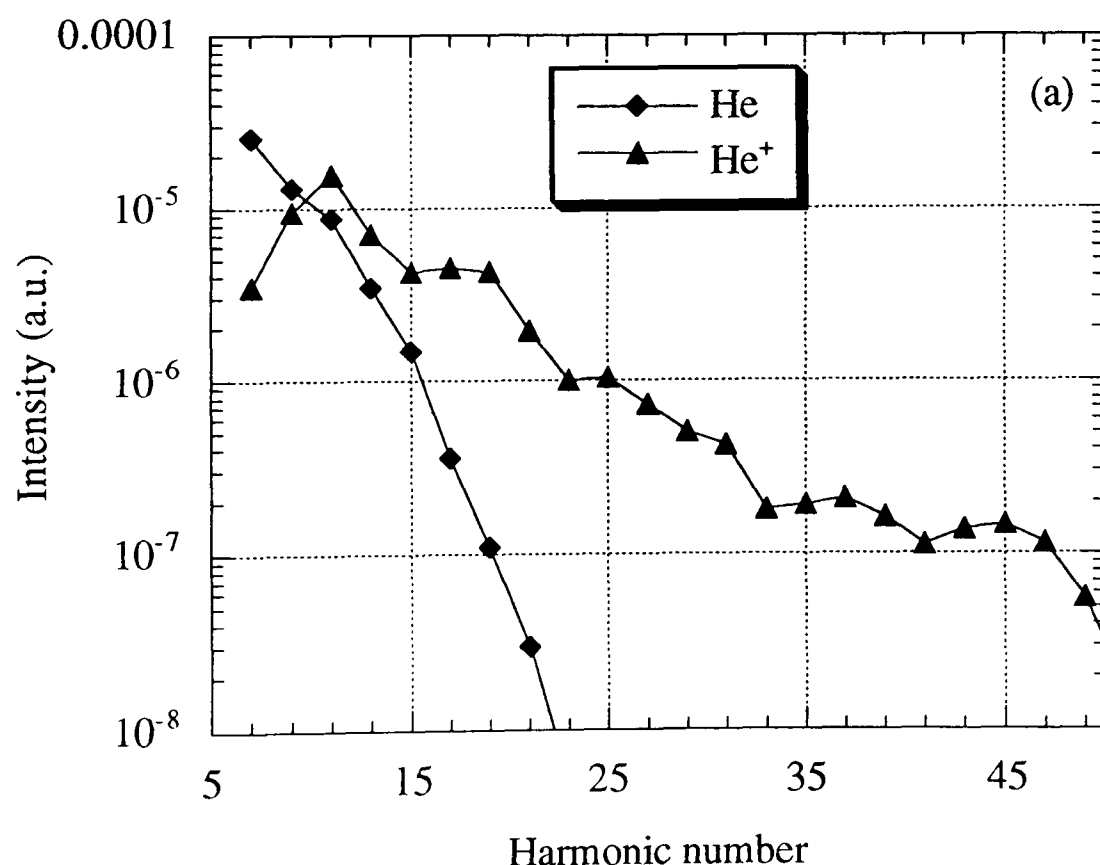
the freed electron experiences is so high that the recollision cross section with the nucleus is extremely low.

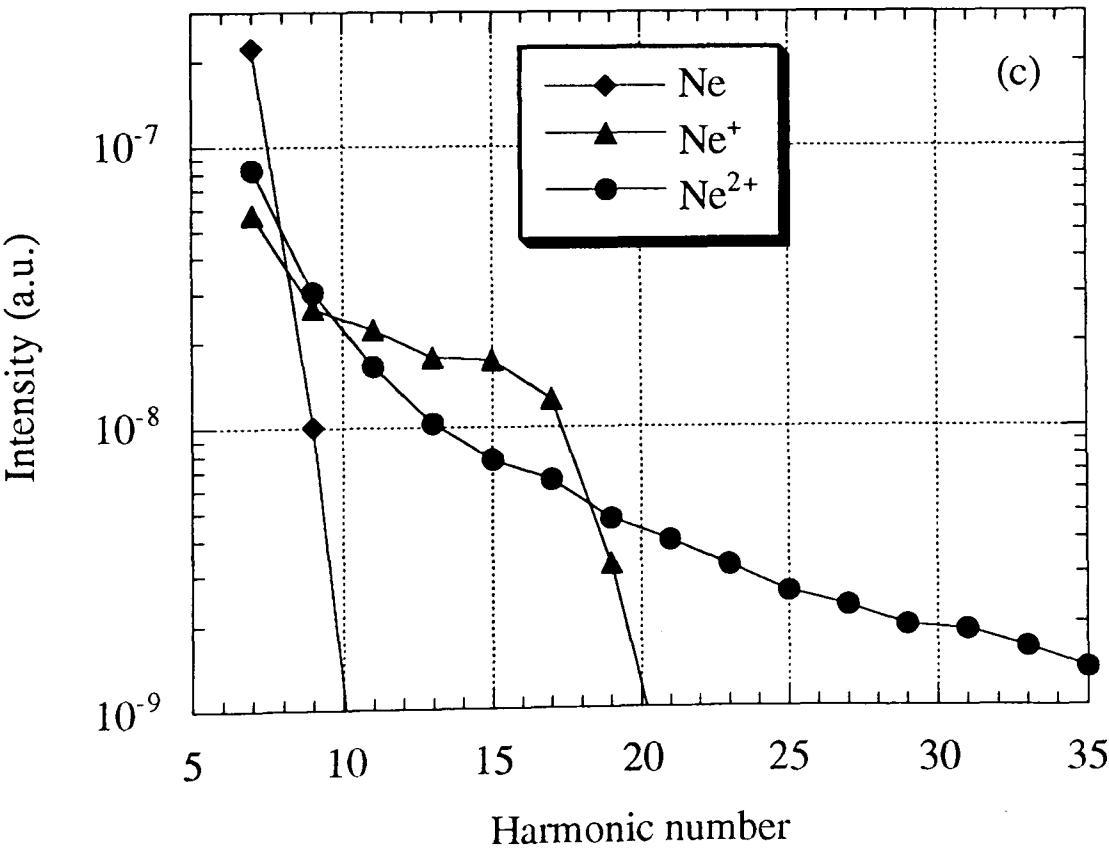
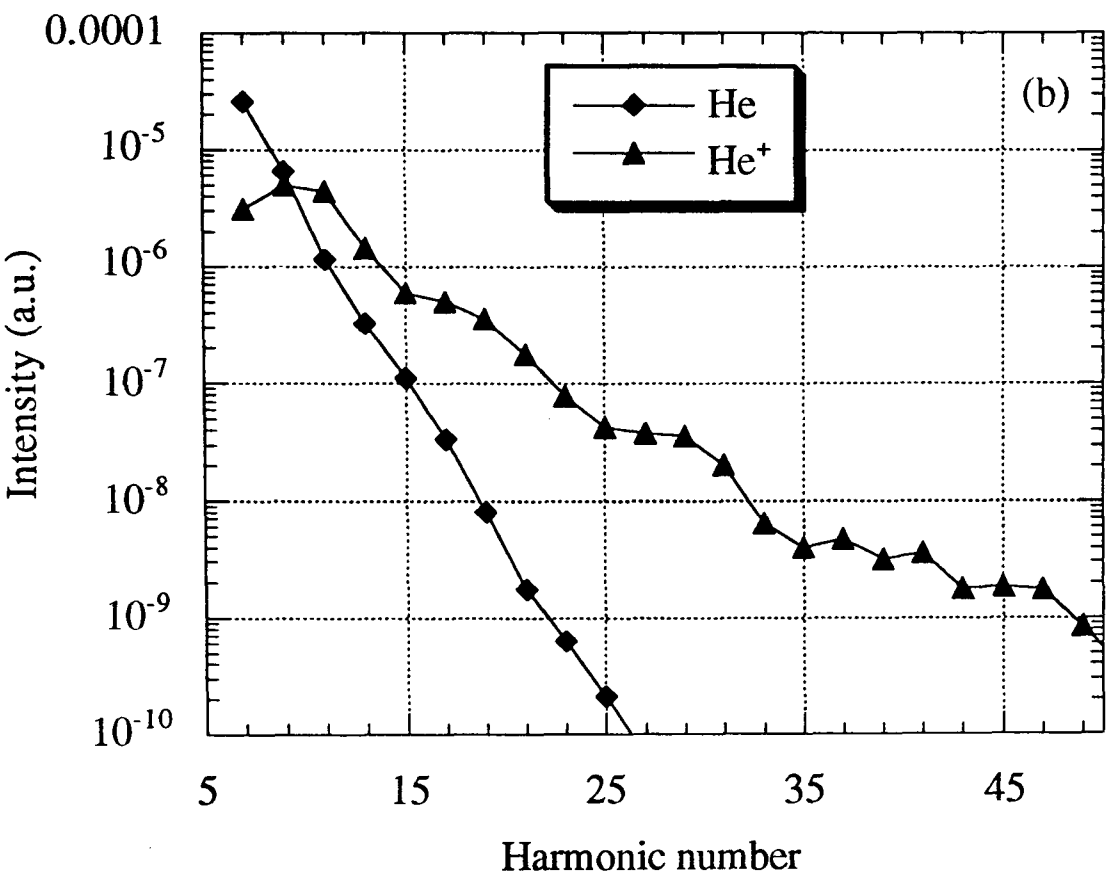
6.1.2. Predictions of the harmonic responses of Helium and Neon.

The simulations include temporal variations in the laser intensity, but ignore spatial variations in the laser intensity and phase. In a real laser focus, not all atoms experience a single, uniform intensity, the laser intensity will vary along and radially outwards from the centre of the focus. To simulate the laser focus more realistically we can make a superposition of the atom/ion response for varying laser intensities, weighted for the relative volume in the focus at which these intensities occur (assuming a Gaussian, TEM₀₀, beam profile). The result of such a superposition is to enhance the generation efficiency for the lower harmonics with respect to the higher harmonics, as they will be produced over a much larger volume. To allow for phase matching effects, we can also weight the response by a q^{-2} factor (where q is the harmonic order). This factor is due to the dependence of the intensity of the q th harmonic on the square of the length of the non-linear medium (equation 3.3.29). This length, however, is determined by the coherence length, which (when phase matching is dominated by free electrons) is proportional to $1/q$ (equation 3.3.35). A full propagation code that includes a simple one-dimensional calculation of the harmonic response has been developed [102], but in its present form it can only predict harmonic response from atoms. This means that it cannot be used to simulate our experiments in which the ion response dominates for most of the harmonic spectrum.

In figure 6.1.1 we present a comparison of the uniform intensity and the weighted harmonic response for both Helium and Neon. Even this simple weighting significantly alters the predicted harmonic response. The higher harmonic efficiencies are reduced by almost two orders of magnitude with respect to the low harmonics when compared to a

uniform intensity response. This effect can, of course, be achieved by selecting a single fundamental intensity that gives the correct response, but this chosen intensity is completely arbitrary in relation to the real intensities experienced by atoms in the focus. The match of the form of this simple Gaussian beam fit to the experimental data (discussed in section 6.2.4) is quite remarkable. The weighted responses of Helium, figure (b), and Neon, figure (d), have been normalised at the 7th harmonic to the single intensity responses (a) and (c). Although the figure seems to indicate a higher harmonic response from Helium, it should be remembered that the Helium response has been calculated using a three-dimensional atomic potential, compared to the one-dimensional potential used for Neon. However, we do expect the general shape and cut off of the harmonic plateau to be reasonably well modelled by both simulations.





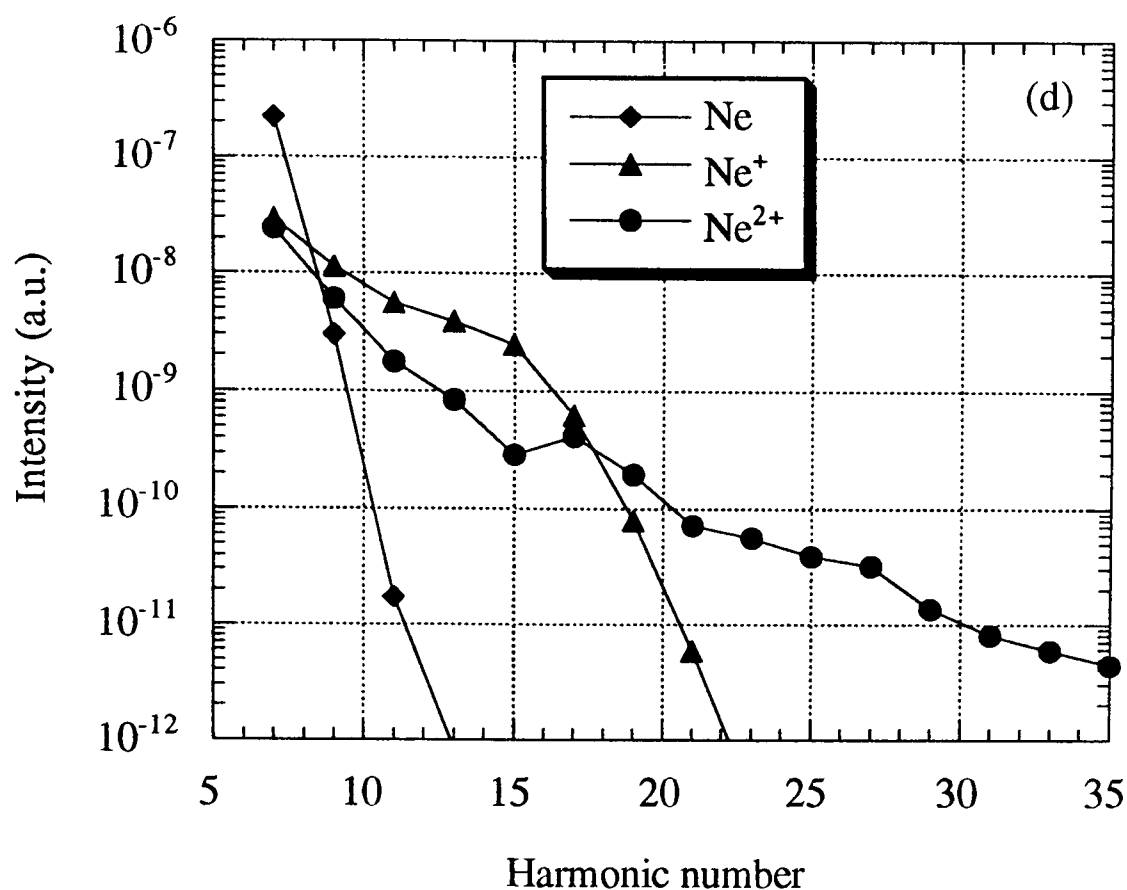


Figure 6.1.1. Comparison of simulations of Helium (a) at a single intensity of 10^{16} W/cm² and (b) with volume and q^{-2} weighting. The same comparison is made for Neon in (c) and (d), but with the single intensity being 1.2×10^{16} W/cm². In both cases the weighted response is normalised to the single intensity response.

To indicate the difference between one- and three- dimensional potentials, we show in figure 6.1.2 a one-dimensional calculation for Helium at 10^{16} W/cm². Note the much sharper cut off of the neutral response in the one-dimensional case. In fact the response of the neutral drops to a negligible level after the 11th harmonic. If we compare this result with the Neon simulation, we see that the comparable simulations now predict that Helium exhibits a higher single atom/ion conversion efficiency. This is an interesting result, as many experiments have indicated that Neon should be the more efficient, but with the Helium response reaching to higher harmonics [97,105]. An explanation of this discrepancy is that the intensities used in these simulations are above the optimum for

Neon. We have optimised our fundamental laser intensity to produce the highest energy harmonics, not the highest efficiencies.

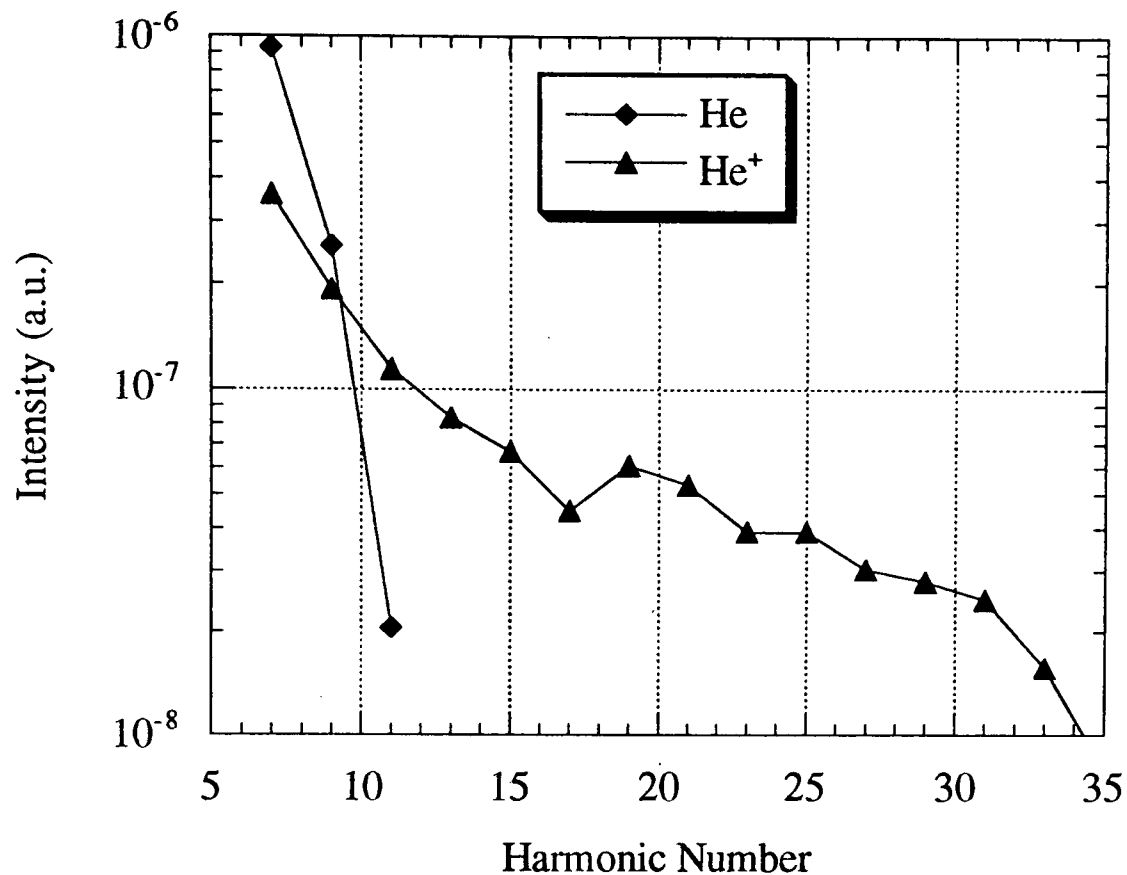


Figure 6.1.2. One-dimensional simulation of Helium and He⁺ at an intensity of 10^{16} W/cm².

Note how the one-dimensional neutral Helium and Neon responses both fall off very rapidly with increasing harmonic order. If the differences seen between the one and three-dimensional Helium simulations can be applied to the case of Neon, then we might expect that the neutral harmonic response will not give way to the ion response as the dominant generator quite as quickly as expected. However, in analogy with the case of Helium, it is reasonable to expect this changeover to occur at nearly the same harmonic.

Another difference between the one- and three- dimensional Helium simulations is the relative magnitudes of the neutral and ion responses. Although it shows a much sharper cut off, the one-dimensional result indicates that the neutral response at the 9th harmonic is much higher than the ion response, whereas the full simulation shows the two to be

much closer. It is this factor which causes the two Helium simulations to show a changeover from neutral to ion response at the same harmonic: the larger difference between the two responses counters the faster fall off. When comparing this aspect of the simulation with experiment, interpretation is somewhat complicated by the possibility that phase matching may be dominated by the laser focusing conditions for the neutral, and by free electrons for the ion. We also note the considerable difference in the plateau cut off harmonic between the two simulations. The one-dimensional simulation cuts off sharply at about the 31st harmonic compared to the three-dimensional case, which shows a slower fall off starting at the 47th harmonic.

