Studies of extreme ultraviolet emission from laser produced plasmas, as sources for next generation lithography

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To my parents
Abstract

The work presented in this thesis is primarily concerned with the optimisation of extreme ultraviolet (EUV) photoemission around 13.5 nm, from laser produced tin (Sn) plasmas. EUV lithography has been identified as the leading next generation technology to take over from the current optical lithography systems, due to its potential of printing smaller feature sizes on integrated circuits. Many of the problems hindering the implementation of EUV lithography for high volume manufacturing have been overcome during the past 20 years of development. However, the lack of source power is a major concern for realising EUV lithography and remains a major roadblock that must be overcome. Therefore in order to optimise and improve the EUV emission from Sn laser plasma sources, many parameters contributing to the make-up of an EUV source are investigated.

Chapter 3 presents the results of varying several different experimental parameters on the EUV emission from Sn laser plasmas. Several of the laser parameters including the energy, gas mixture, focusing lens position and angle of incidence are changed, while their effect on the EUV emission is studied. Double laser pulse experiments are also carried out by creating plasma targets for the main laser pulse to interact with. The resulting emission is compared to that of a single laser pulse on solid Sn.

Chapter 4 investigates tailoring the CO$_2$ laser pulse duration to improve the efficiency of an EUV source set-up. In doing so a new technique for shortening the time duration of the pulse is described. The direct effects of shortening the CO$_2$ laser pulse duration on the EUV emission from Sn are then studied and shown to improve the efficiency of the source.

In Chapter 5 a new plasma target type is studied and compared to the previous dual laser experiments. Laser produced colliding plasma jet targets form a new plasma layer, with densities that can be optimised for re-heating with the main CO$_2$ laser pulse.

Chapter 6 will present some experiments carried out on laser produced gadolinium plasmas, with its photoemission around 6.7 nm seen as a potential beyond EUV source. Three different laser pulse durations and a range of laser intensities are utilised in experiments to try to optimise the in-band emission, while also observing the effect on ion emission from the plasma. Finally, the experiments presented in thesis and their results are summarised in Chapter 7, along with presenting possible future work.
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Chapter 1

Introduction

1.1 Introduction

The semiconductor industry has developed and introduced new technologies at a rate unlike any other industry over the past forty years. The key component responsible for this rate of progress is the transistor, and critically, how many transistors can be fitted onto a single computer chip. The transistor acts as a switch that determines if a current will flow or not. This on or off current is directly related to the ones and zeros of the binary code on which all computer functions are based. In essence, the more transistors that are present, the faster the computer. Intel produced their first commercially available chip in 1971, called the 4004, which contained a total of just 2,300 transistors. Contrast that with the 2.6 Billion transistors of the 10-Core Xeon Westmere-EX IC that Intel released in 2011. This means in the space of forty years the number of transistors on an IC has increased by a factor of a million. Moore’s law has described this steady increase in the number of transistors. The law is based on an article written by Intel co-founder Gordon Moore in 1965 [1]. In it he stated that the number of transistors would roughly double every year, for ten years. He revisited his law in 1975 to update it to a doubling of the number of transistors every two years [2]. Since then, the International Technology Roadmap for Semiconductors (ITRS) has used Moore’s law as a blueprint for the current and future growth in the number of transistors.

The increase in the number of transistors is achieved by progressively reducing the size of the features on the ICs. The half-pitch (half the distance between identical features on a circuit) must therefore be continuously reduced to keep up with Moore’s law. The scale of the shrinkage is clear when the example of the Intel 4004 IC, with a half-pitch of 10 \( \mu \text{m} \), is again
1.2. Current lithography process

compared to the 22 nm half-pitch of the 10-Core Xeon Westmere-EX IC. This constitutes a reduction in size by a factor of over four hundred in that time. However the current techniques used to print ICs are becoming more complex and risk becoming less cost effective [3]. Therefore, in order to continue this reduction, the semiconductor industry will most likely require the introduction of next generation lithography (NGL) technologies to succeed the current lithography processes. One such NGL technology that has been heavily researched and invested in by industry and academia alike is extreme ultraviolet (EUV) sources for extreme ultraviolet lithography (EUVL). The motivation for the research presented in this thesis is the optimisation of EUV sources for lithography.

![Graph showing the change in the half pitch or minimum feature size and its year of introduction, note the log scale on the y-axis [4].](image)

Figure 1.1: Graph showing the change in the half pitch or minimum feature size and its year of introduction, note the log scale on the y-axis [4].

1.2 Current lithography process

The lithography process and improvements made to it, have been integral parts of the continued production of IC chips in accordance with Moore’s law. The basic lithography process involves a few vital components. These are: a light source, a mask, an optical system, a photoresist and a silicon (Si) wafer substrate. A simple lithography layout is shown in Fig. 1.2. Firstly, the light from a source is projected onto a mask. This mask or
1.2. Current lithography process

The photomask is typically made of transparent quartz or fused silica, which has a series of lines and patterns laid out on it. These patterns are defined by a thin film of light-absorbing chrome. Transparent parts of the mask will thus transmit the light, while the rest is opaque. The layout of the optical system used can vary depending on the type of photolithography system (projection, contact or proximity lithography). For the highest resolution and smallest achievable features projection lithography is used. This optical system consists of a condenser and an objective lens. The lenses are used to collimate the light source evenly on the mask and then reduce it, by typically 4:1 demagnification, on to the surface of the Si wafer. The surface of the Si wafer is coated with a photosensitive resist, meaning the portions of the resist that are exposed to the light from the source will become soluble when a solution is applied, while the portions not illuminated remain insoluble. See Fig. 1.3 on the following page for a step by step schematic of the process of printing features on to the Si wafer.

![Figure 1.2: A simple illustration of the photolithography process.](image)
1.2. Current lithography process

In order to increase the number of transistors, the size of the features etched on the silicon wafer must be continuously reduced. The minimum feature size ($F_{\text{min}}$), which is physically limited by the resolution of the optical system, is related to Lord Rayleigh’s equation. This is also known as the Rayleigh criterion:

$$F_{\text{min}} = k \frac{\lambda}{NA}$$  \hspace{1cm} (1.1)

where $k$ is a process constant, $\lambda$ is the exposure wavelength and $NA$, the numerical aperture, is given by Eqn. 1.2. In this equation $\theta_0$ is equal to the minimum angular separation between two objects that will still allow the objects to be resolved and $n$ represents the refractive index of the medium between the projection lens and the wafer.

$$NA = n \sin \theta_0$$  \hspace{1cm} (1.2)

The process parameter $k$ is dependent on resist parameters such as baking time and the coherence of the illumination [6], and it has a physical limit of $\geq 0.25$ [7]. Thus, from the Rayleigh criterion it can be seen that in order to
1.2. Current lithography process

achieve a reduction in feature size there must be a reduction in the wavelength of the light source or an increase in the value of $NA$. This has been the case over the years, as the source wavelength has been reduced. Since the middle of the 1980’s $\lambda$ has been reduced from the 436 nm g-line and 365 nm i-line of mercury vapour lamps, to the 193 nm argon fluoride (ArF) laser used today [Fig. 1.4] [8]. At the same time, various improvements and different techniques have been used to increase the $NA$ of the lithographic systems also. This has increased from the $NA$ of 0.30, used in the mid-1980’s with the 436 nm g-line [9], to 1.35 used in 2011 by the most advanced 193 nm ArF immersion scanners [10].

A limitation was reached using the deep ultraviolet (DUV) ArF systems at 193 nm, meaning it could not be used to print features smaller than 65 nm. Further reduction in the minimum feature size, down to the 45 nm half-pitch node, was thus achieved by the introduction of immersion lithography (193i) [11]. Immersion works by submerging the optical system in a liquid, typically water, with a higher refractive index ($n$) than air. This results in a reduction in the effective wavelength (\(\lambda_{eff}\)) [Eqn. 1.3] and hence a reduction in the feature size printed [Eqn. 1.4].

\[
\lambda_{eff} = \frac{\lambda}{n} \quad (1.3)
\]

\[
F_{min} = k \frac{\lambda}{n} \frac{1}{NA} \quad (1.4)
\]

Further extension of the DUV source down to the 32 nm and beyond to 22 nm, was achieved by implementing double patterning (DP) techniques to print the most critical components by printing them twice [12]. The next step using current wavelengths would then be triple or multiple patterning. This would further increase the complexity and costs of scanners, and it has been suggested by many in the semiconductor industry, that the solution is to implement a NGL scanner by reducing the source wavelength down to 13.5 nm, in the EUV region (EUV is loosely defined as between 50–5 nm [13]).

Another advantage of EUV over DP at 193i is its larger usable depth of focus (DOF), which is defined as [14]:

\[
DOF = k \frac{\lambda}{NA^2} \quad (1.5)
\]

From Eqn. 1.5, it can be seen that for an EUVL scanner at 13.5 nm with a probable $NA$ of between 0.25 and 0.32 [15], a usable $DOF$ of >100 nm can be achieved. Whereas DP with 193i and a $NA$ of 1.35 has a $DOF$ of 50 nm [16].
1.2. Current lithography process

This quote from the 2012 update of the ITRS sums up the current state of the industry and the role EUV is expected to play:

“Extending lithography to smaller dimensions has always been difficult and the 2012 effort is no exception. Single optical exposure has reached its limit at roughly 40 nm half-pitch (hp) using 193 nm wavelength (ArF) exposure tools. Flash devices with 32 nm hp are being manufactured today using double patterning (DP) to reduce half-pitch while keeping the existing exposure NA and wavelength. This approach will be pushed harder as DRAM and MPU drive down to the 32 nm hp and Flash starts to test the limits of double patterning at 22 nm hp in 2013. For even smaller dimensions, extreme ultraviolet lithography (EUVL), multiple patterning (MP) or some non-optical lithography must be introduced. EUVL, which uses light with a wavelength of 13.5 nm, is the clear preference of the semiconductor industry for patterning smaller dimensions. EUVL has been gaining significant momentum with several manufacturers running early EUV pilot lines and some manufacturers have announced plans to purchase production tools that will be delivered in 2013.” [10].

![Diagram showing reduction in source wavelength over time](image_url)
1.3. Extreme ultraviolet lithography

1.3 Extreme ultraviolet lithography

The source wavelength chosen for next generation EUVL is 13.5 nm. The reason for selecting this wavelength was driven by the optics that will be required in an EUVL system. One of the main challenges in moving from the current optical lithography to EUVL is that EUV radiation is strongly absorbed in all materials, including air [17]. This has two major implications for a proposed EUVL source; firstly, the entire system must be kept under high vacuum and secondly, refractive optics can no longer be used, as with 193 nm, but must be replaced with reflective optics. A key step in realising EUVL was the availability of highly reflective multilayer mirrors (MLM) that could be used to reflect the EUV radiation. High reflectance in the EUV region close to 70% had been achieved with molybdenum silicon (Mo/Si) multilayers at 13.5 nm and molybdenum beryllium (Mo/Be) at 11.4 nm [18]. However Mo/Be was not considered for use in the industry due to manufacturing problems and the fact that Be is a carcinogen [19]. Therefore it was the high reflectivity of Mo/Si at 13.5 nm that actually determined the wavelength for a potential future EUV source [13, 17, 19, 20].

1.3.1 Multilayer mirrors

The reflectivities of a single Mo/Si MLM and after 11 reflections are shown in Fig. 1.5. It is expected that 11 mirrors will be required for the illuminator and projection optics of the proposed optical layout of an EUVL scanner [21] [Fig. 1.6]. After these reflections the bandwidth of the mirrors is shown to be reflective between 13.365 and 13.635 nm, in other words the whole system only reflects radiation coming from the source within a 2% bandwidth centered on 13.5 nm. The MLMs are made by building up alternating layers of high-Z (Mo) and low-Z (Si) materials onto a substrate. In theory the thickness of the low-Z and high-Z layers combined should be approximately half the wavelength of the light whose reflection is desired [14]. That means for 13.5 nm radiation, the thickness of each bilayer of Mo/Si is around 6.8 nm (for example Mo = 2.7 nm and Si = 4.1 nm). If these specific thicknesses are used then the 13.5 nm radiation incident at a particular angle will undergo high reflectivity, as the reflections from each interface adds up in phase due to constructive interference [Fig. 1.7] [13]. The light is scattered from the high-Z layer (absorber), while the low-Z layer (spacer) acts as a buffer to minimise the absorption.
1.3. Extreme ultraviolet lithography

Figure 1.5: Mo/Si multilayer mirror reflectivity around 13.5 nm [22].

Figure 1.6: Schematic of the optical layout of an EUV lithography system [23].
1.3. Extreme ultraviolet lithography

The MLM thus acts as a Bragg reflector, with maximum reflectivity achieved according to the Bragg equation [13]:

\[ m\lambda = 2H\sin\theta_m \left( 1 - \frac{2\bar{\delta} - \bar{\delta}^2}{S\sin^2\theta_m} \right)^{\frac{1}{2}} \quad (1.6) \]

where \( m \) is the order of Bragg reflection, \( \lambda \) is the wavelength of the reflected light, \( \theta_m \) is the grazing angle of maximum reflectivity, \( H \) is the thickness of the MLM period given by Eqn. 1.7 and \( \bar{\delta} \) is the weighted-average difference of the refractive index from unity for the spacer and absorber materials in the multilayer structure [Eqn. 1.8] [13].

\[ H = d_{Mo} + d_{Si} \quad (1.7) \]

\[ \bar{\delta} = \frac{\delta_{Mo}d_{Mo} + \delta_{Si}d_{Si}}{d_{Mo} + d_{Si}} \quad (1.8) \]

where \( d_{Mo} \) is the thickness of the Mo layer and \( d_{Si} \) is the thickness of the Si layer.

Figure 1.7: Schematic of the constructive interference of a bi-layer structure (left) and a TEM image of an actual multilayer coating with a period of \( H = 7 \) nm (right) [13]
1.3. Extreme ultraviolet lithography

1.3.2 EUVL source

After the choice of wavelength for EUVL had been made, a viable source of 13.5 nm radiation was required. Plasmas are known to be broad-wavelength range sources of radiation, that depending on the rate of ionisation, emit light from the infrared down through the ultraviolet to the X-ray region of the electromagnetic spectrum [17]. Laser produced plasmas (LPP) [24–27] and discharge produced plasmas (DPP) [28, 29], which include laser-assisted discharge plasmas (LDP) [30–32], became the prominent techniques studied in the field of EUV source development [21, 33]. Alternative sources, which are undulator based sources such as synchrotron radiation and free electron lasers, as well as x-ray lasers, are also potential EUV sources, but are mostly discounted due to their scale and cost. Basic schematics of both LPP and DPP sources are shown in Fig. 1.8 below.

(a)

![LPP schematic](image)

(b)

![DPP schematic](image)

Figure 1.8: Schematic of (a) LPP source and (b) DPP source [33].
1.3. Extreme ultraviolet lithography

The plasma is generated by a high current pinch in the case of DPP and a focussed laser beam for LPP. The EUV radiation from the LPP source is generally collected by a normal incidence multi layer collector, while the plasma produced by a DPP, due to geometrical constraints and the EUV being mostly emitted in the forward direction (less than $2\pi$sr), uses a grazing incidence collector [34]. There is also a high degree of debris mitigation required for both sources as the plasma will generate large amounts of debris in the form of high energy ions and also particulate debris from the fuel that is not used up. This can cause serious damage to optics by bombardment and also coat the collector optic, which will reduce the reflectivity and throughput of the system. For normal incidence collectors, mitigation can include a buffer gas, magnetic field and use of mass limited targets (to reduce excess amounts of fuel), while for grazing incidence collectors a foil trap with gas flow can be used [35, 36]. Spectral purity filters (SPFs) may also be required to remove any out of band (OOB) radiation, from 100–3000 nm, emanating from the source [37]. This requirement is due to the fact that the reflectivity of the MLM is also quite high across this region ($R=40–90\%$) [Fig. 1.9] [19]. OOB radiation emitted in the UV region will cause flare on the wafer, which will effect the resolution of the lines printed due to the resist also being sensitive to radiation in these regions. The infra-red (including lasers used in LPP @ 10.6 $\mu$m) will cause heating of the wafer, resulting in overlay issues.

Figure 1.9: OOB reflectivity of multilayer coated mirrors [19, 37].
1.3. Extreme ultraviolet lithography

The intermediate focus (IF) point, shown in Fig. 1.8, is the point at which the EUV light, after all debris mitigation and SPFs, is focussed by the collector optic and enters the illuminator [19]. The amount of “clean photons” [33] of EUV power generated by an EUVL source is thus referred to as the power at this IF position. This is because a direct comparison can then be made between different LPP and DPP sources which may be using different forms of debris mitigation and spectral filtering. There is another restriction on the EUV source at this point, in that it must meet the etendue limitations. This means the size of the EUV emitting plasma must not be so large that it cannot be accepted by the scanner. The etendue of a source is given by Eqn. 1.9 [7]:

\[ \text{Optic etendue} = \text{Field area} \times \text{acceptance solid angle} \] (1.9)

where the Optic etendue (mm\(^2\) sr) is limited to a maximum accepted etendue of 3.3 mm\(^2\) sr. The Field area (mm\(^2\)) refers to the area of the EUV emitting source and the acceptance solid angle (sr) is the solid angle of collection by the collector optic, which is 2\(\pi\) sr or greater (EUV emission up to 4\(\pi\) sr is possible using droplet targets) for LPP sources but less than 2\(\pi\) sr for DPP due to the proximity of the plasma to the electrodes and the electrodes shape [34]. Using Eqn. 1.9, a calculation of the maximum Field area in 2\(\pi\) sr can be made, which works out as approximately a 720×720 \(\mu m\) sized plasma. LPPs studied for EUV tend to be within this size limit [3, 38], however the plasma created by DPP tend to be larger, especially if the power is increased to try and achieve greater output. This can lead to an etendue mismatch and an inability to collect some of the in-band EUV radiation.

The duty cycle of an EUVL source is also an important characteristic. The heat generated by both sources, but in particular the electrodes of DPP sources [39], means that they can not be run continuously for long periods of time. Often the power of a source is quoted along with the average power, which takes into account the duty cycle of the source. Recently, Cymer Inc. reported very high duty cycles of better than 90% for their 40 W dual-laser pulse, droplet EUVL source for high volume manufacturing (HVM) running for 8 hours [40]. LDP sources, which use a laser to spark the discharge between rotating wheels acting as an anode and cathode, have been shown to have high duty cycles of up to 90% for 20 W power due to the cooling effect of the rotating wheels [31].
1.3. Extreme ultraviolet lithography

A table of the various requirements for an EUVL source are listed in Table 1.1. Much of the source characteristics have already been discussed in this chapter. The source requirement of 10% source cleanliness (as measured by collector reflectivity) after 30,000 hours operation is at the point of IF. The power requirements of 115 W @ 5 mJ/cm\(^2\) and 200 W @ 10 mJ/cm\(^2\) are also both at IF. The minimum power requirement for an EUV source is thus shown to be 115 W at IF. However this is for a resist sensitivity of 5 mJ/cm\(^2\). The smaller resist sensitivity effects the minimum resolution of the lines (i.e. the 22 nm node and beyond) and their line edge roughness (LER). This leads to a resolution-LER-sensitivity (RLS) trade off between the three parameters [7], which means higher source powers at IF will actually be required.

<table>
<thead>
<tr>
<th>Source characteristics</th>
<th>Requirements</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength</td>
<td>13.5 nm</td>
</tr>
<tr>
<td>EUV power (inband)</td>
<td>115 W @ 5 mJ/cm(^2) - 200 W @ 10 mJ/cm(^2)</td>
</tr>
<tr>
<td>Repetition frequency</td>
<td>&gt;10 kHz</td>
</tr>
<tr>
<td>Integrated energy stability</td>
<td>±0.2%, 3σ over 50 pulses</td>
</tr>
<tr>
<td>Source cleanliness (Debris)</td>
<td>Reflectivity degradation ≤ 10% after 30,000 hours operation</td>
</tr>
<tr>
<td>Etendue of source output</td>
<td>3.3 mm(^2) sr maximum</td>
</tr>
<tr>
<td>Max. solid angle input to illuminator</td>
<td>0.03–0.2 sr</td>
</tr>
<tr>
<td>Spectral purity:</td>
<td></td>
</tr>
<tr>
<td>130–400 nm (DUV/VUV)</td>
<td>&lt;1% at wafer</td>
</tr>
<tr>
<td>≥400 nm (IR/Vis) including 10.6 μm</td>
<td>&lt;10% at wafer</td>
</tr>
</tbody>
</table>

Table 1.1: EUV source requirements [41]

Fig. 1.10 on the next page demonstrates the increasing power requirements due to the reduction in sensitivity of the resist and also its effect on wafer throughput, with 100 wafers per hour (wph) being the benchmark requirement from a scanner. The direct effect of resist sensitivity on the LER of the printed features can be seen in Fig. 1.11, also on the next page. Source power and achieving higher power at IF remains a huge focus of efforts in the field of EUVL. Yan Borodovsky of Intel reported at an EUVL meeting in Hawaii in 2012, that in order for EUV light to print critical features, a higher resist dose of 60 mJ/cm\(^2\) is needed, with a predicted requirement of 1 kW source power at IF [42].
1.3. Extreme ultraviolet lithography

Figure 1.10: Wafer throughput dependence on source power and resist sensitivity [41].

Figure 1.11: The effect of resist sensitivity on the line edge roughness of printed features [43].
1.3. Extreme ultraviolet lithography

1.3.3 Source fuel

Lithium (Li) [44–46], xenon (Xe) [28, 29, 47, 48] and tin (Sn) [24–27] have each been studied over the years as a potential fuel for an EUVL source. The interest in Li arises from the fact that it has line emission near 13.5 nm. This is from the $1s^2 \rightarrow 2p$ Lyman alpha transition of the hydrogen-like Li$^{2+}$ ion [45], which results in intense line emission within the 2% bandwidth of the Mo/Si MLMs centered on 13.5 nm. The conversion efficiency (CE), of the energy used to create the plasma source (by either LPP or DPP) into in-band EUV energy, is a commonly used method to describe the applicability of a particular fuel as an EUVL source and compare it with other source materials. Li has been reported as having a CE of between 0.2–2% [44–46]. A benefit of Li as a target is due to its emission being from a single $1s^2 \rightarrow 2p$ line, which means there is not much OOB radiation produced. Another advantage of Li as an EUVL source is that it produces relatively low energy ions during the plasma lifetime, as compared with Sn, which can be more easily deflected by debris mitigation systems [46].

Xe has long been viewed as a strong candidate as the potential fuel of an EUVL source. The emission in Xe around 13.5 nm is due to the radiation emitted from the $4d \rightarrow 5p$ transition array of the Xe$^{10+}$ ion [49]. Xe targets have appeared in many forms including: gas targets, liquid jets and cryogenic solid targets amongst others. The main advantage of Xe as a source fuel comes from the fact that it is an inert noble gas at room temperature, which means that it is an extremely clean plasma, removing the concerns over debris [50]. A maximum CE of between 0.7–1.4% has been reported [51].

The final proposed material for an EUV source is Sn. Sn differs from the other potential source fuels in that its in-band emission is not due to a single ion but many thousands of lines from the $4d^2 4f$, $4p^2 4d$ and $4d^5 5p$ radiation emitted by the Sn$^{7+}$–Sn$^{12+}$ ions [5, 22, 52]. This leads to thousands of overlapping transitions that have such small spacing between each line that they can not be resolved from each other when viewed with a spectrometer, but instead appear as continuum-like emission. This is referred to as an unresolved transition array (UTA). Work carried out by O’Sullivan and Carroll in the 1980’s [53] found this strong band of emission for many materials and observed that it shifted to lower wavelengths for increasing atomic number. In another work [54], the Sn UTA was also observed with its peak emission around 13.5 nm.

Due to this very strong in-band emission, higher CEs up to 4–5% [25, 26] have been achieved by Sn plasmas as compared with Xe or Li, with a theoretical prediction of up to 7% for the optimum plasma parameters [55]. Therefore, due to its advantage as a stronger emitter of radiation at
1.3. Extreme ultraviolet lithography

13.5 nm, Sn has been nominated by many as the fuel of choice for EUVL [21, 33, 56]. However, other materials may still play an important role in EUV research e.g. as possible actinic metrology sources. Some issues persisting with Sn as a source are the debris and OOB radiation it generates. In order to suppress the debris emanating from Sn plasmas, mass limited or droplet targets systems have been utilised. This in effect removes excess Sn from the plasma-generating process that would not be sufficiently ionised and therefore helps limit excess particles that would otherwise become debris. As mentioned earlier in this chapter, problems with Sn as a fuel for an EUVL source still persist, specifically the shortfall in source power that is needed in order to make EUVL a reality for HVM in the semiconductor industry.

1.3.4 Beyond EUVL

The topic of beyond extreme ultraviolet (BEUV) light sources has recently become one of great interest in the field of EUV research. A new shorter wavelength EUVL scanner has been proposed to extend Moore’s law further, by developing future lithography scanners capable of printing features down to the 8 nm half pitch [57]. This is demonstrated in the EUV source roadmap of ASML [Fig. 1.12], a major supplier of photolithography systems to the semiconductor industry.

<table>
<thead>
<tr>
<th>Lens mirrors</th>
<th>0.25 NA</th>
<th>0.33 NA</th>
<th>Under study</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength</td>
<td>13.5 nm</td>
<td>13.5 nm</td>
<td>13.5 nm</td>
</tr>
<tr>
<td>Product</td>
<td>ADT</td>
<td>3100</td>
<td>3300B 3300C</td>
</tr>
<tr>
<td>Introduction year</td>
<td>2006</td>
<td>2010</td>
<td>2012 2013</td>
</tr>
<tr>
<td>Resolution (hp)</td>
<td>32 nm</td>
<td>27 nm</td>
<td>22 nm 18 nm</td>
</tr>
<tr>
<td>Sigma</td>
<td>0.5</td>
<td>0.8</td>
<td>0.2-0.9 OAI</td>
</tr>
<tr>
<td>Overlay (SMO)</td>
<td>7.0 nm</td>
<td>4.5 nm</td>
<td>3.5 nm 3.0 nm</td>
</tr>
<tr>
<td>Throughput (wph)</td>
<td>4 wph</td>
<td>60 wph</td>
<td>125 wph 150 wph</td>
</tr>
</tbody>
</table>

Figure 1.12: Roadmap of EUV and BEUV sources for future lithography nodes down to the 8 nm half pitch [57].
1.3. Extreme ultraviolet lithography

The wavelength choice for a BEUV source was, like that of 13.5 nm before it, driven by the availability of highly reflective MLMs. Lanthanum boron (La/B) based MLMs such as La/B₄C, LaN/B, LaN/B₄C and La₂O₃/B₃C have recently been shown to have measured reflectivity of between 42–54% [58, 59], with a theoretical limit of up to 80% [60]. The peak wavelength of reflectivity varies depending on the MLM, but the 0.6% in-band region of BEUV can be said to lie between 6.4–6.8 nm, hence a strong source of EUV radiation around these wavelengths is required.

![Figure 1.13: Reflectivity of LaN/B MLM of 53.6% at 6.65 nm [59].](image)

Two materials, terbium (Tb) and gadolinium (Gd), have been investigated as potential fuels for a BEUV source [61]. These materials, similar to Sn, have been studied due to their strong narrow-band resonance emission resulting from a UTA that forms around 6.5 nm for Tb and 6.7 nm for Gd, because of the $4p^64d^n - 4p^54d^{n+1} + 4d^{n-1}4f$ transitions in Tb$^{17+}$–Tb$^{28+}$ and Gd$^{16+}$–Gd$^{27+}$ ions respectively [53, 62, 63]. Due to the uncertainty in both the final mirror design and source wavelength, BEUV is often referred to as 6.X nm. LPP experiments of Gd targets to date have shown maximum CEs of 0.4-0.5% [64, 65], while much modelling work has gone into predicting the required laser intensity and plasma temperature for optimised in-band emission [62, 63].

