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Emission and absorption in laser produced plasmas: processes and applications

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Abstract. Laser produced plasmas have been used for many years as intense sources of extreme ultraviolet (EUV) and soft x-ray radiation. Depending on the choice and composition of target the EUV spectra can be dominated by line, unresolved transition array (UTA) or continuum emission. Line and UTA emission have found application in various proposed EUV sources for lithography, which is based on the availability of mirrors with high reflectivity in a 2% bandwidth at a wavelength of 13.5 nm. The results of recent experimental measurements of absolute in-band and out of band intensity, ion distribution and debris are presented. It was found that because of opacity effects, the conversion efficiency is sensitive to ion density and laser wavelength. Various schemes to improve the conversion efficiency are discussed as are the results of recent plasma modelling calculations.

In addition, laser produced plasmas of some high Z elements emit intense line free continua over extensive energy ranges. Some recent results on inner shell photoabsorption spectra of Sn, I and Au ions obtained using these continua are presented also.

1. Introduction

In this paper a brief overview of laser produced plasma properties is given in section 1. Section 2 deals with applications to extreme ultraviolet lithography (EUVL), while section 3 will look at applications of laser produced plasmas to study ionic photoabsorption.

1.1. Laser produced plasmas

When a high power pulsed laser is incident on a solid target a hot transient plasma is produced whose average electron temperature, \( T_e \), is essentially determined by the laser pulse power density \( \Phi \) and wavelength \( \lambda \), according to \( T_e(eV) \approx bA^{1/5}(\lambda^2\Phi)^{3/5} \) while the average charge \( \approx 0.67 (AT_e)^{1/3} \), where \( A \) is the atomic number. Absorption of the laser pulse proceeds in an interaction region typically ~100 \( \mu \text{m} \) deep above the target surface at the so-called critical density \( n_{ec} \) where the plasma frequency matches the laser frequency. Since \( n_{ec} \approx 10^{21}/\lambda^2 \text{ cm}^{-3} \) (\( \lambda \) in \( \mu\text{m} \)) the electron density is thus strongly influenced by laser wavelength, 10¹⁹ cm⁻³ for CO₂ irradiation, 10²¹ cm⁻³ in the case of Nd:YAG. The corresponding maximum ion densities are then typically an order of magnitude lower in each case. Behind the critical density layer, in the denser region, a shock wave propagates into the target and to conserve momentum, the plasma expands rapidly in the opposite direction usually along
the incident beam direction; typical expansion velocities for illumination by a Q-switched laser delivering a few hundred mJ in approximately 10 ns are of the order of $10^6$ cm/s. The ions are accelerated after the lighter more rapidly moving electrons and thus the plasma consists of a hot core with the most highly charged ions emitted strongly in the direction of the incident laser beam and successively lower stages in cones of increased apex angle about this direction. Hence opacity issues are important and absorption by lower ion stages can dramatically reduce the emitted EUV flux. Because of their lower initial densities, CO$_2$ generated plasmas are optically thinner than those from Nd:YAG or visible/UV lasers which is an important consideration in designing sources of line or continuum radiation. Moreover, it has been shown that the EUV and x-ray emission from the plasma core in turn can produce a secondary cold plasma surrounding the main plume and absorption in this plasma can influence radiation transport close to the target surface [1].

1.2 Radiation from Laser Produced Plasmas

The EUV spectrum from a laser produced plasma consists of lines (bound-bound transitions), and because of the high densities encountered, lines from high $n$ states are usually not seen, recombination radiation (bound-free transitions) which scales as $\zeta^4$ where $\zeta$ is the average ionic charge and bremsstrahlung (free-free). For an optically thin plasma: $P_{\text{lines}}:P_{\text{recomb}}:P_{\text{breem}} \sim 100:10:1$. In some cases the lines group together to form an unresolved array (UTA) [2] where the linewidth is typically greater than the spacing between individual line spacing. An example of such an EUV spectrum is shown below.

![Figure 1. EUV spectrum from a target containing Ce in a plastic substrate.](image)

2 Extreme UV lithography

2.1 Background

Conventionally semiconductor devices are fabricated using a lithographic process, the most recent phase of which is referred to as deep-ultraviolet lithography (DUVL). It is well known that the shorter the wavelength of light used in lithography the smaller the feature size that can be fabricated. The next stage in the semiconductor lithography roadmap is Extreme-UV Lithography (EUVL) which utilises a molybdenum silicon (Mo/Si) multi-layer mirror system to collect and focus EUV radiation in narrow band around a wavelength of 13.5nm. The short wavelength will result in a higher density of transistors and therefore faster processors. High temperature plasmas, e.g., Gas Discharge Plasmas (GDP) or Laser Produced Plasmas (LPP) are the sources of choice as they can be tuned to emit at this 13.5nm wavelength. Already it is well known that the optimum electron temperature in such plasmas should be in the 30-40 eV range, the optimum density is around $10^{18}$ ions cm$^{-3}$ and the optimum size should be hundreds of microns [3-5]. For a single pulse typically $10^{18}$ atoms/ions are ablated and can be an important source of contamination in an EUV tool.