Before we move on to the interpretation of experimental results we present in figure 6.1.3 a one-dimensional Helium simulation conducted at a wavelength of 1000 nm (rather than 248 nm used in the above calculations). The most striking feature in the figure is the almost complete disappearance of the ion response. This confirms our simple argument in section 3.3.2.3 that shorter wavelengths are more efficient than long wavelengths for the generation of high harmonics from ions. All parameters in the simulation are the same as for the simulation shown in figure 6.1.2. (It should be noted here that, for an equivalent harmonic number, the 248 nm harmonic will have a wavelength which is a factor of four shorter than the 1000 nm harmonic). The atomic harmonic response for a laser wavelength of 1000 nm is much lower than that predicted for the ionic response for 248 nm. This is mainly due to the intensity of 10^{16} W/cm² being well above the saturation intensity for neutral Helium (as opposed to the saturation intensity for He⁺). To optimise the atomic response would require an intensity of the order of 10^{14} W/cm². A fairer comparison of the neutral response would have to be carried out at the saturation intensity of the neutral for each wavelength. Figures 6.1.2 and 6.1.3 show that short wavelengths must be used for the generation of harmonics from ions.

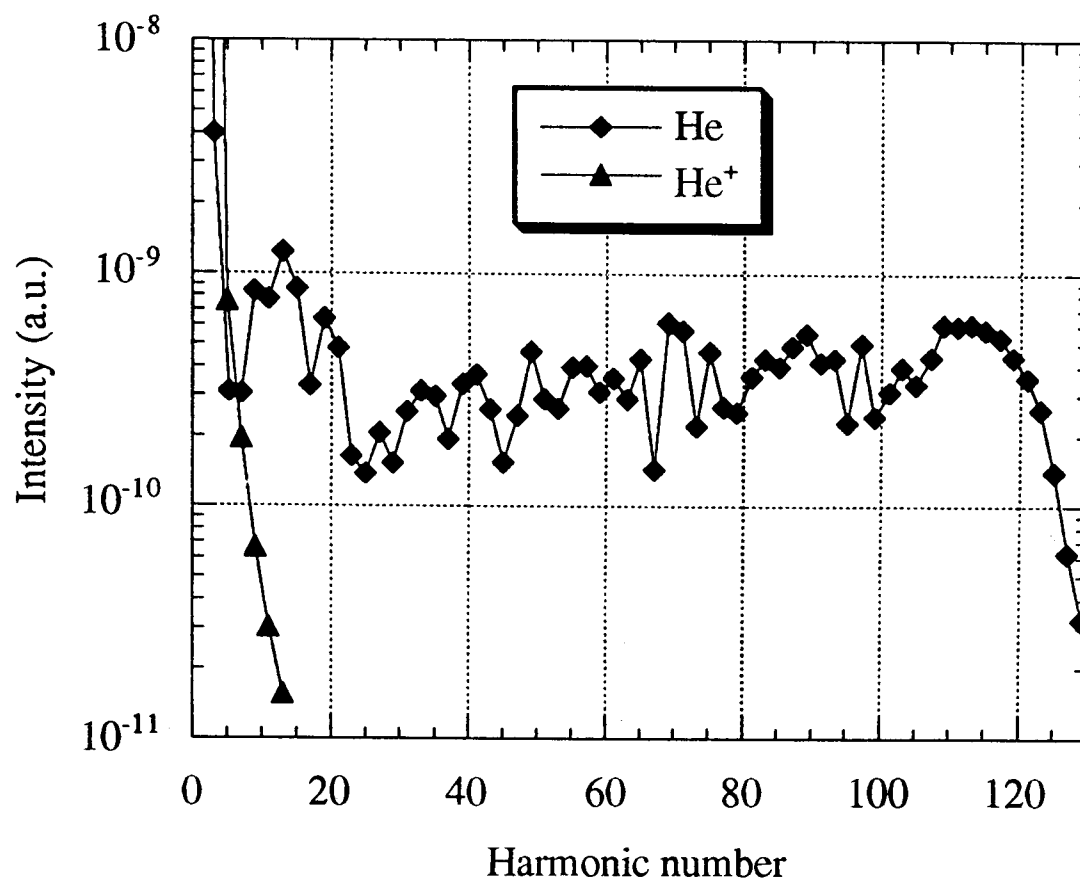


Figure 6.1.3. One dimensional simulation of Helium and He^+ at a fundamental laser wavelength of 1000 nm and an intensity of 10^{16} W/cm^2 . All other conditions in the simulation are as for figure 6.1.2.

6.2. Harmonic generation measurements.

We will now present results of the harmonic generation experiments performed on the Sprite KrF CPA laser system using f/10 and f/3 OAP focusing. The configuration of the experiment is described in section 4.2. The f/10 and f/3 experiment were conducted over a ten week period from October to December, 1994. This was a combined Thomson scattering and harmonic generation experiment, with most emphasis being placed on harmonic generation. The Thomson scattering diagnostic allows us to infer an electron density of the plasma produced during the harmonic generation. A preliminary analysis of the results that follow has been sent to Physical Review A for publication [21].

6.2.1. High harmonic generation in Helium.

6.2.1.1. Harmonic spectra.

The harmonic response of Helium was found to be remarkably independent of the laser focusing and of the gas density. Figure 6.2.1 shows data taken using the f/3 OAP to focus the laser 1 mm above a 1 mm circular aperture nozzle solenoid valve gas jet with a backing pressure of 20 bar gauge. Figure 6.2.2 shows data for the same laser focusing conditions, but with a gas jet backing pressure of 2 bar gauge. Although the backing pressure varies by a factor of ten (2 bar to 20 bar), the highest first order harmonic seen in both images is the 33rd. However, it should be noted that the high noise level to the left of SN19970 is caused by a hole in the Carbon filter letting stray UV and XUV radiation onto the detector which masks the spectrum after the 33rd harmonic.

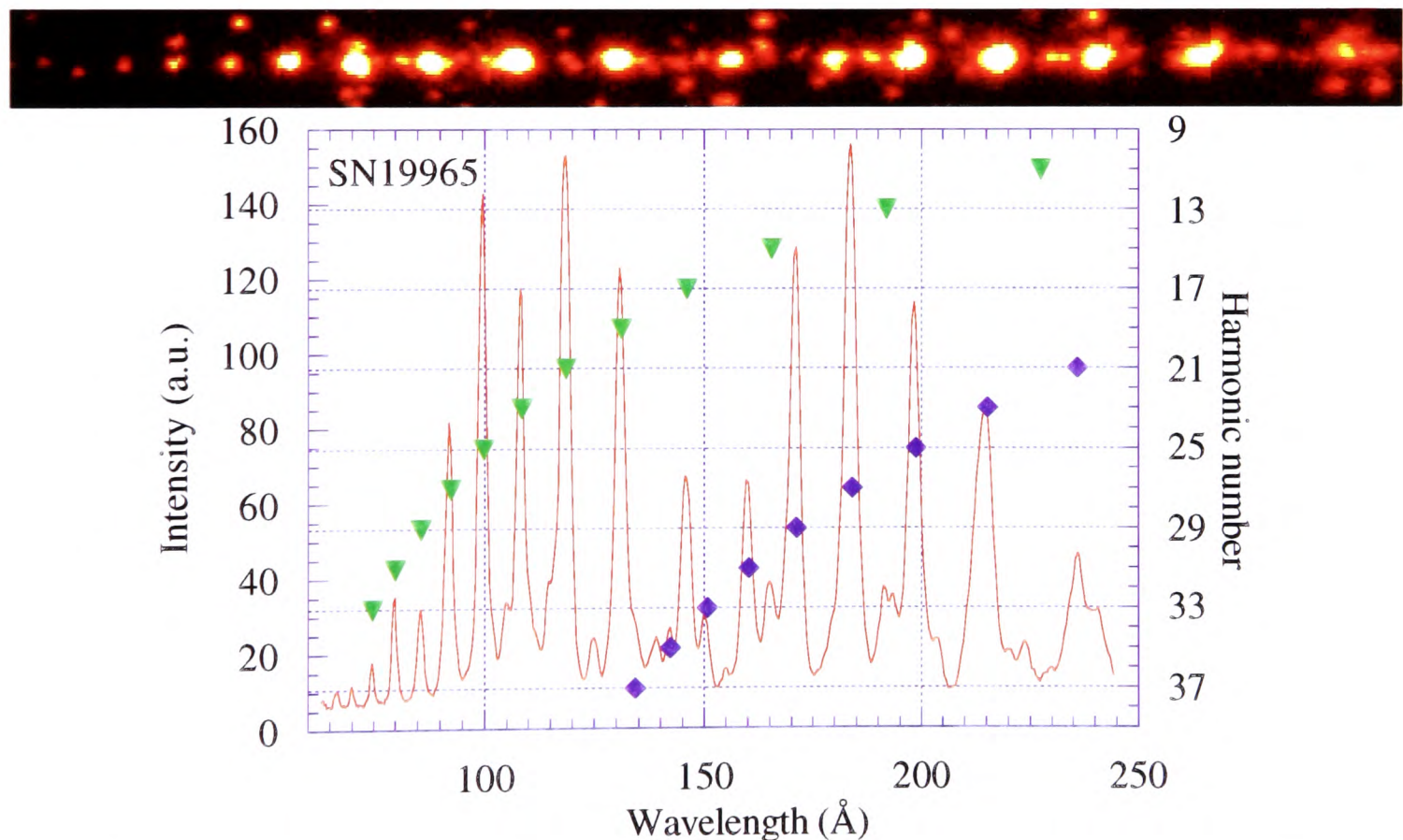


Figure 6.2.1. Image and lineout of a typical harmonic spectrum from Helium using f/3 focusing (SN19965). The backing pressure of the 1 mm circular orifice gas jet was 20 bar gauge. The harmonics are labelled as triangles (first order spectrum) and diamonds (second order spectrum).

The diffracted images also show very high levels of second order spectrum. The reason for this is that the grating blaze angle is optimised for first order diffraction at wavelengths between 100 and 300 Å. This means that at wavelengths below about 100 Å the grating should be optimised for second order diffraction. As we move to higher harmonics in the second order signal, the reduced dispersion combined with the relatively poor instrument resolution causes the second order signal to overlap the first order signal. It can be seen in the figure that, after the 17th harmonic (first order), the second order image appears as wings on the first order (as opposed to separate spots). This makes identification and analysis of higher order harmonics in second order (where the grating efficiency should be higher) rather difficult. In section 6.2.5 we shall devote some time to searching for higher harmonics in this second order signal. In this section we shall concentrate on analysing the first order spectrum.

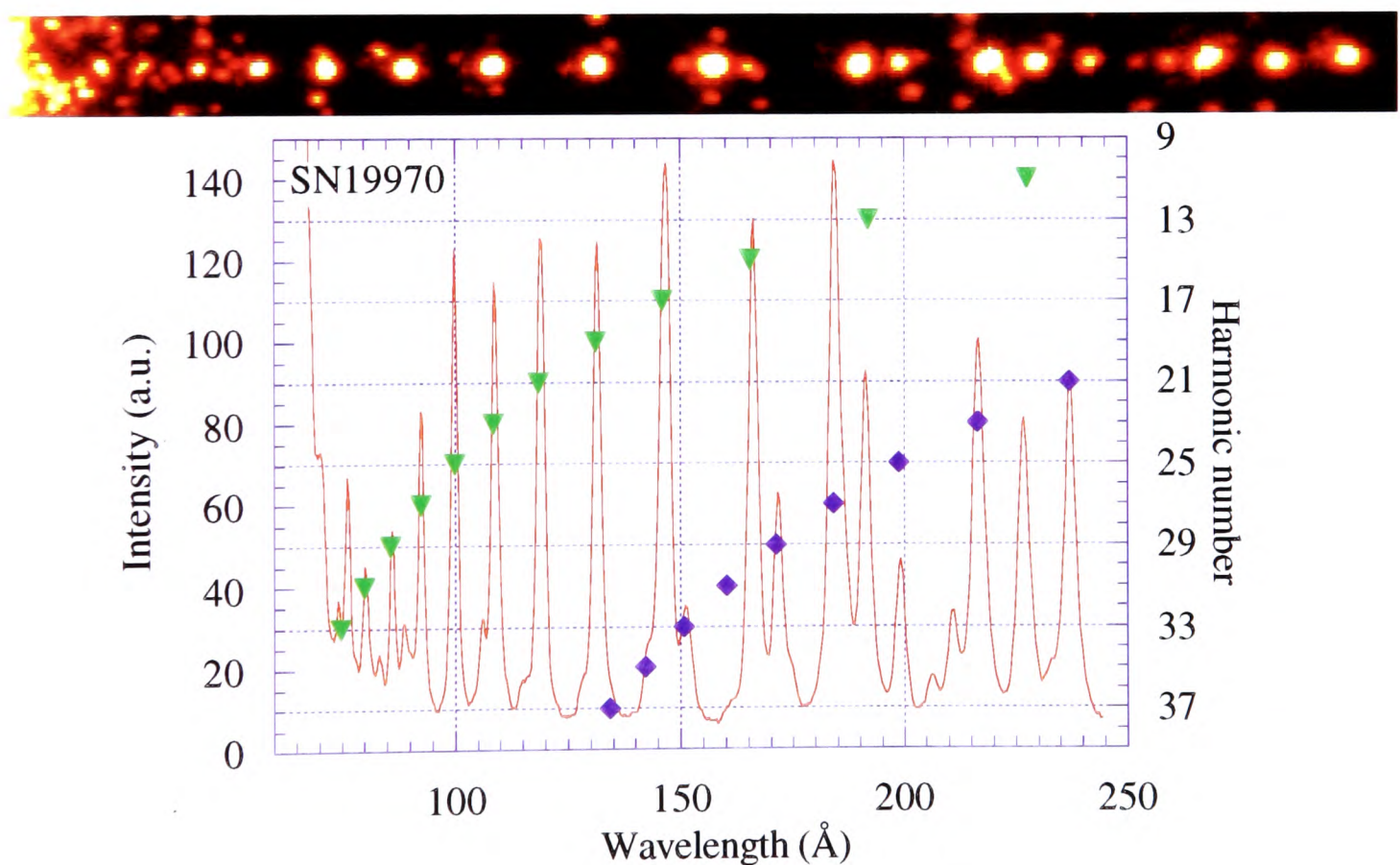


Figure 6.2.2. Image and lineout for $f/3$ focusing (SN19970). The gas jet backing pressure is 2 bar gauge. The harmonics are shown as triangles (first order spectrum) and diamonds (second order spectrum).

Figure 6.2.3 and figure 6.2.4. show the images and lineouts for data using the f/10 OAP. The laser focusing conditions are described in the figure captions. The Helium backing pressures for the two shots are 0.25 and 9.7 bar gauge respectively, giving gas densities at the laser focus of 5×10^{17} and $\sim 5 \times 10^{18} \text{ cm}^{-3}$.

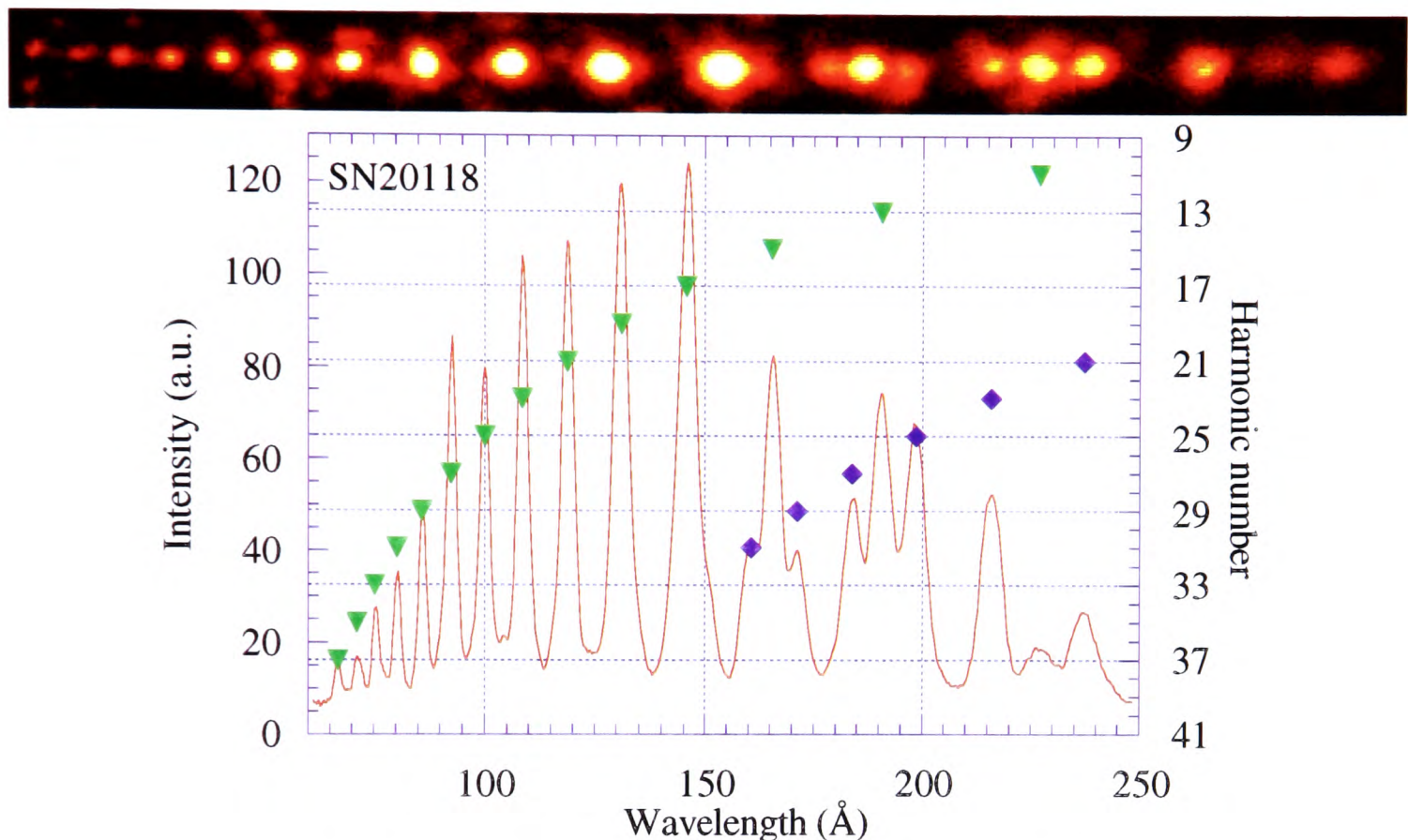


Figure 6.2.3. Image and lineout of SN20118 (Helium). The laser was focused 1 mm above and 0.5 mm beyond the 0.5 mm x 1 mm gas jet nozzle with the 0.5 mm dimension along the laser focus. The gas jet backing pressure was 0.25 bar gauge.

SN20118 is slightly blurred, as the fibre-optic coupler from the MCP to the CCD camera was not quite in contact with the CCD camera. This shot has been shown, however, because it shows most clearly a first order image of the 37th harmonic. Again we see that a factor of approximately ten in gas density does not significantly alter the form of the harmonic spectrum.