17
1.4. Theory of laser-plasma interactions

1.4 Theory of laser-plasma interactions

This section is concerned with the processes involved during the interaction of focussed, high intensity laser pulses with solid targets under vacuum conditions, for the generation of plasmas. Throughout the experimental work laid out in this thesis, the effects and results of these interactions are studied, and therefore a background to the theory of plasmas and laser plasma interaction is given in this chapter. This will begin with the basics of what a plasma is and the properties by which they are defined. Next, the actual formation of a laser produced plasma (LPP) will be discussed, followed by the atomic processes that occur within a plasma, including the different processes of absorption and emission that can occur during the LPP lifetime. Following this is a brief discussion on some of the theoretical models used to describe a plasma. Finally, the topic of colliding plasma targets is introduced, with some detail on their creation and characteristics. The material for this section is heavily referenced from [5, 22, 37, 51, 66–71].

1.4.1 Plasma

![Figure 1.14: Everyday examples of plasmas on Earth [72–74].](image)
1.4. Theory of laser-plasma interactions

1.4.1.1 Definition of a plasma

Plasma is often referred to as the 4\textsuperscript{th} state of matter, as it is neither in a state of purely solid, liquid or gas. As a solid is heated it becomes liquid, as it is heated further it evaporates to become a gas. If it is heated still, then the atoms will begin to dissociate into positive ions and negative electrons, in a process called ionisation. As the plasma is heated further this process increases and so the degree of ionisation in the plasma increases too. The result is a partially or fully ionised gas containing a mixture of positive ions, negative electrons and neutrals. The overall charge however remains neutral, as the ions and electrons act to cancel each other out and the plasma is said to be in a state of quasineutrality \[5\]:

\[ n_e = n_i Z \] \hspace{1cm} (1.10)

where \( n_e \) is the electron density, \( n_i \) is the ion density and \( Z \) is the average charge state.

Plasma is believed to make up a large percentage of matter in the universe, with up to 99\% said to exist in a plasma state. However, it is not so common on Earth, with rare examples of plasmas including lightning and the conducting gas in neon signs. The reason for this can be seen in the Saha equation, which states

\[ \frac{n_i}{n_n} = 2.4 \times 10^{21} \frac{T^{\frac{3}{2}}}{n_i} e^{-U_i/kT} \] \hspace{1cm} (1.11)

where \( n_i \) is the density of ionised atoms, \( n_n \) is the density neutral atoms (both in m\(^{-3}\)), \( T \) is the temperature of the gas (K), \( k \) is Boltzmann’s constant (1.38×10\(^{-23}\) J K\(^{-1}\)) and \( U_i \) (eV) is the ionisation energy required to remove the most loosely bound electron from the outer shell of the atom \[69\]. Eqn. 1.11 can now be used to understand why plasmas are not so common on Earth. At ambient temperature \( n_n \) can be taken as approximately 3×10\(^{25}\) m\(^{-3}\), while \( T \approx 300 \) (°K) and \( U_i \approx 14.5 \) eV. Substituting these values into the Saha equation results in a very small fractional ionisation of only:

\[ \frac{n_i}{n_n} = 10^{-122} \] \hspace{1cm} (1.12)
1.4. Theory of laser-plasma interactions

1.4.1.2 Plasma condition

A main condition for the formation of a plasma is that the average kinetic energy of an electron must overcome the Coulomb force that keeps the electron bound to the ion. This Coulomb force in the atom is given by \( \frac{e^2}{4\pi\varepsilon_0 d^2} \), where \( d \) is the average distance between the ions and electrons, \( e \) is the charge of an electron and \( \varepsilon_0 \) is the permittivity of free space [68]. Three other conditions that a plasma must satisfy are:

\[
\lambda_D \ll L \quad (1.13)
\]

\[
N_D \gg 1 \quad (1.14)
\]

\[
\omega_p \tau > 1 \quad (1.15)
\]

\( L \) in Eqn. 1.13 refers to the dimensions of the plasma, while \( \lambda_D \) is a quantity called the Debye length [Eqn. 1.16]. At distances \( < \lambda_D \) the individual Coulomb forces will dominate, whereas \( > \lambda_D \) refers to distances at which ions and electrons can no longer be distinguished and therefore must be treated collectively [37]. This collective behaviour results in the charged particles arranging themselves so as to shield any electrostatic fields due to a charge within the plasma [70], in what is known as Debye shielding.

\[
\lambda_D = \sqrt{\frac{e_0 kT_e}{n_e e^2}} \quad (1.16)
\]

\[
\lambda_D = 7430 \left( \sqrt{\frac{kT_e}{n_e}} \right) \quad (1.17)
\]

In Eqn. 1.16 \( kT_e \) represents the electron temperature in eV. The equation for \( \lambda_D \) can also be written in the form of Eqn. 1.17. Substituting into this equation for the typical parameters of the laser produced plasmas studied in this thesis, an electron temperature of 30 eV and an electron density of \( 1 \times 10^{19} \) cm\(^{-3} \), results in a \( \lambda_D \) of 12.9 nm.

In order for the Debye shielding effect to take place, the condition in Eqn. 1.13 must have been met i.e. the physical dimensions of the system are very large compared to the \( \lambda_D \), but there must also be enough electrons within a distance \( \lambda_D \) from a disturbance, in order to produce the Debye shielding effect [69]. Therefore the average distance between electrons must be small when compared with the Debye length. This leads to the second plasma
1.4. Theory of laser-plasma interactions

condition, in Eqn. 1.14, which states that the number of electrons, $N_D$ [Eqn. 1.18], that are contained within a sphere of radius $\lambda_D$ must be much greater than 1.

$$N_D = \frac{4\pi}{3} \lambda_D^3 n_e$$  \hspace{1cm} (1.18)

The final criterion for a plasma [Eqn. 1.15], which is another example of its collective behaviour, states that in order for a gas to exhibit behaviour that is characteristic of a plasma as opposed to a neutral gas, $\omega_p \tau$ must be greater than 1 [69]. In this equation, the electron plasma frequency ($\omega_p$) refers to the typical frequency of oscillation of the plasma, which is the collective motion of the negative electrons with respect to the positive ions, while $\tau$ is the mean time between collisions with neutral atoms. The plasma frequency is given by:

$$\omega_p = \sqrt{\frac{e^2 n_e}{\epsilon_0 m_e}}$$  \hspace{1cm} (1.19)

where $m_e$ is the mass of the electron. From Eqn. 1.19 $\omega_p$ is seen to be dependent on $n_e$. The value of $n_e$ in some regions of the plasma will increase until it reaches what is called a critical electron density ($n_{ec}$). For electron densities below $n_{ec}$, the incident laser radiation will be able to penetrate the plasma. However, when the critical electron density of the plasma is reached, the plasma becomes opaque to the radiation, thereby reflecting it [5]. This is due to all of the electrons oscillating together in simple harmonic motion at $\omega_p$ when the density is equal to $n_{ec}$. An expression for $n_{ec}$ is given by Eqn. 1.20:

$$n_{ec} = \frac{4\pi^2 e^2 \epsilon_0}{c^2} \frac{1}{\lambda^2}$$  \hspace{1cm} (1.20)

where $c$ is the speed of light. From Eqn. 1.20 it is observed that $n_{ec}$ has a $1/\lambda^2$ dependence. Therefore increasing the wavelength will result in a lower critical electron density of the plasma. For the two laser systems predominantly used in this thesis, the value of $n_{ec}$ works out as $1 \times 10^{21}$ cm$^{-3}$ for the 1064 nm Nd:YAG laser and $9.9 \times 10^{18}$ cm$^{-3}$ for the 10,600 nm CO$_2$ laser. The other term in Eqn. 1.15 is $\tau$, which is the mean time between collisions with neutral atoms. The value of $\tau$ can be calculated from Eqn. 1.21 [70]:

$$\tau = \frac{4 \times 10^{15}}{n_n T_e^{1/2}}$$  \hspace{1cm} (1.21)
1.4. Theory of laser-plasma interactions

1.4.2 Laser produced plasmas

1.4.2.1 Laser plasma formation

A typical laser produced plasma, as shown in Fig. 1.15 above, is formed by focusing a laser beam of high intensity ($\phi \approx 1 \times 10^9$ W/cm$^2$ for a CO$_2$ laser) onto a target surface. The surface of the metal will begin to heat and this will form a melt layer on the surface. For most of the laser produced plasmas studied in this thesis, the laser pulse duration ($\tau_L$) is of the order of ns or longer, and is therefore much greater than the time scale of the electron-phonon coupling, which is determined by the electron cooling time [Eqn. 1.22] and the lattice (ion) heating time [Eqn. 1.23] [75]:

$$\tau_e = \frac{C_e}{\gamma} \quad (1.22)$$

$$\tau_i = \frac{C_i}{\gamma} \quad (1.23)$$

where $C_e$ and $C_i$ are the heat capacities of the electron and lattice per unit volume respectively and $\gamma$ is the electron-phonon coupling constant. $\tau_e$ and $\tau_i$ are typically 1 and 10 ps respectively for elemental metals [75, 76]. Due to $\tau_L$ being significantly greater than the electron-phonon relaxation time, the hot electrons that have absorbed the laser energy can cool by transferring energy to the lattice, during the lifetime of the pulse. Therefore the electron and
1.4. Theory of laser-plasma interactions

Lattice temperatures will be in equilibrium. By this process, the laser energy is continuously transferred deeper into the material, with a heat penetration depth:

\[ l \sim (Dt)^{1/2} \]  

where \( t \) is the time duration and \( D \) is the heat diffusion coefficient, which is approximately constant [77].

The melt layer that forms on the target surface will also enable further absorption of the laser radiation, as the refractive index of the liquid changes from that of the solid surface [22]. If the laser intensity is above the ablation threshold then the surface continues to heat, the melt layer will eventually reach its boiling point and then evaporation will occur. The threshold laser fluence \( (F_{th}) \) for evaporation to occur is thus given by:

\[ F_{th} = \rho \Omega D^{1/2} \tau_{L}^{1/2} \]  

where \( \rho \) is the density and \( \Omega \) is the specific heat capacity. From Eqn. 1.25 it can be seen that as the laser pulse duration increases, so also does the laser threshold fluence for evaporation [77]. After evaporation, a vapour will form in front of the target, which will continue to grow as it absorbs more of the laser radiation. As more material is removed from the surface to form the vapour, a recoil effect will cause a shockwave to move into the target, thereby compressing it and heating it further. The necessary temperatures will now have been reached to remove electrons from the outer shells of atoms, thus starting the process of ionising the gas cloud, which is assisted by multiphoton absorption. With the ionisation process beginning, the conditions for forming a plasma can then be met.

1.4.2.2 Laser plasma interaction

During the lifetime of the laser pulse the plasma will start to shield the target from further ablation of the laser by either reflection or absorption. The reflection process is from the slightly cooler and much more dense layer that has formed very close to the target surface, called the conduction region. This layer has not begun to expand out from the target yet and therefore is at a higher density. Most of this region is at or above the electron critical density, \( n_{ec} \) [Eqn. 1.20], hence the laser light will not be able to penetrate and will be reflected. Absorption of the laser pulse is also taking place by free electrons in the less dense regions of the plasma further from the surface, in a region called the corona. The region between the conduction layer and the
1.4. Theory of laser-plasma interactions

corona of the plasma is often referred to as the critical density surface [75]. The main process by which the laser photons are absorbed into the plasma is inverse Bremsstrahlung absorption [Eqn. 1.28]. The processes involved in the formation of a laser produced plasma are depicted in Fig. 1.16 below.

Figure 1.16: Formation of a laser produced plasma [78].

As the laser is shielded from the target by the plasma, this will allow for plume expansion, and thus a reduction in density close to the surface below \( n_{ec} \). Thus the laser will again be able to penetrate the plasma to further ablate the target surface. This causes further energy exchange with the surface, which leads to \( n_{ec} \) being reached, and thus plasma shielding occurs again. The distance which the plasma plume will freely expand in one direction, after a time, \( t \), is given by [79]:

\[
l = c_s t = 9.79 \times 10^5 \left( \frac{\gamma Z T_e}{m_i} \right)^{1/2} t \tag{1.26}
\]

\[
c_s = \sqrt{\frac{Z T_e}{m_i}} \tag{1.27}
\]

where \( c_s \) is the ion sound speed [Eqn. 1.27], \( \gamma \) is the ratio of specific heat, \( Z \) is the mean ion charge, \( T_e \) is the temperature, \( m_i \) is the mass of the ion and
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$Z$ is the average charge state.

The absorption and reflection of the laser pulse by the plasma is therefore dictated by the density profile of the plasma. A plot of such a density and temperature profile of a laser produced plasma is shown in Fig. 1.17. The profile shows the outer hotter Corona region with an electron density that is less than the critical density of the plasma ($n_e < n_{ec}$). The profile also shows the region closer to the target, which is cooler and has a density greater than the critical density ($n_e > n_{ec}$).

![Density and temperature profile of a laser produced plasma](image)

Figure 1.17: The typical profile of a laser produced plasma [75].

The main process of absorption of the laser in the plasma, as shown in Fig. 1.16, is by electron-ion collisions via inverse Bremsstrahlung (IB) absorption. This process is heavily dependent on the electron density, ion density and the incident laser wavelength. The co-efficient of IB absorption is given as [5]:

$$\alpha_{IB} = 5.64 \times 10^{-11} \frac{A n_e^2 \ln \Lambda}{\omega^2 T_e^{3/2}} \frac{1}{\left(1 - \frac{\omega^2}{\omega_p^2}\right)^{1/2}}$$  \hspace{1cm} (1.28)

where $n_e$ and $T_e$ is again the electron density and temperature respectively, $\omega$ is the frequency of the incident laser radiation, $\omega_p$ is the plasma frequency (see Eqn. 1.19), $A$ is the atomic number and $\ln \Lambda$ is the Coulomb logarithm. $\ln \Lambda$ is a logarithmic factor that represents the importance of small-angle...
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scattering effects (typically on the order of 10–20) [68], where \( \Lambda \) is given by [5]:

\[
\Lambda = \frac{3}{2A^2} \left( \frac{T_r^3}{\pi e^6 n_e} \right)^2
\]  \hspace{1cm} (1.29)

1.4.3 Atomic processes in plasmas

During the lifetime of the plasma many different atomic processes will occur and in this section an overview of those processes is given. To begin with, when the laser is incident on the target surface and interacts with the plasma, the photons can be absorbed by the following processes:

- **Photoabsorption**: Laser photons are absorbed by atoms and ions, thereby giving their electrons excess energy and allowing the electrons to transition to an excited energy level within the atom or ion.

- **Photoionisation**: In this process when the laser photons are absorbed by the atoms and ions, the electrons acquire energy exceeding that of the binding energy of the atom, molecule or ion. As a result, it escapes and becomes a free electron, with any excess photon energy above the binding energy being transferred to the free electron in the form of kinetic energy.

- **Inverse Bremsstrahlung**: This final process involves the absorption of photons by free electrons in the vicinity of ions, which serves to increase the kinetic energy of electrons.

These processes occurring during the interaction of the laser photons with the atoms, ions and electrons of the plasma are referred to as radiative processes. Once the plasma has been formed, other interactions and transfer of energy then occur due to collisions within the plasma. These collisional and radiative type processes can be grouped together as being either Bound-Bound, Bound-Free or Free-Free. An overview of the different types of transitions and interactions possible are shown in Fig. 1.18. Following that, the full list of processes are summarised in Table 1.1.
1.4. Theory of laser-plasma interactions

Figure 1.18: Example of an energy level diagram. The types of transitions possible are shown with arrows: I–Bound-Bound, II–electron capture, III–photoionisation, IV–Free-Free.

<table>
<thead>
<tr>
<th>Process</th>
<th>Excitation</th>
<th>De-excitation</th>
<th>Type</th>
</tr>
</thead>
<tbody>
<tr>
<td>B-B</td>
<td>Collisional excitation</td>
<td>Collisional de-excitation</td>
<td>Collisional</td>
</tr>
<tr>
<td></td>
<td>Photoabsorption</td>
<td>Spontaneous decay</td>
<td>Radiative</td>
</tr>
<tr>
<td>B-F</td>
<td>Collisional ionisation</td>
<td>3-Body recombination</td>
<td>Collisional</td>
</tr>
<tr>
<td></td>
<td>Photoionisation</td>
<td>Radiative recombination</td>
<td>Radiative</td>
</tr>
<tr>
<td>F-F</td>
<td>Bremsstrahlung</td>
<td>Inverse Bremsstrahlung</td>
<td>Collisional</td>
</tr>
<tr>
<td>F-F</td>
<td></td>
<td></td>
<td>Radiative</td>
</tr>
</tbody>
</table>

Table 1.2: Summary of the main atomic processes occurring in a laser produced plasma. Table key: B-B = Bound-Bound, B-F = Bound-Free, F-F = Free-Free
1.4. Theory of laser-plasma interactions

1.4.3.1 Bound-Bound processes

Radiative

Bound-Bound radiative processes in a laser produced plasma occur in the form of *photoabsorption* (excitation) and *spontaneous decay* (de-excitation). In the case of photoabsorption, an incoming photon from the laser is absorbed by a bound electron in a low-lying energy state of the atom or ion. The energies are quantized, meaning only energies equal to those required for the electron to transition from its initial to final state are accepted [Eqn. 1.30].

The de-excitation or relaxation of an electron from an excited state to a lower state is the inverse process of this, and is called spontaneous decay. As the electron drops from its upper to lower state it will emit a photon of energy equal to the difference between the two states, given by:

\[ \Delta E = E_f - E_i = \frac{hc}{\lambda} \]  \hspace{1cm} (1.30)

where \( h \) is Planck’s constant, \( c \) is the speed of light and \( \lambda \) is the wavelength of the photon of light emitted or absorbed. Photons of only certain discrete energies will be emitted or absorbed during Bound-Bound processes, which means, from Eqn. 1.30, that the emitted or absorbed photons will also only have certain wavelengths. This gives rise to the emission of characteristic emission or absorption line spectra.

![Photoabsorption and spontaneous decay](image)

Figure 1.19: Photoabsorption and spontaneous decay, the radiative Bound-Bound transitions of a laser produced plasma.
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Collisional
Bound-Bound excitation and de-excitation also happen in a plasma due to collisions with free electrons. When a free electron collides with a bound electron, it can transfer some or all of its kinetic energy to it. The energy gained by the bound electron allows it to transition to a higher excited state within the atom or ion, in a process called collisional excitation. The reverse process of this is called collisional de-excitation. In this case a free electron collides with a bound electron that is already in an excited state. The bound electron is demoted to a lower state, while the difference in the energy of its transition is transferred to the free electron in the form of kinetic energy. A simplified schematic of the collisional processes are shown in Fig. 1.20 below.

![Diagram of collisional excitation and de-excitation](image)

Figure 1.20: Collisional excitation and collisional de-excitation, the collisional Bound-Bound transitions of a laser produced plasma.

1.4.3.2 Bound-Free processes

Radiative
A Bound-Free process is so called because it involves an electron transitioning between a bound state of an atom or ion to the continuum. In the radiative process, a photon is absorbed and transfers its energy to a bound electron. However, unlike photoabsorption, the energy acquired by the electron is sufficient to allow the electron to overcome the Coulombic force of the atom and become free. This is called photoionisation and is also known as the photoelectric effect. The inverse effect of this is called radiative recombination. This occurs when a free electron in the continuum recombines with an atom or ion. The electron returns to a bound state and releases a photon in the process. Both types of radiative Bound-Free processes are shown in
1.4. Theory of laser-plasma interactions

Fig. 1.21. These processes are more dominant in less dense plasmas, where collisional effects will be less prevalent. Unlike Bound-Bound processes, the transitions are not between discrete energy levels, therefore the result is continuous absorption and emission spectra.

Collisional
Free electron collisions with bound electrons can result in the transfer of energy, which can be enough to allow the bound electron overcome the binding energy of the atom or ion. The bound electron thus becomes free, while the atom becomes ionised, hence the name collisional ionisation. The inverse process, 3-body recombination, occurs in dense plasma when two electrons can come close enough to an ion (within the Debye sphere). One of the electrons is captured to an outer shell of the ion, thereby reducing the ionisation stage, while the excess energy is transferred to the other free electron in the form of kinetic energy. Both processes are shown in Fig. 1.22 on the next page.
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Figure 1.22: Collisional ionisation and 3-body recombination, the collisional Bound-Free processes of a laser produced plasma.

1.4.3.3 Free-Free processes

Radiative
A free electron, which is close to an atom or ion, increases its kinetic energy when it absorbs a laser photon. In order to obey the law of conservation of momentum, the electron must be in the presence of the ion. This process is called inverse Bremsstrahlung.

Collisional
When a free electron travels close to the electric field of an ion or very close to an atom, it may be deflected by the charge of the nucleus causing it to decelerate and lose some of its kinetic energy, which is then converted into the emission of a photon. The name for this process, Bremsstrahlung, comes from the German “Bremse” meaning brake and “Strahlung” meaning radiation. This photon emission does not involve quantized bound energy levels of the atom or ion, and therefore produces a continuous emission spectrum. Both Free-Free processes are depicted in Fig. 1.23.
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![Image of Bremsstrahlung and inverse Bremsstrahlung processes]

Figure 1.23: The Free-Free processes of a laser produced plasma, Bremsstrahlung and inverse Bremsstrahlung.

1.4.4 Plasma equilibrium models

Three plasma models are typically used to describe a plasma and its behaviour [5]. The use of one of the models to characterise a plasma is dependent on the electron temperature and density. The three models are: local thermodynamic equilibrium (LTE), coronal equilibrium (CE) and the collisional radiative (CR) model. The models can be more or less ascribed to the core (LTE), middle (CR) and outer (CE) regions of a plasma, as depicted by Fig. 1.24.

The LTE model is used to describe a denser plasma, which will therefore have more collisions due to a reduced mean free path. Therefore collisional effects are said to dominate over radiative effects, with collisional de-excitation being the dominant decay process [51]. Due to the high densities, it can be an optically thick plasma, with re-absorption of emitted photons highly probable. The outer region, or corona, of a plasma can be best described by the CE model, as it is much more sparse due to its larger mean free path caused by expansion. As a result, collisions are much less frequent and radiative effects begin to dominate. Unlike the inner region of the plasma, self-absorption of photons is rare due to the optically thin nature of the coronal plasma.

The CR model, also referred to as the non-LTE model, is an intermediate model used to describe a region of the plasma between the high and low electron densities of the core and corona respectively. In this model, a balance is struck between both the collisional and radiative processes as both processes are responsible for excitation and ionisation in the plasma. The CR model was demonstrated, by Colombant and Tonon [80], to be successful
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at describing a laser produced plasma with a temperature above tens of eV. Using the model, a fractional ion population as a function of the electron temperature can be determined. Approximations made by Colombant and Tonon yielded a simplified relation for the average state, $Z$, as:

$$Z = \frac{2}{3} [AT_e^e (eV)]^{1/3}$$

Figure 1.24: Regions where the different plasma models are applied, based upon the densities.
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1.4.5 Colliding laser produced plasmas

Colliding plasmas targets have been predominantly studied in astrophysics since their earliest use in experiments in the 1960s. This was based upon collisions between plasmas found in space such as nebulae and galaxies [67]. More recently, they have become a topic of interest for smaller scale, table top laboratory experiments, with possible applications for pulsed laser deposition (PLD), ion accelerators and EUV sources [81], which is of particular interest in relation to this work.

1.4.5.1 Colliding laser plasma basics

A colliding laser produced plasma is formed by the interaction of two counterpropagating plasmas, called the “seeds” or “seed plasmas”. The seeds are formed by focussing two laser pulses of high intensity ($\phi \approx 10^{10} - 10^{11}$ W/cm$^2$), close to each other on a target surface (typically 0.5–2 mm separation). The initial phase of the plasmas lifetime will be the same as that of a typical laser produced plasma, as discussed earlier. However, if the seed plasmas are sufficiently close to each other, they will eventually begin to interact at an interface as they expand. This interaction between the two seeds leads to two possible outcomes. The first type of interaction, called interpenetration, is when the two fronts of the seed plasmas meet and penetrate through each other. The plasmas behave in effect like dilute gas jets, which will stream past each other, dominated only by binary collisions [66]. In this case the ion–ion mean free path ($\lambda_{ii}$) can be said to be greater than the scale length of the collision front $L$, i.e. $\lambda_{ii} \gg L$ [82]. In the second possible outcome the plasmas collide and interact at their interface, when $\lambda_{ii} \ll L$ [82]. Instead of passing through each other, their velocities drop and a thin layer of quasi-static plasma material is formed at the interface. This process is called stagnation and the layer of plasma that forms, called a stagnation layer, will be a relatively uniform plasma compared to the seed plasmas. More precisely, when the seed plasmas do interact, an outcome somewhere between these two extreme cases is most likely.

The factor that is used to determine what kind of interaction will take place, i.e. soft or hard stagnation, is called the collisionality parameter and is given by:

$$\zeta = \frac{D}{\lambda_{ii}} \quad (1.32)$$

where $D$ is the separation distance between the two seed plasmas and $\lambda_{ii}$ is the ion–ion mean free path, where $\lambda_{ii}$ is:
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\[ \lambda_{ii} = \frac{m_i^2 v_{12}^2}{4 \pi e^2 Z_i n_i \ln \Lambda_{12}} \]  

(1.33)

where \( v_{12} \) is the relative collision velocity of the seeds, and as before \( m_i \) is the mass of the ion, \( e \) is the electron charge, \( Z \) is the average charge state, \( n_i \) is the ion density and \( \Lambda_{12} \) is the Coulomb logarithm for the collision between seed plasma 1 and 2. When \( \zeta \) is low, more interpenetration occurs which leads to a softer stagnation, while a harder stagnation occurs when \( \zeta \) is large. From Eqn. 1.33, it can be seen that \( \lambda_{ii} \) scales as \( v_{12}^2 \), thus slower moving seed plasmas meeting at an interface will have a smaller \( \lambda_{ii} \) and therefore have a greater value of \( \zeta \). Conversely, faster expanding plasmas have a greater \( \lambda_{ii} \) and tend to interpenetrate more before stagnation. This effect is seen in colliding plasma experiments from the PhD thesis of J. Dardis [67], where the stagnation layers produced by wedge targets were observed to be wider than those created by flat targets. The flat target seeds formed a very narrow stagnation layer along the collision front due to the high value of \( \zeta \). In the wedge target set-up the seed plasmas were directed more toward each other and therefore had greater velocities at the point of collision. This produced a smaller value of \( \zeta \) which causes more penetration before the stagnation occurs, thus leading to a wider stagnation layer.

1.4.5.2 Colliding laser plasma formation

Colliding plasma formation can be achieved by splitting a single laser beam using a small angle wedge prism. The prism is placed in the beam path. It is orientated such that approximately half of the beam is transmitted over the prism, while the other half is transmitted through it. The portion of the laser pulse going through the prism is refracted while passing through the medium, due to the acute angle of the wedge. After both portions of the beam have transversed the wedge, they are then both focussed by the same plano-convex lens. The lens focuses the spots onto a surface, with a separation between the lenses that is determined by the acute wedge angle and the focal length of the lens:

\[ D = f \gamma (n - 1) \]  

(1.34)

where \( D \) is the separation between the centers of the seed plasmas, \( f \) is the focal length of the lens, \( \gamma \) is the acute angle of the wedge and \( n \) is the refractive index of the wedge.