Initially, the need to reduce particulate emission led to the choice of xenon, which as an inert gas should provide a debris-free source. There is a line group in the spectrum of xenon which has been shown by a number of researchers to arise from 4d$^8$-4d$^7$5p transitions in Xe XI by comparison with atomic structure calculations [6]. The population of this ion stage needs to be optimised to attain maximum intensity. Note that, depending on the type of source and hence ion density, optimisation is not always necessarily the same as maximising a given ion population because of opacity effects. Considerable work has been expended on exploring the feasibility of using laser produced plasmas of xenon clusters, produced by supersonic jets or gas puffs from nozzles and solid xenon targets [7]. The
optimum in-band efficiency obtainable from laser produced plasmas appears to be of the order of 1%; therefore xenon is not the optimum choice if higher conversion is required. In studies on Xe spectra the bulk of the resonance emission was always found to lie in the 10–12-nm region and it was shown by Fahy et al. [8] that potentially 5 times as much flux could be obtained within a 2% bandwidth at 11 nm compared to 13.5 nm. The transitions responsible arise from $4p^54d^{n} - 4p^54d^{n+1} + 4d^{n-1}4f$ lines which essentially overlap in adjacent ion stages and merge to form a UTA [9].

The same unresolved transition array as that in xenon at 11 nm is also observed in adjacent elements. From work, ongoing at UCD since the late 1970s, tin was already identified as potentially the strongest emitter at 13.5 nm since the UTA is centred near that wavelength. It was further shown that if the tin concentration was reduced to approximately 10%, the peak brightness actually increased due to the reduction in opacity effects. Furthermore, if the remaining 90% of the constituents were low-Z materials the radiation emitted was concentrated in a band 1–2-nm wide centred near 13.4 nm and the ordinary recombination continuum from the plasma was suppressed in comparison with that from a pure tin target. In fact if pure targets are used the recombination continuum is greatly enhanced as is the contribution from satellite emission which has been shown to be particularly important at higher ion densities [10].

The current status of EUVL is that the initial prototype or $\alpha$-tools have been supplied by ASML, Nikon and Canon. These tools, originally based on xenon plasmas, will all run with tin discharge plasmas because of their superior conversion efficiency (CE). However, although the $\alpha$-tools are based on discharge produced plasmas (DPP), it is not certain that DPP sources will be able to deliver the powers, >115 W into $2\pi$ sr at intermediate focus (IF), that will be required for a manufacturing tool. Consequently, laser-produced plasma (LPP) sources, which are more readily scalable and permit an increase in collection efficiency by up to a factor of three, since they can use normal rather than grazing incidence collection remain of significant interest to EUVL source developers.

One drawback however, is that normal incidence systems are vulnerable to sputtering and erosion by fast ions. However these can be deflected by electromagnetic fields while plasma curtains have been shown to be quite effective in removing neutrals. To operate at high repetition rates, the use of mass limited droplet systems has been pioneered by Richardson et al. [11]. In this approach the number of tin ions is $\sim 10^{17}$ and all are ionised and emit a number of times during the laser pulse. 100 kHz injection has been successfully demonstrated for such droplet based systems while laser operation has been achieved at frequencies up to 30 kHz. Moreover, CE values of 4.5% have been obtained using CO$_2$ laser based systems, compared with typical values of 2.5% for DPP sources [12].

2.2 Experimental Results from Nd:YAG Plasmas

2.2.1 Conversion Efficiencies: Using a Jenoptic 0.25 m Spectrograph with an absolutely calibrated CCD detector, spectra were recorded at a range of laser pulse power densities from both pure tin and glass targets containing 5% tin by number. This concentration had been shown to be optimum in the
trade-off between emission which in an optically thin regime scales linearly with number density and absorption which scales exponentially. The results are shown in Figure 2 and translate to maximum in-band CEs of 2.3% into $2\pi$ sr for $\Phi = 1.6 \times 10^{11} \text{ Wcm}^{-2}$ for pure tin and 2.9% into $2\pi$ sr for $\Phi = 2 \times 10^{11} \text{ Wcm}^{-2}$ for the target containing 5% tin [13].