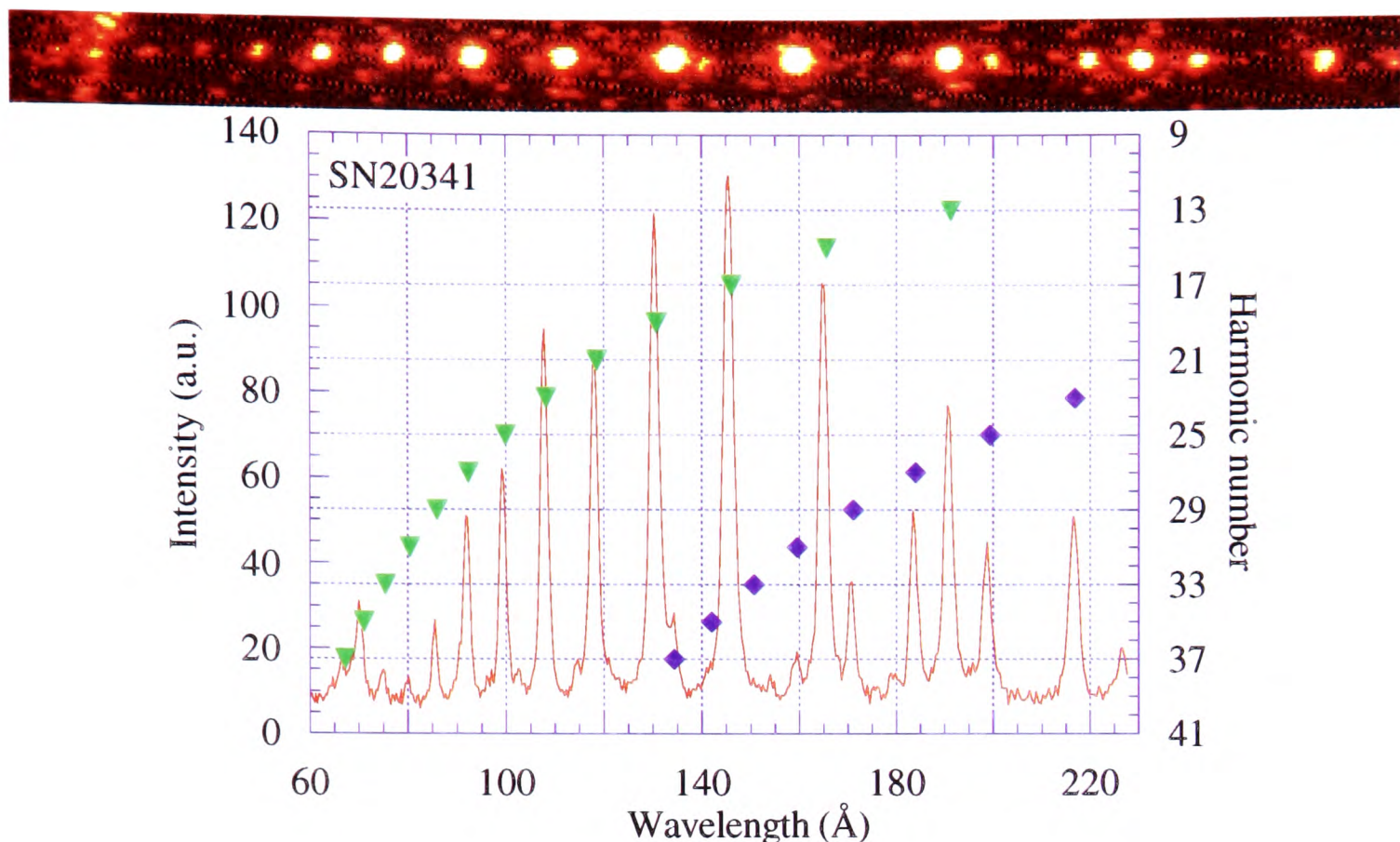


Figure 6.2.4. Image and lineout of SN20341. The laser focusing is the same as for SN20118 (figure 6.2.3), but the gas jet was oriented with its 2 mm dimension along the laser focus and the backing pressure was 9.7 bar gauge.

6.2.1.2. Harmonic efficiencies.

To analyse the harmonic response more quantitatively we must integrate the total energy of each harmonic. In order to compare harmonic spectra produced by different laser intensities (caused by unavoidable shot-to-shot variations in the laser energy and focal spot profile) we will present here the conversion efficiencies for each harmonic. Conversion efficiencies are calculated assuming that the total solid angle of emission of the harmonic was equal to that of the laser beam. Further calibration factors came from the absolute response of the MCP [122], the grating reflectivity [123], the filter transmission, the collection solid angle of the spectrometer, and the focusing mirror reflectivity and its collection solid angle. These factors are discussed in section 4.3.2. The angular distribution of the harmonics was measured in a separate series of experiments using the 33 cm OAP, and without the focusing mirror [150]. The absence

of the mirror reduced the overall detection sensitivity, so that in these experiments only harmonics up to the 19th were observed. However, all the harmonics observed covered the full solid angle of the laser beam and showed no reproducible angular structure. (This is contrary to the results of experiments using a diffraction limited beam [151] which see angular structure and significant narrowing of harmonic emission near the cut-off.) An image of the emission and harmonic spectrum from low pressure Argon is shown below in figure 6.2.5, in which the harmonic lines cover the full aperture of the detector. For this reason the energy conversion measurements assume harmonic emission into the full beam angle. The assumption is validated for harmonics up to the 19th, but conversion into higher harmonics could be over-estimated if the angular distribution of the beam narrows significantly. An upper bound of a factor of 40 on the possible error for data recorded with the 100 cm OAP is given by the fact that the acceptance solid angle of the spectrometer was 1/40th that of the f/10 laser beam. The possible error on the f/3 data is a factor of $(10/3)^2$ higher. Another slight error comes from the second order harmonic spectrum which, after the 17th harmonic (in first order), cannot be separated from the first order spectrum and will contribute slightly to the integrated pixel count of each harmonic. Figure 6.2.6 shows the absolute harmonic conversion efficiencies for the data presented in figures 6.2.1 to 6.2.4. The harmonic conversion efficiency is the ratio of the total energy of a given harmonic (calculated from integrating the pixel count above noise) to the laser energy delivered on target.

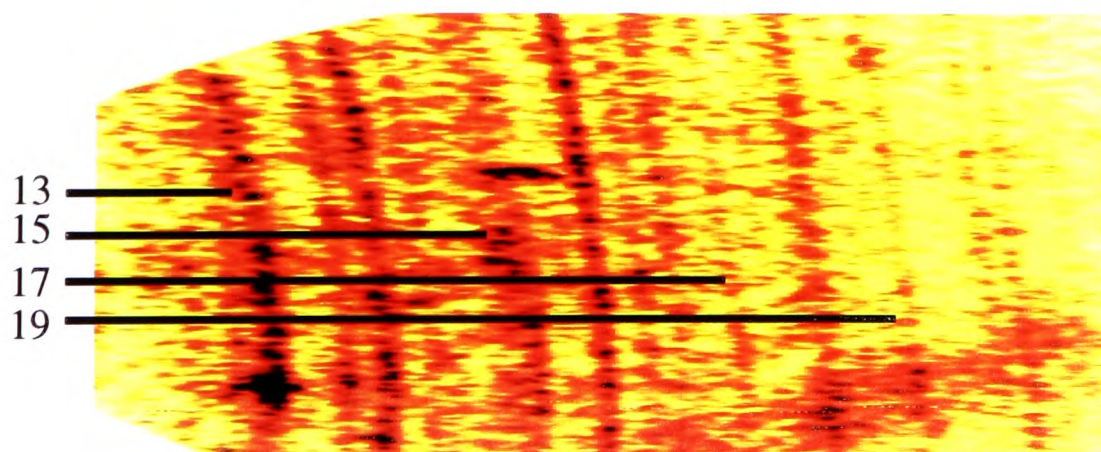


Figure 6.2.5. *Harmonic and line emission spectrum from low pressure Argon without a focusing mirror.*

The differences between the two focusing configurations now become apparent. The efficiencies from the f/10 focusing are between a factor of 3 and 10 higher than for the f/3 focusing. Gas density appears to have no real effect for the f/3 data, whereas there seems to be a very slight enhancement of the high pressure f/10 data over the low pressure data. If harmonic generation is unsaturated (i.e. is generated over less than a coherence length) we would expect the harmonic yield to scale as the square of the density. This would give a difference in efficiency of about two orders of magnitude between the low and high density data. The actual difference is no more than a factor of three, showing that we are in a heavily saturated regime where the coherence length is much smaller than the laser confocal parameter.

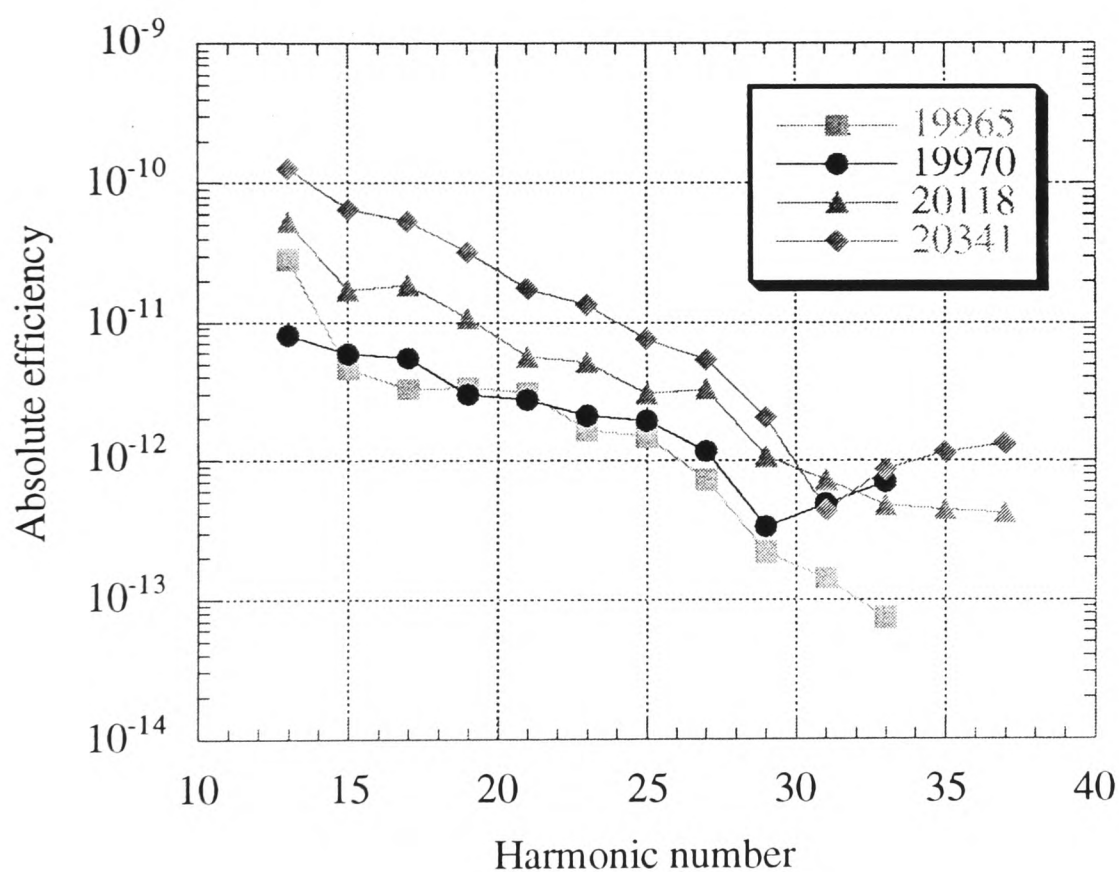


Figure 6.2.6. The absolute conversion efficiencies for harmonics generated from Helium. The data is for the same laser shots as shown in figures 6.2.1 to 6.2.4.

The dependence on focusing can be attributed to two primary effects. The first is that with a longer confocal parameter a larger volume of gas experiences intensities high

enough for high harmonic generation to occur. At the lower densities, this longer confocal parameter may also extend the effective coherence length over which harmonic generation occurs. For the conditions in our experiment, we expect that the coherence length will be determined by the free electron refractive index (equation 3.3.35). This means that the coherence length will not increase with increasing confocal parameter. However, the area of our focused is determined by our focused spot size, which we measure to have a FWHM of $\sim 9 \mu\text{m}$ for f/3 focusing and $\sim 18 \mu\text{m}$ for f/10 focusing (see section 4.1.3). This should cause efficiencies produced from f/10 focusing to be about four times those with f/3 focusing. This is consistent with the difference seen in figure 6.2.6. The second reason for the higher efficiencies with f/10 focusing is that the longer focal length OAP, in producing a larger focal spot size, also creates a lower intensity at focus. This means that the ionisation rate of Helium and He^+ will be somewhat lower than for the f/3 OAP. This will cause harmonic generation to occur over a longer fraction of the laser pulse before all the Helium gas is fully ionised to He^{2+} (which obviously does not generate harmonics).

6.2.1.3. Intensity dependence of the 7th to 13th harmonics.

The wavelength coverage of the spectrometer and MCP is such that the spectrometer can cover any harmonic above the 7th, but the active area of the MCP only covers about $200 \text{ \AA}$. So, in order to study the lower harmonics (7th to 13th) the MCP has to be moved. This means that conversion efficiencies for these lower harmonics are from a different sequence of shots than the higher harmonics. Figure 6.2.7 shows the dependence of the 7th to 13th harmonics to a focal scan where the gas jet is driven along the direction of the laser (which is focused using the f/10 OAP). Figure 6.2.7.(a) shows the intensity dependence on gas jet position where positive numbers indicate that the gas jet was upstream of the focus (i.e. the laser focuses beyond the gas jet). Figure 6.2.7.(b) shows the focal scan as an intensity variation seen at the centre of the gas jet where the

intensities when the gas jet is positioned at the focus (0 mm) are calculated assuming a Gaussian focus, and the intensities when the gas jet is further than 1 mm away from the laser focus have been calculated using geometric optics. The gas jet nozzle used in this scan has a 2 mm x 0.5 mm rectangular aperture aligned with the 2 mm dimension along the direction of the laser. The gas jet backing pressure is 0.2 bar gauge.

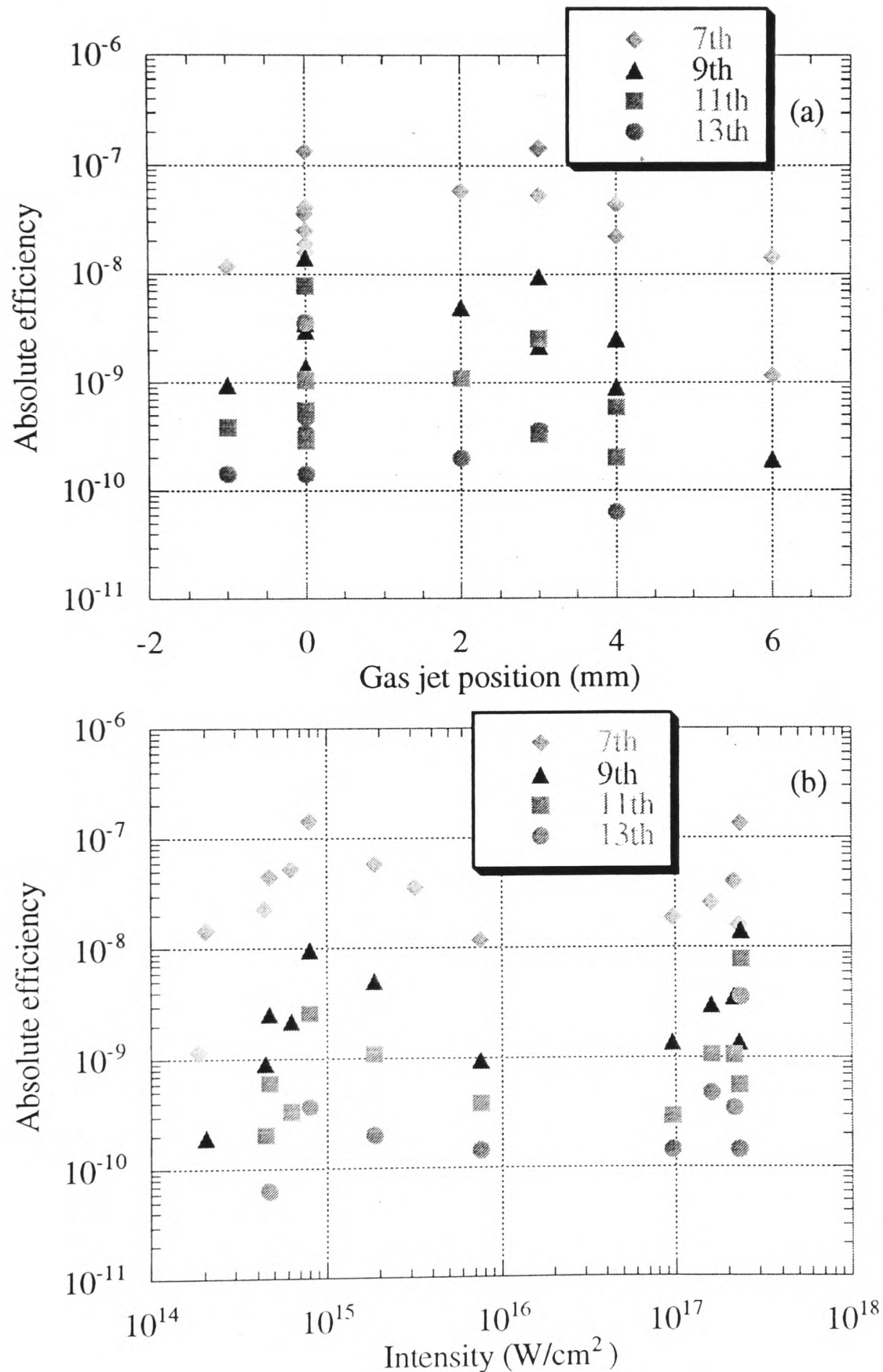


Figure 6.2.7. Harmonic conversion efficiencies in Helium as a function of (a) gas jet position and (b) laser intensity for the 7th to 13th harmonics.

It is interesting to see from figure 6.2.7 that a simple calculation of the laser intensity from geometric optics indicates that (apart from one laser shot which may have had a very poor focal spot) the highest efficiency harmonic generation from neutral Helium is at an intensity of approximately 10^{15} W/cm². Neutral Helium is the dominant generator of the 7th, 9th and possibly 11th harmonics and our ionisation rate plot in figure 2.2.2 shows that the threshold intensity for the complete ionisation of Helium to He⁺ in a 380 fs pulse is at about 2×10^{15} W/cm². This highlights the fact that harmonic generation in the strong field limit is a consequence of optical ionisation. The electron tunnels out of the atomic potential, and then emits a harmonic photon when it is later re-incident on the atomic core and has some probability of recombining with it. If recombination does not occur then the atom has been optically-ionised. Hence, the optimum intensity for harmonic generation is the threshold intensity for optical ionisation. An alternative argument is that we are seeing some sort of fringe structure in the non-linear polarisation of the Helium target [96,98]. However, the large scale of this variation compared to the laser confocal parameter (~ 300 μ m) makes this very improbable.

Using the f/10 OAP we generate over 30 nJ at 355 Å (7th harmonic), i.e. a conversion efficiency of 10^{-7} . If we assume that this energy is generated over the entire length of the laser pulse (380 fs) this corresponds to a power of over 80 kW. In reality He⁺ is fully ionised long before the peak of the pulse which implies that harmonic powers are much higher than this and that the pulse length of the harmonic is less than the laser pulse length. It is interesting to compare these efficiencies with the highest reported elsewhere (by Ditmire et al. [107]) which were obtained using 526 nm light. Our efficiency at 355 Å is approximately the same as that at 251 Å from 526 nm, and at approximately the same XUV wavelength our efficiencies are about a factor of two lower than Ditmire et al's results. It remains to be explored whether further increases in efficiency with the KrF laser would be possible with a larger focal spot and longer confocal parameter (f/70 focusing) as used in the 526 nm work. The present results certainly suggest that the

trend towards better conversion efficiency at a given harmonic wavelength using a shorter primary laser wavelength may continue beyond 526 nm.