From time gated imaging of colliding plasma formation in the PhD thesis of P. Hough [66], it was shown, for a separation of 1.3 mm and on-target
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laser intensity of approximately $1 \times 10^{11}$ W/cm$^2$ from a 6 ns pulse duration, that stagnation between the seeds will begin to form after around 30 ns. The imaging also shows the seeds dissipating to leave only the stagnation layer after 55 ns. These images are shown in Fig. 1.25, and can be used as a basis for the timing of the colliding plasmas that will be formed later in Chapter 5, as almost identical experimental parameters of seed separation, pulse duration and laser intensity are used. The target is different however as Al is used in Fig. 1.25, where as Sn colliding plasmas are the focus of Chapter 5 in this work. The increased mass of the Sn ions (almost 4.5 times that of Al) means that the expansion velocity will be slower and that stagnation layer formation as a result takes longer.

During the experiments reported here the colliding plasmas are formed first on a flat target and then later on wedge targets. The flat target is initially used for ease of alignment and for maximising the number of data acquisitions that can be made on the target. Later experiments are carried out using wedge shaped targets. These type of wedge target set-ups have previously been studied for colliding plasma experiments [83–86]. The plasma expansion velocities normal to the target surface are as much as an order of magnitude greater than lateral expansion velocities [86]. That means a greater amount of plasma will expand in that direction, making for a larger region of interaction for producing the stagnation layer. As discussed, the

![Figure 1.25: Time sequenced optical images of Al$^+$ emission at 460 nm from colliding laser plasmas [66].](image)
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increased ion velocity component in Eqn. 1.33 also means that the collisionality parameter [Eqn. 1.32] is smaller for wedge targets. This means a softer stagnation layer is created, with more interpenetration taking place before stagnation occurs, which will lead to a larger interaction region. It must be noted that for very small laser spot sizes, the lateral expansion is shown to be of greater significance [87].
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Chapter 2

Apparatus

2.1 Introduction

This chapter describes the experimental equipment that was used during the majority of experiments reported in this thesis. It contains four sections, which include descriptions of the three main components of the laboratory equipment and a final section on additional equipment used. The first section gives details of the carbon dioxide (CO$_2$) laser system, which was the main laser used for creating tin (Sn) plasmas for extreme ultraviolet (EUV) studies. It was also used as the second laser or “pump” laser in dual laser pulse experiments for re-heating Sn plasma targets. Following this is a section reporting on the details of the neodymium-doped (Nd$^{3+}$) yttrium aluminum garnet (Y$_3$Al$_5$O$_{12}$) or Nd:YAG laser. This laser was primarily used as the pre-pulse for creating a plasma target, which was then re-heated by the pump laser in the dual pulse experiments. The third section gives details pertaining to the Jenoptik EUV spectrograph, which was the main diagnostic tool used throughout the 13.5 nm experiments. Finally, the last section in this chapter gives a brief description of all other apparatus employed in experiments.

2.2 TEA CO$_2$ laser

This is the primary laser used in the majority of experiments in this thesis. A basic description of the operation of the transversely excited atmospheric (TEA) CO$_2$ laser is detailed next, with the excellent “The CO$_2$ laser” book by Walt Witteman [1] used as a considerable source of information in this chapter. The first CO$_2$ laser was demonstrated in 1963 by Kumar Patel in Bell Laboratories [2]. The CO$_2$ laser has since become very prominent and widely used in various applications especially materials processing [1]. One
2.2. TEA CO₂ laser

of the main advantages for this is its efficient conversion of electrical energy into laser radiation, the theoretical limit of which is 38% but typical systems achieve 20% [1]. The lasing action of the CO₂ laser is achieved within a mixture of gases, which represent the amplifying medium of the process. The majority of systems will contain CO₂, helium (He) and nitrogen (N₂). Other systems may contain some hydrogen (H₂), water vapor (H₂O) or possibly xenon (Xe). Two mirrors at either end of the medium form the optical resonator, along which the emission from the gas medium will oscillate. The pumping action of the medium is formed by an electrical discharge across the transverse length of the cavity, hence the name TEA. This can be achieved by direct current (DC) or alternating current (AC) or even using radio frequency (RF). In order to pump the gas medium longitudinally a very high voltage would be required as the voltage needed for gas breakdown increases linearly with density. Therefore the voltage is applied transversely across the length of the optical axis.

2.2.1 Rotational-vibrational structure of CO₂

![Figure 2.1: Normal modes of vibrational motion of CO₂ [3].](image)


2.2. TEA CO\textsubscript{2} laser

The main emission from the CO\textsubscript{2} laser is at 10.6 µm and 9.6 µm in the infra-red region of the electromagnetic spectrum. This is due to the lasing action in all CO\textsubscript{2} lasers occurring between low-lying vibrational-rotational levels of the ground electronic state, and because the dipole moment created by these vibrational motions are said to be “infra-red active” [3]. The normal modes of vibrational motion of the CO\textsubscript{2} molecule are shown in Fig. 2.1. The normal mode of motion means that as the molecule vibrates all of the atoms will pass through their equilibrium position simultaneously and oscillate at the same frequency. The unexcited CO\textsubscript{2} molecule is shown in (A), while the three normal modes of vibrational motion are shown in (B–D). However, the symmetric stretching mode (\(\nu_1\)) of the CO\textsubscript{2} molecule will produce no change in the dipole moment, therefore its vibration is not infra-red active. An example of the dipole moment created by the asymmetric stretching mode (\(\nu_3\)) is shown in Fig. 2.2 taken from Ref [3].

![Diagram of CO\textsubscript{2} molecule showing normal modes of vibration and dipole moment](image)

Figure 2.2: Asymmetric stretching vibrations of CO\textsubscript{2} [3].
2.2. TEA CO₂ laser

2.2.2 Lasing in CO₂

The process of lasing in a CO₂ laser is discussed next. The N₂ molecule is used in most CO₂ laser systems as a means of transferring energy by vibration to the CO₂ molecule. The diatomic molecule N₂ is said to be homonuclear as both of its nuclei are identical. Therefore dipole radiation is forbidden and it can only decay by colliding with the laser chamber wall or with the other gases contained in the medium. In the lasing process, electrons are produced by the ionisation of the gases, which in turn excite the bond-stretching mode of the N₂ molecules. This promotes them from the ground state \( (v = 0) \) to its very long lived first excited vibrational level \( (v = 1) \). This lasing process is shown in Fig. 2.3 taken from Ref. [1] and is in essence a four level laser system.

![Diagram showing the vibrational levels of the CO₂ and N₂ molecules involved in the lasing process.](image)

Figure 2.3: Diagram showing the vibrational levels of the CO₂ and N₂ molecules involved in the lasing process. The ground state \( (v = 0) \) and the first excited state \( (v = 1) \) of N₂ are shown, which play an important role in the selective excitation of the \( (00^01) \) level of CO₂ [1].

The figure shows that the first excited N₂ level lies very close to the asymmetric stretching mode, \( \nu_3 \), vibrational level of CO₂. This \( (00^01) \) level is only \( \Delta E = 18 \, \text{cm}^{-1} \) higher than the \( v = 1 \) vibrational level of N₂. Thus collisions allow the transfer of energy from the N₂ first excited level to the excited vibrational state \( (00^01) \) of the CO₂ molecule [4]. The N₂ molecules then return to their
2.2. TEA CO$_2$ laser

The CO$_2$ laser emission at 10.6 $\mu$m is produced by the transition from the excited asymmetric stretching mode $\nu_3$ (00$^01$) to the symmetric stretching mode $\nu_1$ (10$^00$). However, strong emission also occurs due to the (00$^01$) excited level also relaxing to the bending mode $\nu_2$ (02$^00$), which produces emission at 9.6 $\mu$m. The lifetime of the CO$_2$ upper level is approximately 0.4 ms and the lower levels are 20 $\mu$s [5]. The transfer of energy occurs not just at the first excited states of N$_2$ but also exists up to the values of $\nu = 4$ in N$_2$ as the quantum values still do not differ from the (00$^01$) level more than the average kinetic energy [1]. The symbols representing the laser transitions such as (00$^01$) and (10$^00$) denote the set of the vibrational quantum number, $\nu$, i.e. (100) is the $\nu_1$ mode, (010) is the $\nu_2$ mode and (001) is the $\nu_3$ mode, while the upper indices refer to the angular momentum excited in the $\nu_2$ vibrational level.

During the process of the discharge, dissociation of the CO$_2$ molecule can occur if the bond between the atoms is stretched to the point of breaking, resulting in the production of carbon monoxide (CO) molecules. Other constituents used in some laser systems such as H$_2$ or H$_2$O can help (particularly in sealed-tube lasers) to reoxidize CO back into CO$_2$. In sealed systems without their presence, the amount of CO being produced during the discharge can be up to 50% [1]. However, CO can also transfer energy to CO$_2$ by means of vibrational energy exchange much the same as N$_2$ does. This process is not as efficient as the energy exchange by N$_2$ though and excited CO molecules also experience spontaneous decay as it is a heteronuclear diatomic molecule with a dipole moment. He is also used in many TEA CO$_2$ laser systems as a buffer gas. It serves to depopulate the lower laser level and to remove heat from the process by acquiring low level energies, which it then loses through collisions with the laser chamber wall. He has a relatively high thermal conductivity (k) of 0.1513 Wm$^{-1}$ K$^{-1}$ at room temperature compared with 0.0146 Wm$^{-1}$ K$^{-1}$ for CO$_2$ and N$_2$ with k = 0.0258 Wm$^{-1}$ K$^{-1}$. This helps to cool the system as the temperature difference between gas and the laser chamber wall is inversely proportional to the conductivity of the gas.

2.2.3 InfraLight SP10 gas flow TEA CO$_2$ laser

The CO$_2$ laser used in the experimental sections of this thesis is the InfraLight SP10 gas flow TEA CO$_2$ laser from Optosystems. The operating wavelength of this CO$_2$ laser is 10.6 $\mu$m and the maximum output energy is close to 1.4 J. The output energy can vary depending on the ratio of the laser gas mix (He:N$_2$:CO$_2$) used and the discharge voltage across the gases contained in the laser chamber, which ranges from 18 to 27.5kV. As a result, energies from
2.2. TEA CO\textsubscript{2} laser

around 100 mJ up to 1.4 J can be achieved, with varying pulse temporal profiles. Typically if the laser is operated with a standard gas mix of 6:1:3 (He:N\textsubscript{2}:CO\textsubscript{2}) and at a discharge voltage 27.5 kV, it will produce an output energy of around 1–1.2 J. Fig. 2.4 shows the effect of increasing the discharge voltage on the output energy. Note these energies were recorded after several reflections, which explains the lower reading for the maximum voltage. The CO\textsubscript{2} laser temporal profile is characterized by an initial large sharp peak of 50 - 100 ns pulse duration, which is followed by a low energy, long duration N\textsubscript{2} tail. The overall pulse duration is of the order of 1-3 \(\mu\)s. The pulse temporal profile is discussed in more detail in Chapter 4.

![Figure 2.4: Plot of laser output energy as the discharge voltage is increased.](image)

Inside the laser [Fig. 2.5], a series of specially shaped electrodes are placed parallel to each other, at a distance of 15 mm apart, transversely along the length of the gas discharge chamber. Supplementary electrodes are placed periodically either side of the main electrodes. These auxiliary electrodes are used to preionise the gas mixture by means of a spark discharge. A regenerator unit is connected to the laser, so that the gas in the laser chamber flows through it and back into the chamber. The reason for this, is to allow recombination of the CO\textsubscript{2} molecule following its dissociation into CO, by
2.2. TEA CO₂ laser

Figure 2.5: The Infralight SP10 gas flow TEA CO₂ laser head.

providing water vapour for it to bond with. When the regenerator is switched on, so is its water supply. It is connected to a flow switch and so will only activate if a sufficient flow of water is supplied to it. The gas lines to the He, N₂ and CO₂ all connect with the rear of the laser head from canisters. A vacuum pump is also connected to the laser chamber, which is used to evacuate the chamber before the changing or refreshing the gas mixture.

2.2.4 Laser pulse energy measurements

Accurate on-target laser energy measurements and estimates are very important. From a knowledge of the laser energy, the laser intensity of the beam can be determined (W/cm²). This is extremely useful when it comes to developing a model to explain the behaviour of a laser produced plasma (LPP). In this work, the energy is also used to help define the conversion efficiency (CE) of an EUV source system, by finding what percentage of the laser energy going into a system is then measured as re-emitted in-band EUV radiation. To achieve this, regular energy measurements of the lasers are made with energy heads and power heads from Gentec.

The energy head, QE25SP-H-MB, is constructed from a pyroelectric crystal that works by generating a small temporary voltage when heat is applied i.e. by irradiating them with the laser pulse. As the pyroelectric crystal is heated, modification of the crystal structure occurs, this results in a change in the polarisation of the material. The process is based on this linear vari-
2.2. TEA CO$_2$ laser

The spontaneous polarisation $P_s$ with the applied temperature $T$, given by [6]

$$\Delta P_s = \gamma \Delta T$$ (2.1)

where $\gamma$ is the pyroelectric coefficient of the material (C m$^{-2}$ K$^{-1}$). The surface charge density ($I_P$) of the sample changes due to the variations in $P_s$. This produces a pyroelectric current in the crystal, which is proportional to both the time derivative power of the laser pulse and of the temperature [6]

$$I_P \sim \frac{dP_s}{dt} \sim \frac{dT}{dt}$$ (2.2)

The power head detector, UP50N-40S-IR, used in experiments is basically a thermopile, where a temperature difference is used to create a voltage. The laser energy that is incident on the detector surface is converted into heat, while the other side of the material acts as a heat sink. A temperature difference is set-up across the thermoelectric device due to the absorber on one surface and the heat sink on the other. This results in a voltage that is proportional to the temperature difference, which is also proportional to the power of the laser.

![Figure 2.6: Schematic showing energy head location during experiments.](image)

Due to the variation in the shot to shot stability of the energy of the CO$_2$ laser, the Gentec energy head is placed permanently in the set-up so as to record in-situ energy measurements for every laser pulse, as shown in Fig.
2.2. TEA CO$_2$ laser

2.6. A ZnSe window is placed in the beam path at 45° with respect to the direction of the laser pulse which acts as a ∼97:3 beam splitter. The ∼3% reflection is then directed to the energy head where it is recorded. The power head is temporarily placed in the path of the full laser energy and the system is run at 1 Hz. This way the full laser energy can be determined and the energy head can be calibrated, so that for each laser pulse the full energy can be determined. A typical measurement of the CO$_2$ laser energy and power recorded using the two detectors found the percentage of energy detected on the energy head to be 2.97% of the full beam.

2.2.5 Temporal and spatial pulse profile

Temporal profile measurements of the CO$_2$ laser beam were also made during experiments. An alteration was made to the set-up shown above [Fig. 2.6] to include a second ZnSe window and a photon drag detector that can be used to record the temporal profile by connecting it to an oscilloscope [Fig. 2.7]. The drag detector operates by means of the photon drag effect acting on the germanium (Ge) material inside the detector. When the 10.6 μm photons of the CO$_2$ laser are incident on the Ge surface they interact with free electrons and holes. The electrons and holes acquire the momentum of the photons, ħk [7]. This transfer of momentum causes the carriers created by the photons to be dragged in the same direction as the photons. This transfer of momentum in the material produces an electromotive force (emf) [8]. A simple classical approximation of the photon-drag voltage under open-circuit conditions between the ends of a sample length $L$ is given as [9]

$$V = \frac{\mu \rho K_h}{c} \int_0^L I(x) dx$$

(2.3)

where $\mu$ is the hole mobility, $\rho$ is the sample resistivity, $c$ is the velocity of light, $K_h$ is the absorption coefficient due to holes and $I(x)$ is the intensity distribution in the propagation direction $x$. A more detailed discussion of the CO$_2$ temporal profile can be found in Chapter 4.

An image of the spatial profile of the CO$_2$ beam was recorded using an 140×140 (25 mm × 25 mm) infra-red thermal imaging vanadium detector (8–14 μm) and is shown in Fig. 2.8. This shows the asymmetry in the beam profile and the region of peak beam intensity. The spatial profile can clearly be seen to be neither Gaussian nor flat-top, as is the case with many other commercial lasers, such as Nd:glass lasers.
2.2. TEA CO\textsubscript{2} laser

Figure 2.7: Schematic showing location of the temporal profiler (photon drag detector) during experiments.

Figure 2.8: CO\textsubscript{2} laser spatial profile recorded with infra-red detector.
2.2. TEA CO\textsubscript{2} laser

![CO\textsubscript{2} laser spatial profile recorded with infra-red detector.](image)

2.2.6 Knife edge technique for spot size measurements

In order to precisely quote the laser intensity of the CO\textsubscript{2} incident on a target surface, the beam spot size at various focal positions must be known. The energy and pulse duration can both be measured as previously discussed in this section. Knife edge experiments [10, 11] have been demonstrated as a method of determining beam diameters. An experiment similar to that described in the PhD thesis work of A. O’Connor [12] was set-up [Fig. 2.9].

A razor blade was mounted on top of \( x \) and \( y \) linear motion stages controlled by actuators. The stages allowed movement of the blade along the axis of beam and also across its diameter so as to completely occlude it. The beam passes first through a partial reflector (ZnSe window). When it is placed at 45° it acts as a \( \sim 97:3 \) beamsplitter. The transmitted portion of the beam is sent to energy detector 2 and is used as a reference energy for each shot. The \( \sim 3\% \) reflected portion of the beam is focussed by a ZnSe lens and measured by energy detector 1. Energy measurements are made by detector 1 for radial scans (\( r \)) of the knife edge along the optical axis (\( z \)) of the beam. Once a full radial scan is complete, the blade was moved along the optical axis, and then scanned again across the beam. This was carried out for the unfocussed and focussed beam using a 20 cm lens. The energy on detector 1 could be
2.2. TEA CO$_2$ laser

corrected for variations in pulse stability by dividing by the corresponding reference energy taken by detector 2.

Figure 2.9: Schematic of Knife Edge experiment set-up.

The energy values recorded for a radial scan of the unfocussed beam were carried out first and are plotted in Fig. 2.10 as a function of the actuator position. A theoretical fit is applied to the data as described by Khosrofian et al. [13–15]. The fitting function is given by

\[ f(s) = \frac{1}{1 + \exp[p(s)]} \]  

(2.4)

where

\[ p(s) = \sum_{i=0}^{m} a_i s^i \]  

(2.5)

\[ s = \frac{\sqrt{2}(x - x_0)}{w} \]  

(2.6)

Khosrofian et al. limited the polynomial to the third order so that

\[ f(s) = \frac{1}{1 + \exp(a_0 + a_1 s + a_2 s^2 + a_3 s^3)} \]  

(2.7)
2.2. TEA CO₂ laser

In Eqn. 2.6, \( x-x_0 \) refers to the radial distance from the optical axis and \( w \) is the beam radius at focus. The coefficients \( a_i \) of the polynomial in Eqn. 2.7 are constants given as \( a_0 = -6.71387 \times 10^{-3} \), \( a_1 = -1.55115 \), \( a_2 = -5.13306 \times 10^{-2} \) and \( a_3 = -5.49164 \times 10^{-2} \).

![Figure 2.10: Normalised energy profile of the unfocussed beam using a razor blade. The Khrosrofian fit is applied to the data.](image)

Two methods exist for taking a direct measurement of the beam radius. The first method (Eqn. 2.8) equates the beam radius \( r \) to the translation distance (radially) for which the beam intensity falls from 84.1 to 15.9%. The second method uses the translation distance between 10 and 90% (Eqn. 2.9) [13] of the beam intensity, where \( \beta^{-1} \) is the radius at which cross-sectional profile of a Gaussian laser beam falls to \( 1/e \) times its maximum value (~37%). However, as the spatial beam profile in Fig. 2.8 showed, the beam is not Gaussian and so this is only an approximation of the true beam radius.

\[
r = x_{15.9} - x_{84.1}
\]

\[
\beta^{-1} = 0.552(x_{10} - x_{90})
\]

\[
r = \sqrt{2}\beta^{-1}
\]
2.2. TEA CO$_2$ laser

Using method 1 (Eqn. 2.8) the beam radius of the unfocussed CO$_2$ pulse works out to be 7.60 mm and from method 2 (Eqn. 2.9) the measured beam radius is 7.57 mm. This gives a beam diameter of 15.2 mm and 15.14 mm using methods 1 and 2 respectively. This compares well with a measurement made of the burn pattern that the unfocussed beam produces on thermal printer paper as shown in Fig. 2.11. Measurements of this pattern give a beam diameter of approximately 16 mm for the unfocussed CO$_2$ laser beam.

![Figure 2.11: Burn pattern taken of full CO$_2$ beam on thermal printer paper.](image)

Knife edge scans were also carried out at various axial positions after focussing with a 20 cm focal length lens. Some of these scans obtained are shown in Fig. 2.12. In each plot the Khosrofian fit is applied and therefore the minimum spot size can be extrapolated using the methods described in Eqn. 2.8 and Eqn. 2.9. After the minimum spot size was determined it could be plotted as a function of axial position to which a hyperbola is fitted [Fig. 2.13]. This fit is given by the following expression [Eqn. 2.10] for the beam waist at a distance $z$ from focus [15]

$$\omega_M(z) = \omega_{\text{min}} \sqrt{1 + \left( \frac{z\lambda M^2}{\pi \omega_{\text{min}}^2} \right)}$$

(2.10)

$$M^2 = \frac{\theta \pi \omega_0}{\lambda}$$

(2.11)

where $\omega_{\text{min}}$ is the beam radius at focus, $\lambda$ is the laser wavelength and $M^2$ is the beam quality factor [Eqn. 2.11] [16], $\theta$ is the half angle beam divergence and $\omega_0$ is the unfocussed beam waist radius.
2.2. TEA CO$_2$ laser

![Graphs showing energy distribution along various axial positions of CO$_2$ beam.](Figure_2.12)

Figure 2.12: Knife edge scans along various axial positions of CO$_2$ beam, focussed by a 20 cm lens.
2.2. TEA CO₂ laser

Figure 2.13: Beam waist using the $x_{15.1}-x_{84.9}$ method, after focussing with a 20 cm lens.

After fitting Eqn. 2.10 to the data, a value of the minimum beam radius and quality factor could be determined. Fig. 2.13 shows the data obtained after applying the $x_{15.1}-x_{84.9}$ method. All of the data is presented below for both the $x_{15.1}-x_{84.9}$ and $x_{10}-x_{90}$ methods [Table 2.1].

<table>
<thead>
<tr>
<th>Measurement</th>
<th>$\omega_{\text{min}}$ (µm)</th>
<th>$M^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$x_{15.1}-x_{84.9}$</td>
<td>0.370±0.068</td>
<td>4.22±0.76</td>
</tr>
<tr>
<td>$x_{10}-x_{90}$</td>
<td>0.359±0.058</td>
<td>3.98±0.64</td>
</tr>
</tbody>
</table>

Table 2.1: Results of knife edge measurements on focussed beam
2.3 Nd:YAG laser

2.3 Nd:YAG laser

In the double pulse and colliding plasma experiments described later in this work, a Continuum Surelite III Nd:YAG laser is employed as a pre-pulse laser and to form the “seeds” of the colliding plasmas. A brief description of the laser is given next, followed by information on the pulse energy, temporal profile and finally the triggering system used to time both the Nd:YAG and CO$_2$ laser in dual laser pulse experiments. It has a central wavelength of 1064 nm, maximum output energy of 1 J and a temporal pulse length of 5 ns.

2.3.1 Laser operation

The Nd:YAG is referred to as a solid-state laser because the YAG crystal is the gain medium of the laser. The energy level diagram of the lasing process in the Nd:YAG laser is shown in Fig. 2.14, taken from Ref [17].

![Energy Level Diagram](image)

Figure 2.14: The transitions responsible for 1064 nm radiation in Nd:YAG laser [17].

The lasing process involves four energy levels: E1–E4. The Nd ions are optically pumped from the ground state (E1) to an excited state (E4) by flash-lamps running at a repetition rate of 10 Hz. This state only has a short lifetime, and there is a very fast radiationless decay to the metastable state,
2.3. Nd:YAG laser

E3. The transition lifetime $\tau_{32}$ from the E3 to E2 level is long compared to that of E4 to E3 transition ($\tau_{43}$) and a population accumulates in the lasing level for about 100 $\mu$s. Thus population inversion takes place between the E3–E2 levels, and between these states relaxation can occur through spontaneous or stimulated emissions. This results in the characteristic 1064 nm wavelength radiation of the Nd:YAG laser. The lower laser level, E2, quickly relaxes to the ground state in a fast decay time $\tau_{21}$.

The $Q$-switch technique is used in the lasing process of the Nd:YAG laser. The name for the technique arises from the quality factor, $Q$, which is the ratio of the energy stored in the cavity to the energy loss per cycle [4]. The energy in the upper level, E3, builds up until the $Q$-switch is activated and is then released as a single high energy pulse. However, lasing can be prevented initially by maintaining controlled losses in the system by use of a low $Q$ factor. The gain is high but so also are the losses and therefore lasing does not occur. The Nd ions are kept in the E3 level by the continuous pumping of the flash-lamps until a high $Q$ factor is activated when the gain reaches its peak, thereby opening the cavity to allow oscillation.

The process works by means of an electro-optic switch and a description of the operation taken from the laser manual is given here. It consists of a Pockels cell, a plate polarising element and a quarter-wave ($\lambda/4$) plate. A schematic of the arrangement inside the laser is shown in Fig. 2.15 on the next page. In a closed cavity [Fig. 2.15 (a)], the beam propagating within the oscillator cavity makes a double pass through the Pockels cell and the $\lambda/4$ plate. With 0 volts applied to the Pockels cell there is no rotation added, while the $\lambda/4$ plate will add $45^\circ$ with each pass, thereby giving a total rotation of $90^\circ$. Therefore the horizontally polarised beam that was transmitted through the plate polariser has been rotated to vertical polarisation. This is rejected by the polariser and no lasing occurs. In the case of Fig. 2.15 (b), the beam makes a double pass through the Pockels cell and the $\lambda/4$ plate. With 3600 volts on the Pockels cell there will be $45^\circ$ rotation added while the $\lambda/4$ plate adds $45^\circ$ with each pass, this will result in a total rotation of the beam of $180^\circ$. Therefore the output is returned to horizontally polarised light so that it can be transmitted by the polariser and lasing can occur.
2.3. Nd:YAG laser

Figure 2.15: Nd:YAG laser cavity when (a) closed with no lasing and (b) open with lasing.

2.3.2 Laser pulse energy and temporal profile

The Continuum Surelite III is designed for higher energy output and as such does not contain two additional $\lambda/4$ plates and dielectric polariser to improve the polarisation of the laser output, as is the case with other models of the Nd:YAG laser. As a result the peak output energy is as high as 1 J, however this also means the laser light is not as fully polarised and
2.3. Nd:YAG laser

will contain additional modes. The inclusion of extra modes will reduce the quality of the pulse slightly as well as increase the energy. This is evident in the structure of the temporal pulse profile [Fig. 2.16], where additional structure can be seen in the pulse profile when viewed with a fast photodiode with a sub nanosecond rise-time.