6.2.2. High harmonic generation in Neon.

6.2.2.1. Harmonic spectra.

The harmonic response in Neon was found to be much more dependent on focusing than that of Helium. Unfortunately, we only have data taken with 20 bar backing pressure with the f/3 OAP, and so we cannot comment on the density scaling at the shorter focal length. A typical image and lineout of such a spectrum is shown in figure 6.2.8. This data was taken with the laser focused 1 mm above a 1 mm circular aperture gas jet nozzle and with a gas jet backing pressure of 20 bar gauge.

A problem that arises with Neon is that the many ionisation stages that can be produced with a focused intensity of $>10^{17}$ W/cm² emit numerous spectral lines in the wavelength region we are studying. These spectral lines form a fairly uniform background to the harmonic spectrum, as can be seen in figures 6.2.8 to 6.2.10. This causes problems when calculating the absolute efficiency of each harmonic, as great care must be taken when determining the level of this background radiation. Background radiation also increases the detection threshold of our spectrometer, preventing us from detecting harmonics that may be present if their intensity is less than that of the spectral line emission.

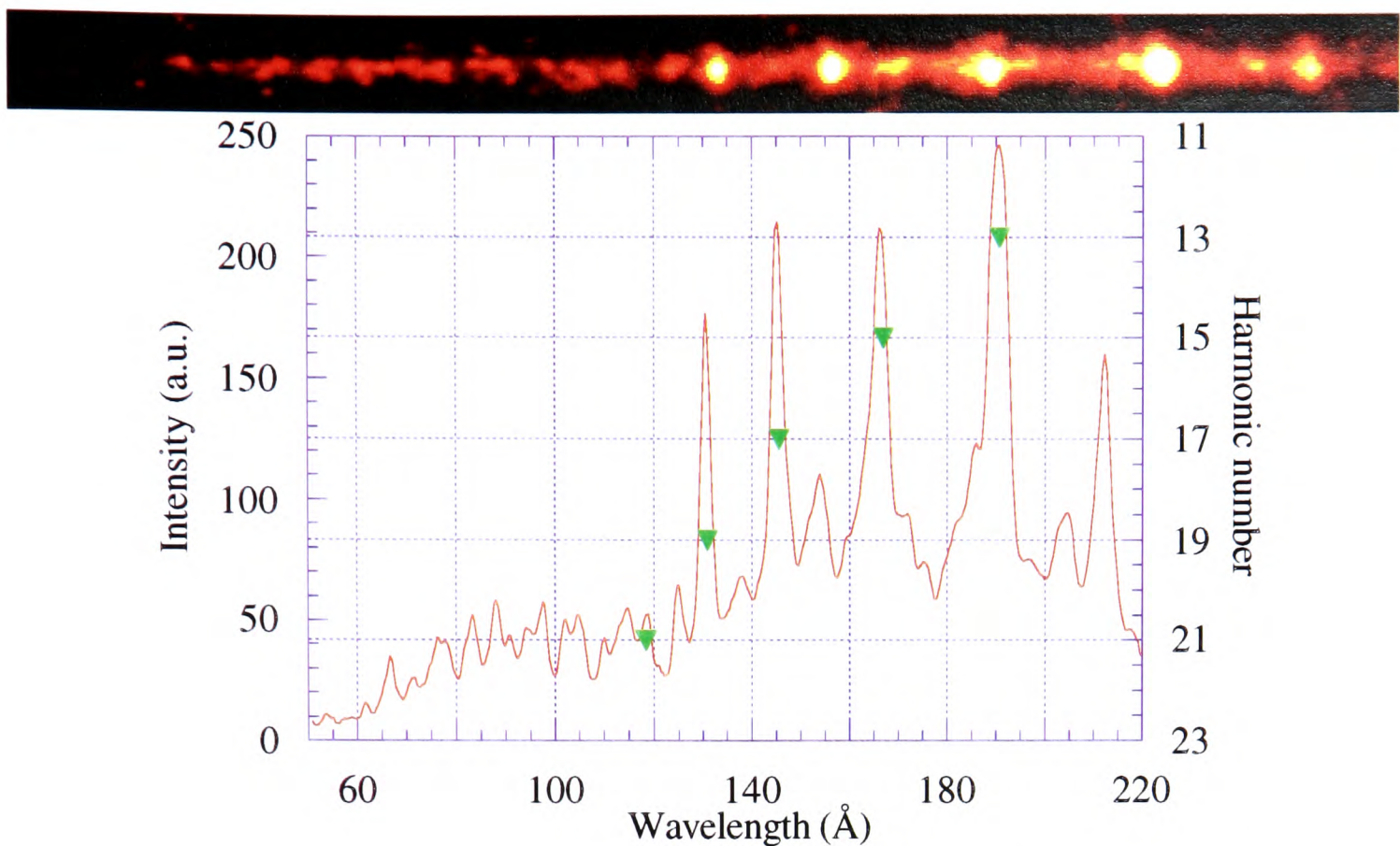


Figure 6.2.8. Image and lineout of Neon harmonic spectrum using $f/3$ focusing (SN19944).

As can be seen in figure 6.2.8 the harmonic cut off is reduced from that of Helium when using the short focal length OAP. Moving to the longer focal length ($f/10$) OAP we see that the harmonic emission greatly increases. Figures 6.2.9 and 6.2.10 show low and high pressure data respectively using the $f/10$ OAP. For the data in figure 6.2.9, the laser was focused 1 mm above a 2 mm x 0.5 mm rectangular aperture gas jet nozzle with the 2 mm dimension in the laser direction and a gas jet backing pressure was 0.3 bar gauge. For figure 6.2.10 the laser focusing and gas jet were the same as for figure 6.2.8, but the backing pressure was 26 bar gauge. The most noticeable change in the form of these two spectra is that the brightness of the plasma emission compared to the harmonic signal seems to reduce with increased pressure. This can be attributed to the fact that the plasma remains optically thin at the harmonic wavelengths, whereas the plasma becomes optically thick to its own emission as the density increases.

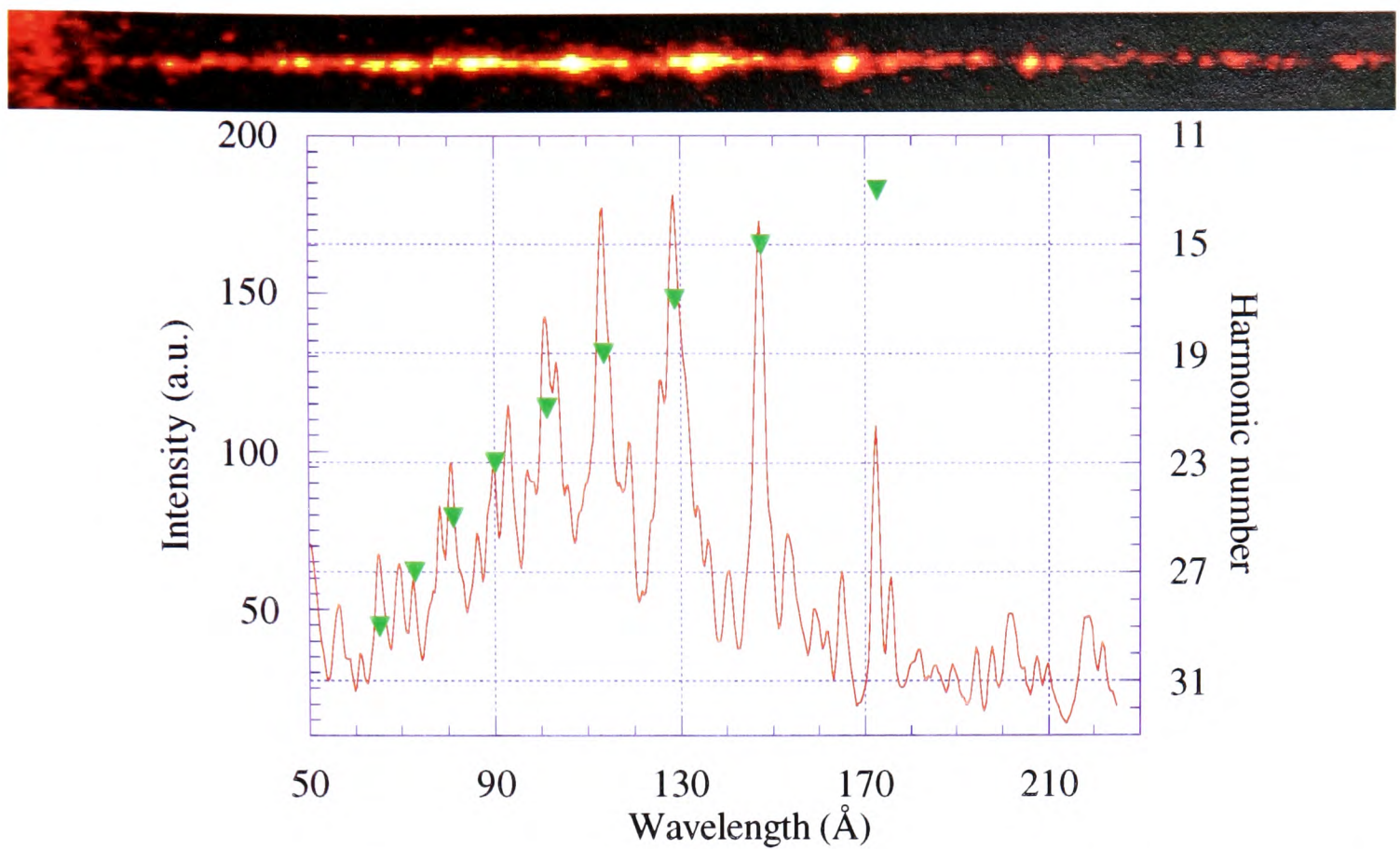


Figure 6.2.9. Image and lineout of low pressure Neon harmonic spectrum using $f/10$ focusing (SN20316).

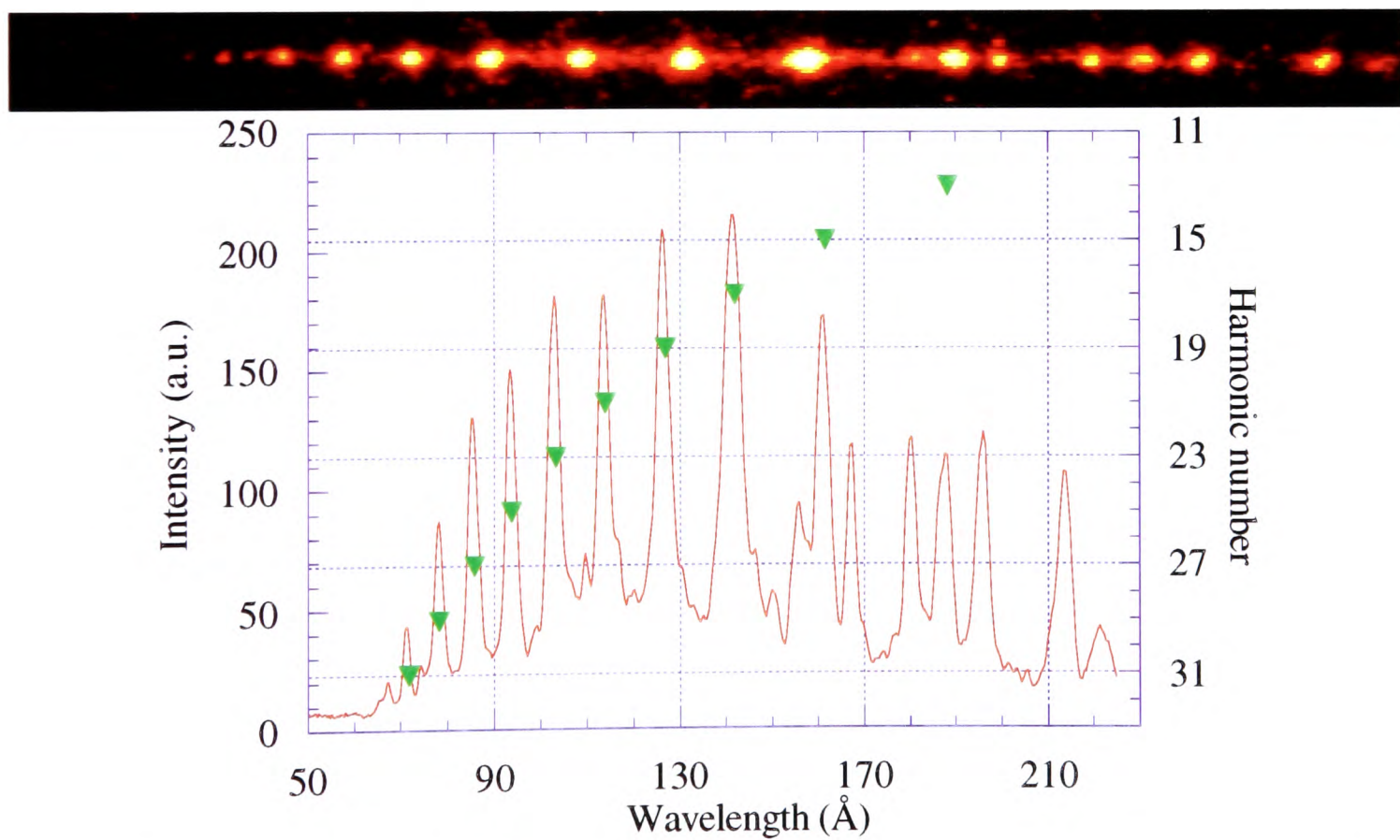


Figure 6.2.10. Image and lineout of high pressure Neon harmonic spectrum using $f/10$ focusing (SN20329).

6.2.2.2. Harmonic efficiencies.

To compare the above data quantitatively we show their absolute conversion efficiencies in figure 6.2.11. This figure also includes an extra data set (SN20317) which shows a harmonic signal up to the 35th. The conditions for this shot were essentially the same as for SN20316 but the gas jet was positioned 0.5 mm upstream of the laser focus. The difference is probably due primarily to variation in the focal spot profile. The harmonic response is essentially independent of pressure. Apart from the early cut off of the f/3 spectrum and possibly a higher efficiency for the 13th harmonic, the response is also independent of focusing. This is probably due to filamentation of the laser beam as it passes through the higher electron density caused by the optical-ionisation of Neon compared to Helium. The laser intensity may also be too high to efficiently generate harmonics from Neon and Ne^+ which have lower ionisation potentials than Helium and He^+ .

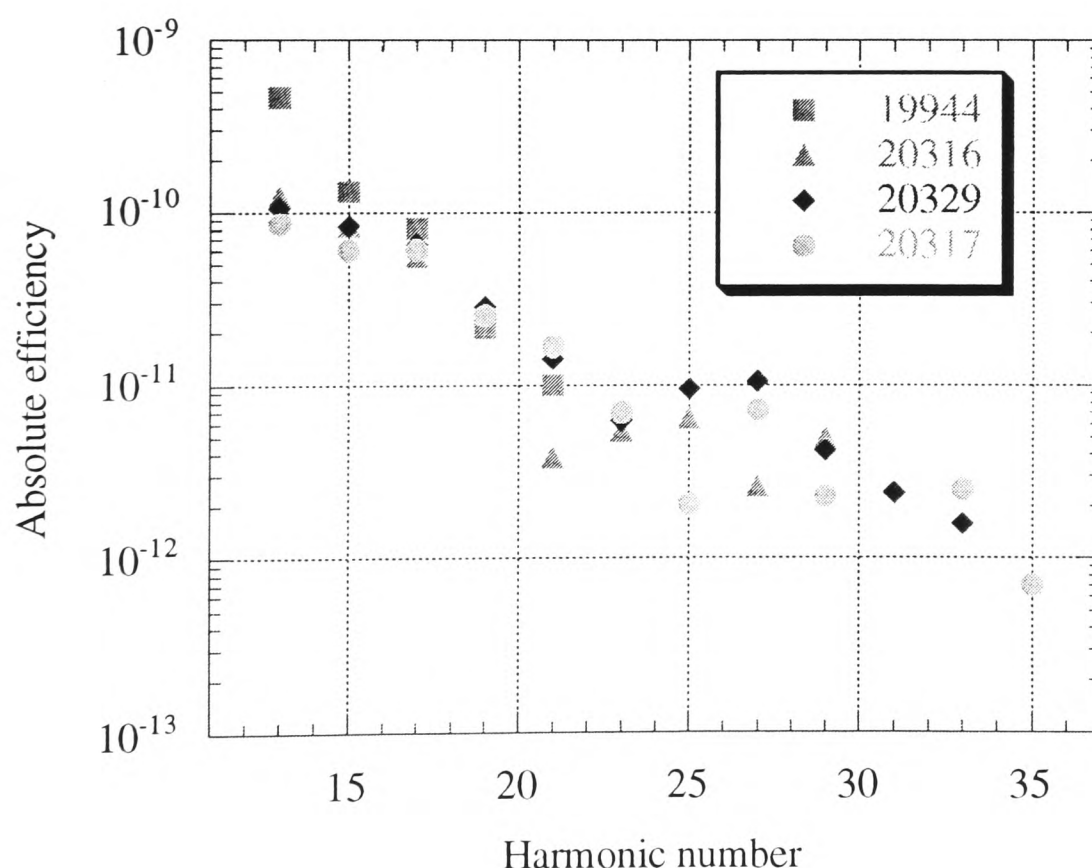


Figure 6.2.11. The absolute conversion efficiencies corresponding to the data shown in figures 6.2.8 to 6.2.10. An extra data set (SN20317) has been included to show a laser shot that produced 35th harmonic.

6.2.2.3. Intensity dependence of the 7th to 13th harmonics.

We also moved the MCP detector to study the scaling of generation efficiency with gas jet position. The data taken is somewhat limited, and essentially consists of four shots at various gas jet positions upstream of the laser focus (figure 6.2.12). The gas jet configuration was the same as for the Helium scan (see figure 6.2.7) and the backing pressure was 2 bar gauge.

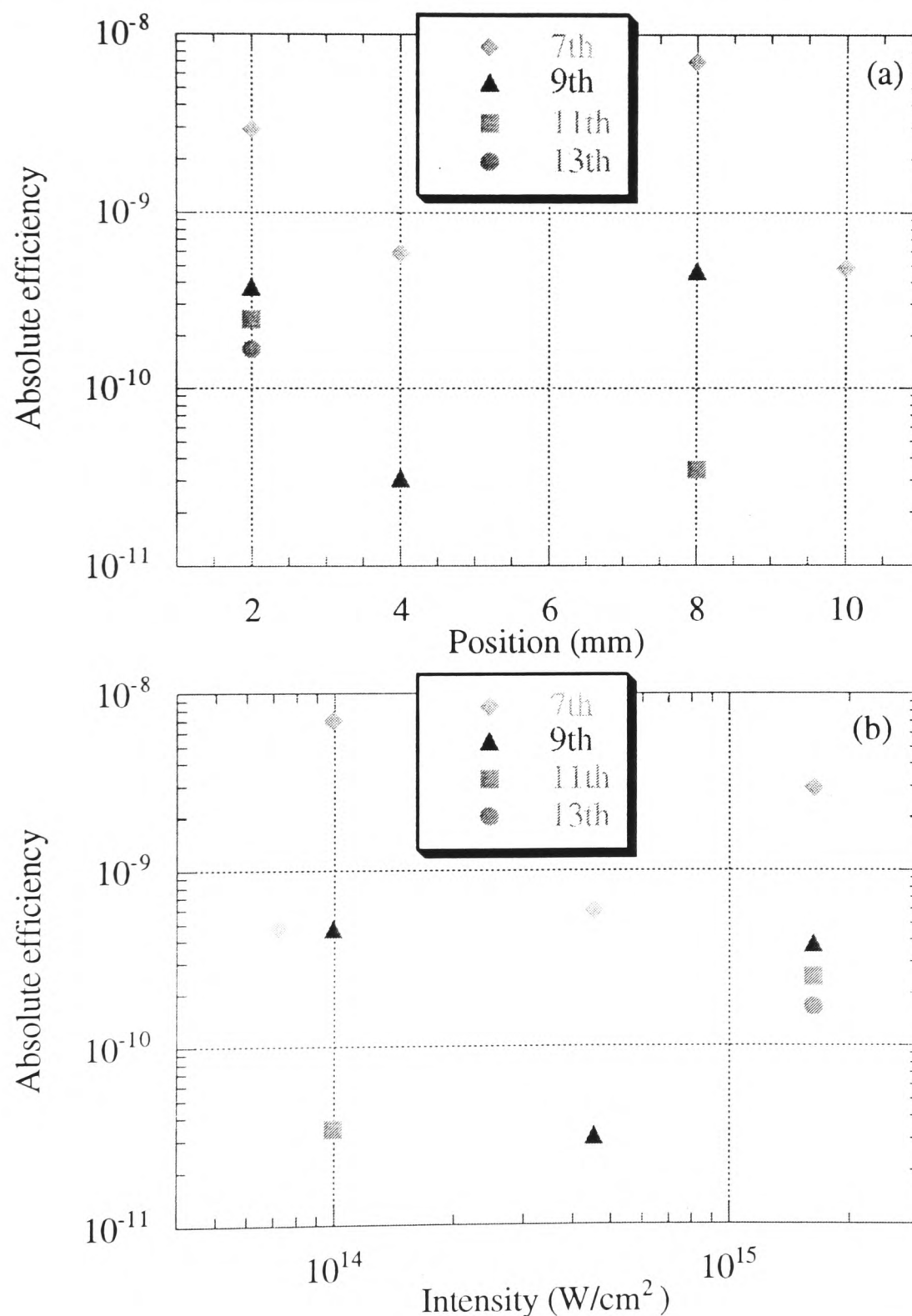


Figure 6.2.12. Harmonic conversion efficiencies in Neon as a function of (a) gas jet position and (b) laser intensity for the 7th to 13th harmonics.

This scan appears to indicate a maximum for generation of the 7th and 9th harmonics at around 10^{14} W/cm² (8 mm upstream). The 11th and 13th harmonics are brightest at the highest intensity in the scan. However, it is possible that the data points at 4.5×10^{14} W/cm² are from a shot with a particularly unusual focal spot. The plot of Neon ionisation rates in figure 2.2.2 shows that the threshold intensity for the complete ionisation of Neon to Ne⁺ in a 380 fs pulse is 3×10^{14} W/cm². The threshold intensity for ionisation Ne⁺ to Ne²⁺ in the same time is intensity of 2×10^{15} W/cm² which approximately corresponds to the highest generation efficiencies of the 11th and 13th harmonics. Figure 6.1.1 indicates that the 11th and 13th harmonics are the first to be generated by Ne⁺. This again highlights that, in the strong field limit, optimum harmonic generation occurs at the same intensity as the threshold for optical ionisation.

6.2.3. Comparisons of Helium and Neon harmonic responses.

Having reviewed harmonic generation of the 248 nm Sprite CPA laser system from both Helium and Neon we must now compare these results with each other and with the single atom computer simulations described in section 6.1. This will enable us to draw a conclusion as to which gas is the better choice for the production of the highest harmonic and the highest efficiency and it will enable us to determine how accurate the predictions of our simple simulations are.

6.2.3.1. *f/3 focusing.*

First let us compare the Helium and Neon data using the *f/3* OAP. Figure 6.2.13 shows a selection of data, some of which were presented previously, others of which are shown for the first time. The Neon spectrum cuts off much sooner than the Helium spectrum. In fact, all the Neon harmonics shown here could be attributed to neutral Neon and Ne⁺ without having to invoke generation from Ne²⁺. The generation efficiencies are divided

into three distinct bands, with high pressure Neon showing the highest efficiencies, followed by high pressure Helium and one of the lower pressure Helium shots, and then the other lower pressure Helium.

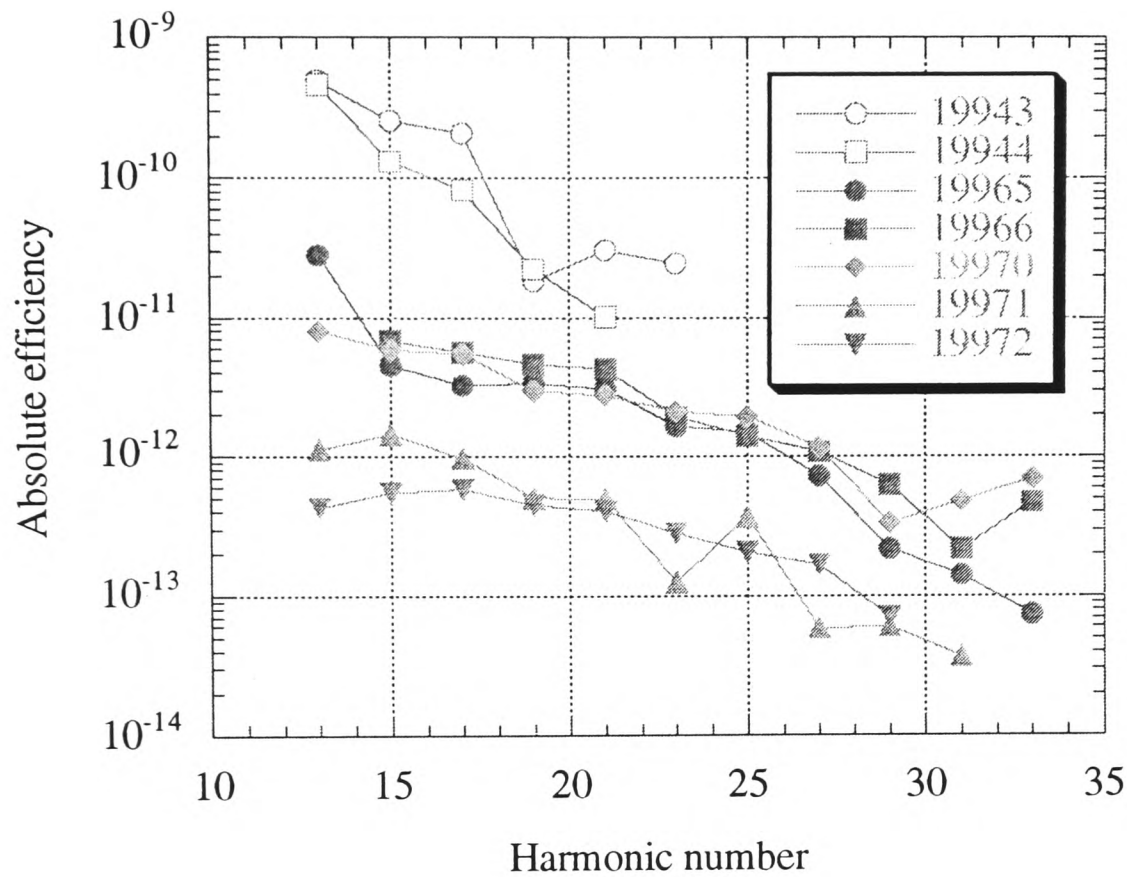


Figure 6.2.13. Absolute efficiencies of Neon (open symbols) and Helium (filled symbols) with $f/3$ focusing 1 mm above a 1mm circular aperture gas jet nozzle with 20 bar (squares and circles) and 2 bar (diamonds and triangles) gauge backing pressures.

The large variation in the lower pressure Helium shots is due to the variation in laser energy incident on target. This effect may be exacerbated by variation in the focal spot quality. The laser energy on target for each shot is tabulated in table 6.2. We can see that the two shots with the lowest conversion efficiency also had the lowest laser energy. From this we infer that, all else being equal, the Helium response with $f/3$ focusing is not strongly dependent on pressure. The high generation efficiencies in Neon compared to Helium are somewhat contrary to our expectations since our simulations predict that Helium should exhibit the stronger single atom response, but other experiments have

shown the same results [97,105]. The laser energies for the 20 bar Neon data are similar to those for Helium, so there is perhaps some beam propagation effect that favours phase matching for Neon over Helium in this configuration.

<i>Shot number</i>	<i>Gas</i>	<i>Backing pressure (bar)</i>	<i>Laser energy (mJ)</i>
19943	Neon	20	121
19944	Neon	20	155
19965	Helium	20	177
19966	Helium	20	144
19970	Helium	2	180
19971	Helium	2	67
19972	Helium	2	104

Table 6.2. *Summary of the conditions for the data presented in figure 6.2.12.*

6.2.3.2. *f/10 focusing.*

Figure 6.2.14 shows the generation efficiencies for Helium and Neon with *f/10* focusing. Filled symbols denote data from Helium, open symbols denote data from Neon. Squares denote high gas jet backing pressures and circles denote lower backing pressures. These backing pressures were 0.3, 20.7, 0.3 and 26 bar gauge for SN20315, SN20343, SN20317 and SN20329 respectively. All laser energies were similar (200 to 230 mJ) except SN20317 which was 98 mJ. In this case the energy fluctuations do not appear to affect the generation efficiencies. From this figure we see that the harmonic response of Helium and Neon is essentially the same from the 13th to the 35th harmonic regardless of gas density (given that we see shot-to-shot variation that is at least as large as the spread of data here). It is also obvious that the response is unaffected by a change in density of approximately a factor of 20. The highest efficiencies with *f/10* focusing for the 13th to

17th harmonic are perhaps slightly lower than those for f/3 focusing, but the difference is slight.

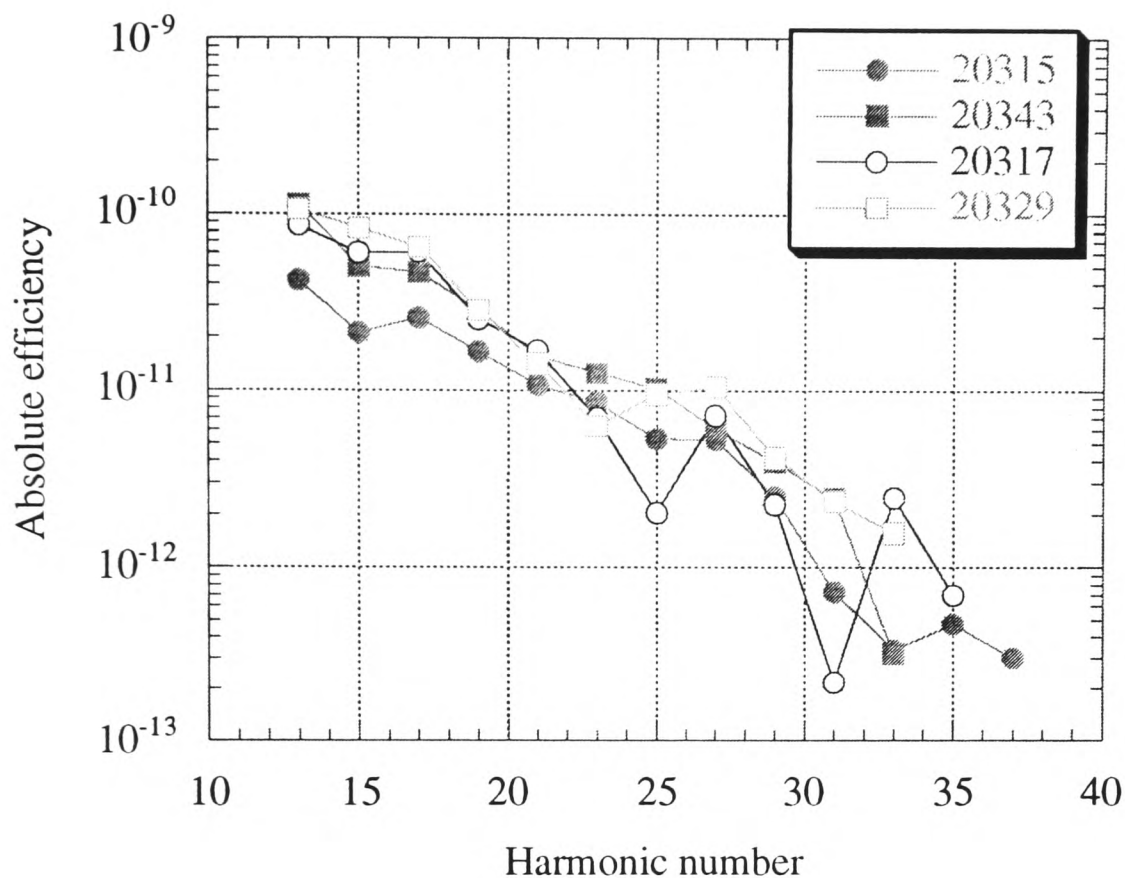


Figure 6.2.14. Absolute efficiencies of Neon (open symbols) and Helium (filled symbols) with f/10 focusing showing high (squares) and low (circles) backing pressures.

At very low gas densities we do see a pressure dependence of the harmonic response. This occurs when the coherence length becomes longer than the confocal parameter and/or the gas length. The reason for this is that if the coherence length is less than the confocal parameter or the gas length the harmonic signal tends to saturate at a level that is approximately that which it would reach after one coherence length. If the coherence length is longer than this the harmonic signal will never reach saturation and the conversion efficiency reduces. Since the coherence length is determined by the free electron density, and optical ionisation is essentially sequential, the electron density during harmonic generation from He^+ will be somewhere between 1 and 2 times the gas density. For Ne^{2+} the electron density will be somewhere between 2 and 3 times the gas density. If we take the electron density to be twice the gas density in both cases we

calculate that the coherence length of the q th harmonic will become less than the confocal parameter ($\sim 300 \mu\text{m}$ for $f/10$ focusing) at an electron density of $A/q \text{ cm}^{-3}$ where $A = 1.5 \times 10^{19} \text{ cm}^{-3}$ for a 248 nm fundamental and a confocal parameter of $300 \mu\text{m}$. This is consistent with the electron density at which this was seen to occur (e.g. $\sim 5 \times 10^{17} \text{ W/cm}^2$ for the 31st harmonic). Below this density the harmonic intensity is expected to vary as the square of the number of atoms/ions for a fixed length of gas.

Next we must compare the generation efficiencies of the lower harmonics (7th to 13th). It is here that we see the largest difference between the harmonic responses of Helium and Neon. Figure 6.2.15 shows a selection of data shots in both gases. The data covers a range of intensities and densities. The Helium generation efficiencies are consistently higher than those for Neon. This indicates the potential of Helium for generating high powers at the lower harmonic wavelengths when some consideration is given to optimising irradiance and density.

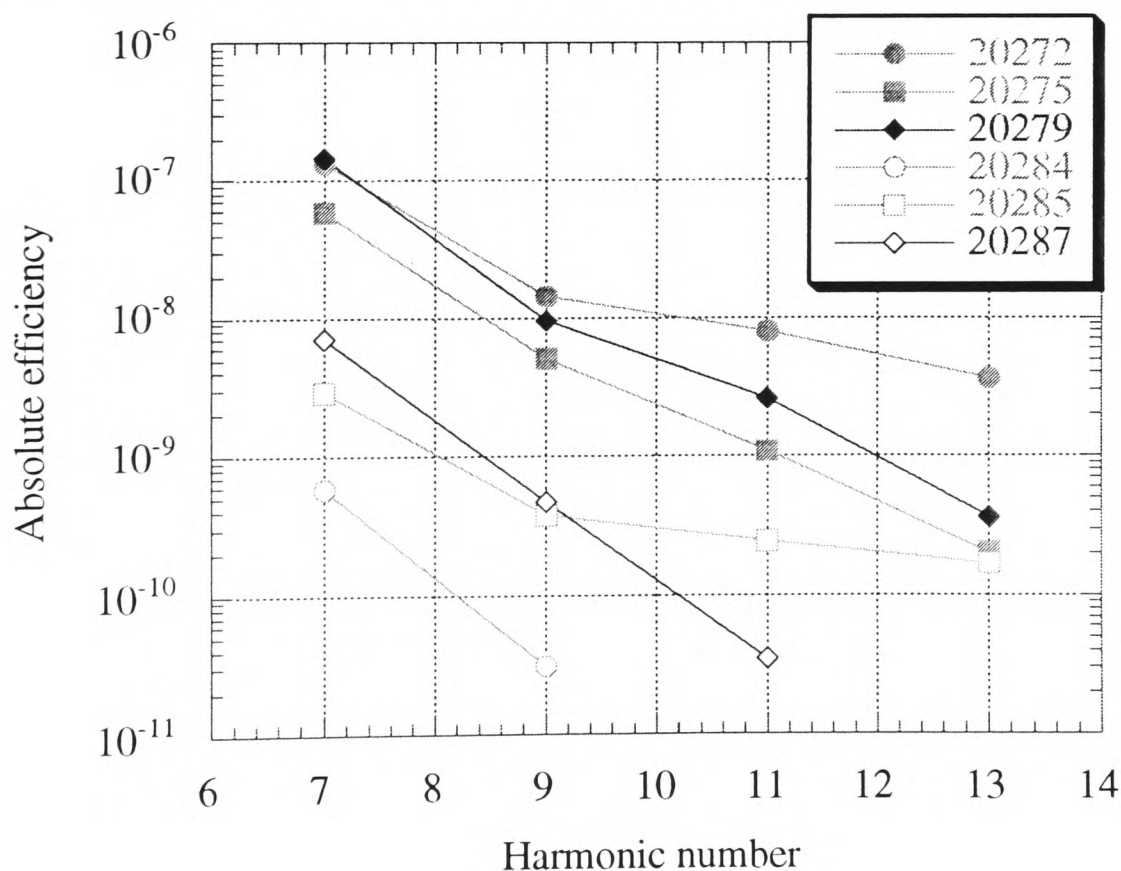


Figure 6.2.15. Absolute efficiencies of Neon (open symbols) and Helium (filled symbols) with $f/10$ focusing 1 mm above a 0.5 mm x 2 mm rectangular aperture gas jet nozzle with the 2 mm dimension in the laser direction.

6.2.4. Comparisons of experiment and theory.

All that remains now is to compare our simulations with the experimental data. Our simulations only predict the harmonic response of a single, isolated atom. They do not include any beam propagation or profile effects at all. This type of simulation is commonly used to model experimental harmonic responses. The usual method of obtaining a good fit between the real data and the simulation is to vary the laser intensity in the simulation until the cut off of the experimental harmonic plateau is matched. When this procedure is followed, the modelled intensity bears absolutely no relationship to the actual intensity experienced by the atoms in the laser focus.

To try to make the simulations a more accurate representation of the conditions in our experiments, we have run a number of simulations at laser intensities that atoms in the focus actually experience and produced a weighted sum of these intensities. The weighting takes account of the volume of a Gaussian (TEM₀₀) focus that actually experiences each intensity and introduces a q^{-2} phase matching dependence, where q is the harmonic order. This is because the harmonic intensity is proportional to the square of the coherence length which is inversely proportional to the harmonic order in the free electron phase matching case (see section 3.3.3). The simulations are compared with a selection of experimental data for (a) Helium, and (b) Neon in figure 6.2.16.

We see in the figures that both the full Helium and the one-dimensional Neon simulations provide an excellent fit to the experimental data. The experimental data shown represents a range of gas densities and gas jet nozzle configurations. The neutral Helium simulation models the form of the data very well, but appears to be about an order of magnitude too low. This can be explained by the increased phase matching during generation from a neutral when the coherence length is dominated by Gaussian focusing and by dispersion in the gas compared to free electrons for the ions. If we were to multiply the neutral

Helium simulation by a factor of about ten, then the changeover from neutral to ionic response would occur near the 15th to 17th harmonic rather than the 9th or 11th which is currently predicted.