In experiments it is important to be able to control the Nd:YAG laser output energy as this will vary the on-target laser intensity. This was achieved by passing the beam through a half-wave (\(\lambda/2\)) plate and a linear polariser. By varying the angle of the \(\lambda/2\) plate, the polarisation state of the beam changes. The linear polariser is placed following the wave plate at the Brewster angle (57° in air) with respect to the laser beam. The light polarised in the plane (\(P\)-polarised) will be transmitted by the polariser while light polarised perpendicular to the plane (\(S\)-polarised) will be reflected. Therefore the laser beam can be varied between these polarisation states by rotation of the \(\lambda/2\) plate and the result is a range of energies between the maximum (fully \(p\)-polarised) and minimum (fully \(s\)-polarised). Due to the additional modes mentioned above, the beam can not be fully polarised and some energy

Figure 2.16: Sample temporal profiles recorded of the Surelite III Nd:YAG laser.
2.3. Nd:YAG laser

will still be transmitted when the laser pulse is s-polarised.

2.3.3 External triggering set-up for dual laser pulse experiments

The Continuum Surelite III Nd:YAG laser was triggered using the direct access triggering (DAT) Mode 2. This is reported to have an inherent jitter of ±1 ns according to the laser manual. This meant that the CO₂ and Nd:YAG lasers could be set to trigger with varying delays between them, all controlled by the delay generator. The overall recorded jitter of such a system was about 10 ns. The timing in experiments was achieved by using a Stanford delay generator along with a 10 Hz frequency generator. A schematic of the timing circuit is shown in Fig. 2.17.

Figure 2.17: Schematic of timing set-up used to trigger both lasers externally in dual laser pulse experiments.
2.3. Nd:YAG laser

The full procedure of operating the Continuum Surelite III Nd:YAG laser in DAT 2 mode can be found in the laser operation manual (pages 45-46), but a brief description is given here. The Q-switch toggle at the rear of the laser controller was set to EXT. A D style male connector was connected to the EXTERNAL port on the front panel. The BNC cables used to trigger the flashlamp and Q-switch are both soldered to the connector. A 15 V DC power supply powers the 10 Hz frequency generator which produces a negative going 5 V TTL signal 10 µs wide that triggers the flashlamps of the Nd:YAG laser. The flashlamp sync out, which is a 10 Hz negative going 3.5 V TTL signal 20 µs wide, is taken from the front panel of the Nd:YAG laser controller and sent to a “pulse extractor”, also powered by the 15 V power supply.

When the red trigger button is pushed on the pulse extractor, a 20 µs 5 V TTL pulse is generated when the next input trigger from flashlamp sync out has been received. This push button on the pulse extractor box is thus the master of the whole timing set-up. It outputs a positive 5 V TTL signal 20 µs wide which externally triggers the Stanford delay generator DG645. The AB output on the delay generator is sent to trigger the Q-switch of the Nd:YAG laser resulting in lasing. Output A has a delay of 243 µs set to it as this was found to be the optimum delay between the flashlamp sync out and the Q-switch for producing the maximum laser output energy. A delay of A + 10 µs is set on output B, meaning AB produces a negative going 10 µs 5 V TTL pulse necessary for triggering the Q-switch.

By observing the temporal profiles of both the Nd:YAG and CO₂ laser on an oscilloscope the time delay between them could be varied using the Stanford DG645 until the rising edge between both matched up temporally. This was assumed to be a 0 ns delay between both laser pulses, not withstanding the ±10 ns jitter. Therefore the time delay between the lasing action of both lasers was determined to be approximately 241.825 µs. The delay on output E was now set to this value of 241.825 µs. Output C for triggering the CO₂ laser was now set to E + ∆t, therefore the delay between the lasers will be equal to the value set for ∆t on output C. Finally, output D was set to C + 10 µs and produced a positive going 5 V TTL pulse necessary for externally triggering the CO₂ laser.
Spectroscopy and EUV in-band measurements are almost exclusively carried out in experiments using the Jenoptik [19], 0.25 m absolutely calibrated EUV Spectrograph. The Jenoptik is a grazing incidence flat-field spectrometer with an operating wavelength range of 9.6 to 18 nm. A schematic of the spectrometer is shown above in Fig. 2.18 (reproduced from the PhD Thesis of P. Hayden [18]) and consists of an entrance slit, an Hitachi spherical variable line spacing grating (1200 lines per mm), a zero order stop, a number of apertures for blocking stray light and a charge coupled device (CCD) camera for detecting the image. The vacuum vessel containing the spectrograph is maintained under a high vacuum on the order of \(1 \times 10^{-7}\) mbar.

The entrance slit has a fixed horizontal length of 4 mm and a maximum height of 36 \(\mu\)m when fully opened. It is opened by a piezoelectrical actuator by applying a voltage, which is controlled by the Princeton Instruments [20] ST-133A camera controller. When an open slit is required for an exposure of the CCD, the ST-133A camera controller sends a TTL high signal to the piezo controller which then supplies a voltage of between 0 V and 150 V (at 150 V the vertical slit separation is 36 \(\mu\)m). When the slit is closed the controller sends a TTL low signal to the piezo controller. A -10 V signal is then sent to the slit to ensure it is fully shut [18]. It takes 2.5 ms for the slit to be fully opened and another 2.5 ms for it to fully closed. Therefore
2.4. Jenoptik extreme ultraviolet spectrograph

exposure times of less than 5 ms should not be used. The slit opening time is synchronised to the readout of the CCD, to ensure a well defined exposure of the CCD array.

The zero order stop, along with the apertures, prevent scattered light from being captured by the CCD and ensure that the spectra recorded are entirely from the first order diffraction from the grating. The stop is coated with a Mo/Si multilayer which is highly reflective around 13.5 nm as well as reflecting visible light. This means that an alignment laser can be very easily used to align the spectrometer along one axis of the target chamber. The laser light is reflected by the zero stop and up to a small viewing port above as shown in the schematic. However, placing a photodiode on the port, coupled with a filter for suppressing the visible emission, can allow in-band EUV measurements to be made also.

Due to the variable line spacing of the spectrometer grating, a spectrum is formed along a flat focal plane, which can be recorded directly by the Princeton Instruments PI•MAX:1K SX 1024EB/TE [33] CCD. The CCD chip consists of a 1024 × 1024 pixel array, with each pixel being 13 µm × 13 µm in size. The CCD is highly sensitive as it is a thinned backside illuminated CCD. It has sensitivity from 0.51 counts/photon at 18 nm to 0.94 counts/photon at 12.5 nm. However, the readout time of the array is relatively long at approximately 1 s. The camera continues to be sensitive to radiation during the readout time, thus the need for the piezoelectric slit [18]. WinSpec32 [21] software is used to interface with the controller and operate the CCD.

2.4.1 Spectrograph calibration

The Jenoptik spectrometer was absolutely wavelength and intensity calibrated at the PTB radiometry beamline at the BESSY II electron storage ring in Berlin. A calibration file supplied with the spectrometer details the wavelength (λ), the change in wavelength (δλ) and the number of counts per photon for each pixel of the CCD [Fig. 2.19] [18]. An additional feature, which is not part of the intensity calibration, appears in the spectra produced around 12.3 nm. The feature is an anomaly that was most likely caused by degradation of the front illuminated Si CCD, as it is close to the Si L–absorption edge. However, it lies far enough away from the in-band 13.5 nm region to not be of major concern for this work.
2.4. Jenoptik extreme ultraviolet spectrograph

The solid angle ($\Omega$) for the collection of EUV radiation by a pixel of the spectrometer is given by:

$$\Omega = \left( \frac{\text{Slit Width}}{L_1} \right) \times \left( \frac{\text{Pixel Width}}{L_2} \right)$$  \hspace{1cm} (2.12)

where the Slit Width is 0.036 mm, the Pixel Width is 0.013 mm, $L_1$ is the distance from the plasma formed in the centre of the chamber to the entrance slit of the spectrometer and $L_2$ is the distance from the plasma to the CCD detector [18]. During experiments the set-up varied so that a few different lengths for $L_1$ and $L_2$ were used. However, it was calculated to be $2.4 \times 10^{-10}$ sr for the colliding plasma experiments carried out in this work. Once the solid angle has been calculated and knowing the calibration data [Fig. 2.19], the pulse energy ($E_{\text{Pulse}_i}(\lambda)$) in J/(sr nm) for pixel $i$ per wavelength interval of width $\Delta \lambda_i$ can be calculated as
2.5 Experimental equipment

\[ E_{\text{Pulse}_i}(\lambda) = \left( \frac{N_{\text{counts}_i}}{\Delta \lambda_i} \right) \times \left[ \frac{1.602 \times 10^{-19} \text{ (J/eV)} \times \frac{1239.8 \text{ eV nm}}{\lambda_i}}{N_p} \right] \times \left[ 1 + \frac{1}{0.94} \right] \quad (2.13) \]

where \( N_{\text{counts}_i} \) is the average number of counts in pixel \( i \) and \( N_p \) is the number of pulses. The \( \frac{1+0.94}{2} \) term arises due to the difference in grating reflectivity for \( S \)- and \( P \)-polarised light. The 2\% in-band energy (J/sr) is determined by integrating \( E_{\text{Pulse}_i}(\lambda) \) between 13.365 and 13.635 nm (which spans 33 pixels of the CCD). The conversion efficiency (CE) is then calculated by multiplying by 2\( \pi \), and dividing by the on-target laser energy. This is assuming the emission is isotropic in a solid angle of 2\( \pi \) sr.

\[ CE(\% \text{ per } 2\pi \text{ sr}) = \left( \int_{\text{2\% bandwidth}} E_{\text{Pulse}_i}(\lambda) \right) \times \frac{2\pi}{E_{\text{Laser pulse}}} \times 100 \quad (2.14) \]

2.5 Experimental equipment

2.5.1 Motorised motors and stages for target mount and lenses

The \( x \), \( y \) and \( z \) positioning of the targets used in experiments were controlled by using a series of Zaber linear actuators and stages, that could be controlled while under vacuum, from a laboratory computer. The advantage of these actuators is they can be connected in series, linking all of them together, meaning only one electrical connection to the chamber was required to control them. Several programmes were written using Visual Basic to control the positioning of each actuator during experiments. The 3-D motion of the target position was very important as it enables precise alignment of the plasma, formed near the target surface, with the optical axis of the spectrometer. It also allowed the surface to be replenished by moving to a new spot when desired. The orientation of the target inside the chamber could be changed so as to vary the angle of incidence of the laser beams or spectrometer viewing angle. Lenses are also mounted on linear motion stages with actuators. This allows variation of the focussed beam position with respect to target surface for laser intensity variations. Fig. 2.20 shows the view from inside the chamber, showing the various actuators and demonstrates how they are utilised in experiments.
2.5. Experimental equipment

![Image of experimental setup](image)

Figure 2.20: Image of a view from inside the chamber of typical set-up.

2.5.2 Optics

The CO$_2$ beam is directed into the target chamber using 25.4 mm diameter gold coated copper mirrors. Mirrors can be quite easily fabricated for the CO$_2$ laser as most metals are highly reflective in the far infra-red region of the spectrum. The beam enters the chamber through a zinc selenide (ZnSe) window that is mounted onto a flange on one of the chamber ports, in order to maintain the chamber under vacuum. Inside the chamber ZnSe is again used, this time a ZnSe plano-convex lens is used to focus the laser beam. The transmission curve of uncoated ZnSe is shown in Fig. 2.21 [22]. ZnSe reflection of light begins to increase in the visible region, with transmission dropping to 60% at 600 nm and 10% below 500 nm. This effect is observed later on in this work when visible images of the plasma are recorded in Chapter 5.
2.5. Experimental equipment

![ZnSe transmission curve](image1)

Figure 2.21: ZnSe transmission curve [22].

![Nd:YAG laser line mirror transmission curve](image2)

Figure 2.22: Nd:YAG laser line mirror transmission curve [22].

Coated UV grade fused silica or BK7 glass substrate mirrors are used for the Nd:YAG laser at 1064 nm. The transmission of the mirrors around 1064 nm is shown in Fig. 2.22 above [22]. Substrate materials constructed from BK7 with anti-reflection coatings serve as plano-convex lenses and flat windows that are also used for the Nd:YAG laser.
2.5. Experimental equipment

2.5.3 Vacuum chambers and pumps

All experiments were carried out in the target chamber shown in Fig. 2.23. The drawing of the chamber was made using the Solid Edge software package [23] and was used to visualise the actuators, stages, breadboard and target layout before purchasing all the individual elements. As previously stated, the experiments were all carried out under vacuum to prevent the EUV light being absorbed in air, therefore the target chamber and spectrometer needed to be maintained at low pressure. The need for this can be demonstrated by calculating the transmission of photons in the EUV region through air, on the website of the Center for X-ray Optics (CXRO) [24]. The calculation is carried out for a distance of 120 cm, as this equates approximately to the distance the EUV photons need to travel in the experimental set-up from the plasma to the CCD. The strong absorption of EUV at higher pressures is found, and as the number of particles is reduced the transmission is seen to increase. At $1 \times 10^{-4}$ mbar the transmission around 13.5 nm is almost 100%. To achieve the necessary pressure, a water cooled Leybold Turbovac TMP 151 turbo-molecular pump [25] with a pump speed of 145 l/s backed by an oil free Scrollvac SC 5D roughing pump [26] was used.

![Schematic of the experimental set-up made using Solid Edge.](image)

Figure 2.23: Schematic of the experimental set-up made using Solid Edge.
Bibliography


Bibliography


Chapter 3

Study of EUV emission from CO$_2$ laser produced tin plasmas

3.1 Introduction

In this chapter results are presented on experiments carried out to investigate the 10–18 nm extreme ultraviolet (EUV) emission from laser produced tin (Sn) plasmas formed with a CO$_2$ laser. The experiments investigate the effect of various laser parameters on the efficiency of the 13.5 nm in-band emission. Parameters such as the lens focus position, the on-target laser energy, the gas mixture in the laser chamber, the angle of laser incidence and spectrometer viewing angle were all varied. The second section of this chapter reports on results of double pulse experiments, in which a laser pre-pulse is used to form a plasma target, which the CO$_2$ laser pulse then interacts with.

3.2 Experimental set-up

For single pulse CO$_2$ EUV experiments the laboratory layout is as shown in Fig. 3.1. In Fig. 3.1 (a), the CO$_2$ laser is shown on the far right of the image, and its beam is directed into the target chamber via three gold coated mirrors. The first two mirrors act as a periscope to bring the beam down to the same height as that of the entrance window port of the chamber. A red alignment laser (operating wavelength $\approx 630$ nm) on a linear track [labeled in Fig. 3.1 (b)] is placed at the same height as the beam and the entrance window. The alignment laser is mounted on top of a stage on the linear track so that it can be slid in and out of the beam path when needed. The red laser spot can be aligned to the CO$_2$ laser spot and therefore is used to align the CO$_2$ laser onto the target surface. Diagnostics of each laser
3.2. Experimental set-up

pulse are made using the reflections from ZnSe windows placed at 45° in the beam path. The windows act as approximately 97:3 beam splitters and allow in-situ measurement of the pulse temporal profile and laser energy with the photon drag detector and energy head respectively [Fig. 3.1 (b)], as described in Chapter 2. In Fig. 3 (a) the Jenoptik EUV spectrometer is seen to be orientated at the chamber port 45° with respect to the incoming laser pulse. The effective viewing angle of the spectrometer and angle of incidence of the laser were thus changed by rotation of the target surface.

Figure 3.1: Laboratory layout for EUV experiments.
3.3. Effects of varying CO$_2$ laser parameters on EUV emission

3.3 Effects of varying CO$_2$ laser parameters on EUV emission

3.3.1 Varying the lens position to change the beam spot size

The first parameter that was varied was the lens to target distance. Movement of the lens position with respect to the target is of relevance as it changes the spot size of the laser pulse on the target surface, which will vary the laser intensity $\phi$ (W/cm$^2$) and also effect the size of the plasma source. The lens used was a 10 cm focal length plano-convex ZnSe lens, which was mounted on a linear stage controlled by a motorised actuator. The laser was directed at normal incidence onto the target surface and the spectrometer viewed the plasma at 45$^\circ$ with respect to this. As the lens is moved, its position can be recorded by the control panel of the Visual Basic program used to control its motion. Fig. 3.2 shows the result of one such lens scan. At each lens position three spectra are taken, allowing an averaged in-band energy to be determined and hence conversion efficiency (CE), by using the process set out in Chapter 2. The actuators have a maximum range of motion of 60 mm and the minimum step size can be altered. In this case there are a maximum of 60 steps, meaning that each step gives 1 mm of motion.

![Figure 3.2: CE as a function of the lens position.](image-url)
3.3. Effects of varying CO₂ laser parameters on EUV emission

Two positions of peak emission are observed from the lens scan, with a maximum CE of approximately 1.6% (13.4 mJ in-band energy in 2π sr from \(~800\) mJ laser energy), but also a dip in the emission is observed. This dip in the centre of the lens scan is assumed to be the location of the position of tightest focus of the lens [1] and appears in all lens scans. By assuming the centre point to be the focus, the lens scan can be replotted as in Fig. 3.3, showing the CE as a function of the distance from focus. The positive scale indicates when the lens is further from the target surface and it becomes negative as the lens moves through focus closer to the target. Thus the peak emission occurs at 2–3 mm from focus, which, using information on the beam determined from the knife edge experiment in Chapter 2, corresponds to a beam diameter of 496 \(\mu\)m at 2 mm and 612 \(\mu\)m at 3 mm.

![Figure 3.3: CE as a function of the distance from focus.](image)

Previous investigations into the effects of laser spot size have shown that it has a large influence over the shape of the plasma [2] and also the EUV emission [1]. In these works, Harilal et al. observed that a small spot size results in a spherically shaped plasma, while the plasma expansion becomes more cylindrical for a larger laser spot size. Therefore smaller plasmas formed at tighter focus are observed to have more lateral expansion. There are several reasons why the drop in emission may be occurring at the position of tightest focus, such as overheating due to too high a laser intensity, self-absorption caused by opacity effects in the plasma or the size of the laser spot being coupled into the plasma.
3.3. Effects of varying CO$_2$ laser parameters on EUV emission

![Graph showing EUV emission intensity vs. wavelength for two positions: best CE and focus.](image)

Figure 3.4: Three shot averaged Sn EUV spectra measured with the lens at the position of best CE and tightest focus, using approximately 800 mJ laser energy.

The idea that the plasma is being overheated is an obvious solution, as the smaller focus spot size will mean a higher on-target intensity which could result in the dominant ion stages present in the plasma being above those of Sn$^{7+}$–Sn$^{13+}$ necessary for in-band emission. However, if the spectra at these two positions are compared, as in the three shot averaged spectra in Fig. 3.4, it can be seen that they are in fact very similar. The intensity at the position of focus is less than that for peak emission, but there does not appear to be clear differences in the spectral features, which would suggest a similar temperature plasma and not one that is being overheated. This argument is further supported by the fact that the dip is also observed when a lower laser energy input is used, such as $\sim$700 mJ and $\sim$450 mJ, shown in Fig. 3.5. In these instances the peak laser intensity could not have been achieved because of the lower energy input, and therefore the plasma could not be overheated. The spectra produced by both of these laser energies at focus and peak emission are then shown in Fig. 3.6 and Fig. 3.7, confirming the conclusions made regarding the spectra of higher energy laser input in Fig. 3.4.
3.3. Effects of varying CO\textsubscript{2} laser parameters on EUV emission

Figure 3.5: CE as a function of the distance from focus for \sim 700 mJ and \sim 450 mJ CO\textsubscript{2} laser energy.

Figure 3.6: Three shot averaged Sn EUV spectra, using approximately 700 mJ laser energy.
3.3. Effects of varying CO$_2$ laser parameters on EUV emission

The spectra taken at the peak emission and at focus in Fig. 3.4, Fig. 3.6 and Fig. 3.7 appear to be optically thin, with very little absorption effects that are usually associated with an overheated plasma. Therefore, the reason for the dip at focus can most likely be attributed to the loss of laser energy due to lateral expansion of the spherical plasma formed with a smaller spot size [3].

Another feature observed in the lens scans was that one of the peaks either side of the focus was higher than the other. This was seen to be the case for the three different laser energies used in Fig. 3.3 and Fig. 3.5. However, the focussed spot size will be the same for two points equi-distant from focus, so it is not just the size of the spot that is influencing the in-band energy emitted. Fig. 3.8 helps visualise what is happening in both cases. In Fig. 3.8 (a) the plasma is formed by a certain spot size and as it becomes more dense, it expands and the absorption front moves outwards and interacts with the laser pulse. However, the size of the laser cone that is coupled into the plasma is quite small, as the plasma will first need to pass through the laser focus. In part (b) the plasma is formed with the same spot size, but this time there is a much larger laser cone present to interact with as the plasma expands. This will mean better coupling of the laser by inverse Bremsstrahlung and lead to greater emission of EUV photons.

Figure 3.7: Three shot averaged Sn EUV spectra, using approximately 450 mJ laser energy.
3.3. Effects of varying CO$_2$ laser parameters on EUV emission

Figure 3.8: CO$_2$ laser focusing onto a Sn target. The lens positions shown are for (a) the lens further from the target and (b) closer to the target. Both result in the same focused spot size but different in-band EUV energies.

3.3.2 The effect of laser energy

The laser energy can be varied by adjusting the high voltage supply to the discharge across the gas chamber containing the CO$_2$, He and N$_2$ gases. Fig. 3.9 shows the range of CO$_2$ laser energies achievable by varying the laser discharge voltage. These energies were typical of the output measured from the laser, however from week-to-week operation of the laser with changing gas mixtures, these energies were also seen to increase and decrease in value. The previous section has already shown some of the results from studying the effect of laser energy on the EUV emission from Sn plasma. Laser voltages of 21, 24 and 27 kV were chosen, which corresponded to laser energies of the order of 450, 700 and 800 mJ respectively. The variation of the energy stability from shot to shot for the three voltages is shown in Fig. 3.10. A discharge voltage of 21 kV results in an output energy of 473.5 ± 21.7 mJ, the average energy produced by a 24 kV discharge is 690 ± 27.8 mJ while 846.5 ± 37.4 mJ was recorded for a laser discharge voltage of 27 kV. Fig. 3.11 shows the recorded temporal profiles for the three voltages, with the profiles due to the higher laser voltage seen to have a higher intensity as expected. Note that the profiles have been smoothed to allow the three profiles to be plotted together.
3.3. Effects of varying CO$_2$ laser parameters on EUV emission

![Figure 3.9](image1.png)

Figure 3.9: Plot of laser output energy as the discharge voltage is increased.

![Figure 3.10](image2.png)

Figure 3.10: CO$_2$ laser shot to shot energy stability for voltages of 21, 24 and 27 kV.
3.3. Effects of varying CO$_2$ laser parameters on EUV emission

![Figure 3.11: Temporal profiles recorded for voltages 21, 24 and 27 kV.](image)

Table 3.1 is presented next and shows the results of varying the laser input energy on EUV emission. The laser intensity is calculated in each case only for the spike in the temporal profile ($\approx$60 ns duration) and therefore an energy of 40% of the total energy is used, along with a focal diameter of 600 $\mu$m. As will be discussed in Chapter 4, a large amount of the energy present (as much as 60–70%) in the laser pulse is actually contained in the N$_2$ tail.

<table>
<thead>
<tr>
<th>V (kV)</th>
<th>Energy (mJ)</th>
<th>$\phi$ (W/cm$^2$)</th>
<th>IBE (mJ)</th>
<th>CE (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>21</td>
<td>473.5 ± 21.7</td>
<td>$1\times10^9$</td>
<td>5.75</td>
<td>1.24</td>
</tr>
<tr>
<td>24</td>
<td>690 ± 27.8</td>
<td>$1.5\times10^9$</td>
<td>10.13</td>
<td>1.44</td>
</tr>
<tr>
<td>27</td>
<td>846.5 ± 37.4</td>
<td>$2\times10^9$</td>
<td>13.37</td>
<td>1.63</td>
</tr>
</tbody>
</table>

Table 3.1: Effects of laser energy. Table key: V = voltage, $\phi$ = laser intensity, IBE = in-band energy and CE = conversion efficiency.
3.3. Effects of varying CO$_2$ laser parameters on EUV emission

3.3.3 CO$_2$ laser gas mixture

Following on from this, tests were made on the effect of the gas mixture ratio (He:N$_2$:CO$_2$) within the laser chamber. These mixes include the standard mix of 6:1:3 which was recommended on the installation of the laser for energy stability and a N$_2$ heavy mixture of 5:2:3. The laser intensity falling on the target surface was varied by scanning the lens as before. The lens position was then fixed at the position just after focus, where the optimum emission is observed. At this position five spectra were taken and averaged for a range of different gas mixtures. The results of this are shown in Table 3.2 below. As stated previously, the standard mixture of 6:1:3 is the most stable, whereas higher CE values are obtained when more N$_2$ is added to the gas mixture. This is particularly seen to be the case for the 5:2:3 mixture, which produces an average CE of around 1.95% and maximum values above 2%. The higher percentages of N$_2$ and reduction of the buffer gas, are seen to lead to higher energy laser output that produce a greater laser intensity. This increasing intensity is seen to increase EUV emission.

<table>
<thead>
<tr>
<th>He:N$_2$:CO$_2$</th>
<th>Energy (J)</th>
<th>IBE (mJ)</th>
<th>CE (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>7:1:2</td>
<td>0.87 ± 0.01</td>
<td>13.57 ± 0.21</td>
<td>1.62 ± 0.04</td>
</tr>
<tr>
<td>6:1:3</td>
<td>0.91 ± 0.02</td>
<td>15.04 ± 0.32</td>
<td>1.66 ± 0.04</td>
</tr>
<tr>
<td>4:2:4</td>
<td>0.94 ± 0.05</td>
<td>16.59 ± 0.86</td>
<td>1.76 ± 0.12</td>
</tr>
<tr>
<td>4:3:3</td>
<td>1.09 ± 0.07</td>
<td>20.05 ± 1.28</td>
<td>1.85 ± 0.07</td>
</tr>
<tr>
<td>5:2:3</td>
<td>1.07 ± 0.07</td>
<td>20.82 ± 1.31</td>
<td>1.95 ± 0.19</td>
</tr>
</tbody>
</table>

Table 3.2: Effects of laser gas mixture. Table key: IBE = in-band energy and CE = conversion efficiency.

Five shot averaged spectra for both the standard gas mix of 6:1:3 and the higher energy gas mix 5:2:3 are shown next [Fig. 3.12].
3.3. Effects of varying CO$_2$ laser parameters on EUV emission

![Normalized intensity vs Wavelength (nm)](image)

Figure 3.12: Five shot average spectra recorded using 5:2:3 and 6:1:3 gas mixtures.

3.3.4 Incoming laser beam angle of incidence and spectrometer viewing angle

To study the effect of angle, both the laser incidence and the spectrometer viewing angle were altered in the next investigation. It has been observed, for Nd:YAG LPPs, that there is a significant change in the Sn spectrum as a function of angle of incidence (AOI) for detection and irradiation [4–7]. The changes seen for the Nd:YAG laser are quite pronounced with a significant reduction in CE occurring, when viewed at 90$^\circ$ to the target normal as compared to 45$^\circ$, due to the higher optical thickness at the cold side lobes of the plasma. The distribution of ions emitted from the plasma is also seen to be strongly angularly dependent [8, 9], as is the emission of out of band (OOB) radiation [10]. However, this effect is not as significant for CO$_2$ laser generated Sn plasmas due to reduced opacity. This is due to the critical electron density ($n_{ec}$) of the plasma being inversely dependent on the laser wavelength ($\lambda$) [11]:

$$n_{ec} \propto 1/\lambda^2$$

(3.1)

From this equation the critical density of a CO$_2$ ($\lambda$=10.6 $\mu$m) LPP is deter-
3.3. Effects of varying CO\textsubscript{2} laser parameters on EUV emission

mined to be $9.9 \times 10^{18} \text{ cm}^{-3}$, while a Nd:YAG ($\lambda=1.06$ $\mu$m) LPP is $1 \times 10^{21} \text{ cm}^{-3}$. This is why the Sn spectrum produced by the CO\textsubscript{2} is seen to have a lot more observable line features, a characteristic associated with an optically thin plasma.