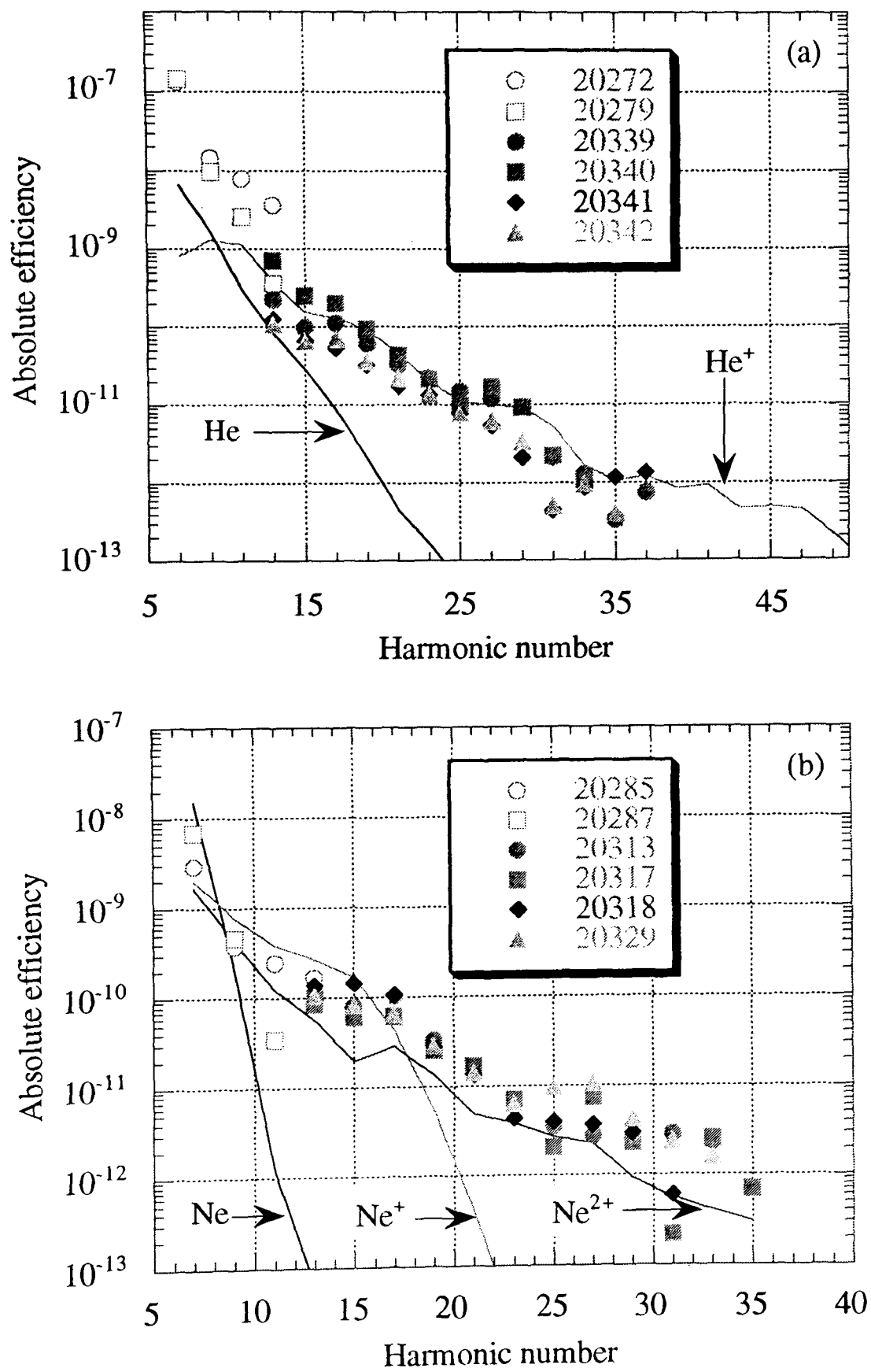


Figure 6.2.16. A comparison of experimental data with the weighted simulated response for (a) Helium, and (b) Neon. The simulation has been multiplied by an arbitrary constant.

The one-dimensional Neon simulation provides a remarkably good fit to the experimental data. Not only does it exactly predict the cut off at the 35th harmonic, but it models the changes in the slope of the response at the 9th, 17th, 21st and 27th harmonics. This good match implies that the harmonic signal we are seeing is truly from Neon and its ions up to the doubly ionised stage. This means that harmonics above the 9th are primarily due to Ne^+ , and harmonics above the 19th are primarily due to Ne^{2+} . Harmonic efficiencies above the 27th do appear to be approximately a factor of two or three higher than the simulation predicts. This is not surprising given the shot-to-shot variation in signal. However, it is interesting that above about the 31st harmonic the response of Ne^{3+} becomes approximately the same as Ne^{2+} until the 35th harmonic when the Ne^{2+} response falls off. The results as they stand can all be explained without having to invoke response from a third ionisation stage of Neon, but had any harmonic been detected above the 35th we would have had a very strong case for attributing it to Ne^{3+} .

6.2.5. Harmonics higher than 37th in Helium.

Considering the Helium data in figure 6.2.16.(a) we see that the ion response matches the form of the experimental data very well, although the predicted cut off doesn't occur until the 47th harmonic compared to the 37th which was the highest harmonic seen in the experimental first order diffraction signal. However, in section 6.2.1 we commented on the high levels of the second order diffracted signal for wavelengths below about 100 Å. The spatial resolution of the MCP detector is rather poor and introduces an instrument function with a FWHM of about 4.5 pixels (which corresponds to a wavelength spread of over 1 Å). This poor resolution causes the second order signal to merge with the wings of the brighter first order signal. However, several images do show signs of wings on the first order diffracted image which, if real, indicate a harmonic response that could reach as high as the 53rd harmonic. To verify this we will attempt to deconvolve the MCP instrument response from some lineouts which show these wings. The data

will be presented as an indication that these high harmonics exist, no attempt will be made to calculate their generation efficiencies.

Figure 6.2.17 shows a deconvolution of SN19965, which was a laser shot using $f/3$ focusing with a vacuum focused intensity of around $6 \times 10^{17} \text{ W/cm}^2$. The deconvolution removes the MCP instrument function which is a Gaussian with a FWHM of 4.5 pixels. We are still left with a considerable spectrometer instrument function caused by the grating not focusing the wide wavelength range exactly on the detector plane. The instrument width of the spectrometer varies along the detector and is different for the first and second order. For this reason we cannot fully deconvolve the instrument width. The raw data is first smoothed with a first order Butterworth low pass filter function with a cut off frequency of a quarter of the sample frequency, and the deconvolution is performed using a fast Fourier transform method. Despite this rather crude deconvolution, we can see indications of second order signal up to the 53rd harmonic (47 Å). These are labelled in the figure. Several other harmonic spectra also exhibit the same (second order) features, but are not shown here. This indicates that the signal is not due to random noise. Figure 6.2.18 shows another deconvolved harmonic spectrum (deconvolved with a Gaussian with a FWHM of 4 pixels) taken with $f/10$ focusing. The first order spectrum only extended out to the 27th harmonic for this laser shot, and so we would not expect the harmonic response to extend as far as for figure 6.2.17 which extended to 33rd harmonic in first order. This is a further indication that the second order spectrum (when it appears) is real. The deconvolution routine used on this data is very basic and has not been optimised. More work must be done to analyse this signal, or another experiment must be performed using a grating with a higher first order reflectivity in the 40 - 70 Å region in order to verify this result.

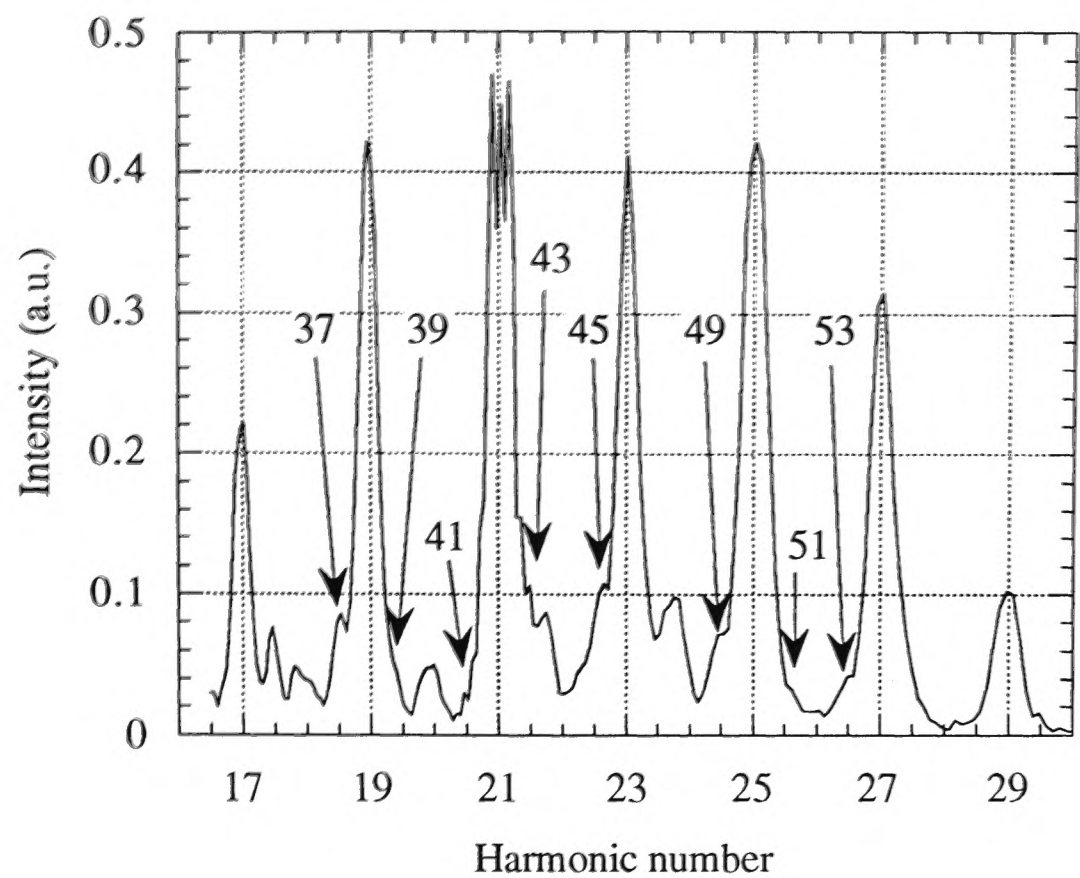


Figure 6.2.17. Deconvolved lineout from SN19965 showing indications of harmonics as high as 53rd (47 Å).

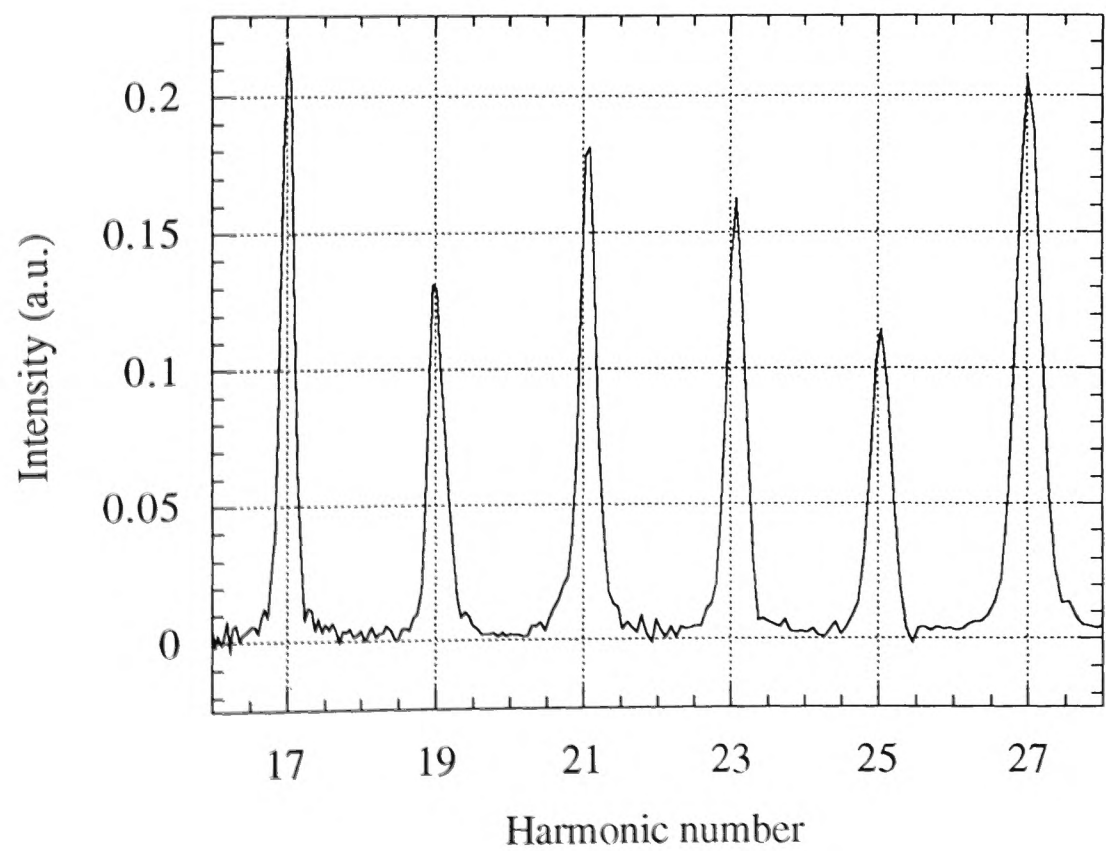


Figure 6.2.18. Deconvolved lineout of SN20122. This time there is no second order signal.

6.3. Summary.

In the experiments presented in this chapter we have demonstrated and quantitatively analysed the production of up to 35th harmonic (71 Å) in Neon and up to 37th harmonic (67 Å) in Helium. These are some of the shortest harmonic wavelengths generated to date, corresponding to the 148th and 157th harmonics respectively of a 1053 nm Nd:Glass laser, or 113th and 119th of 800 nm Ti:Sapphire laser. There are indications that harmonics as high as the 53rd (47 Å) are present in the second order diffracted spectrum for some conditions. This is tantalisingly close to the harmonic order required to produce XUV radiation in the water window (57th harmonic). The highest harmonics reported from long wavelength systems to date are 141st of 1053 nm from Helium [19] and 109th of 800 nm from Neon [20]. Recent work with an ultra-short pulse (25 fs) Ti:Sapphire laser by Murnane and co-workers [152] shows indications of the generation of harmonic wavelengths as short as 60 Å (~133rd harmonic) in Neon. However, using such a short laser pulse produces harmonics with a very large bandwidth. We have therefore shown that a short wavelength KrF laser at moderate powers (~0.6 TW) can generate harmonics from ions at shorter wavelengths than can be generated from atoms using a long wavelength laser.

We also show that a KrF laser can produce high harmonic generation efficiencies for the lower harmonics. The 7th harmonic (355 Å) is produced with efficiencies higher than 10^{-7} at intensities of the order of 10^{15} W/cm². This is comparable to experiments performed with a 526 nm frequency doubled Nd:Glass laser [107] which used much longer confocal lengths produced with f/70 focusing. It remains to be seen whether a similar scaling of the KrF results with focal length will lead to further enhancements in the generation efficiency of the lower harmonics.

Comparisons of the experiment with simulations show the need for an appropriate weighting of the simulated data to produce a reasonable match. This highlights the need for a code that includes beam propagation effects as well as the single atom/ion response.

Finally, experimental studies of both Helium and Neon show a marked difference in their harmonic response with $f/3$ focusing (Neon being higher) and for the lower harmonics at lower intensities with $f/10$ focusing (Helium being higher). The response of the two gases for higher harmonics using $f/10$ focusing is essentially independent of density above about 10^{17} cm^{-3} (for the 31st harmonic).

Chapter Seven

Conclusions and Suggestions for Further Work

In this chapter we will review the main conclusions of this thesis and their implications for the generation of coherent XUV generation. We will then briefly discuss possibilities for future extensions to this work: both in the field of OFI X-ray lasers; and in the field of harmonic generation.

7.1. Conclusions of this thesis.

7.1.1. Plasma temperature measurements.

In chapter five we present Thomson scattered and SRS spectra from Helium and Neon and discuss the electron temperatures and densities we infer. We support our analysis of SRS reflectivities with Faraday cup data and an X-ray spectrum showing line emission from Helium- and Hydrogen- like Neon. The conclusions of this investigation are that the scheme proposed by Eder et al. [44] should be possible for a laser pulse length of 100 fs and a focused intensity of $3 \times 10^{17} \text{ W/cm}^2$ provided the laser beam quality is good. However, the onset of SRS and the increase in IB with electron density will limit the system to elements with $Z < 12$. This restricts the shortest wavelength that can be produced by this scheme to about 60 Å. We also note that more recent calculations of gains in these systems, that take heating by three body recombination into account [81], suggest a much lower gain than suggested by Eder et al. Experimental evidence also suggests that the three body recombination rate in a non-thermal OFI plasma is

considerably reduced from that of a thermal system [82]. Combining these low gains with the limits placed on the length of any longitudinally pumped system (caused by the XUV pulse catching up the pump laser pulse in the plasma) will severely restrict the power of this OFI recombination laser scheme. This will be so even if a method can be found to guide the pump laser pulse over several centimetres in a plasma channel. Hence, the OFI recombination laser scheme will be in direct competition with harmonic generation for 'table top' production of coherent XUV radiation.

7.1.2. Harmonic generation.

Chapter six presents computer simulations of harmonic generation in Helium and Neon which predict that high harmonic efficiencies can be obtained from ions at a fundamental laser wavelength of 248 nm. This contrasts with longer wavelength lasers which produce a negligible ion response. This indicates that, contrary to previous assumptions, shorter wavelength harmonics can be generated from ions using a short wavelength laser than from neutrals using a long wavelength laser. The simulations also predict that, in our experiment (in the absence of phase matching effects), Helium should show a higher response than Neon.

We present experimental data that reveal that the harmonic responses from Helium and Neon plasmas are essentially the same, although Helium exhibits a higher response than Neon for the 7th to 13th harmonic at lower intensities away from the laser focus. Comparisons of the experiments with simulations show a good match, but indicate the need to allow for spatial variation of the laser phase and amplitude at the focus rather than just considering the single atom/ion response.

The highest harmonics which were quantitatively analysed were the 35th (71 Å) in Neon which we attribute to Ne^{2+} , and 37th (67 Å) in Helium (attributed to He^+). These are the

shortest wavelength harmonics generated to date by either short or long wavelength fundamental lasers and confirm the predictions of our simulations that short wavelength fundamental lasers are better suited to generating high photon energies than long wavelength lasers. There is some indication of harmonics higher than 37th in the second order diffracted signal from Helium (up to 53rd harmonic) which suggest that the cut off of the harmonic plateau in Helium is well matched to that predicted by the simulations. The cut off of the Neon response exactly matches the simulation and is the first unambiguous evidence of harmonic generation from doubly-ionised Neon.

Finally, we note that studies of lower (7th to 13th) harmonics from neutral Helium at lower intensities show a generation efficiency of 10^{-7} which matches the best seen elsewhere [107] which used a much longer confocal parameter. This indicates that shorter wavelength fundamental lasers do lead to a considerably greater harmonic generation efficiency. It remains to be seen whether this can be further scaled by moving to a much longer confocal parameter than that produced by the f/10 OAP used in these experiments.

7.2. Suggestions for future work.

7.2.1. Optically-ionised X-ray lasers.

The temperature measurements described in chapter five are aimed at determining the feasibility of creating a recombination X-ray laser by optical-ionisation. We conclude that the specific scheme modelled by Eder et al. [44] is possible, but that severe limits will be placed on the efficiency of such a system. Observations of gain in non-resonance transitions in OFI systems have been reported on the 3d-5f transition in N^{4+} (512 Å) and on the 2s2p²-2s2p3d transition in N^{2+} (472 Å) [153,154] but these are at relatively long wavelengths. Another claim has been made of gain on the 1s-2p transition in Li^{2+}

(135 Å), i.e. a hydrogen-like scheme [83,84]. This system uses an initial 20 ns laser pulse to create a plasma of Li^+ from a solid target, before Li^{3+} is created by OFI from a 500 fs laser pulse. Both these claims must be treated very carefully, because of the difficulty in fitting a gain curve to a very small plasma length. The plasma lengths in the two experiments were claimed to be approximately 800 μm and 2 mm respectively.

To further extend the work in this thesis we must move to shorter (probably less than 100 fs) laser pulses to see if we can produce a working OFI laser scheme in Neon, or we must work with elements of lower atomic number over longer interaction lengths to look for gain. This must be combined with research into guiding of the laser over a long length in the plasma and preventing the laser beam from breaking up and forming filaments. We will then be able to determine whether the OFI recombination laser is worth pursuing as a source of coherent XUV radiation.

7.2.1.1. *The Titania laser.*

The Sprite laser, used for the experiments in this thesis, has now been decommissioned and is being upgraded by the addition of a new 40 cm aperture, e-beam pumped amplifier. The new laser will be known as Titania [26]. The output of this laser in CPA mode is predicted to be of the order of 1 J in 300 fs, although this will depend on the availability of good quality compression gratings. The laser pulse length is not expected to be shorter than that of Sprite, but the extra energy (and possibly better beam quality) will make it possible to optically-ionise materials higher than Neon to the Helium-like state. It will also be possible to generate much longer confocal parameters at lower intensities to study the OFI scheme in Neon and lower elements over a much longer plasma length and with a much larger focal spot.

7.2.2. Harmonic generation.*7.2.2.1. Improving our results with higher power lasers.*

The field of harmonic generation is still in its infancy. The push of research will inevitably lead in the same two directions as for X-ray lasers. The first will be to increase efficiencies, and the second will be to produce shorter wavelengths. The advantage of short wavelength lasers for both of these has been demonstrated in this thesis. When the Titania laser comes on-line, we will be able to continue our present research by scaling to higher intensities. More importantly, we will be able to generate the same intensities as those used in the experiments we have performed to date, but over a much longer interaction length, and with a much larger focal spot (produced with a much longer focal length OAP). For example, the work of Ditmire et al. [107] produced efficiencies comparable to ours using $f/70$ focusing (as opposed to our $f/3$ and $f/10$ focusing). If our results scale similarly, we should see considerable increases in harmonic generation efficiency.

7.2.2.2. Harmonic generation from plasmas and excited states.

Using a second laser beam to create a Lithium plasma will enable us to investigate whether we can scale the cut off of our harmonic plateau to much shorter wavelengths, by using the higher ionisation potentials of Helium- and Hydrogen- like Lithium. There is also the possibility of generating lower harmonics with much higher efficiencies from the excited states of ions or atoms. If the ion is in an excited state, it requires a much lower laser intensity to ionise, but the electron will only recombine to the excited state when it is re-incident on the atomic core. The plateau cut off will therefore reduce with the lowered ionisation potential.

7.2.2.3. *Two colour harmonic generation.*

The probability that an electron will recombine with the atomic core and emit a harmonic photon is determined by the collision cross section. This cross section is dependent on the electron velocity as it is re-incident on the core: the lower the electron velocity, the higher the probability of harmonic emission. If the electric field driving the electron is sinusoidal (from a monochromatic laser), then the electron will always return to the atomic core when its velocity is near to maximum. However, it is possible to combine two or more colours in the laser pulse (fundamental and third harmonic, for example) to modify the electron's motion so that it re-approaches the atomic core with a much lower velocity. This leads to much higher harmonic generation efficiencies. Such a system has been modelled theoretically [155], and a preliminary experiment has been performed [156] showing an order of magnitude increase in the harmonic response.

7.2.2.4. *Harmonic generation from coherent states.*

Another proposal for increasing harmonic generation efficiencies is to prepare a population of atoms in a coherent superposition of the ground state and an excited state. An electron can then be ionised from the excited state and recombine with the ground state on its return to the atomic core. This can easily be done for states that can be excited by a single photon, and just involves irradiating the atoms with laser radiation whose frequency exactly matches the transition frequency between the ground and excited states. To make this method useful for harmonic generation, however, we would need to produce the coherent superposition by exciting a multiphoton resonance.



References

1. F.F. Chen. *Introduction to plasma physics*; Plenum Press: New York (1974).
2. D. Mihalas. *Stellar atmospheres*. 2nd Edition; W.H. Freeman and Company: San Francisco (1978).
3. S.J. Rose. *High-power laser-produced plasmas and astrophysics*. *Laser and Particle Beams*, **9** (4), pp 869 (1991).
4. M.H. Key and R.G. Evans. *Inertial confinement of fusion*. *Nature*, **313** (5998), pp 94-95 (1985).
5. R.G. Evans and M.H. Key. *Lasers - fusion turned inside out*. *Nature*, **320** (6058), pp 106 (1986).
6. R.G. Evans. *The basic physics of laser fusion*. *Canadian Journal of Physics*, **64**, pp 893 (1985).
7. S. Ichimaru. *Nuclear-fusion in dense-plasmas*. *Reviews of Modern Physics*, **65** (2), pp 255-299 (1993).
8. T.W. Johnston. *Plasma effects in laser fusion: past and present*. *Canadian Journal of Physics*, **64**, pp 1006 (1986).
9. *Laser-induced plasmas and applications*; Ed. L.J. Radziemski and D.A. Cremers; Marcel Dekker Inc.: New York (1989).

10. R.C. Elton. *X-ray lasers*; Academic Press: New York, (1990).
11. M.H. Key. *Laboratory production of X-ray lasers*. *Nature*, **3** (16), pp 314 (1985)
12. J.S. Wark, R.R. Whitlock, A.A. Hauer, J.E. Swain and P.J. Solone. *Subnanosecond X-ray diffraction from laser-shocked crystals*. *Physical Review B*, **40** (8), pp 5705 (1989).
13. D.E. Sayers and E.A. Stern. *New technique for investigating noncrystalline structures: Fourier analysis of the extended X-ray absorption fine structure*. *Physical Review Letters*, **27** (18), pp 1204 (1971).
14. P.A. Lee, P.H. Citrin, P. Eisenberger and B.M. Kincaid. *Extended X-ray absorption fine structure - its strengths and limitations as a structural tool*. *Reviews of Modern Physics*, **53** (4), pp 769 (1981).
15. M.R. Howells, J. Kirz and D. Sayre. *X-ray microscopes*. *Scientific American*, **264** (2), pp 88 (1991)
16. J.E. Trebes, S.B. Brown, E.M. Campbell, D.L. Matthews, D.G. Nilson, G.F. Stone and D.A. Whelan. *Demonstration of X-ray holography with an X-ray laser*. *Science*, **238**, pp 517 (1987).
17. M. Howells, C. Jacobson, J. Kirz, R. Feder, K. Mcquaid and S. Rothman. *X-ray holograms at improved resolution: a study of zymogen granules*. *Science*, **238**, pp 514 (1987).

18. L.B. Da Silva, T.W. Barbee Jr, R. Cauble, P. Celliers, D. Ciarlo, S. Libby, R.A. London, D. Matthews, S. Mrowka, J.C. Moreno, D. Ress, J.E. Trebes, A.S. Wan and F. Weber. *Electron density measurements of high density plasmas using soft X-ray laser interferometry*. Physical Review Letters, **74** (20), pp 3991-3994 (1995).

19. M.D. Perry and G. Mourou. *Terawatt to petawatt subpicosecond lasers*. Science, **264**, pp 917-924 (1994).

20. J.J. Macklin, J.D. Kmetec and C.L. Gordon III. *High order harmonic generation using intense femtosecond pulses*. Physical Review Letters, **70** (6), pp 766 (1993).

21. S.G. Preston, A. Sanpera, M. Zepf, W.J. Blyth, C.G. Smith, K. Burnett, M.H. Key, J.S. Wark, D. Neely and A.A. Offenberger. *High-order harmonics of 248.6 nm KrF laser from Helium and Neon ions*. Physical Review A, (January 1996).

22. T.H. Maiman. *Stimulated optical radiation in Ruby*. Nature, **187** pp 493 (1960)

23. R.W. Hellwarth. *Control of fluorescent pulsations in Advances in Quantum Electronics*; Ed. J. R. Singer; Columbia University Press: New York, pp 334-341 (1961).

24. *Ultrashort Light Pulses: Picosecond Techniques and Applications*; Ed. S.L. Shapiro; Springer-Verlag: Berlin (1977).

25. D. Strickland and G. Mourou. *Compression of amplified chirped optical pulses*. Optics Communications, **56**, pp 219-221 (1985).

26. J.M.D. Lister, E.J. Divall, S.W. Downes, C.B. Edwards, G.J. Hirst, C.J. Hooker, M.H. Key, I.N. Ross, M.J. Shaw and W.T. Toner. *Sprite - A very high brightness, ultraviolet laser system*. Journal of Modern Optics, **41** (6), pp 1203-1215 (1994).

27. C.N. Danson, L.J. Barzanti, Z. Chang, A.E. Damerell, M.H.R. Hutchinson, M.H. Key, S. Luan, R.R. Mahadeo, I.P. Mercer, P. Norreys, D.A. Pepler, D.A. Rodkiss, I.N. Ross, M.A. Smith, R.A. Smith, P. Taday, W.T. Toner, K.W.M. Wigmore, T.B. Winstone, R.W.W. Wyatt and F. Zhou. *High-contrast multi-terawatt pulse generation using chirped pulse amplification on the Vulcan laser facility*. Optics Communications, **103** (5-6), pp 392-397 (1993).

28. W.H. Knox, R.L. Fork, M.C. Downer, R.H. Stolen, C.V. Shank and J.A. Valdmanis. *Optical pulse-compression to 8 fs at a 5-khz repetition rate*. Applied Physics Letters, **46** (12), pp 1120-1121 (1985).

29. W.J. Blyth. *Temperature measurements of optically ionised plasmas*. D.Phil Thesis, University of Oxford, (1994).

30. R.R. Freeman and P.H. Bucksbaum. *Investigations of above-threshold ionization using subpicosecond laser pulses*. Journal of Physics B, **24**, pp 325-347 (1991).

31. S. Augst, D. Strickland, D.D. Meyerhofer, S.L. Chin and J.H. Eberly. *Tunneling ionization of Noble-gases in a high-intensity laser field*. Physical Review Letters, **63**, pp 2212 (1989).

32. N.B. Delone and V.P. Krainov. *Energy and angular electron spectra for the tunnel ionization of atoms by strong low-frequency radiation*. Journal of the Optical Society of America B, **8**, pp 1207 (1991).
33. M.V. Ammosov, N.B. Delone and V.P. Krainov. *Tunnel ionisation of complex atoms and of atomic ions in an alternating electromagnetic field*. Soviet Physics JETP, **64** (6), pp 1191-1194 (1986).
34. S.C. Rae. *Short-pulse laser-plasma interactions*. D.Phil Thesis, Oxford, (1991).
35. A. Djaoui and A.A. Offenberger. *Heating of underdense plasmas by intense short-pulse lasers*. Physical Review E, **50** (6), pp 4961-4968 (1994).
36. *Physics of fully ionised gases*; Ed. L. Spitzer; Interscience: New York (1956).
37. T.W. Johnston and J.M. Dawson. *Correct values for high-frequency power absorption by inverse bremsstrahlung in plasmas*. Physics of Fluids, **16** (5), pp 722 (1973).
38. N.H. Burnett and G.D. Enright. *Population inversion in the recombination of optically-ionized plasmas*. IEEE Journal of Quantum Electronics, **26** (10), pp 1797-1808 (1990).
39. A.Y. Polishchuk and J. Meyer-Ter-Vehn. *Electron-ion relaxation in a plasma interacting with an intense laser field*. Physical Review E, **49** (1), pp 663 (1994).
40. G.J. Pert. *Inverse bremsstrahlung in strong radiation fields at low temperatures*. Physical Review E, **51** (5), pp 4778-4789 (1995).

41. J.P. Christiansen, D.E.T.F. Ashby and K.V. Roberts. *Medusa; A one-dimensional laser fusion code*. Computer Physics Communications, **7**, pp 271 (1974).

42. T.P. Hughes, *An introduction to the absorption of laser light in plasmas* in The Twentieth Scottish Universities Summer School in Physics, St. Andrews, Scotland, Ed. R. A. Cairns and J. J. Sanderson, Scottish University Summer Schools in Physics, pp 1-86 (1979).

43. D.W. Forslund, J.M. Kindell and E.L. Lindman. *Theory of stimulated scattering processes in laser-irradiated plasmas*. Physics of Fluids, **18** (8), pp 1002-1016 (1975).

44. D.C. Eder, P. Amendt and S.C. Wilks. *Optical-field ionized plasma x-ray lasers*. Physical Review A, **45** (9), pp 6761-6772 (1992).

45. S.G. Preston, W.J. Blyth, A.E. Dangor, A. Djaoui, M.H. Key, A. Modena, Z. Najmudin, A.A. Offenberger and J.S. Wark, *Electron temperature measurements of optically ionised plasmas* in XXIII European Conference on Laser Interactions with Matter, St. John's College, Oxford, U.K., Ed. S. J. Rose, IOP Publishing, **140**, pp 149-152 (1994).

46. B.M. Penetrante and J.N. Bardsley. *Residual energy in plasmas produced by intense subpicosecond lasers*. Physical Review A, **43** (6), pp 3100 (1991).

47. S.C. Rae. *Ionization-induced defocusing of intense laser pulses in high pressure gases*. Optics Communications, **97**, pp 25-28 (1993).

48. P. Sprangle, E. Esarey and A. Ting. *Nonlinear interaction of intense laser pulses in plasmas*. Physical Review A, **41** (8), pp 4463-4469 (1990).
49. T. Tajima and J.M. Dawson. *Laser electron accelerator*. Physical Review Letters, **43** (4), pp 267-270 (1979).
50. T.M. Antonsen and P. Mora. *Self-focussing and Raman scattering of laser pulses in tenuous plasmas*. Physical Review Letters, **69** (15), pp 2204 (1992).
51. W.B. Mori, C. Joshi, J.M. Dawson, D.W. Forslund and J.M. Kindel. *Evolution of self-focusing of intense electromagnetic waves in a plasma*. Physical Review Letters, **60** (13), pp 1298-1301 (1988).
52. P. Sprangle, E. Esarey, J. Krall and G. Joyce. *Propagation and guiding of intense laser pulses in plasmas*. Physical Review Letters, **69** (15), pp 2200 (1992).
53. X. Liu and D. Umstadter, *Self-focusing of intense subpicosecond laser pulses in a low pressure gas* in Optical Society of America - Shortwavelength V, Ed. P. B. Corkum and M. D. Perry, Optical Society of America, **17**, pp 45-49 (1993).
54. A. Sullivan, H. Hamster, S.P. Gordon, R.W. Falcone and H. Nathel. *Propagation of intense, ultrashort laser-pulses in plasmas*. Optics Letters, **19** (19), pp 1544-1546 (1994).
55. A.B. Borisov, A.V. Borovskiy, V.V. Korobhin, A.M. Prokhorov, O.B. Shiryayev, X.M. Shi, T.S. Luk, A. McPherson, J.C. Solem, K. Boyer and C.K. Rhodes. *Observation of relativistic and charge-displacement self-channeling of intense*

- subpicosecond ultraviolet (248 nm) radiation in plasmas*. Physical Review Letters, **68** (15), pp 2309-2312 (1992).
56. S.C. Rae and K. Burnett. *Detailed simulation of plasma-induced spectral blueshifting*. Physical Review A, **46** (2), pp 1084 (1992).
57. J.B. Watson, A. Sanpera and K. Burnett. *Pulse-shape effects and blueshifting in the single-atom harmonic-generation from neutral species and ions*. Physical Review A, **51** (2), pp 1458-1463 (1995).
58. C.E. Hall. *Electron microscopy*; McGraw-Hill: New York (1966).
59. A. Baez. *A study in diffraction microscopy with special reference to X-rays*. Journal of the Optical Society of America, **42**, pp 756 (1952).
60. D. Sayre, M. Howells, J. Kirz and H. Rarback. *X-ray microscopy II*; Springer-Verlag: New York, (1988).
61. C.H. Skinner, D. Diccico, L. Kim, C.H. Meixler, W. Nam, W. Tighe and S. Suckewer. *Towards shorter wavelength lasers and soft X-ray laser microscopy*. IEEE Transactions in Plasma Science, **16**, pp 512 (1988).
62. T.W. Ford, A.D. Stead and R.A. Cotton. *Soft X-ray contact microscopy*. Electron Microscopy Reviews, **4**, pp 269 (1991).
63. *Synchrotron radiation research*; Ed. M. Winick and S. Doniach; Plenum Press: New York (1980).

64. R.A. London, M.D. Rosen and J.E. Trebes. *Wavelength choice for soft X-ray laser holography of biological samples*. Applied Optics, **28** (15), pp 3397 (1989).

65. M.H. Key, T.W. Barbee, L.B. Da Silva, S.G. Glendinning, D.H. Kalantar, S.J. Rose and S.V. Weber. *New plasma diagnostic possibilities from radiography with XUV lasers*. Journal of Quantitative Spectroscopy and Radiative Transfer, **54** (1/2), pp 221-226 (1995).

66. B.J. MacGowan, S. Maxon, L.B. Da Silva, D.J. Fields, C.J. Keane, D.L. Matthews, A.L. Osterheld, J.H. Schofield, G. Shimkaveg and G.F. Stone. *Demonstration of X-ray amplifiers near the carbon K-edge*. Physical Review Letters, **65** (4), pp 420 (1990).

67. L.B. DaSilva, J.E. Trebes, R. Balhorn, S. Mrowka, E. Anderson, D.T. Attwood, T.W. Barbee, J. Brase, M. Corzett, J. Gray, J.A. Koch, C. Lee, D. Kern, R.A. London, B.J. Macgowan, D.L. Matthews and G. Stone. *X-ray laser microscopy of rat sperm nuclei*. Science, **258**, pp 269-271 (1992).

68. D.L. Matthews, P.L. Hagelstein, M.D. Rosen, M.J. Eckart, N.M. Ceglio, A.U. Hazi, H. Medeck, B.J. MacGowan, J.E. Trebes, B.L. Whitten, E.M. Campbell, C.W. Hatcher, A.M. Hawryluk, R.L. Kauffman, L.D. Pleasance, G. Rambach, J.H. Schofield, G. Stone and T.A. Weaver. *Demonstration of a Soft X-ray Amplifier*. Physical Review Letters, **54** (2), pp 110 (1985).

69. M.D. Rosen, P.L. Hagelstein, D.L. Matthews, E.M. Campbell, A.U. Hazi, B.L. Whitten, B. MacGowan, R.E. Turner, R.W. Lee, G. Charatis, G.E. Busch, C.L. Shepard and P.D. Rockett. *Exploding foil technique for achieving a soft X-ray laser*. Physical Review Letters, **54**, pp 110 (1985).

70. D. Matthews, M. Rosen, S. Brown, N. Ceglio, D. Eder, A. Hawryluk, C. Keane, R. London, B. MacGowan, S. Maxon, D. Nilson, J. Scofield and J. Trebes. *X-ray laser research at the Lawrence Livermore National Laboratory Nova laser facility*. Journal of the Optical Society of America B, **4** (4), pp 575 (1987).