The set-up for angular studies is shown in Fig. 3.13 below. The CO\textsubscript{2} laser was orientated [Fig. 3.13 (a)] at 45\textdegree to the target surface and also [Fig. 3.13 (b)] along the normal incidence of the target surface. The position of the Jenoptik spectrometer was kept fixed while the target was rotated, this had the effect of changing the angle at which the emission was observed from normal incidence (0\textdegree) to 45\textdegree.

![Figure 3.13: Spectrometer orientated at (a) 0\textdegree and (b) 45\textdegree with respect to the target normal.](image)

Spectra recorded using both orientations are shown in Fig. 3.14. The energy was fixed at around 900 mJ and spectra were taken at three different lens positions for both viewing angles of the spectrometer. The spectra show there is no observable change in shape or absorption between viewing at (a) 0\textdegree and (b) 45\textdegree.
3.3. Effects of varying CO$_2$ laser parameters on EUV emission

Figure 3.14: Spectra recorded at $0^\circ$ and $45^\circ$ spectrometer viewing angles, for three different laser intensities: (a) $7\times10^7$ W/cm$^2$, (b) $1\times10^8$ W/cm$^2$ and (c) $5\times10^9$ W/cm$^2$. 
3.4. Double laser pulse experiments

3.4 Double laser pulse experiments

An improvement in the coupling of the CO\textsubscript{2} pulse with the target can be achieved by reducing the concentrations of Sn in the target to the minimum number of atoms required for EUV emission. Mass limited targets [5, 6, 12–14] as well as liquid droplets of Sn [15] and other materials [16, 17] have been used as such in previous studies. These methods have the advantage of suppressing out of band radiation and reducing self-absorption by creating a less dense plasma. Dual laser pulses have also been shown to enhance soft x-ray emission in laser produced plasmas [18, 19]. The use of dual laser illumination also has the advantage of reducing fast ions [12, 14, 20, 21], particulate debris and increasing the rate of ionisation in the entire plasma. The former is advantageous for mitigating debris emanating from the EUV source from damaging optics, by ionising as much of the particles in the plasma as possible so that magnetic fields can be used to deflect all of the debris [22]. The plasma targets also have preferable electron densities over solid targets, which are closer to the critical density of the CO\textsubscript{2} laser and allow for better absorption. In the experiments described here, the Nd:YAG laser is used to produce an initial plasma, which is then re-heated by the CO\textsubscript{2} laser beam. After a time delay and with certain initial Nd:YAG laser conditions, an optimised plasma target can be formed to be re-heated with the CO\textsubscript{2} laser. To optimise absorption the plasma size also needs to be tailored to match that of the focussed CO\textsubscript{2} beam diameter.

3.4.1 Double pulse set-up

The set-up of the equipment for the double pulse experiments was as shown in Fig. 3.15. The lasers were incident at 90° with respect to each other and the Jenoptik spectrometer was set-up so that it was viewing at 45° to the target normal and hence the Nd:YAG laser pulse. Alignment lasers were aligned to both laser pulses and then used to direct the two beams into the chamber and intersect each other. The external triggering and delay set between both lasers was controlled by the method set out in Chapter 2. The position of the laser pulses in time was determined by recording the temporal pulse profiles of both on a digital oscilloscope. The infra-red temporal profiler was used to determine the timing of the CO\textsubscript{2} pulse and a fast photodiode recorded the timing of the Nd:YAG laser, with an inherent timing jitter of 10–15 ns due to the triggering of the CO\textsubscript{2} laser.
3.4. Double laser pulse experiments

Figure 3.15: Lab set-up for dual laser pulse experiments.
3.4. Double laser pulse experiments

3.4.2 Double pulse results

The spectra recorded show an increase in in-band emission after re-heating the prepulse plasma with the main laser pulse. Fig. 3.16 demonstrates this by showing the emission spectrum due to the Nd:YAG laser only and the spectrum after it has been re-heated by the CO$_2$ laser approximately 80 ns later. This shows the interaction between the laser and the plasma is resulting in enhanced EUV emission. The Nd:YAG laser lens position was fixed at 4 mm from focus, while the on-target energy was 491.9 ± 8.2 mJ, which resulted in an estimated laser intensity of 1.15×10$^{11}$ W/cm$^2$. The full CO$_2$ pulse was not used, as was the case in the previous CO$_2$ studies in this chapter. Instead, a plasma pinhole shutter [23] described in Chapter 4, was used to clip the N$_2$ tail from the pulse. As a result of this, the on-target energy was approximately 200 mJ in 34.1 ± 3.8 ns, which corresponds to an intensity of 4×10$^9$ W/cm$^2$. However, exact energy measurements of the CO$_2$ pulse were also made for each shot by using the in-situ energy head, and these values were used in CE calculations.

![Figure 3.16: Averaged emission spectra produced by re-heated plasma and Nd:YAG only.](image-url)
3.4. Double laser pulse experiments

Figure 3.17: Averaged emission spectra produced by long pulse CO$_2$ only and re-heated plasma.

Figure 3.18: Averaged emission spectra produced by re-heated plasma and short pulse CO$_2$ only.
3.4. Double laser pulse experiments

The spectrum produced by the re-heat is compared to the CO$_2$ only spectrum in Fig. 3.17. The higher energy emitted in the EUV region is explained by the fact that almost four times as much laser energy went into generating the plasma. For a more direct comparison, Fig. 3.18 then shows the re-heated spectrum with that of a similar shortened CO$_2$ laser pulse on solid Sn. The opacity of the re-heated spectrum looks more like that of the Nd:YAG and not as optically thin as the CO$_2$. However self-absorption effects are clearly observable in the Nd:YAG spectrum that are not present in the re-heat. The dual laser pulse emission spectrum is also seen to be greater than the summation of the Nd:YAG only and shortened CO$_2$ only, as Fig. 3.19 demonstrates.

![Emission spectra comparison](image)

Figure 3.19: Averaged emission spectra produced by re-heated plasma and Nd:YAG summed with short pulse CO$_2$ only.

To optimise the emission in double pulse experiments, the plasma was re-heated at various positions along its expansion length ($z$). As the two lasers were incident at 90° with respect to each other, this could be achieved by stepping the Sn target backwards. The Nd:YAG lens was moved forward at the same rate to maintain consistent focussing conditions. The result of re-heating along the plasma length is presented in Fig. 3.20. Close to the target surface ($z < 500 \mu m$) the greatest coupling of the CO$_2$ into the plasma target is observed.

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3.4. Double laser pulse experiments

Figure 3.20: CE by re-heating at various distances along the plasma, also showing the positions (a–d) where the effect of time delay was tested.

The next parameter varied was the time delay between the two lasers. Four positions along the length of the plasma were chosen for testing the effects of the time delay, labeled (a), (b), (c) and (d) in Fig. 3.20. The results of the four time delay tests are shown in Fig. 3.21 on the next page. From this, the maximum CE observed is approximately 2.6% with a delay of 80 ns between the Nd:YAG and CO\textsubscript{2} lasers. The full results for the different laser systems used are tabulated on the next page [Table 3.3]. The Nd:YAG only CE of about 2.2% compares well with previous similar set-ups, where greater than 2% was achieved for solid Sn at 45° and laser intensities of $1\times10^{11}$ [6] and $3\times10^{11}$ W/cm\textsuperscript{2} [24]. DP w/o Nd:YAG, or double pulse without Nd:YAG, in Table 3.3 was calculated by excluding both the Nd:YAG laser energy and the amount of in-band energy the Nd:YAG generates by itself. Therefore the efficiency of the plasma as a target is being compared directly to the solid Sn as a target for EUV emission.
3.4. Double laser pulse experiments

Figure 3.21: Time delay tests at four positions from the target surface.

<table>
<thead>
<tr>
<th>Laser set-up</th>
<th>Energy (J)</th>
<th>IBE (mJ)</th>
<th>CE (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nd:YAG only</td>
<td>0.491 ± 0.008</td>
<td>14.29 ± 0.48</td>
<td>2.24 ± 0.07</td>
</tr>
<tr>
<td>Long CO₂ only</td>
<td>1.070 ± 0.070</td>
<td>20.82 ± 1.31</td>
<td>1.95 ± 0.19</td>
</tr>
<tr>
<td>Short CO₂ only</td>
<td>0.148 ± 0.003</td>
<td>4.69 ± 0.23</td>
<td>3.18 ± 0.22</td>
</tr>
<tr>
<td>Double pulse</td>
<td>0.774 ± 0.010</td>
<td>19.73 ± 0.72</td>
<td>2.55 ± 0.06</td>
</tr>
<tr>
<td>DP w/o Nd:YAG</td>
<td>0.138 ± 0.010</td>
<td>5.44 ± 0.19</td>
<td>3.94 ± 0.12</td>
</tr>
</tbody>
</table>

Table 3.3: Comparison between single and dual laser experiments. Table key: DP = double pulse, IBE = in-band energy and CE = conversion efficiency.
3.5 Summary and conclusions

A range of parameters were investigated to determine their effect on EUV emission from Sn LPP’s. Focussed spot size, laser energy, gas mixture, viewing angle, angle of incidence and use of plasma targets are all seen to impact on the emission. The optimisation of these parameters demonstrated that the use of a plasma target results in improved efficiency of the source. A maximum CE of around 4% was recorded during experiments, which agrees extremely well with maximum values observed by Fujioka et al., who reported a 4% CE by carrying out double pulse irradiation on pure Sn microdroplet targets, even though the Nd:YAG laser or EUV energy was not included for these maximum values reported here. Previous studies have shown that the pulse duration and mass density, are also seen to heavily influence the efficiency of in-band energy emitted from the plasma. Ando et al. [25] reported that a pulse duration of 2.3 ns was the optimum duration for a laser produced Sn plasma using a Nd:YAG laser, giving a 2% CE. For the CO₂ laser, modeling by Sunahara et al. demonstrated that, under certain experimental parameters, a pulse duration of 10 ns would be optimal for the most efficient 13.5 nm emission [26].

Studies of the mass density of Sn have also showed that using mass limited targets causes the self absorption effects in the plasma to be suppressed by reducing the amount of material present in the plasma [5, 6, 13]. The results of the initial studies of laser produced Sn plasmas in this chapter have demonstrated that in order to improve the CE of the laboratory source, these two parameters would need to be investigated further. Firstly, the low CE values have shown that the CO₂ temporal profile needs to be altered in order to remove any portions that don’t effectively contribute to the EUV emission and secondly the double pulse experiments showed that a low density, mass limited plasma is a preferred target for efficient interaction with the CO₂ laser. Chapter 4 “Plasma shutter device for CO₂ laser pulse shortening” and Chapter 5 “EUV emission from re-heating colliding plasma jet targets” investigate these two parameters.
Bibliography


Bibliography


Bibliography


Chapter 4

Plasma shutter device for CO$_2$ laser pulse shortening

4.1 Introduction

In this chapter two methods of reducing the CO$_2$ pulse duration are reported. Due to the low (2%) conversion efficiencies (CE) obtained with the long CO$_2$ pulse in Chapter 3, a shorter pulse duration was desired. Firstly, a new technique for shortening the temporal pulse length of the CO$_2$ laser is presented [Section 4.3], as outlined in the work by Donnelly et al. [1]. This novel method of clipping the laser pulse involves focussing the laser output onto a highly reflective metal target so that plasma is formed, which then operates as a shutter due to strong laser absorption and scattering. Precise control of the focussed laser intensity allows for timing of the shutter in that different temporal portions of the pulse can be reflected from the target surface before plasma formation occurs. This type of shutter enables the reduction of the pulse duration down to $\sim$2 ns. It also allows the removal of the low power, long duration tails that are present in transversely excited atmospheric (TEA) CO$_2$ laser pulses. A simple laser heating model was used to verify that the pulse shortening depends directly on the plasma formation time, which in turn is dependent on the applied laser intensity. Later in the chapter [Section 4.4], an alternative pulse shortening device, developed by Hurst and Harilal [2], is investigated. The effect of the shortened laser pulse on the EUV emission from Sn plasmas using this technique is studied and improvements in the CE up to 3–4% are observed.
4.2 TEA CO\textsubscript{2} laser temporal profile

The CO\textsubscript{2} laser used in experiments [see Chapter 2] employs the transverse electric atmospheric (TEA) configuration, which consists of a pulsed electrical discharge through a gaseous laser medium close to atmospheric pressure, generating a high power laser pulse. The laser gas medium is a combination of CO\textsubscript{2}, N\textsubscript{2} and He. The relative abundance of each gas determines the laser pulse energy and temporal profile. High volume manufacturing (HVM) suitable CO\textsubscript{2} lasers for extreme ultraviolet (EUV) lithography are still under development [3], but TEA CO\textsubscript{2} lasers are routinely used for research studies into the EUV emission of laser produced plasmas (LPPs). A TEA CO\textsubscript{2} laser temporal profile is characterised by an initial large peak of 50-100 ns pulse duration, followed by a low energy tail that is up to 5 \(\mu\)s in duration. The presence of a small amount (10\%) of N\textsubscript{2} in the gas mix allows for improved excitation of the upper level of the CO\textsubscript{2} molecule thus increasing the energy yield from the lasing process, but N\textsubscript{2} also has a longer de-excitation time, resulting in the low energy tail that follows the initial laser pulse peak. A large portion (50%-70\%) of the energy contained in the laser pulse is in this tail. An example of the TEA CO\textsubscript{2} laser pulse profile is shown in Fig. 4.1 (a), while Fig. 4.1 (b) shows the same pulse over a shorter scale to demonstrate the shape of the main pulse in more detail.

![Figure 4.1](image.png)

Figure 4.1: (a) Typical TEA CO\textsubscript{2} temporal profile showing the main spike and the \(\mu\)s N\textsubscript{2} tail. (b) A close up of the profile.
4.2. TEA CO$_2$ laser temporal profile

![Temporal profile graph](image)

Figure 4.1: (a) Typical TEA CO$_2$ temporal profile showing the main spike and the $\mu$s N$_2$ tail. (b) A close up of the profile.

The initial peak and long tail are observable in the pulse profile. In Fig. 4.1 (a) the tail of the pulse is seen to extend out to 1 $\mu$s in duration, while Fig. 4.1 (b) shows the main spike of the pulse lasting for about 50–100 ns. The laser pulse temporal profile is problematic for two reasons. Firstly, it is very difficult to assign a meaningful laser intensity value to the overall pulse when it is focussed onto a target. This issue is further compounded by the oscillatory nature of the pulse power during the laser pulse. Secondly, the low energy tail is of insufficient power to sustain the required electron temperature in the plasma. This means the ion stages required for efficient in-band EUV emission in Sn cannot be maintained, resulting in a poor CE. For these reasons, it is desirable to find a technique to remove the tail and also to tune the length of the pulse in order to improve the CE. The shortened pulses, if amplified, would also serve to generate plasmas with a higher electron temperature. Moving to shorter wavelength sources for lithography beyond 13.5 nm, amplified, short CO$_2$ pulses could be used to excite plasmas containing gadolinium for example, as 6.7 nm sources [4].
4.3. Plasma shutter experiments

4.2.1 Existing techniques for laser pulse shortening

Several methods of producing shortened CO$_2$ laser pulses have been previously demonstrated [2, 5–13] with varying degrees of complexity and success. Earlier techniques relied on focussing the laser pulse in a background gas to form a plasma shutter [5–7]. Plasma formation occurs at a definite time during the laser pulse and the remaining part of the pulse is absorbed in the plasma thus modifying the pulse length. Further development of this simple technique followed which saw the addition of an electrical discharge [8, 9] or a second laser pulse [10] to aid breakdown in the background gas. These procedures are limited to low repetition rates due to the background gas renewal time and control of spatial stability can also be an issue. Q-switched pulse shortening [11, 12] has also been demonstrated, but the output energies need to be small to avoid damage to optical components. Short pulse production by mode locking [13] schemes also suffer from this drawback. More recently, an aluminium pinhole target plasma shutter [2] has been demonstrated where the pulse is shortened by an expanding plasma, formed around the periphery of the pinhole. This technique will be discussed in more detail later in this chapter. The minimum reported duration achieved from these techniques, with the exception of Q-switched and mode locked pulses, is 10 ns [8].

4.3 Plasma shutter experiments

The TEA-CO$_2$ laser used in these experiments is the Optosystems Infralight SP10 gas flow laser system described in Chapter 2. The operating wavelength is 10.6 $\mu$m and the total energy output was measured to be 1–1.2 J for a laser gas mix ratio of 6:1:3 (He:N$_2$:CO$_2$) and a discharge voltage of 27 kV. Due to losses at several optical components the on-target laser energy was approximately 0.8–1 J. To estimate the laser intensity, the laser pulse profile is smoothed to remove the oscillating structure. Fig. 4.2 (a) shows the profile after the intensity has been smoothed. A Gaussian fitting function is then applied to the profile and superimposed onto the smoothed profile [Fig. 4.2 (b)]. From the fit, the pulse profile can be viewed in its two main components, i.e. the spike and tail. By comparing the ratio of their areas, by integrating under both curves, the spike is determined to be $\sim$ 45% of the total area, thus for a pulse of 1 J, the spike will contain approximately 0.45 J of the energy. However this calculation was carried for a 2$\mu$s pulse duration, therefore if the pulse was measured out further in time e.g. 5$\mu$s, the ratio of the spike to tail would reduce further. Fig. 4.3 shows a close-up of the fit in the region of the main spike, from which a full width half max (FWHM)
4.3. Plasma shutter experiments

pulse duration of $\sim 50$ ns was determined.

Figure 4.2: (a) Smoothed CO$_2$ pulse temporal profile (blue) and (b) Gaussian fit applied to the spike and tail (red).

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4.3. Plasma shutter experiments

Figure 4.3: Close up of Gaussian fit (red) applied to the spike region (blue) to allow an estimate of pulse duration.

4.3.1 Design and operation of plasma shutter

A schematic (not to scale) of the experimental set-up is shown in Fig. 4.4. The laser pulse to be shortened entered a collimating lens configuration where a 500 nm thick gold-coated silicon target was placed close to the collimator focus. The target was placed at 45° to the incident laser pulse. Motorised stages and actuators were used to move the target within the lens collimator set-up so as to change the focussed laser spot size on the reflective gold (Au) coated silicon (Si) target and thus change the applied laser intensity. A CO\textsubscript{2} laser mirror was then used on the same translation stage to direct the beam to the second lens of the fixed optical collimator. Zinc selenide (ZnSe) windows were placed in the beam path before and after the plasma shutter at 45°, which acted as approximately 97:3 beam splitters for in-situ measurement of the laser pulse energy and temporal profile. The beam energy was measured using calibrated energy monitors and the temporal profiles were recorded using the infrared photon drag detectors (described in Chapter 2) with rise-times of ∼1 ns. The jitter in timing measurements using these detectors is also ∼1 ns. The target set-up is maintained under vacuum with a chamber pressure of 5×10^{-6} mbar. As the ZnSe lenses used in the set-up have a focal length of 10 cm the peak CO\textsubscript{2} intensity is estimated to be 5×10^{9} W/cm\textsuperscript{2} at focus. This intensity was approximated for the spike by taking 0.45 J as the energy and 50 ns as the pulse duration, which were the
4.3. Plasma shutter experiments

values determined from the Gaussian fit in Fig. 4.3. This intensity is above the threshold of \( \sim 1 \times 10^9 \) W/cm\(^2\) required for air breakdown caused by the CO\(_2\) laser [14], therefore justifying that the plasma shutter be kept under vacuum throughout experiments.

![Plasma shutter set-up](image)

**Figure 4.4:** Plasma shutter set-up. Note: Not drawn to scale.

As the high intensity laser pulse is incident on the target surface a plasma will form. For high laser intensities (CO\(_2\) > \( 10^9 \) W/cm\(^2\)), a plasma is typically produced during the first 100 ps of the laser pulse. The remainder of the laser pulse is then strongly coupled into this plasma by inverse Bremsstrahlung, inducing significant heating and ionisation of the plasma. For dense low temperature plasmas, far infrared radiation is efficiently absorbed by inverse Bremsstrahlung (section 1.4) [15]. If the applied laser intensity is lowered to a value close to the ablation threshold, then plasma formation does not take place until later in the irradiation process. Therefore, before plasma formation occurs, the laser pulse can be efficiently reflected from the metal
4.3. Plasma shutter experiments

surface. Thus, the plasma formation time can be controlled by adjusting
the laser intensity falling on the target. This variation in the intensity was
achieved by moving the Au coated Si target very precisely with respect to the
laser focus, thereby increasing or reducing the laser spot size as the target
moves through the axis of laser focus.

4.3.2 Results of shortening on pulse duration and energy

Example of the reflected CO\textsubscript{2} laser pulses obtained are presented in Fig.
4.5, showing four reflected pulses, as well as the laser pulse measured before
shortening, for increasing laser intensity. In Fig. 4.5 (a), the applied laser
intensity is below the ablation threshold and the metal film acts as a mirror.
As the laser intensity is increased plasma is formed and efficiently absorbs the
4.3. Plasma shutter experiments

remaining part of the laser pulse, including the low energy long duration tail. This can be seen in Fig. 4.5 (b). Increasing the laser intensity still further results in earlier plasma formation and therefore a shorter duration reflected pulse as shown in Fig. 4.5(c). Eventually, the laser intensity becomes so high that very little of the incident pulse is reflected [Fig. 4.5(d)]. All spot sizes and hence laser intensity values are estimates, based on the knife edge experiment presented in Chapter 2. For a laser intensity below $2.5 \times 10^7$ W/cm$^2$ there is no pulse shortening, indicating that this value is close to the plasma formation threshold of the Au coated Si target. In the laser intensity range $2.5 \times 10^7$-1.0$ \times 10^8$ W/cm$^2$, the laser pulse is shortened from 50 to 10 ns. Increasing the intensity beyond this value shortens the pulse further down to a minimum value of approximately 2 ns at $1.8 \times 10^8$ W/cm$^2$.

A scan of the variation of pulse duration with incident laser intensity was performed by moving the target position relative to the focus of the beam collimator. Close to the laser focus a short pulse length is observed which increases in length the further from the focus that the target is located. The variation of pulse duration with laser intensity is shown next in Fig. 4.6.

![Figure 4.6: Variation of reflected pulse duration as a function of applied laser intensity.](image_url)

The variation of total reflected energy corresponding to the pulse durations given in Fig. 4.6 was also measured and is shown in Fig. 4.7. For incident laser intensities below the plasma formation threshold almost all of the in-
4.3. Plasma shutter experiments

Incident energy is reflected. As the laser intensity is increased, the reflected energy is reduced by plasma absorption. For an intensity of $3.4 \times 10^7 \text{ Wcm}^{-2}$ (Fig. 4.5 (b)), 270 mJ is reflected in a pulse of duration 22 ns. At $8.1 \times 10^7 \text{ Wcm}^{-2}$ [Fig. 4.5(c)], 63 mJ is reflected in 10 ns, while at the minimum pulse duration measured of 2.5 ns the energy reflected from the target was measured to be 35 mJ. For fixed conditions the shortened pulse duration and energy varies on a shot to shot basis but this is due to the variation of the initial input pulse from the TEA CO2 laser. The variation is ±10% for longer pulse durations (~10-50 ns) and less than ±20% for shorter pulse durations (<10 ns). The instantaneous power for each of these pulses is determined and also shown in Fig. 4.7. It is observed that the power is relatively stable around a value of 10 MW.

![Graph showing variation of reflected pulse energy and total reflected power as a function of applied laser intensity.](image)

Figure 4.7: Variation of reflected pulse energy and total reflected power as a function of applied laser intensity.
4.3. Plasma shutter experiments

4.3.3 Comparison between results and target surface heating model

To explore the mechanisms responsible a simple laser heating model [16] was used to explain the behavior of the plasma shutter. The model assumes heating of a semi-infinite, homogeneous, and isotropic metallic sample by a laser pulse with a rectangular (i.e., top-hat) temporal profile. Solving the heat conduction equation [16, 17] gives an analytic expression for the temperature distribution during the laser pulse, inside the metal target, $T(z,t)$ as

$$ T(z,t) = \left( \frac{2(1-R)I_0}{\rho C} \right) \sqrt{\frac{t}{\kappa}} \text{erfc} \left[ \frac{z}{\kappa t^{1/2}} \right] + T_i \quad (4.1) $$

In Eqn. 4.1, $I_0$ is the laser intensity, $R$ is the target reflectivity at the laser wavelength, $\rho$ is the density, $C$ is the specific heat capacity and $\kappa = (k/\rho C)$ is the thermal diffusivity ($k$ being the thermal conductivity). $T_i$ is the initial temperature of the system. The $\text{erfc}(x)$ integral error function is described in Ref. [17] and for small values of $x$ it can be given as [18]:

$$ \text{erfc}(x) = \frac{1}{\sqrt{\pi}} - \frac{x^2}{\sqrt{\pi}} - \frac{x^4}{6\sqrt{\pi}} + ... \quad (4.2) $$

At the surface (i.e., $z = 0$) $\text{erfc}(x)$ reduces to $\frac{1}{\sqrt{\pi}}$. Thus Eqn. 4.1 becomes:

$$ T(0,t) = \left( \frac{2(1-R)I_0}{\rho C} \right) \sqrt{\frac{t}{\pi \kappa}} + T_i \quad (4.3) $$

The above equation is valid for laser heating when the target remains in the solid state. When laser heating is sufficient to induce surface melting, Eqn. 4.3 becomes:

$$ T(0,t) = \left( \frac{2(1-R)I_0}{\rho C} \right) \sqrt{\frac{t}{\pi \kappa}} + T_i - T_q \quad (4.4) $$

where $T_q = q/C$ and $q$ is the latent heat of melting. The operation of plasma shutter induced pulse shortening depends on the plasma formation time during laser irradiation. If we consider that the plasma formation time is approximately equal to the time it takes for the surface temperature to reach the boiling point, then equation 4.4 can be used to predict the plasma formation time and thus the reflected pulse duration, as a function of laser laser intensity. Rearranging equation 4.4 gives:
4.3. Plasma shutter experiments

\[ t_B = \pi \kappa \left[ \left( \frac{\rho C}{2(1 - R)I_0} \right) (T_B - (T_i - T_q)) \right]^2 \]  (4.5)

where \( t_B \) is the time it takes for the surface temperature to reach the boiling point and \( T_B \) is the boiling temperature of the metal. Using Eqn. 4.5 above with the room temperature, bulk thermal properties of Au [19], \( t_B \) can be estimated as a function of laser intensity. This is shown in Fig. 4.8 and compared to the reflected pulse duration measurements. The thermal properties used in the calculation are shown in Table 4.1.

![Figure 4.8: Comparison of the measured reflected pulse durations (square symbols) and the calculated boiling times (dashed line) as a function of laser intensity.](image)

The calculated boiling times show the same general trend as observed for the measured pulse durations with increasing laser intensity. The measured pulse duration and calculated boiling time both drop from 50 ns to less than 10 ns with one order of magnitude increase in intensity. However, the laser intensity range where the calculated boiling time is comparable to the shortened pulse durations is approximately an order of magnitude larger for
4.3. Plasma shutter experiments

the calculated values. For the comparison shown in Fig. 4.8, the calculated values were shifted to lower intensity to overlap the experimental values. There are several reasons that higher calculated laser intensity values are required to reproduce the experimental trend. Hot spots are present in the beam and the experimental intensity is averaged over the fluctuations evident in Fig. 4.1. Taking these factors together leads to spatial and temporal instantaneous values of laser intensity significantly in excess of the average. Second, the laser heating model assumes a semi-infinite slab, whereas for this experiment Au thin films of \( \sim 500 \text{ nm} \) thickness deposited on Si were used as reflective targets. The heat penetration depth, \( l_{th} = (\kappa \tau_p)^{1/2} \) for the CO\(_2\) laser is about 2.5 \( \mu \text{m} \), taking the pulse duration as 50 ns. This is significantly larger than the Au layer thickness. As the thermal conductivity of Si is half that of Au, the heat conduction away from the target surface is larger in the model thus underestimating the surface temperature rise. Thirdly, the thermal conductivity of gold at 1400 \( ^\circ \text{C} \) is almost one third of that at room temperature, leading to a faster temperature rise than predicted by the model. The comparison also does not take account of the ionisation phase in the plasma formation that follows the boiling. Including the temperature dependence of the thermal parameters in the laser heating model is difficult and numerical solutions are required. The comparison of this simple laser heating model was not meant as a rigorous quantitative treatment but rather to examine the link between the laser heating dynamics and the reflected pulse duration. From the comparison it can be clearly seen that the reflected pulse duration is directly dependent on the plasma formation time which can be controlled by adjusting the applied laser intensity.

<table>
<thead>
<tr>
<th>Bulk room temperature properties of Au</th>
</tr>
</thead>
<tbody>
<tr>
<td>Specific heat (C) – 130 (J kg(^{-1}) K(^{-1}))</td>
</tr>
<tr>
<td>Thermal conductivity (k) – 315 (W m(^{-1}) K(^{-1}))</td>
</tr>
<tr>
<td>Thermal diffusivity ((\kappa)) – 1.28\times10^{-4} (m(^2) s(^{-1}))</td>
</tr>
<tr>
<td>R (\oplus) 10600 nm – 0.9</td>
</tr>
<tr>
<td>Latent heat of melting (q) – 6.3\times10^4 (J kg(^{-1}))</td>
</tr>
<tr>
<td>Density ((\rho)) – 19 300 (kg m(^{-3}))</td>
</tr>
<tr>
<td>Melting temperature ((T_m)) – 1337 (K)</td>
</tr>
<tr>
<td>Boiling temperature ((T_B)) – 3129 (K)</td>
</tr>
</tbody>
</table>
4.4. CO$_2$ laser pulse shortening using a pinhole plasma shutter

In this section, the effect of the CO$_2$ temporal pulse length on the emission of EUV photons from the plasma was observed by using a photodiode (PD) with a very fast temporal response. The photodiode used in experiments was the AXUVHS5 from IRD [20]. The PD has a 10–90% rise-time of 700 ps and a sensitivity area of 1 mm$^2$. The extremely quick response of the PD means that it should give good temporal information on the emission of photons from the plasma. This will also allow the emission to be compared to the temporal profile recorded of the CO$_2$ laser pulse. A schematic of the PD is shown in Fig. 4.9.

![Figure 4.9: Schematic of AXUVHS5 photodiode [20].](image)

The photodiode was placed inside the vacuum chamber approximately 15 cm away from the target and at an angle of 45° with respect to the direction of laser incidence and the target normal. The AXUVHS5 PD has quite a broad wavelength region of response, so in order to reduce the effects of out-of-band radiation on the detector a zirconium (Zr) filter was used. The Zr filter was 0.5 μm in thickness and its transmission as a function of wavelength is presented in Fig. 4.10. As a multilayer mirror was not used in the set-up, the signal recorded by the PD will therefore be from a broad range of
4.4. CO$_2$ laser pulse shortening using a pinhole plasma shutter

wavelengths (6–18 nm) and will not just be the 13.5 nm in-band emission. However, this can still be used to give information on the EUV emission from the plasma and its dependence on the CO$_2$ temporal profile.

![Graph showing transmission through a 500 nm Zr filter.](image)

**Figure 4.10:** Transmission through a 500 nm Zr filter.

The CO$_2$ pulse was shortened by using the pinhole plasma shutter [2]. The shutter was set-up in air outside the target chamber. It operates by first focussing the CO$_2$ laser pulse with a 20 cm focal length ZnSe plano-convex lens onto a pinhole. A piece of 1 mm thick aluminium, 2.54 cm in diameter was placed along the optical axis of the laser, very close to the position of focus i.e. 20 cm away. The pinhole was formed by drilling a 1.5 mm hole in the centre of the aluminium piece. An estimation of the laser intensity is made for the main spike of the pulse, which does not include the N$_2$ tail. Of the approximately 850 mJ of the output energy, the main spike contains about 340 mJ as it accounts for 35–40% of the total laser energy in about 50–70 ns. The minimum focal spot size $\omega(z)$ at a distance $z$ from the point of focus was determined by using the method set out in Chapter 2. The pinhole was set-up at a distance of 15 mm from the point of focus, which corresponds to a theoretical beam diameter of $\omega(z)=1.4$ mm and beam area of 0.015 cm$^2$. The laser pulse intensity of the pulse spike (i.e. not including the tail) at the pinhole was therefore estimated to be $5.5 \times 10^8$ W/cm$^2$. As
4.4. \textit{CO}_2\textit{ laser pulse shortening using a pinhole plasma shutter}

the intensity of the pulse is below the threshold of that required for the laser pulse to breakdown in air ($\sim 1 \times 10^9$ W/cm$^2$) \cite{14}, a plasma is instead formed around the periphery of the pinhole. Lateral expansion of the plasma will cause the remaining portion of the pulse to interact with the plasma formed instead of transmitting through the pinhole. This renders the pinhole opaque to the rest of laser and shortens the pulse length of the output laser beam. The position of the pinhole was set 5 mm from focus because at this position about 40–50 ns of the pulse was transmitted, which also contained about 250 mJ of energy. The diverging beam that is transmitted through the pinhole is then collimated by a second plano-convex ZnSe 20 cm focal length lens, placed 40 cm away from the focusing lens. A picture of the plasma pinhole shutter is shown Fig. 4.11.

![Figure 4.11: Image of the pinhole plasma shutter set-up.](image)

The fast PD was now used to record the time resolved EUV signals produced by the long and short CO$_2$ pulses as shown in Fig 4.12 and 4.13. The laser was focussed onto a planar solid Sn target at normal incidence using a 10 cm focal length ZnSe lens inside the chamber. Ten measurements were made with the PD for both pulses and all ten are plotted. First the unclipped long CO$_2$ pulse and PD signal are shown in Fig. 4.12. Following this Fig. 4.13 shows the short pulse temporal signal after clipping along with the PD signal. In both figures the CO$_2$ profile is offset vertically for clarity.
4.4. CO$_2$ laser pulse shortening using a pinhole plasma shutter

![Graphs showing CO$_2$ profile and EUV response over time.](image)

Figure 4.12: PD EUV response using the long CO$_2$ pulse duration.
4.4. CO$_2$ laser pulse shortening using a pinhole plasma shutter

Figure 4.12: PD EUV response using the long CO$_2$ pulse duration.

Figure 4.13: PD EUV response using the clipped CO$_2$ pulse duration
4.4. CO$_2$ laser pulse shortening using a pinhole plasma shutter

Figure 4.13: PD EUV response using the clipped CO$_2$ pulse duration.

Observing the PD response of both pulses it can be seen that there is a delay of approximately 10 ns between the peak intensity of the laser profile and the EUV signal. However, this is effected by lengths of cables or the distance between the two detectors (approx. 1 ns per 30 cm). There is also interference from the plasma with the PD signal which causes a negative going peak to form between the main spike and the tail, as seen in each figure. Taking this feature into account, the area underneath the PD response curve of both the long and short CO$_2$ pulses were determined. The area integrated
4.4. CO2 laser pulse shortening using a pinhole plasma shutter

under the voltage-time curve is proportional to the photo-generated charge \( Q \), which is given by:

\[
Q = \int V(t)dt
\]  

(4.6)

Where \( V \) refers to the voltage signal recorded on the \( y \)-axis of the oscilloscope and \( t \) refers to the time on the \( x \)-axis. Using this and knowing information such as the resistance of the scope and filter transmission, the total energy of the photons could be determined. However, because the Zr filter transmits photons over a wide range in the EUV region [Fig. 4.10], the absolute value of the area under the curve of the long and short pulses will be compared directly.

The area under the signal of the CO2 laser is first determined for ten shots of the long and short CO2 pulse profiles. The integration is carried out from \( t=0 \) ns until \( t=450 \) ns. By comparing the areas, averaged over ten shots, of both long and short pulses, it was found that the percentage area of the shortened laser pulse transmitted was 66.6% of the original laser pulse. Note that this was only carried out for the first 450 ns of the pulse. As the long lived \( \text{N}_2 \) tail exists for \( >2 \) \( \mu \)s, it is expected that this percentage would then be much lower (30–40%). An example of the short and long CO2 pulse profiles are shown in Fig 4.14, and from these plots the ten shot averaged full width at half maximum of the shortened pulses was found to be 34.1 ± 3.8 ns. Following this is a plot of the normalised areas under the long and short CO2 profiles for ten shots [Fig. 4.15]. The same procedure was carried out for the area under the PD signal. However the ten shot averaged area under the signal of the PD from the short CO2 pulse was 84.7% of the PD signal obtained from the long pulse. From this it can be inferred that excess amount of laser energy contained within the tail of the CO2 temporal profile is not actually contributing to efficient EUV emission from the plasma. The change in the area of the PD signal as a function of the change in area of the pulse duration gives a 0.85/0.67 = 1.3 times increase in efficiency for the short laser pulse.

Similar structure to that in the CO2 profile is observable in the diode response for the EUV emission as Figs. 4.12 and 4.13 show. This means the \( \sim 2 \) ns spikes of the CO2 laser profile act like a train of pulses instead of one uniform profile, which causes the plasma emission to rise and fall around what appears to be a threshold for EUV emission. Therefore a more uniform flat-top or Gaussian CO2 pulse profile could keep the plasma above this threshold level and result in more efficient emission.
4.4. CO₂ laser pulse shortening using a pinhole plasma shutter

Figure 4.14: Long and clipped CO₂ pulse temporal profiles corresponding to shots 1-4.

Figure 4.15: Normalised area under the temporal profiles recorded of the long and short CO₂ pulse.
4.4. CO$_2$ laser pulse shortening using a pinhole plasma shutter

4.4.1 Effect of CO$_2$ pulse shortening on EUV emission

The effect of reducing the pulse duration on the in-band emission was now investigated by observing the EUV spectra. The typical duration of the shortened CO$_2$ pulse was 34.1 ± 3.8 ns and the average energy contained in each pulse was 144.3 ± 21.2 mJ. Using theoretical estimates of 500 µm for the spot size 2 mm from focus, this corresponded to a peak on-target laser intensity of $4 \times 10^9$ W/cm$^2$. Using these shortened pulses, the 2% in-band CE at 13.5 nm was determined and compared with previous results using the long duration CO$_2$ pulse. The absolutely calibrated Jenoptik spectrograph was used to observe the spectrum at 45° with respect to the laser incidence and target normal. A sample spectrum is shown for the long pulse along with one produced with the short CO$_2$ pulse in Fig. 4.16.

![EUV spectra produced by long (above) and short (below) CO$_2$ pulses.](image)

Figure 4.16: EUV spectra produced by long (above) and short (below) CO$_2$ pulses.
4.4. CO$_2$ laser pulse shortening using a pinhole plasma shutter

From the spectra in Fig. 4.16 it is observed that there is less line emission at longer wavelength for the short CO$_2$ pulse than the long pulse. This longer wavelength line emission is due to transitions in the lower ion stages of Sn [15]. Thus an improvement in the spectral purity for the short pulse is observed.

The energy on-target contained in the long CO$_2$ pulse, including the N$_2$ tail, is between 0.9-1.2 J. The maximum in-band EUV energy produced in a solid angle of 2 $\pi$sr by these pulses, taken from the results in Chapter 3, was approximately 20 mJ, which corresponds to a maximum CE of $\sim$ 2% for the long pulse. By switching to the shorter duration pulses the peak in-band energy is observed to be 4.5 mJ for pulse energies between 150-300 mJ. This equates to a CE of about 3% using the 34.1 $\pm$ 3.8 ns CO$_2$ laser pulse. These results show the improved efficiency of coupling CO$_2$ laser energy by clipping the pulse length.

![Figure 4.17: The measured CE found by repeatedly forming a plasma with the shortened CO$_2$ pulse on the same target spot.](image)

The measured CE is shown in Fig. 4.17 for the short pulse by repeatedly forming the plasma on the same target spot for an estimated laser intensity of $4 \times 10^9$ W/cm$^2$. The first shot records a CE of 3% and as the plasma continues to form in a crater for each subsequent shot, the in-band emission increases initially and then plateaus. The first shot is a 1.5 times increase on the CE achieved with the long pulse, which is close to the estimate of
4.5. Summary and conclusions

1.3 made for the improvement in efficiency. These irradiation conditions resulted in a peak in-band energy of 7.5 mJ and an average CE of 4.5%. These results agree very well with the previous study carried out by Harilal et al. [21] who observed a CE of 2.7% for a single shot and an average CE of 4.5% by forming the plasma in a crater, for a reported laser intensity of $6 \times 10^9$ W/cm$^2$. In this work they determined the reason for the enhancement in emission to be from the restriction of the plasma expansion by the sides of the crater, which leads to confinement of the plasma. This in turn causes prolonged heating of the confined regions of the plasma to the optimum plasma temperature for more efficient emission [21]. The instability of the values could be due to a combination of the pulse to pulse instability of the laser and the reproducibility of the shortened pulses generated by the pinhole shutter.

4.5 Summary and conclusions

A repeatable and flexible technique for pulse shortening of laser pulses was applied to TEA CO$_2$ laser pulses. The reflected pulse duration was shown to be controlled by changing the on-target laser intensity, which is easily achieved by adjusting the focussing lens position. Using this plasma shuttering technique, pulse durations down to $\sim 2$ ns were demonstrated. The transmitted energy is also reduced as the pulse duration is decreased, but the reflected power remains stable at around 10 MW for all pulse durations. A laser heating model verified the pulse shortening dependence on the plasma formation time. The major drawback of this technique is that after each pulse shortening event, a new ablation site must be used as the reflective coating is destroyed at the point of laser incidence. Once the metal coating is removed, the target must be changed. For continuous operation of this shutter a regenerating, liquid metal-coated target could be used in place of the gold-coated silicon target. This technology has been developed by researchers in spectroscopy group at UCD [22]. This plasma shutter could thus be potentially used as a tool for pulse shaping in the search for laser pulse conditions to optimise CE from laser energy to useable EUV radiation for EUV source development. The effect of the CO$_2$ laser pulse duration on the CE of EUV emission was then demonstrated using a simple pinhole plasma shutter. This showed the advantage of removing the N$_2$ tail for improving the EUV emission. The variation in the results of CE can be ascribed to the instability in the energy of the CO$_2$ laser and how repeatable the pulse duration was by the clipping the pulse with the pinhole plasma shutter.
Bibliography


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Bibliography


Chapter 5

EUV emission from re-heating colliding plasma targets

5.1 Introduction

In this chapter results of the extreme ultraviolet (EUV) emission from the re-heating of colliding plasma targets are presented [1]. The theoretical background of colliding plasmas as well as their potential benefits as an EUV source were discussed in the first chapter of this thesis. However they can be briefly described as two counter propagating plasma plumes, which with a certain collisionality parameter ($\zeta$) [Eqn. 5.1], will result in the plasma material stagnating at their interface to produce a quasi-static plasma layer, referred to as a stagnation layer [2, 3].

$$\zeta = \frac{D}{\lambda_{ii}}$$ (5.1)

In Eqn. 5.1 $D$ is the separation between the two colliding plasmas and $\lambda_{ii}$ is the mean free path. If $\zeta$ is small, then the two plasmas will pass through each other without stagnating in a process called interpenetration [4]. This interaction is dependent on a number of parameters including the orientation of the plasmas, as well as irradiation conditions [5]. One key advantage of the stagnation layer as an EUV source is the ability to tailor the target density [6] for optimal absorption of the re-heating laser pulse. It has also been shown to be a stable target, by producing a consistent flat target during the life time of the stagnation layer, as observed from visible imaging in previous works [7, 8]. In these experiments, the stagnation layers of tin (Sn) plasmas are investigated as potential EUV sources for a very wide parameter range spanning both time and space.
5.2. Experimental set-up

The apparatus used in the colliding plasma experiments was almost the same as that used in experiments in Chapter 3. The procedure for externally triggering both lasers is as laid out in the “External triggering set-up for dual laser pulse experiments” section of Chapter 2. A change to the set-up however was made to the target used in the colliding plasma experiments from single pulse experiments. During experiments a circular Sn disk target was used instead of a flat Sn target. The reason for this was the orientation of the incoming laser pulses. The Nd:YAG laser irradiated the target along normal incidence and the incoming direction of the CO$_2$ pulse was at 90° to normal incidence. This meant the CO$_2$ pulse would be occluded by the target holder and flat slab of Sn itself before being able to interact with the plasma created by Nd:YAG laser. Therefore the flat target was replaced with a Sn disc target which could also be rotated on a small stepper motor to allow a fresh spot for each shot to prevent forming the plasma in a crater. The full experimental set-up is shown in Fig. 5.1.

![Figure 5.1: Schematic of colliding plasma set-up.](image-url)
5.2. Experimental set-up

The polarisation and energy of the Nd:YAG laser pulse were varied by use of a half-wave ($\lambda/2$) plate and a linear polariser at the Brewster angle. The output energies at various angles were recorded using the Litron energy detector. The output energy varied from 150 - 900 mJ and was plotted as a Sine function shown in Fig. 5.2.

![Figure 5.2: Variation in laser energy for changing $\theta$.](image)

The other main additions to the experimental set-up were a wedge prism placed in the Nd:YAG beam path, a visible camera used for alignment and a pinhole plasma shutter [9] used to clip the CO$_2$ pulse, which is described in Chapter 4. The wedge prism was placed in the beam path of the Nd:YAG laser just in front of the entrance window to the target chamber, as shown in Fig. 5.1. The prism was used to dissect the laser beam close to half way across its spatial profile, so that roughly half the laser energy will pass directly over it and the other half will pass through the prism. Depending on the orientation of the prism, the portion of the beam that transmits through the optic will be either refracted upwards or downwards. Two laser spots are thus created and can then be focussed using a lens and form what will be referred to as the “seeds” or “seed plasmas”. Depending on the alignment of the prism and how precisely it dissects the center of the beam, the two seeds can contain almost the same amount of energy. The seed plasmas are balanced by moving the wedge prism vertically upwards and downwards as shown in Fig. 5.3. The rotational orientation of the prism will also influence the positioning of the seeds. By rotating the prism, the portion
of the beam passing through it will be refracted at varying angles. With the correct rotation, it can be ensured that the two spots will be close to vertically aligned. This orientation was chosen so that both seeds could be clearly imaged by the visible camera observing their expansion horizontally from the target surface. The separation, $D$, between the two seeds can be determined from the following relation:

$$D = f \gamma (n - 1)$$  \hspace{1cm} (5.2)

where $n$ is the refractive index ($n = 1.5$), $\gamma$ is the acute angle of the wedge prism and $f$ is the focal length of the lens. In the experiments that will be discussed in this chapter the $\gamma$ of the prism was $2^\circ$ and the focal length of the lens used was 100 mm. This works out as a separation of 1.75 mm between the center of both the seed plasmas at focus. However the separation is seen to change as the lens target distance changes [10]. The lens position was fixed at 4 mm outside the focus point, which formed a separation of 1.3 mm used throughout the experiments.

Figure 5.3: Schematic of wedge prism used for forming colliding plasmas.
5.2. Experimental set-up

5.2.1 Flat target

As mentioned, the Sn slab target was replaced with a round Sn wheel target [Fig. 5.4]. In this chapter this round target will be referred to as a flat target because the two seed plasmas will form vertically on the same plane on the surface of the wheel which can be assumed to be flat on the scale of the plasma. The target consisted of three 4 mm thick Sn disks with an M6 hole drilled through their centre. The disks were held together by a nut that was tightened onto a screw passing through the hole which also fixed the target to a hollow brass mount. This could then be mounted on to the rotating axle of the stepper motor, which is slotted into the hollow of the brass mount. The target is held in place by a small grub screw in the side of the brass mount that will fix the target to the rotating axle of the stepper motor.

Figure 5.4: The flat target made up of three Sn discs stacked on top of each other.

5.2.1.1 Imaging the stagnation layer

The CCD used for observing visible emission from the colliding plasmas was set up as shown in Fig. 5.1. The camera used for imaging was a Princeton Instruments CCD which was connected by USB to a computer and operated via the WinSpec software. It was orientated approximately 90° with respect to the target surface normal. In initial alignment tests, a small torch was
5.2. Experimental set-up

used to illuminate the target chamber and the Sn wheel target. The two lenses in front of the CCD were adjusted in order to form a collimated sharp image of the front of the target with almost 1 to 1 magnification. Next a ruler was placed beside the target surface in order to determine what the pixel to length ratio is for each image so as to determine the scale of images produced. These two images are shown in Fig. 5.5 (a) and (b) below.

![Image](a)

![Image](b)

Figure 5.5: Image of (a) Sn wheel target and (b) the wheel with a ruler showing the scale of the image.

Another addition to the experimental set-up was the inclusion of a knife edge mounted onto a linear motorised actuator and stage. This was used to aid in the visible imaging of the stagnation layer. While observing the whole plasma the seeds can be clearly seen, which could saturate the CCD. In order to isolate only the stagnation layer for imaging, the knife edge was moved across the plasma in 0.1 mm steps until the seeds had been fully occluded and only the image of the stagnation layer remained. A demonstration of this process is shown on the next page in the four images in Fig. 5.6, as the knife edge is moved further along the plasma. Note false colouring has been used on the imaging for clarity. In the optical path a 450 +/- 50 nm filter was used to reduce the intensity of emission. While imaging the seeds, neutral density filters were also used in the path of the CCD in order to view them without saturating the camera.
5.2. Experimental set-up

Figure 5.6: Adjusting the position of the knife edge to reduce the seed intensity, allowing the stagnation layer to be observed.

An important application of the imaging was also to ensure that the two seeds of the Nd:YAG pulse were equal in size and brightness. This was important as it then produced a more balanced and stable stagnation layer as could be observed from the images. By adjusting the vertical position of the wedge prism up and down through the path of the Nd:YAG laser pulse, the energy distribution in the seeds could be varied as demonstrated on the next page [Fig. 5.7]. In the final figure [Fig. 5.7(d)] the seeds have been balanced and are equivalent in size and intensity, thereby creating a flatter stagnation layer.
5.2. Experimental set-up

Figure 5.7: Varying stagnation layer direction by adjusting the seed balance with wedge prism.

5.2.1.2 Colliding plasma re-heat alignment on flat target

The other main use of the visible imaging was to align the incoming CO\textsubscript{2} pulse spatially with the stagnation layer. This could be used to determine if interaction between the laser pulse and plasma was actually taking place. Alignment lasers were used to direct the beams as closely together as possible, however for much finer spatial alignment the CCD was used. To help carry out the process of alignment a thin (\sim 650 \, \mu m) straight piece of metal wire was attached to the top of the Sn disk target before pumping down the chamber. The Nd:YAG laser was first fired alone creating a stagnation layer from the colliding plasma forming on the target surface. The image of the stagnation layer is recorded in Fig. 5.8.
5.2. Experimental set-up

![Figure 5.8: Visible image of the stagnation layer formed by the Nd:YAG laser](image)

Figure 5.8: Visible image of the stagnation layer formed by the Nd:YAG laser

![Figure 5.9: Image of (a) the wire and (b) the wire irradiated with the CO\textsubscript{2} pulse used for alignment.](image)

Figure 5.9: Image of (a) the wire and (b) the wire irradiated with the CO\textsubscript{2} pulse used for alignment.

Next a small torch was used to illuminate the chamber and observe the wire that is placed above the target surface. Using the linear actuators, which control the three dimensional motion of the target, the wire was positioned to the same $y$ (vertical) coordinate on screen as the stagnation layer was formed. Next the CO\textsubscript{2} laser was fired into the chamber creating a plasma on the surface of the wire. The height of the incoming CO\textsubscript{2} laser was then adjusted vertically in order to fine tune its alignment with the wire. This is demonstrated in Fig. 5.9 which shows an image of the wire [Fig. 5.9 (a)] and then the wire with the plasma formed on its surface by the CO\textsubscript{2} laser [Fig. 5.9 (b)]. They are assumed to be aligned when the wire is approximately 136
5.2. Experimental set-up

bisecting the plasma in the images, which is limited by the minimum step (0.05 mm) of the vertical \( y \) actuator. Therefore the position of the stagnation layer and the centre of the incoming CO\(_2\) laser pulse should be close to vertically aligned.

![Image of the re-heating of the stagnation layer with the CO\(_2\) laser.](image)

The final alignment check was then to reposition the \( y \) actuator so that the stagnation layer is once again being formed on the target surface. This is now re-heated by the CO\(_2\) laser and the image is observed. By studying the visible imaging alone it can be determined that there has in fact been interaction between the stagnation layer and the pump laser as can be seen from the increase in visible emission between the re-heat shown in Fig. 5.10 and the stagnation layer shown alone in Fig. 5.8. This whole process is demonstrated in Fig. 5.11 (a), (b) and (c) on the next page. In Fig. 5.11 (a) an image is recorded of the stagnation layer, in (b) the \( x \), \( y \) and \( z \) actuators are moved to locate the wire in the same position as the stagnation layer. This is then irradiated with the CO\(_2\) pulse to check its alignment. Finally in (c), the target is returned to its original position and the stagnation layer is re-heated by the CO\(_2\) pulse.
5.2. Experimental set-up

The spatial alignment of the CO\textsubscript{2} laser was therefore complete and the time delay between the pulses was also controlled. The delay between the two pulses was measured by observing the CO\textsubscript{2} and Nd:YAG laser pulse temporal profiles on a digital oscilloscope. The CO\textsubscript{2} pulse was observed using the infrared temporal profiler and the Nd:YAG was detected with a photodiode, placed behind the turning mirror that directs the laser pulse into the vacuum chamber. Using this method the time delay between both lasers using the Stanford delay generator could be quantified. The time delay between the stagnation layer and the pump laser is an important variable in the experiment and its effect on the emission could be observed by recording visible images of the re-heating for various delays as shown in the series of images in Fig. 5.12. A negative time delay means that the CO\textsubscript{2} laser pulse arrives at the target before the seed plasmas are formed.
5.2. Experimental set-up

Figure 5.12: Visible imaging of the stagnation layer re-heated by the pump laser for various delays between the two laser pulses.
5.2. Experimental set-up

Figure 5.12: Visible imaging of the stagnation layer re-heated by the pump laser for various delays between the two laser pulses.
5.2. Experimental set-up

5.2.2 Wedge target

After experiments had been carried out with the flat Sn disks, the target was switched to a wedge target. The wedge target was made by cutting a 90° angle into the surface of each of three Sn disks that make up the wheel. It is thought that by forming the seed plasmas in such a way would cause greater interaction due to a larger component of the plasmas moving towards each other. This would potentially lead to a larger region of interaction and hence stagnation layer volume, which would mean a larger target for the pump laser pulse to interact with when re-heating. The target that was used, with its three 90° wedges, is shown in Fig. 5.13.