71. J.A. Plowes, G.J. Pert and P.B. Holden. *Refraction compensation by target curvature in x-ray lasers*. Optics Communications, **116** (1-3), pp 260-268 (1995).

72. A. Carillon, H.Z. Chen, P. Dhez, L. Dwivedi, P. Jaegle, G. Jamelot, J. Zhang, M.H. Key, A. Kidd, A. Klisnick, R. Kodama, J. Krishnan, C.L.S. Lewis, D. Neely, P. Norreys, D.M. O'Neill, G.J. Pert, S.A. Ramsden, J.P. Raucourt, G.J. Tallents and J. Uhomoibhi. *Saturated and near diffraction limited operation of an XUV laser at 23.6 nm*. Physical Review Letters, **68** (19), pp 2917 (1992).

73. M.H. Key, W.J. Blyth, G. Cairns, A.R. Damerall, A.E. Dangor, C.N. Danson, J.M. Evans, G.J. Hirst, M. Holden, C.J. Hooker, J. Houliston, J. Krishnan, C.L.S. Lewis, J.M.D. Lister, A.G. McPhee, Z. Najmudin, D. Neely, P.A. Norreys, A.A. Offenberger, K. Osvay, G.J. Pert, S.G. Preston, S.A. Ramsden, I.N. Ross, W. Sibbett, G.J. Tallents, C. Smith, J.S. Wark and J. Zhang, *Development of injector/amplifier lasers and initial studies of ultrashort pulse UV multiphoton ionisation* in Ultrashort Wavelength Lasers II, San Diego, California, SPIE - The International Society for Optical Engineering, **2012**, pp 22-31 (1993).

74. J. Zhang and M.H. Key. *High-gain recombination O-VIII X-ray laser*. Journal of Applied Physics, **74** (12), pp 7606-7608 (1993).

75. J. Zhang and M.H. Key. *Hydrogen-like recombination X-ray lasers using ps pulse drivers*. Applied Physics B, **58** (1), pp 13-18 (1994).

76. J. Zhang, M.H. Key, P.A. Norreys, G.J. Tallents, A. Behjat, C. Danson, A. Demir, L. Dwivedi, M. Holden, P.B. Holden, C.L.S. Lewis, A.G. MacPhee, D. Neely, G.J. Pert, S.A. Ramsden, S.J. Rose, Y.F. Shao, O. Thomas, F. Walsh and Y.L. You. *Demonstration of high gain in a recombination XUV laser at 18.2nm driven by a 20J, 2ps glass laser*. Physical Review Letters, **74** (8), pp 1335-1338 (1995).

77. N.H. Burnett and P.B. Corkum. *Cold-plasma production for recombination extreme-ultraviolet lasers by optical-field-induced ionisation*. Journal of the Optical Society of America B, **6**, pp 1195 (1989).

78. P. Amendt, D.C. Eder and S.C. Wilks. *X-ray lasing by optical-field-induced ionization*. Physical Review Letters, **66** (20), pp 2589-2592 (1991).

79. P. Amendt, D.C. Eder, R.A. London and M.D. Rosen. *Plasma dispersion in ultra-short pulse X-ray lasers*. Physical Review A, **47** (2), pp 1572-1575 (1992).

80. D.C. Eder, P. Amendt, L.B. DaSilva, R.A. London, B.J. MacGowan, D.L. Matthews, B.M. Penetrante, M.D. Rosen, S.C. Wilks, T.D. Donnelly, R.W. Falcone and G.L. Strobel. *Table top X-ray lasers*. Physics of Plasmas, **1** (5), pp 1744-1752 (1994).

81. T.D. Donnelly, R.W. Lee and R.W. Falcone. *Dynamics of optical-field-ionized plasmas for x-ray lasers*. Physical Review A, **51** (4), pp R2691-R2694 (1995).

82. T.E. Glover, J.K. Crane, M.D. Perry, R.W. Lee and R.W. Falcone. *Measurement of velocity distributions and recombination kinetics in tunnel-ionized Helium plasmas*. Physical Review Letters, **75** (3), pp 445-448 (1995).

83. Y. Nagata, K. Midorikawa, S. Kubodera, M. Obara, H. Tashiro and K. Toyoda. *Soft-X-ray amplification of the Lyman-alpha transition by optical-field-induced ionisation*. Physical Review Letters, **71** (23), pp 3774 (1993).

84. Y. Nagata, K. Midorikawa, S. Kubodera, M. Obaru, H. Tashiro, K. Toyoda and Y. Kato. *Production of an extremely cold plasma by optical-field-induced ionisation*. Physical Review A, **51** (2), pp 1415-1419 (1995).

85. B.E. Lemoff, C.P.J. Barty and S.E. Harris. *Femtosecond-pulse-driven, electron-excited XUV lasers in eight-times-ionized noble gases*. Optics Letters, **19** (8), pp 569-571 (1994).

86. B.E. Lemoff, G.Y. Yin, C.L. Gordon III, C.P.J. Barty and S.E. Harris. *Demonstration of a 10-Hz, femtosecond-pulse-driven XUV laser at 41.8nm in Xe IX*. Physical Review Letters, **74** (9), pp 1574-1577 (1994).

87. M. Castro-Collin, *Personal communication*. (1995).

88. P.A. Franken, A.E. Hill, C.W. Peters and G. Weinreich. *Generation of optical harmonics*. Physical Review Letters, **7**, pp 118-119 (1961).

89. K.C. Kulander, K.J. Schafer and J.L. Krause. *Dynamics of short-pulse excitation, ionization and harmonic conversion*. in *Super-intense laser-atom physics*; Ed. B. P. e. al., ; Plenum Press: New York, ; pp 95-110 (1993).

90. J.L. Krause, K.J. Schafer and K.C. Kulander. *High-order harmonic generation from atoms and ions in the high intensity regime*. Physical Review Letters, **68** (24), pp 3535 (1992).
91. J.K. Crane, M.D. Perry, S. Herman and R.W. Falcone. *High-field harmonic generation in helium*. Optics Letters, **17** (18), pp 1256-1258 (1992).
92. N. Sarukura, K. Hata, T. Adachi, R. Nodomi, M. Watanabe and S. Watanabe. *Coherent soft x-ray generation by the harmonics of an ultrahigh-power KrF laser*. Physical Review A, **43** (3), pp 1669 (1991).
93. A. Sanpera, P. Jonson, J.B. Watson and K. Burnett. *Harmonic generation beyond the saturation intensity in Helium*. Physical Review A, **51** (4), pp 3148-3153 (1995).
94. P.B. Corkum. *Plasma perspective on strong-field multiphoton ionization*. Physical Review Letters, **71** (13), pp 1994-1997 (1993).
95. A. L'Huillier, K.J. Schafer and K.C. Kulander. *High-order harmonic generation in Xenon at 1064 nm: the role of phase matching*. Physical Review Letters, **66** (17), pp 2200 (1991).
96. A. L'Huillier, P. Balcou and L.A. Lompre. *Coherence and resonance effects in high order harmonic generation*. Physical Review Letters, **68** (2), pp 166 (1992).
97. A. L'Huillier and P. Balcou. *High order harmonic generation in rare gases with a 1-ps 1053-nm laser*. Physical Review Letters, **70** (6), pp 774 (1993).

98. P. Balcou and A. L'Huillier. *Phase-matching effects in strong-field harmonic generation*. Physical Review A, **47** (2), pp 1447-1457 (1993).
99. C.-G. Wahlstrom, S. Borgstrom, J. Larsson and S.-G. Pettersson. *High-order harmonic generation in laser-produced ions using a near-infrared laser*. Physical Review A, **51** (1), pp 585-591 (1995).
100. C.-G. Wahlstrom, J. Larsson, A. Perrson, T. Starczewski, S. Svanberg, P. Salieres, P. Balcou and A. L'Huillier. *High-order harmonic generation in rare-gases with an intense short-pulse laser*. Physical Review A, **48** (6), pp 4709-4720 (1993).
101. C.G. Wahlstrom. *High-order harmonic-generation using high-power lasers*. Physica Scripta, **49** (2), pp 201-208 (1994).
102. J.E. Muffett. *A numerical model of high harmonic generation in the rare gases*. Ph.D Thesis, Imperial College of Science and Technology and Medicine, (1994).
103. P. Dietrich, N.H. Burnett, M. Ivanov and P.B. Corkum. *High-harmonic generation and correlated two-electron multiphoton ionization with elliptically polarized light*. Physical Review A, **50** (5), pp R3585-R3588 (1994).
104. P. Salieres, A. L'Huillier and M. Lewenstein. *Coherence control of high-order harmonics*. Physical Review Letters, **74** (19), pp 3776-3779 (1995).
105. K. Kondo, T. Tamida, Y. Nabekawa and S. Watanabe. *High-order harmonic generation and ionization using ultrashort KrF and Ti:sapphire lasers*. Physical Review A, **49** (5), pp 3881-3889 (1994).

106. C.R. Vidal. *4-wave frequency mixing in gases in Tunable Lasers, Topics in Applied Physics*; Ed. L. F. Mollenauer and J. C. White, ; Springer: Heidelberg, ; Vol. 59; pp 57-113 (1987).

107. T. Ditmire, J.K. Crane, H. Nguyen, L.B. DaSilva and M.D. Perry. *Energy-yield and conversion-efficiency measurements of high-order harmonic radiation*. Physical Review A, **51** (2), pp R902-R905 (1995).

108. R. Haight and D.R. Peale. *Tunable photoemission with harmonics of subpicosecond lasers*. Review of Scientific Instruments, **65** (6), pp 1853-1857 (1994).

109. J. Larsson, E. Mevel, R. Zerne, A. Lhuillier, C.G. Wahlstrom and S. Svanberg. *2-color time-resolved spectroscopy of helium using high-order harmonics*. Journal of Physics B, **28** (3), pp L53-L58 (1995).

110. T. Ditmire, M.H.R. Hutchinson, M.H. Key, C.L.S. Lewis, A. MacPhee, I. Mercer, D. Neely, M.D. Perry, R.A. Smaith, J.S. Wark and M. Zepf. *Amplification of XUV harmonic radiation in a Gallium amplifier*. Physical Review A, **51** (6), pp R4337-R4340 (1995).

111. C.E. Webb. *Undergraduate lecture notes*. University of Oxford, (1992).

112. E.C. Harvey, C.J. Hooker, J.R. Houlston, M.H. Key, A.K. Kidd, J.M.D. Lister, I.N. Ross and M.J. Shaw. *12ps operation of the Sprite KrF pumped Raman laser at exceptionally high brightness* . CLF Annual Report to the Laser Facility Committee, RAL-91-025, RAL, (1991).

113. I.N. Ross, A.R. Damerall, E.J. Divall, J. Evans, G.J. Hirst, C.J. Hooker, J.R. Houliston, M.H. Key, J.M.D. Lister, K. Osvay and M.J. Shaw. *A 1 TW KrF laser using chirped pulse amplification*. Optics Communications, **109**, pp 288 (1994).
114. I.N. Ross, A.R. Damerall, E. Divall, J.M. Evans, G.J. Hirst, C.J. Hooker, J.R. Houliston, M.H. Key, Z. Najmudin, P. Norreys, K. Osvay and J.M.D. Lister. *1-terawatt, 300 femtosecond pulses by chirped pulse amplification in Sprite*. CLF Annual Report to the Laser Facility Committee, RAL-93-031, RAL, (1993).
115. A. Migus, *Femtosecond and subpicosecond ultra-intense lasers* in SUSSP - Laser-Plasma Interactions, St. Andrews University, (1994).
116. M.J. Shaw, E.J. Divall, G.J. Hirst, C.J. Hooker and A. Kvaran. *Bandwidth limitations in KrF lasers*. Office of Naval Research, Contract Award N68171-95-C-9040, Rutherford Appleton Laboratory, (1995).
117. J.M.D. Lister, *Personal communication*. (1994).
118. A.E. Siegman. *Pulse breakup in practical laser systems: the B integral in Lasers*; University Science Books: Mill Valley, California, ; pp 385-386 (1986).
119. Y.M. Li and R. Fedosejevs. *Density-measurements of a high-density pulsed gas-jet for laser-plasma interaction studies*. Measurement Science and Technology, **5** (10), pp 1197-1201 (1994).
120. T. Kita, T. Harada, N. Nakano and H. Kuroda. *Mechanically ruled aberration-corrected concave gratings for a flat-field grazing-incidence spectrograph*. Applied Optics, **22** (4), pp 512-513 (1983).

121. B.L. Henke, P. Lee, T.J. Tanaka, R.L. Shimabukuro and B.K. Fujikawa. *Low-energy X-ray interaction coefficients: Photoabsorption, scattering, and reflection*. Atomic Data and Nuclear Data Tables, **27** (1), pp 1-144 (1982).

122. M. Zepf, A.G. MacPhee, C.L.S. Lewis and J.S. Wark. *Comparison of X-ray detectors*. CLF Annual Report to the Laser Facility Committee, To be published, DRAL, (1995).

123. J. Edelstein, M.C. Hettrick, S. Mrowka, P. Jelinsky and C. Martin. *Extreme UV measurements of a varied line-space Hitachi reflection grating: efficiency and scattering*. Applied Optics, **23** (19), pp 3267-3270 (1984).

124. D. Neely, *Personal communication*. (1995).

125. K.L. Bowles. *Radio wave scattering in the ionosphere*. Advances in Electronics and Electron Physics, **19**, pp 55 (1964).

126. A.A. Offenberger, W. Blyth, A.E. Dangor, A. Djaoui, M.H. Key, Z. Najmudin and J.S. Wark. *Electron temperature of optically ionised gases produced by high intensity 268 nm laser radiation*. Physical Review Letters, **71** (24), pp 3983-3986 (1993).

127. W.J. Blyth, S.G. Preston, A.A. Offenberger, M.H. Key, J.S. Wark, Z. Najmudin, A. Modena and A.E. Dangor. *Plasma temperature in optical field ionisation of gases by intense ultrashort pulses of ultraviolet radiation*. Physical Review Letters, **74** (4), pp 554-557 (1995).

128. W.E.R. Davies and S.A. Ramsden. *Scattering of light from the electrons in a plasma*. Physics Letters, **8**, pp 179 (1964).

129. A.W. DeSilva, D.E. Evans and M.J. Forrest. *Observation of Thomson and co-operative scattering of ruby laser light*. Nature, **203**, pp 1321 (1964).

130. H.J. Kunze, A. Eberhagen and E. Funfer. *Electron density and temperature measurements in a 26 kJ θ -pinch by light scattering*. Physics Letters, **13**, pp 38 (1964).

131. E.T. Gerry and D.J. Rose. *Plasma diagnostics by Thomson scattering of a laser beam*. Journal of Applied Physics, **37**, pp 2715 (1966).

132. T.E. Glover, T.D. Donnelly, E.A. Lipman, A. Sullivan and R.W. Falcone. *Subpicosecond Thomson-Scattering Measurements of Optically Ionised Helium Plasmas*. Physical Review Letters, **73** (1), pp 78-81 (1994).

133. J. Sheffield. *Plasma scattering of electromagnetic radiation*; Academic Press: London (1975).

134. D.E. Evans and J. Katzenstein. *Laser light scattering in laboratory plasmas*. Reports of Progress Physics, **32**, pp 207-271 (1969).

135. E.E. Salpeter. *Electron density fluctuations in a plasma*. Physical Review, **120** (5), pp 1528-1535 (1960).

136. B.D. Fried and S.D. Conte. *The plasma dispersion function*; Academic Press: New York (1961).

137. J.S. Wark, A.A. Offenberger, W.J. Blyth, M.H. Key, S.G. Preston, Z. Najmudin, A.E. Dangor and A. Djaoui, *Thomson scattering measurements of optically ionised plasmas produced by intense KrF radiation: the observation of high electron to ion temperature ratios* in Applications of laser plasma radiation, San Diego, California, Ed. M. C. Richardson, SPIE - The International Society for Optical Engineering, **2015**, pp 270-276 (1993).

138. A. Djaoui. *Short pulse high intensity laser interaction with gas targets* . CLF Annual Report to the Laser Facility Committee, RAL-93-031, RAL, (1993).

139. A. Djaoui, *Personal Communication*. (1995).

140. L. Spitzer and R. Härm. *Transport phenomena in a completely ionized gas*. Physical Review, **89**, pp 977-981 (1953).

141. P.A. Holstein, J. Delettrez, S. Skupsky and J.P. Matte. *Modelling nonlocal heat-flow in laser-produced plasmas*. Journal of Applied Physics, **60** (7), pp 2296-2300 (1987).

142. E.M. Epperlein and R.W. Short. *A practical nonlocal model for electron heat transport in laser plasmas*. Physics of Fluids B, **3** (11), pp 3092 (1991).

143. J.F. Luciani, P. Mora and R. Pellat. *Quasistatic heat front and delocalized heat flux*. Physics of Fluids, **28** (3), pp 835 (1985).

144. M. Dunne, T. Afshar-Rad, J. Edwards, A.J. Mackinnon, S.M. Viana and O. Willi. *Experimental observations of expansion of an optical-field-induced ionization channel in a gas jet target*. Physical Review Letters, **72** (7), pp 1024-1027 (1994).

145. H.R. Griem. *Spectral line broadening by plasmas*; Academic Press: New York and London, Vol. 39 (1974).

146. W.L. Kruer, *Theory and simulation of laser plasma coupling-II* in Laser-plasma interactions II, Edinburgh, Ed. R.A.Cairns, Scottish Universities Summer School in Physics, pp 185-204 (1982).

147. B.L. Henke, J.Y. Uejio, G.F. Stone, C.H. Dittmore and F.G. Fujiwara. *High energy X-ray response of photographic films. Models and measurements.* Journal of the Optical Society of America B, **3**, pp 1540 (1986).

148. A. Djaoui, *Personal Communication.* (1995).

149. H. Xu, X. Tang and P. Lambrououlos. *Nonperturbative theory of harmonic generation in helium under a high-intensity laser field: The role of intermediate resonance and of the ion.* Physical Review A, **46** (5), pp R2225-R2228 (1992).

150. M. Zepf, W. Blyth, M.H. Key, M. Nakai, S.G. Preston and J.S. Wark, *High harmonic generation with a CPA KrF laser* in XXIII European conference on Laser Interactions with Matter, St. John's College, Oxford, Ed. S. J. Rose, IOP Publishing, **140**, pp 467-470 (1994).

151. J.W.G. Tisch, R.A. Smith, J.E. Muffett, M. Ciarrocca, J.P. Marangos and M.H.R. Hutchinson. *Angularly resolved high-order harmonic generation in helium.* Physical Review A, **49** (1), pp R28-R31 (1994).

152. J. Zhou, J. Peatross, M.M. Murnane and H.C. Kapteyn. *Enhanced high-harmonic generation using 25 femtosecond laser pulses*. Submitted to Physical Review Letters, , pp (1995).
153. S. Borgstrom, E. Fill, J. Larsson, T. Starczewski, J. Steingruber, S. Svanberg and C. Wahlstrom, *X-ray spectra of optical-field ionized plasma* in XXIII European Conference on Laser Interactions with Matter, St. Johns College, Oxford, Ed. S. J. Rose, IOP Publishing, **140**, pp 141-144 (1994).
154. E. Fill, S. Borgstrom, J. Larsson, T. Starczewski, C.-G. Wahlstrom and S. Svanberg. *XUV spectra of optical-ionized plasma*. Physical Review E, **51** (6), pp 6016-6027 (1995).
155. T. Zuo and A.D. Bandrauk. *Control of high-order harmonic generation in strong laser fields*. Physical Review A, **51** (5), pp 3991-3998 (1995).
156. S. Watanabe, K. Kondo, Y. Nabekawa, A. Sagisaka and Y. Kobayashi. *Two-color phase control in tunnelling ionization and harmonic generation by a strong laser field and its third harmonic*. Physical Review Letters, **73** (20), pp 2692-2695 (1994).