Figure 5.13: The wedge target made up of three Sn discs with a 90° wedge cut into the face of each piece.

5.2.2.1 Imaging the stagnation layer

The alignment process for the wedge target was almost identical to that of the flat target. The wire placed on top of the Sn disks was used to align the position of the stagnation layer and the position of the CO\textsubscript{2} laser pulse. The only difference to the procedure was the vertical positioning of the Sn target. When using the flat target the vertical (y actuator) position was not important as the seed plasmas would be formed in the same way at any point along the flat surface. However this vertical motion of the target was more critical to the alignment and stability of the stagnation layer on the wedge. There is a point on the wedge where the seed plasmas will be formed
5.2. Experimental set-up

along the same vertical plane. Producing the seed plasmas at this point will ensure a balanced and flat stagnation layer. The visible imaging was once again used to align the seeds. As the emission from the seeds was enough to saturate the camera the \( Q \)-switch delay on the Nd:YAG laser was sent out to 485 \( \mu s \) to reduce the laser energy to only a few mJ. The flashlamps were run on internal triggering mode and the “single shot in” mode was used to activate the \( Q \)-switch. The camera was set to acquire an image and then the laser was fired. With each image the alignment was checked and the vertical position was adjusted until the two seeds appeared vertically aligned. Fig. 5.14 shows the varying positioning of the two seeds as the \( y \) actuator is moved in a minimum of 0.05 mm steps until they are balanced [Fig. 5.14(d)].

![Figure 5.14](image)

Figure 5.14: Vertical adjustment of the wedge target in order to vertically align the position of both seeds.

Some images recorded of the wedge and plasma during alignment are shown in Fig. 5.15. A small light was again used to illuminate the chamber in order to observe the target surface [Fig. 5.15 (a)]. The seed plasmas and stagnation layer were also imaged [Fig. 5.15 (b)] and by combining the image with that
5.2. Experimental set-up

of the wedge target using the Spectra Math application of WinSpec, the position of the seeds on the target could be confirmed [Fig. 5.15(c)].

Figure 5.15: Visible imaging used to align the colliding plasma with a 90° wedge target.

5.2.2.2 Colliding plasma re-heat alignment on wedge target

The CO$_2$ position was checked again using the wire as previously carried out for the flat target. The stagnation layer was then re-heated by the CO$_2$ laser for the final step in alignment of the wedge target set-up. Several time integrated visible images of the re-heating process using the wedge target are presented next in Fig. 5.16, showing visible enhancement of the plasma. Beginning with the stagnation layer produced by the Nd:YAG pulse only and then re-heating with the CO$_2$ for various time delays from the CO$_2$ arriving 100 ns before the seed plasmas up to 500 ns after.
5.2. Experimental set-up

Figure 5.16: Visible imaging of the stagnation layer re-heated by the pump laser for various delays between the two laser pulses.
5.2. Experimental set-up

Figure 5.16: Visible imaging of the stagnation layer re-heated by the pump laser for various delays between the two laser pulses.
5.3. Impact of CO$_2$ re-heating on EUV emission

In this section the results of the study of the EUV emission from a re-heated stagnation layer will be presented. The parameter space that could be varied was very large for this experiment. With the vertical alignment of the CO$_2$ with the stagnation layer complete, various parameter scans could thus be carried out in order to optimise the emission. Linear motorised actuators connected to translation stages inside the chamber were used to vary the $x$, $y$ and $z$ position of the Sn disk target, the Nd:YAG and CO$_2$ lens focus positions and the knife edge position described in the alignment process. Microsoft Visual Basic was used to interface with the actuators, which were all connected in the chamber via a daisy chain. The control panel on the lab PC created to operate the actuators in this experiment is shown in Fig. 5.17.

![Control panel](image)

**Figure 5.17:** Screen shot of the panel used to control the motion of the target, lenses and knife edge.

Three actuators with maximum extended length of 60 mm and three with a maximum length of 28 mm were used. The minimum step size of all the actuators was set to 0.1 mm, therefore meaning the limit of movement for the 60 mm was 600 steps and 280 steps for the 28 mm actuators. Additional experimental information could be entered into the text boxes such as the angle set for the $\lambda/2$ plate relating to the Nd:YAG energy, the time delay between the Nd:YAG and CO$_2$ (if applicable) and comments on the experiment, which were useful if checking back over old results. There is also an
5.3. Impact of CO$_2$ re-heating on EUV emission

“Energy” button which allowed the actuator programme to interface with the S-Link controller of the Gen-Tec energy head in the same way as it was used previously in the knife edge experiment described in Chapter 2. A new dialog box was opened as shown in the screen shot in Fig. 5.18. In this control panel the wavelength of the detector and energy scale could be set. Then it could be set to continuous polling mode which read in the energy value from the energy head each time it was triggered. On the main panel screen in the CO$_2$ energy text box the energy was thus displayed each time the laser was fired. After each spectrum or image had been recorded the print button was pressed, which writes a line to a text file with all of the information contained in each text box for that particular data point. When large amounts of data files are being analysed this improves the efficiency of the process as all of the experimental data can be associated with a line in the text file.

Figure 5.18: Screen shot of the panel used to communicate with the energy head and record CO$_2$ energy for every shot.

The main diagnostic tool for this experiment was the EUV spectrograph from Jenoptik. Throughout the colliding plasma experiments the spectrometer was positioned at $45^\circ$ to the target normal as in Fig. 5.1. It was important to align the spectrometer so that it was observing the emission from the point on the stagnation layer that would be pumped by the CO$_2$ laser. The wire
5.3. Impact of CO$_2$ re-heating on EUV emission

was once again used in the alignment process. The actuators positioned it again in the same position as the stagnation layer would form and was then irradiated by the CO$_2$ laser. The spectrometer was thus aligned to this point.

5.3.1 Flat target

5.3.1.1 Optimisation of experimental parameters

The first parameter that was optimised was the focal position of the Nd:YAG lens. The actuator controlling the lens for focussing the Nd:YAG laser pulse was moved in 2 mm steps and at each position a spectrum was recorded from the plasma produced by the colliding plasmas only. The $\lambda/2$ plate was set at 30$^\circ$ which corresponded to an on-target energy of $491.9 \pm 8.2$ mJ contained in the two seeds and an estimated laser intensity of $2.7 \times 10^{11}$ W/cm$^2$ for each seed. The in-band energy was calculated using the procedure in Chapter 2 and an estimation of the conversion efficiency (CE) was made. For each shot a fresh piece of target was used by rotating the motor. In Fig. 5.19 the result of the lens scan is shown. From this scan, the actuator position for the lens was chosen from the point of peak CE and was thus set as this for the rest of the experiment, i.e. 4 mm from focus, corresponding to a 1.3 mm seed separation.

![Figure 5.19: Lens scan of the seed plasmas produced by the Nd:YAG laser.](image)

10 shots were then taken on a fresh target each time at this lens position to determine the stability of emission. The average CE was determined to be 2%
5.3. Impact of CO\textsubscript{2} re-heating on EUV emission

which compares well with previous works [11–13] using similar parameters for a single laser pulse experiment without the colliding plasmas.

The spectra produced by the Nd:YAG only and by re-heating the stagnation layer with the CO\textsubscript{2} laser are shown in Fig. 5.20, both spectra shown were three shot averages. An enhancement in the intensity of the emission around 13.5 nm is observed, meaning the interaction of the CO\textsubscript{2} pulse with the plasma target is causing a greater emission of EUV photons than the colliding plasmas alone. The most observable feature in the Nd:YAG spectrum is the dip around 13.5 nm mostly likely caused by self absorption effects in the plasma [11]. However the similarity between the spectra indicate that the emission is being dominated by the Nd:YAG laser plasma and the CO\textsubscript{2} re-heat is not having much effect on the emission.

![Spectra produced by colliding plasmas of Nd:YAG only and by re-heating the colliding plasmas with the CO\textsubscript{2} laser.](image)

Figure 5.20: Spectra produced by colliding plasmas of Nd:YAG only and by re-heating the colliding plasmas with the CO\textsubscript{2} laser.
5.3. Impact of CO\textsubscript{2} re-heating on EUV emission

Since the interaction of the CO\textsubscript{2} pulse with the stagnation layer was shown to increase the emission of EUV, the next parameter that was to be varied was the CO\textsubscript{2} lens position, which determines the laser intensity on-target (\(\phi\)) and hence the plasma temperature (\(T\)) [Eqn. 5.3] [14], thus leading to greater ionisation of the plasma.

\[
T (eV) \propto \lambda^2 \phi (W/cm^2)^{3/2}
\]

The actuator controlling the CO\textsubscript{2} lens was moved in 2 mm steps and at each position three spectra were recorded. The CO\textsubscript{2} energy on-target for each shot was approximately 200 mJ in 34.1 \(\pm\) 3.8 ns by using the pinhole plasma shutter (4\(\times\)10\textsuperscript{9} W/cm\textsuperscript{2}), however exact energy measurements were also made for each shot and are used in CE calculations. The focussing conditions that corresponded to maximum emission were found and the lens was thus fixed at this position.

With the focal positions of both lenses now fixed the next parameter scan could be carried out. The \(z\) actuator was now moved forwards and backwards so that target was moved left to right with respect to the incoming CO\textsubscript{2} laser pulse. This meant that a study of the effects of the CO\textsubscript{2} pulse re-heating at various points along the length of the stagnation layer could thus be carried out. To begin, the CO\textsubscript{2} laser alone was fired into the target chamber while the target surface was slowly stepped in towards it. This was observed using the visible imaging. When the target surface was moved into the path of the incoming CO\textsubscript{2} pulse a small plasma was produced. Then the \(z\) actuator was stepped forward 200 \(\mu\)m which is approximately half the CO\textsubscript{2} focussed beam diameter. The \(z\) actuator position at this point was noted and was taken as the CO\textsubscript{2} interacting with the target surface at 0 mm. The Nd:YAG laser was fired again to create a stagnation layer for the CO\textsubscript{2} laser to irradiate. The \(z\) actuator was moved back and three spectra were recorded at various positions out to 2 mm from the target surface, at which point the CO\textsubscript{2} appeared to no longer be interacting with the stagnation layer.

A fixed delay of 80 ns was kept between the two lasers for the scan, with the Nd:YAG arriving before the CO\textsubscript{2} pulse. Spectra of the Nd:YAG only and CO\textsubscript{2} only were also recorded for each \(z\) actuator position. Spectra recorded of the CO\textsubscript{2} laser pulse by itself were taken to ensure that all emission was resulting from re-heating the stagnation layer and not the solid Sn target surface. As the \(z\) actuator was stepped backwards the actuator controlling the position of Nd:YAG lens was stepped forward at the same rate to keep the focussing conditions of the colliding plasmas constant throughout. The result of the scan of the \(z\) actuator is shown in Fig. 5.21 for a fixed time.
5.3. Impact of CO$_2$ re-heating on EUV emission

delay of 80 ns.

![Figure 5.21: Re-heating along the length of the stagnation layer. Showing the positions (a), (b), (c) and (d) along the stagnation layer that time scans were carried out.](image)

From the plot it is observed that region of greatest interaction between the plasma target and the pump laser is close to the target surface ($z<0.5$ mm). This is possibly due to an optimum density profile forming close to the target where the seed interaction is greatest. This region then strongly absorbs the CO$_2$ laser pulse via electron-ion collisions, in the inverse Bremsstrahlung process [15]. The size of the plasma target is also greater at this point as the laser can interact with both the seeds and the stagnation layer. During various times in the lifetime of the laser pulse, the densities at the target will be such that the plasma will become opaque to the CO$_2$ laser radiation, i.e. the critical density is reached [16]. As the plasma expands, the absorption front between the higher and lower density regions moves further outwards from the target surface. This effect is supported by observing the EUV energy emitted when the stagnation layer is re-heated for longer time delays between the seeds and the pump laser. At the longer time delays the peak emission is observed to be at further $z$ distances from the target surface. These results are presented next in “Time delay scans”.

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5.3. Impact of CO\textsubscript{2} re-heating on EUV emission

5.3.1.2 Time delay scans

The regions of the stagnation layer that the pump laser was interacting with were now better understood from the results of changing the \( z \) actuator position. Next the effect of the varying time delay between the two laser pulses was investigated. In order to determine the influence of time delay on the EUV emission, four positions from the \( z \) scan in Fig. 5.21 were chosen. These positions refer to distances approximately (a) 0.25 mm, (b) 0.45 mm, (c) 0.85 mm and (d) 1.65 mm from the target surface.

![Figure 5.22: Conversion efficiency of re-heating stagnation layer for different delays between seed and pump lasers, at points (a), (b), (c) and (d) along the stagnation length.](image)

At each of these positions a time scan was carried out. The delay on the output of the Stanford delay generator that externally triggered the CO\textsubscript{2} laser was varied from -20 ns to 300 ns with respect to the Nd:YAG laser pulse. The delay was changed in steps of 20 ns and at each timing step three spectra were obtained. The results of the four time scans are presented in Fig. 5.22. The jitter of approximately 10 ns recorded for the laser timing resulted in the uncertainty in the timing measurements of ± 5 ns. The peak
5.3. Impact of CO$_2$ re-heating on EUV emission

EUV emission of around 2% occurred around 0.25–0.45 mm from the target and for a time delay of 20-80 ns.

5.3.2 Wedge target

The following results are those obtained using the 90° Sn wedge target. These experiments followed the same procedure as those for the flat target previously reported but were carried out for three Nd:YAG laser energies for both seeds combined: 491.9 ± 8.2 mJ, 269.1 ± 4.5 mJ and 134.7 ± 4.0 mJ. These energies corresponded to estimated laser intensities of $2.7\times10^{11}$ W/cm$^2$, $1.5\times10^{11}$ W/cm$^2$ and $7.4\times10^{10}$ W/cm$^2$ for the two seeds respectively. These are only estimated values and most likely over estimate the actual laser intensity.

5.3.2.1 Optimisation of experimental parameters: position and time

As the target had been changed from the flat disks to the wedge, lens scans of both the Nd:YAG and CO$_2$ lasers were again carried out to identify the optimum focus positions. A $z$ scan of the pump laser through the length of the stagnation layer was then produced for the three laser energies. Once again this was taken at a fixed time delay of 80 ns and three spectra were recorded for each shot as well as an Nd:YAG and CO$_2$ only spectrum. This is shown in Fig. 5.23 for an Nd:YAG energy of 134.7 ± 4.0 mJ ($3.09\times10^{10}$ W/cm$^2$), Fig. 5.25 for an energy of 269.1 ± 4.5 mJ ($6.20\times10^{10}$ W/cm$^2$) and finally the $z$ scan for an Nd:YAG on-target energy of 491.9 ± 8.2 mJ ($1.15\times10^{11}$ W/cm$^2$) is shown in Fig. 5.27. The results of CE % as a function of time delay between the two laser is also shown for each of the three Nd:YAG energy inputs for the wedge target. As before the positions of the re-heat along the stagnation length are shown before each time scan plots.
5.3. Impact of CO$_2$ re-heating on EUV emission

Figure 5.23: Positions along the stagnation layer that time scans were carried out for an Nd:YAG energy of $134.7 \pm 4.0$ mJ and $\phi = 7.4\times10^{10}$ W/cm$^2$.

Figure 5.24: CE of re-heating stagnation layer for different delays between seed and pump lasers, at points (a) and (b) along the stagnation length.
5.3. Impact of CO$_2$ re-heating on EUV emission

Figure 5.25: Positions along the stagnation layer that time scans were carried out for an Nd:YAG energy of 269.1 ± 4.5 mJ and $\phi = 1.5 \times 10^{11}$ W/cm$^2$.

Figure 5.26: CE of re-heating stagnation layer for different delays between seed and pump lasers, at points (a), (b), (c) and (d) along the stagnation length.
5.3. Impact of CO$_2$ re-heating on EUV emission

Figure 5.27: Positions along the stagnation layer that time scans were carried out for an Nd:YAG energy of $491.9 \pm 8.2$ mJ and $\phi = 2.7 \times 10^{11}$ W/cm$^2$.

Figure 5.28: CE of re-heating stagnation layer for different delays between seed and pump lasers, at points (a), (b), (c) and (d) along the stagnation length.
5.3. Impact of CO$_2$ re-heating on EUV emission

The maximum observed CE was 3.5% for the Nd:YAG input energy of 269.1 ± 4.5 mJ with a delay of 60 ns between the seed plasmas and the pump laser. Below are the three shot averaged spectra recorded of the re-heat for 0 ns, 60 ns, 100 ns and 200 ns time delays of this Nd:YAG energy, which refer to position (a) of the z scan in Fig. 5.25. Along with each re-heated spectrum is a three shot averaged spectrum of the Nd:YAG only for the same set-up parameters. The enhancement in the in-band emission is clearly observable in Fig. 5.29. It is again noted that no spectra were observable when the CO$_2$ laser was fired by itself.

Figure 5.29: Three shot averaged spectra of the Nd:YAG laser only and the CO$_2$ laser re-heating the seed plasmas for 0 ns, 60 ns, 100 ns and 200 ns time delays.
5.3. Impact of CO$_2$ re-heating on EUV emission

5.3.3 Horizontal seed plasmas

5.3.3.1 Target and wedge prism orientation

The final experiment in this chapter was to observe if there is a change in the emission from the re-heated stagnation layer if the seeds are formed horizontally to each other, as opposed to vertically, as was the case in the earlier study in this chapter. The target set-up was changed by rotating the Sn wheel target by 90° with respect to its previous orientation, as shown in Fig. 5.30. The next change in the set-up involved rotating the position of wedge prism, also by 90°, so that it would now split the Nd:YAG laser beam along its vertical diameter [Fig. 5.31], thereby creating two seed plasmas in a horizontal plane. The alignment process described previously was repeated for the new set-up. However, for imaging purposes the stagnation layer could not be isolated from the seeds using the knife edge, as the knife edge would also have occluded the stagnation layer. Therefore, use of the visible camera was more limited than in the previous studies. Images of the plasma after it was re-heated by the CO$_2$ could still be recorded and from these visible images the effect of the CO$_2$ re-heat was still observed.

Figure 5.30: Set-up of target for experiment with horizontal seeds.
5.3. Impact of CO\textsubscript{2} re-heating on EUV emission

Figure 5.31: Set-up of wedge prism for experiment with horizontal seeds.

5.3.3.2 Optimisation of experimental parameters: position and time

The same optimisation of the experimental parameter space was carried out for the horizontal seeds set-up. After determining the optimum focal positions of both lasers, the CO\textsubscript{2} laser was focussed into the stagnation layer at various points along its length. This z scan was produced for the combined seeds lower energy 269.1 ± 4.5 mJ Nd:YAG input in Fig. 5.32 and the higher energy 491.9 ± 8.2 mJ input in Fig. 5.34. The time delay scans were then produced for both the lower and higher Nd:YAG energy inputs. It was observed that the length of interaction of the CO\textsubscript{2} with the stagnation layer was a lot shorter for the flat target compared to the wedge target. On the flat target, for an Nd:YAG energy of 269.1 ± 4.5 mJ, the interaction length was around 0.5 mm for the horizontal seeds compared to approximately 2 mm using the same energy with the wedge target. For the higher energy input the interaction length was 1.5 mm for both horizontal and vertical seeds but was close to 3 mm using the wedge target. Therefore time delay scans at fewer positions along the stagnation layer are now shown. The positions along the stagnation layer in Fig. 5.32 and Fig. 5.34 are followed by results of these time delay scans in Fig. 5.33 and Fig. 5.35 for lower and higher Nd:YAG energies, respectively.
5.3. Impact of CO$_2$ re-heating on EUV emission

![Graph showing CE % per 2π sr vs. Distance from centre of wedge (mm).](image)

Figure 5.32: Re-heating along the length of the stagnation layer, Nd:YAG energy = 269.1 $\pm$ 4.5 mJ and $\phi = 1.5 \times 10^{11}$ W/cm$^2$.

![Graph showing CE % per 2π sr vs. Time delay (ns).](image)

Figure 5.33: Conversion efficiency of re-heating stagnation layer for different delays between seed and pump lasers, at point (a) along the stagnation length.
5.3. Impact of CO$_2$ re-heating on EUV emission

Figure 5.34: Re-heating along the length of the stagnation layer, Nd:YAG energy = 491.9 ± 8.2 mJ and $\phi = 2.7 \times 10^{11}$

Figure 5.35: Conversion efficiency of re-heating stagnation layer for different delays between seed and pump lasers, at points (a) and (b) along the stagnation length.
5.4. Summary and conclusions

5.4 Summary and conclusions

The effects of re-heating a colliding plasma target with a CO\(_2\) laser at an 90° angle of incidence were investigated in this chapter. EUV spectra were recorded for a very wide parameter space. The optimum coupling occurred close to the target surface (z<0.5 mm) for time delays between 60–100 ns. Work from the PhD thesis of P. Hough has previously shown from optical images with a time gated camera, that a stagnation layer produced on a flat Al target, was fully formed after approximately 55 ns, while the emission from the seeds was minimal [2].

Initial tests on a flat target showed an enhancement of EUV emission, demonstrating its interaction with the plasma target. However it was found not to be an efficient process as the CE calculated, which included the energy of both lasers, was 2%. This corresponded to 12–13 mJ of in-band energy generated by both lasers. It must be noted that only the maximum input Nd:YAG laser energy was used in tests with the flat target and therefore the density of the plasma target may not have been optimised. Also, the Nd:YAG laser lens position was not varied but instead fixed throughout experiments for a separation of 1.3 mm between the seeds. The inefficiency of the process could be explained by the target geometry. The flat surface means that, as the plasma will expand normal to the target surface, the two plumes of the seed plasmas are not been directed towards each other but instead the majority of the expansion is the axial direction perpendicular to the surface [17]. This will result in a smaller area of collision between the seeds and hence a smaller target will be generated for the pump laser to re-heat. There is also overheating of the seed plasma caused by the Nd:YAG due to the high laser intensity of 1x10\(^{11}\) W/cm\(^2\), which could make coupling with the pump laser less efficient. This is supported by the self-absorption effects seen in the spectra of the Nd:YAG laser only for this intensity in Fig. 5.20.

A greater improvement in CE was then observed by using the 90° wedge target. A maximum average CE of 3.5% was recorded by a colliding plasma formed with an estimated Nd:YAG laser intensity of 1.5x11\(^{10}\) W/cm\(^2\) and re-heated by a CO\(_2\) pulse with typically ~ 250 mJ in 34.1 ± 3.8 ns or \(\phi = 4x10^9\) W/cm\(^2\). The improvement could now be due to the two seed plumes being directed towards each other, as the bulk of the plasma outflow is in the axial direction [17]. There may also be some confinement effects caused by the wedge that prevents the plasma expansion thereby maintaining temperatures and densities, similarly to experiments with planar Sn groove targets [18]. A 3-D map of the results is shown for the CE as a function of time delay and distance from the target in Fig. 5.36.
5.4. Summary and conclusions

The maximum value compares well with a previous study using a conventional double pulse scheme by Fujioka et al. [19], where a maximum CE of 4% was achieved using a mass limited 20 µm droplet target. If the Nd:YAG energy is excluded and the stagnation layer is treated as target to be directly compared to solid Sn, a CE of 4.9% is calculated, with a peak in-band energy of 13.18 mJ recorded. Assuming all of this energy can be collected and brought to the point of intermediate focus (IF), for a requirement of 180 W at IF [20] this amount of output energy would require a 14 KHz repetition rate source. A summary of the 3–5 shot averaged results achieved in this section is compiled in Table 5.1, along with results for the CO₂ only, Nd:YAG only and double pulse experiments from Chapter 3. DP w/o Nd:YAG and CP Wedge w/o Nd:YAG, in Table 5.1 were calculated by excluding both the Nd:YAG laser energy and the amount of in-band energy the Nd:YAG generates by itself. Therefore the efficiency of the plasma and stagnation layer are being compared directly to the solid Sn as a target for EUV emission.

This set-up could be further optimised as the approximate 500 µm diameter of the CO₂ pulse does not match well with the actual size of the stagnation
5.4. Summary and conclusions

<table>
<thead>
<tr>
<th>Laser set-up</th>
<th>Energy (J)</th>
<th>IBE (mJ)</th>
<th>CE (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nd:YAG only</td>
<td>0.491 ± 0.008</td>
<td>14.29 ± 0.48</td>
<td>2.24 ± 0.07</td>
</tr>
<tr>
<td>Long CO₂ only</td>
<td>1.070 ± 0.070</td>
<td>20.82 ± 1.31</td>
<td>1.95 ± 0.19</td>
</tr>
<tr>
<td>Short CO₂ only</td>
<td>0.148 ± 0.003</td>
<td>4.69 ± 0.23</td>
<td>3.18 ± 0.22</td>
</tr>
<tr>
<td>Double pulse (DP)</td>
<td>0.774 ± 0.010</td>
<td>19.73 ± 0.72</td>
<td>2.55 ± 0.06</td>
</tr>
<tr>
<td>DP w/o Nd:YAG</td>
<td>0.138 ± 0.012</td>
<td>5.44 ± 0.19</td>
<td>3.94 ± 0.12</td>
</tr>
<tr>
<td>CP Flat</td>
<td>0.636 ± 0.017</td>
<td>12.7 ± 0.47</td>
<td>2 ± 0.1</td>
</tr>
<tr>
<td>CP Wedge</td>
<td>0.507 ± 0.027</td>
<td>17.06 ± 1.16</td>
<td>3.33 ± 0.16</td>
</tr>
<tr>
<td>CP Wedge w/o Nd:YAG</td>
<td>0.238 ± 0.03</td>
<td>11.54 ± 0.02</td>
<td>4.85 ± 0.1</td>
</tr>
</tbody>
</table>

Table 5.1: Comparison between single, dual laser experiments [from Chapter 3] and colliding plasma experiments. Table key: DP = double pulse, CP = colliding plasma, w/o Nd:YAG = not including Nd:YAG laser energy, IBE = in-band energy and CE = conversion efficiency.

layer, which was observed to have a width of 250 µm from the visible imaging. This means that part of the CO₂ energy is not being fully utilised, which could be improved with tighter focussing of the CO₂ beam. When the seeds were orientated horizontally to each other, a slight enhancement in the EUV emission was observed at 2.5%, as compared with 2% for the vertical seeds. This could be due to the stagnation layer appearing larger when re-heated in this orientation, as the visible imaging would suggest, which would support the argument that part of the energy is not been coupled into the target i.e. the CO₂ laser is overfilling the target. The angle of incidence of the pump laser could also have a significant effect on its absorption by the stagnation layer. By re-heating at 90° there is a very steep electron density gradient across the stagnation layer, however it is much more gentle along its length as observed from optical interferometry performed by Hough et al. [21]. Further studies into the affects of the CO₂ laser angle of incidence on emission from a re-heated stagnation layer at 45° to normal incidence can be found in the PhD thesis of C. O’Gorman [10].
Bibliography


Bibliography


Chapter 6

Investigating the effects of laser intensity and pulse duration on EUV emission and ion time of flight from a laser produced gadolinium plasma

6.1 Introduction

In this chapter the effects of varying laser intensity and pulse duration on the extreme ultraviolet (EUV) emission of Gd targets at 6.7 nm and also the emission of ions from its plasma are investigated. The results are based upon the work published by Cummins et al. [1] and is similar to previous investigations [2–6]. In the experiments three pulse durations; 140 fs, 150 ps and 10 ns are used at various input energies to produce a laser intensity range of $10^{11} - 10^{15}$ W/cm$^2$. An EUV energy monitor was used to record in-band emission from the plasma, while a Faraday cup was used to observe ion yield and time of flight signals for ions from plasmas generated by each laser.

6.2 Experimental set-up and apparatus

The experiments were carried during a two month research visit to Utsunomiya University, Utsunomiya, Japan as part of an ongoing collaboration with UCD. The experimental apparatus consisted of a nanosecond (ns), picosecond (ps) and femtosecond (fs) laser, along with target chamber. All
6.2. Experimental set-up and apparatus

Three laser systems used in the experiment, EUV energy monitor and Faraday cup will be discussed in this section.

6.2.1 Target chamber, EUV photodiode and Faraday cup

In order to align the target chamber, two alignment lasers (operating wavelength \( \approx 540 \text{ nm} \)) were incident across the axis of the chamber perpendicular to each other as shown in Fig. 6.1. One of the beams was aligned to the optical axis of the incoming lasers used in the experiments, while the other beam was incident at 90° with respect to this. Assuming symmetry of the chamber ports, the point where the two alignment lasers met was taken to be the centre of the chamber. The Gd, which was attached to a linear feed-through, could then be positioned so that the face of the target was at the point where the two alignment beams met.

![Figure 6.1: Alignment process for Gd target.](image)

A full schematic of the experimental set-up and target chamber is shown in Fig. 6.2 on the next page. In the experiments a 99% pure planar Gd target was used. The linear feed-through it was used to allow the target to be moved after each shot to prevent cratering from occurring. The lens used was plano-convex fused silica with a 10 cm focal length and 5 cm diameter.
6.2. Experimental set-up and apparatus

The lens was mounted on a small stage whose position could be finely adjusted using a micrometer screw, allowing the focus of the lens to be varied. For experiments with the fs laser the set-up was changed by removing the 10 cm lens and replacing it with a 40 cm focal length lens placed outside the target chamber. The EUV energy monitor was then attached to the port orientated at 45° to the target normal and the axis of laser incidence. A schematic of the energy monitor is shown in Fig. 6.3. EUV light produced by the plasma emitted at 45° is incident onto a 25.4 mm diameter molybdenum boron carbide (Mo/B₄C) mirror coated for 6.7 ± 0.05 nm with a reflectivity of 23% and a collection angle of incidence of 5°. This directed the EUV photons through a zirconium (Zr) filter [7] with a thickness of 200 nm onto a photodiode. Given the density of Zr at room temperature is 6.49 g/cm³ [8] and the filter has a thickness of 200 nm, the transmission can thus be determined [Fig. 6.4] [9]. For a Zr filter of 200 nm thickness this equates to a transmission of 0.47 at 6.7 nm. The photodiode used to detect the transmitted photons was the AXUV100G from IRD [10]. The diode has an active area of 100 mm² and a rise time of 10 μs.
6.2. Experimental set-up and apparatus

Figure 6.3: Schematic of the EUV energy monitor used for measurements at 6.7 nm.

Figure 6.4: Transmission of 200-nm Zr filter in the soft x-ray/EUV region.
6.2. Experimental set-up and apparatus

The electrical signal produced by the photodiode was transmitted via BNC cable to a BT-250 bias tee. As the photodiode would saturate when exposed to pulse energies greater than 10 $\mu$J, a 12 V reverse bias voltage applied raises the saturation threshold to a higher level so that it can detect the photons from the plasma. From the bias tee, the signal is sent to a digital oscilloscope where the EUV signals were observed. According to the technical specifications of the photodiode [10] there is an almost flat diode response of around 0.26 A/W across most of the soft x-ray/EUV wavelength region [Fig. 6.5].

![Figure 6.5: Responsivity of the AXUV100G photodiode [10].](image)

The other diagnostic used in this experiment was a Faraday cup (FC) [11]. The FC was placed inside the target chamber at the other 45° degree angle with respect to the target normal. It was attached to an optical post and mounted approximately 10 cm away from the target surface. A very simple design for the FC was used, which consisted of a cylindrical cup made of a electrically conductive metallic material, over which a small aperture was placed. A bias of -17 V used to deflect electrons produced from the plasma and attract the beam of ions, was applied to the cup using a simple RC electrical circuit, which produces a time dependence as the resistor will control the rate at which the capacitor is discharged. In theory, the current passing through the cup, as the ions impinge on its surface, is the charged particle beam and the FC acts as a resistor, therefore the current read from the FC is a direct measurement of the beam current [12]. The current that corresponds to the ion signals from the plasma could then be displayed on the digital oscilloscope. The temporal response of the ion signals gives information on the time of flight (TOF) of the ions from the plasma.
6.2. Experimental set-up and apparatus

6.2.2 Laser systems

Three lasers were employed in the experiments to achieve the desired variation of laser pulse duration and intensity. The full laboratory layout with the target chamber, energy monitor and three laser systems is shown in Fig. 6.6. The details of the three laser systems used are listed then in Table 6.1.

![Figure 6.6: View of target chamber, EUV energy monitor and laser systems.](image)

<table>
<thead>
<tr>
<th>System</th>
<th>τ</th>
<th>λ</th>
<th>$E_{\text{max}}$</th>
<th>$\phi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nd:YAG</td>
<td>10 ns</td>
<td>1064 nm</td>
<td>420 mJ</td>
<td>$10^{11} - 10^{12}$ W/cm$^2$</td>
</tr>
<tr>
<td>Nd:YAG</td>
<td>150 ps</td>
<td>1064 nm</td>
<td>210 mJ</td>
<td>$10^{13} - 10^{14}$ W/cm$^2$</td>
</tr>
<tr>
<td>Ti:sapphire</td>
<td>140 fs</td>
<td>800 nm</td>
<td>30 mJ</td>
<td>$10^{14} - 10^{15}$ W/cm$^2$</td>
</tr>
</tbody>
</table>

Table 6.1: Tabulated details of the three laser systems used in experiments. Table key: $\tau$ = pulse duration, $\lambda$ = laser wavelength, $E_{\text{max}}$ = the maximum laser energy and $\phi$ = the laser intensity range.
6.2. Experimental set-up and apparatus

The fs laser system [Fig. 6.7] utilised in experiments was a titanium-sapphire (Ti:sapphire), so called because the lasing medium is a sapphire crystal (Al₂O₃) doped with titanium. The laser oscillator is driven by a frequency doubled (λ = 532 nm) Nd:YAG pump laser operating at a 10 Hz repetition rate. A regenerative amplifier is then used to amplify the pulse. This is achieved by placing the gain medium in an optical resonator along with an optical switch, which allows control over the number of passes made. The optical switch consists of an electro optic modulator and polariser. The final output from the amplifier has a pulse duration of 140 fs, a maximum output energy of 30 mJ and 10 Hz repetition rate. The Ti:sapphire emits over a broad wavelength range but operates most efficiently at λ = 800 nm.

![Figure 6.7: The Ti:sapphire femtosecond laser system at Utsunomiya University.](image)

The ps laser used was the EKSPLA SL312 Nd:YAG, operating at a wavelength of λ = 1064 nm. A schematic of the laser can be seen in Fig. 6.8. A full description of the laser operation is given in the EKSPLA laser user manual, which is briefly described here. The laser head consists of three main parts; the master oscillator, a system of pulse compression and a system of pulse amplification.
6.2. Experimental set-up and apparatus

Figure 6.8: Schematic of EKSPLA SL312 picosecond Nd:YAG.

The output from the master oscillator stage of the laser head is 4 - 5 mJ in ~ 2 ns and this energy is monitored by a photodiode at PD1. Compression of the pulse is carried out in the second stage of the laser head. It is achieved by stimulated Brillouin scattering (SBS). The Brillouin scattering effect occurs when an incident photon, travelling through a non-linear medium, is converted into a scattered photon of lower energy and is usually directed backwards in the direction of incidence. SBS occurs when the input beam is above a certain power threshold and as a result most of the input beam is reflected [13]. The pulse is focussed into the SBS cell containing carbon tetrachloride (CCl₄), with the focussing arranged to compress the pulse via the SBS process. Finally three passes are made through a flash lamp pumped Nd:YAG rod, the gain medium of the amplifier, to produce a 150 ps pulse with a maximum energy of 190 mJ.

The final laser system used in experiments was the ns Nd:YAG. It was similar to the the ns laser described in Chapter 2, which was used in other experiments in this thesis. It had a 10 ns pulse duration with a maximum energy of 420 mJ, also at a wavelength of \( \lambda = 1064 \text{ nm} \). In order to improve the focussing conditions of both the ns and ps lasers, the beam spots were expanded using a beam expander with 2X magnification. Firstly, they were expanded from an initial beam diameter of 7 mm using a 150 mm focal length diverging lens. The beam was then collimated using a 300 mm focal length lens to a beam diameter of 14 mm. This then reduced the final focal spot size of laser pulse when focussed with the 100 mm lens in the chamber, from the equation:

\[
 w_0 = \frac{4M^2\lambda f}{\pi D}
\]  
(6.1)
6.3. EUV in-band measurements

where \( w_0 \) is the focussed beam diameter, \( M \) is laser beam quality, \( \lambda \) is the laser wavelength, \( f \) is the focal length of the lens and \( D \) is the unfocussed beam diameter. From this relation it is observed that a small spot size and hence a greater laser intensity (W/cm\(^2\)) is achievable by increasing the unfocussed beam diameter as such. The focal spot diameter of the ns and ps laser on-target was estimated to be 30–40 µm and \( \sim 50 \) µm for the fs laser.

6.3 EUV in-band measurements

During experiments each laser beam was incident normally onto the planar Gd target and after each laser pulse the target surface was replenished by moving the target using the linear feed-through. The output energy of each laser was changed during experiments in order to vary the laser intensity. The \( Q \)-switch delay on both the ns and ps lasers were gradually increased to reduce the lasing energy, while a polariser was used to control the output from the fs laser. The EUV photons were detected by the energy monitor and multiple shot averaged signals were recorded on the oscilloscope. The results of investigating the influence of laser intensity on the conversion efficiency (CE) of laser energy to in-band EUV emission are presented.

6.3.1 Nanosecond pulse

The energy of the ns laser was varied from 30–420 mJ, which corresponded to a laser intensity scan of \( 4 \times 10^{11}–6 \times 10^{12} \) W/cm\(^2\). Five shot averaged EUV signals were recorded for nine different laser intensities, each on a fresh target spot. The averaged EUV signals are presented below:

![Graphs showing averaged EUV signals for different laser input energies](image)

Figure 6.9: Five shot averaged in-band EUV response from photodiode using the 10-ns laser, as recorded on oscilloscope for various laser input energies.
6.3. EUV in-band measurements

Figure 6.9: Five shot averaged in-band EUV response from photodiode using the 10-ns laser, as recorded on oscilloscope for various laser input energies.
6.3. EUV in-band measurements

6.3.2 Picosecond pulse

The energy of the ps laser was varied from 20–180 mJ, which corresponded to a laser intensity scan of $1 \times 10^{11} - 2 \times 10^{12}$ W/cm$^2$. Three shot averaged EUV signals were recorded for eleven different laser intensities, presented next:

![Graphs showing Voltage vs. Time for different laser energies.](image)

Figure 6.10: Three shot averaged in-band EUV response from photodiode using the 150-ps laser, as recorded on oscilloscope for various laser input energies.
6.3. EUV in-band measurements

Figure 6.10: Three shot averaged in-band EUV response from photodiode using the 150-ps laser, as recorded on oscilloscope for various laser input energies.
6.3. EUV in-band measurements

6.3.3 Femtosecond pulse

Finally, the energy of the fs laser was varied from 5–20 mJ, corresponding to an intensity scan of $1 \times 10^{15} - 6 \times 10^{15}$ W/cm$^2$. The laser was firing at a 10 Hz repetition rate and the oscilloscope was set to record a 20 shot averaged signal. The averaged EUV signals of six laser energies are presented below:

![Figure 6.11: Averaged in-band EUV response from photodiode using the 140-fs laser, as recorded on oscilloscope for various laser input energies.](image-url)
6.3. EUV in-band measurements

6.3.4 Results

The in-band energy was determined from the following relation:

\[ EUV_{\text{energy}} = \frac{BW \cdot 2\pi V_s}{B_M \times R_M \times T_F \times T_G \times R_{\Omega} \times Bias\% \times PD\% \times \Omega} \]  \hspace{1cm} (6.2)

where the bandwidth available determined by currently available multilayer optics (BW) is 0.6, \( V_s \) is the summed voltage from integrating under the curve of the EUV signal recorded on the oscilloscope, the mirror bandwidth \( (B_M) \) is 0.4478, the mirror reflectivity \( (R_M) \) is 0.25, the filter transmission \( (T_F) \) is 0.4765, the gas transmission \( (T_G) \) is 1, the oscilloscope resistance \( (R_{\Omega}) \) is 50 \( \Omega \), the bias electronics efficiency \( (Bias\%) \) is 1 and the photodiode efficiency \( (PD\%) \) is 0.257. The solid angle \( (\Omega) \) is given by:

\[ \Omega = 2\pi \left(1 - \frac{1}{\sqrt{1 + \left(\frac{r}{l}\right)^2}}\right) \]  \hspace{1cm} (6.3)

where the radius \( (r) \) of the Mo/B_4C multilayer mirror that is reflecting the EUV photons is 5 mm and the distance from the plasma to the mirror \( (l) \) is 88 mm.

Using equation 6.2, the CE could thus be determined for each laser input energy by dividing the EUV energy by the laser energy falling on the target. The laser intensity at each input energy could also be determined. The result of CE plotted as a function of the laser intensity is shown in Fig. 6.12 on the next page. A maximum CE, for the case of the 150 ps pulses, of 0.4% was observed compared to values of 0.3% and 0.15% for 10 ns and 140 fs laser irradiation, respectively. This is equivalent to a 1.33% CE if it is calculated in a 2% bandwidth, for comparison with tin (Sn) sources at 13.5 nm. The optimum laser intensity for efficient 6.7 nm EUV emission was found to be around \( 7 \times 10^{13} \) W/cm\(^2\). For 10 ns pulses, a peak CE at a flux close to \( 4 \times 10^{12} \) W/cm\(^2\) was observed. The drop in CE between these two laser intensity values may be ascribed to both pulse duration and plasma temperature effects. The latter influences the peak emission wavelength as described previously [14, 15]. In the case of the fs laser pulses, the CE had a maximum of only around 0.15%, as the laser intensity was too high and plasma volume too small for efficient emission of photons around 6.7 nm. The high intensity likely results in an overheating of the plasma, which must then expand and cool before the emission from the desired ion stages occurs.
6.4 Ion time of flight measurements

Figure 6.12: Conversion efficiency for a range of intensities using ns, ps and fs pulse durations. A polynomial fit is applied to that data to help the viewer to observe the trends.

6.4 Ion time of flight measurements

Information on the ion yield and ion TOF are presented in this section. The ion yield as a function of the flight time for various laser intensities of ns, ps and fs pulse durations are shown next:

6.4.1 Nanosecond pulse

Figure 6.13: Sample TOF signals recorded from ns LPP.
6.4. Ion time of flight measurements

![Sample TOF signals recorded from ns LPP.](image1)

**Figure 6.13:** Sample TOF signals recorded from ns LPP.

6.4.2 Picosecond pulse

![Sample TOF signals recorded from ps LPP.](image2)

**Figure 6.14:** Sample TOF signals recorded from ps LPP.
6.4. Ion time of flight measurements

6.4.3 Femtosecond pulse

![Graphs showing ion yield vs. time of flight for different laser intensities and pulse durations.]

Figure 6.15: Sample TOF signals recorded from fs LPP.

6.4.4 Results

The ion kinetic energy was lower for the shorter pulse durations, which also produced higher electron temperatures, due to their high laser intensity. The ion TOF at the laser intensity for optimum CE of the ps LPP was $9 \mu$s, compared to $2 \mu$s for the corresponding optimum intensity of the ns LPP. The time durations of each TOF signal are lengthened by the continuous supply of material from the wide planar Gd target. The accelerating length, which originates from the plasma expansion [16], is present for a much shorter time in the 150 ps case. As a result, the ion yield at the time corresponding to the peak signal amplitude was seen to be lower for ps than ns Nd:YAG laser irradiation. The peak TOF as a function of laser intensity is presented in Fig. 6.16, which shows longer flight times for ions from the fs and ps LPPs compared with the ns LPP. Due to the very short pulse duration of the fs laser, a change in the trend is observed for TOF as a function of laser intensity compared to the ns and ps LPPs. This is as a result of different ion
6.5. Summary and conclusions

Acceleration processes beginning to dominate. The extremely short time scale of the fs plasma lifetime means hydrodynamic motion is suppressed, due to the fs pulse duration being shorter than the electron phonon relaxation time (typically 10 ps for elemental metals) [17, 18] and hence charge separation dominates.

![Figure 6.16: TOF as a function of laser intensity for ns, ps and fs pulse durations. A fit is applied to help the viewer observe the trend.](image)

Figure 6.16: TOF as a function of laser intensity for ns, ps and fs pulse durations. A fit is applied to help the viewer observe the trend.

6.5 Summary and conclusions

In this work, processes involved in the laser-plasma interaction for varying laser pulse durations and intensities were investigated. Information on TOF signals was recorded to help better understand the effect of pulse duration on ion energy emission from the plasma. By studying the effects of laser intensity on CE, the optimum laser intensity, for irradiating with a given focal spot size, could also be determined. In these experiments, the laser intensity was varied from $10^{11}$-$10^{15}$ W/cm$^2$ to produce emission from Gd$^{12+}$ to Gd$^{25+}$, whose resonant emission is around 6.7 nm [14, 15, 19-21]. To achieve efficient EUV emission and high CE, both the laser intensity and the laser pulse duration were optimised. Peak laser intensities for maximum CE from the ns and ps lasers were observed. The ps laser was found to
6.5. Summary and conclusions

be the most efficient at generating 6.7 nm EUV source emission, with a maximum CE of 0.4% recorded in a 0.6% bandwidth for an intensity of $6 \times 10^{13}$ W/cm$^2$. This is comparable to the maximum reported values of 0.5% using a double pulse and 0.45% achieved using mass limited Gd targets [22]. The ps also resulted in a longer peak ion TOF time of 9 $\mu$s, compared to 2 $\mu$s for the ns at peak CE. From a simple one-dimensional hydrodynamic code simulation [1], the electron temperature dependence on the laser intensity for the interaction of a laser at $\lambda=1.06$ $\mu$m with high Z plasma is shown. From this, the peak laser intensity obtained in experiments corresponds to an electron temperature of 130 eV. This estimated temperature is close to previous theoretical predictions of the optimum Gd electron temperatures of 144 eV [14] and 110 eV [23].

In summary, an increased CE was observed using 150 ps Nd:YAG laser pulses as compared with ns and fs pulses. The short pulse duration of the ps also has the effect of increasing ion TOF and reducing peak ion kinetic energy. The electrostatic field induced by the formation of the plasma determines the TOF and maximum kinetic energy of ions produced [16]. The smaller electrostatic potential as created by the ps laser results in a longer TOF and a reduction in the peak ion kinetic energy. The EUV emission, however, is not fully optimised due to strong self-absorption in the optically thick plasma that is created. This self-absorption can be observed from spectra obtained of the Gd plasma by O’Gorman et al. [24] using the same ps laser with similar experimental conditions. These results point towards the use of a pulse duration of the order of hundreds of ps in order to obtain greater EUV emission. CO$_2$ laser systems with ps pulse duration could therefore be of great interest for BEUV sources. The emission would not only increase due to the improved opacity effects, due to the reduction in plasma critical density with the use of longer laser wavelength ($\lambda=10.6$ $\mu$m) [25], but the plasma would also produce ions with lower kinetic energy due to the ps pulse duration, which would assist with debris mitigation and reduce the damage to optical components by fast ion bombardment.
Bibliography


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Chapter 7

Thesis summary and conclusions

7.1 Summary of experimental results

This chapter is a summary of the preceding work presented in this thesis, beginning with the introductory chapter, which sets out the motivation for the studies undertaken, as well as giving details of the theory relevant to the work. The motivation for this thesis was stated as being a study of the extreme ultraviolet (EUV) emission from laser produced plasmas (LPP), including the experimental parameters involved in producing the plasma, which can be used to optimise its emission at certain wavelengths, in particular, optimising emission at 13.5 nm desired for EUV lithography and future lithography wavelengths around 6.7 nm. The theory discussed is primarily related to plasmas, including a basic definition, the process of how they are formed by laser ablation and what properties they exhibit. Importantly, which properties can be tailored to achieve maximum emission of the desired EUV radiation. Following this, Chapter 2: “Apparatus” gave a detailed account of the majority of experimental equipment used in this work including laser systems, spectrometer and optics. Particular mention was made of the CO₂ laser, which was the primary laser used to generate EUV radiation via ablation of target materials.

The first experimental chapter of this thesis, Chapter 3: “Study of EUV emission from CO₂ laser produced tin plasmas”, reported on the results of varying multiple experimental parameters on the EUV emission from laser produced tin (Sn) plasmas. The influence of the CO₂ laser energy, gas mixture, focus lens position, angle of incidence of the laser pulse, laser temporal pulse duration and laser wavelength were all investigated. From studies of
7.1. Summary of experimental results

the CO$_2$ laser pulse duration it was found that a large percentage of the CO$_2$
lar pulse was not used to heat the plasma sufficiently for in-band emission
at 13.5 nm. The existence of a long tail at the end of the CO$_2$ temporal pro-
file meant that a large amount of energy contained in the pulse was reducing
overall efficiency. For the EUV industry this additional laser radiation should
thus be avoided as interaction of the tail with the target may produce addi-
tional Sn debris, if the energy is not sufficient to be transferred into creating
plasma. Additionally, excess infra-red radiation not used in the laser target
interaction can propagate through to the scanner, causing potential heating
and thus overlay problems at wafer level [1]. The results of this extensive
survey of parameters affecting the plasma emission was the motivation for
experiments carried out in the following two chapters. Chapter 4: “Plasma
shutter device for CO$_2$ laser pulse shortening” presented a new technique
for shortening the duration of the CO$_2$ laser pulse [2], which avoids the is-
issue associated with the longer pulse. By using this new method, CO$_2$ laser
pulses of duration down to 2 ns were achieved. The effectiveness of such a
device was studied by observing the impact of CO$_2$ laser pulses on the EUV
emission from Sn plasmas. It was demonstrated that an improvement of 1.5
times in conversion efficiency (CE) could be achieved when comparing longer
and shorter pulse durations.

The extensive study of different experimental parameters in Chapter 3
also included dual laser pulse experiments. These tests were carried out by
producing pre-formed plasma targets with a pre-pulse laser, which were then
re-heated by the CO$_2$ laser main pulse. From the work carried out with the
dual laser pulses it was found that there was an improvement in the efficiency
of the in-band EUV radiation as compared with a single laser pulse on a solid
target. This pre-formed plasma resulted in a lower density, mass limited tar-
get into which the CO$_2$ pulse was more efficiently coupled. The results of this
chapter indicated that low density plasma targets are more efficient sources
of EUV radiation, compared with solid Sn. This result was investigated fur-
ther, when a systematic study of low density colliding plasma targets was
carried out in Chapter 5: “EUV emission from re-heating colliding plasma
targets”. This chapter proposed colliding plasmas as potential sources of ef-
ficient EUV emission [3]. Details were given on the set-up of the colliding
plasma experiments and how they are formed. Following this was a detailed
study of a wide range of parameters that effect the EUV emission. The pre-
pulse laser energy was varied, as well as the spatial and temporal alignmen
t of the main pulse. The more efficient short pulse profile of the CO$_2$ laser,
studied in Chapter 4, was also utilised during the experiments. During this
systematic investigation, an enhancement of the EUV emission was observed
as compared with a range of potential laser and target configurations studied
7.2. Future work

in Chapter 3. Both single laser pre-pulse and colliding plasma targets were shown to be preferred sources for absorbing CO\textsubscript{2} laser radiation efficiently and re-emitting as EUV. However for very similar experimental inputs, the colliding plasma target demonstrated a 1.3 times increase in efficiency when compared to the plasma target generated by a single pre-pulse laser spot. When the input energy of the pre-pulse laser as well as the EUV it emits is excluded from calculations of efficiency, the CE of the colliding plasma target peaked close to 5\%, this value being comparable to current maximum reported values [4, 5].

Finally, Chapter 6: “Investigating the effects of laser intensity and pulse duration on EUV emission and ion time of flight from a laser produced gadolinium plasma” reported on experiments that were carried out on gadolinium (Gd) targets, a potential source for beyond EUV. As was discussed in the thesis introduction, Gd plasma is known to emit strong in-band radiation around 6.7 nm when ions in the range of Gd\textsuperscript{16+–Gd\textsuperscript{27+}} are produced [6]. In order to achieve this high rate of ionisation in the plasma, a significant plasma temperature of greater than 100 eV has been predicted for optimum emission [6]. To achieve these temperatures a sufficient input laser intensity is required [7]. Therefore, in this chapter both the effect of laser pulse duration and on-target energy were studied as factors effecting the emission from the plasma due to changes in the laser intensity. This was carried out for a range of energies for nanosecond, picosecond and femtosecond laser systems [8]. The experiments indicated that the higher intensity picosecond laser was better suited for generation of 6.7 nm emission via ablation of Gd targets. In addition to this, the ion emission from the plasmas was also studied. The ion time of flight was monitored by means of a Faraday cup. From this the picosecond laser was also found to have the additional advantage of slower ion flight times, beneficial when considering bombarding of optical components in a potential industrial scale application.

7.2 Future work

The colliding plasma targets have been demonstrated to be an extremely promising source of EUV emission. Previous work has shown that given the optimum plasma parameters, a CE up to 8\% may be achievable [9]. With that in mind, colliding plasma targets can be considered a source that could potentially deliver this high level of EUV efficiency. The study presented in Chapter 5 only reports on some of the possible adjustments that could be made to optimise the experiment. Future work on this experiment would involve further target optimisation by means of seed separation, which will
7.2. Future work

affect the size and timing of the formation of the stagnation layer. In addition to this, variation of the wedge target angle on which the seeds are formed. Both of these changes will affect the collisionality parameter, i.e. if more interpenetration or stagnation occurs at the interface of the two seed plasmas. In effect the size and density of the stagnation layer can be finely tuned until it is optimal for coupling the main pulse laser.

Another experimental parameter that could be improved from this study was the focussing conditions of the CO\textsubscript{2} laser. From imaging of the plasma, the diameter of the stagnation layer region, which was irradiated by the CO\textsubscript{2} laser, was determined to be approximately 350 \(\mu\)m. However, the laser beam diameter at focus was calculated to be around 520 \(\mu\)m. This means that the laser pulse was overfilling the target and therefore only 70–80\% of the beam interacts with the target. With improvements to the beam matching to target, as well as further fine tuning of the stagnation layer, the higher limits of CE up to 8\% could be achievable. To further study colliding plasmas potential for EUV industry a full systematic study of the debris emitted from the target surface also needs to be carried out. This study is important for validation of colliding plasma targets as solid Sn targets will inherently produce much greater debris than Sn droplet targets.
Bibliography

Bibliography


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"Dann, Blümlein alle,
Heraus, heraus!
Der Mai ist kommen,
Der Winter ist aus."