2.2.2. Isotropy of emission: The above data were recorded from slab targets irradiated at normal incidence and viewed at 45% to the target normal. Isotropy of emission was assumed. However in the introduction it was noted that the ion distribution varies with angle and consequently one would expect both emission and absorption to be sensitive functions of angle. Moreover, the plasma core is typically 220-300 $\mu$m in diameter but only $\sim 100 \mu$m deep so geometrical effects are important also. In a detailed study it was found that maximum emission was obtained when the plasma was viewed close to normal incidence and the intensity dropped by a factor of 2 in going from 20° to 90°. The average integrated flux was essentially the same as that recorded at 45°. Thus care is needed in performing CE measurements and comparisons and the disagreement among a number of published CEs.

2.2.3. Effect of a prepulse: Previous work with laser produced plasmas has shown that increased UV and x-ray flux may be obtained if a prepulse is used to produce a seed plasma that is then rapidly heated by the main pulse [14]. In an experiment to explore the effects of a prepulse on inband emission from a tin plasma, a 170 ps , 500 mJ, 1.06 $\mu$m pulse was divided into differing pre-/ main pulse fractions and the delay between them was varied using an optical delay line. The results are shown in Figure 3 where it is seen that at an interpulse delay of 5.5ns, for a pre-/main pulse ration of 1/9, an increase in intensity of 80% was achieved.

![Figure 3. Effects of using prepulses: EUV intensity as a function of interpulse delay and fraction.](image)

In a subsequent experiment the effects of heating a plasma produced by a 14 ns pulse with a 170 ns one was also investigated. In this case a 75% increase of in-band CE was obtained using a pure tin target pointing to the possibility of obtaining an overall efficiency of $\sim 4\%$.

2.3. Theoretical modelling of LPPs for EUVL applications:
Considerable effort has been expended on modeling the emission from LPPs in order to identify the optimum set of operating parameters to optimize the in-band CE. These have included both steady state and time dependent approaches.
2.3.1. *Steady state plasmas.* In this approach ion fractions as a function of electron temperature were calculated assuming a collisional radiative equilibrium (CRE) [15]. The emission spectra from $4p^6 4d^n - 4p^5 4d^{n+1} + 4d^{n-1} 4f + 4d^{n-1} 5p$ transitions were calculated for each ion stage and then weighted with the ion fraction appropriate to a particular electron temperature and summed to give a spectrum for that temperature. The results, with the line distributions fitted to Gaussians and assuming optical transparency are shown in Figure 4. From this figure it is seen that the optimum electron temperature is in the 30-40 eV range.

![Figure 4: Calculated emission from an optically thin Sn plasma as a function of electron temperature.](image)

2.3.2. *Time dependent modeling:* For a more realistic description, one has to allow for plasma expansion and optical opacity effects and perform a full time dependent calculation. A number of different results using 1- and 2-D models to describe the plasma expansion have been obtained and all point to a CE of about 3% being attainable with Nd:YAG plasmas, which is limited by absorption in the outer layers. If the opacity could be reduced and the most intense emission zones imaged directly, then a CE of 5-6% should be achievable [3, 4]. One way to reduce the amount of self absorption in the plume could be to use a modified laser pulse profile. Indeed a recent calculation using the Z* code has shown that if a flat-top rather than Gaussian profile were used the CE is enhanced by almost 100% [5].

An alternative approach would be to use a lower density plasma and this scenario is realized in a CO2 plasma. Here, because of the lower cutoff density, electron and ion densities are only 1% of those of Nd:YAG plasmas and the emission approximates better to that shown in Figure 2. for 5% Sn targets. CEs of 4-4.5% [16,17] have been measured with such plasmas and the best laser source now available comprises mass limited droplet injection, with Nd:YAG pumping to vapourise and ionize it that then expands to a size where the average electron density matches the CO2 pulse which then illuminates it [17]. The disadvantage is that currently CO2 lasers have pulse lengths in the 40 - 50 ns
range and the plasma expands to a lower suboptimal density within this time frame so only the early part of the pulse is fully utilised. For bulk targets, preliminary Z* calculations show that the EUV output remains essentially flat regardless of the length of the laser pulse for pulses in excess of 10 ns duration, while for significantly shorter pulses the EUV CE is reduced, presumably because of the importance of reflection at the beginning of the ablation process.

Figure 5. EUV emission from slab targets produced by Nd:YAG pulses with different spatial profiles: Flat-top (left) and Gaussian (right). Note that for observation from the sides, the distance to the peak emission region is shorter for the flat-top pulse. This leads to an enhancement of EUV CE by almost a factor of 2.

3. Photoabsorption studies.

The EUV spectra of the elements from Z = 62 to 74 (i.e. samarium to tungsten) produced by focusing the output pulse of a Q-switched laser onto a solid target at power densities in the range 10^{11} to 10^{12} W cm^{-2} contain extensive regions of line free continua [17]. In particular the spectrum of samarium is line free from 3 –200 nm, while with increasing Z the long wavelength limit decreases, until at tungsten the continuum is line free from 3-13 nm although the line density is very low below 20 nm. Since the intensity of the recombination radiation scales with <\zeta>^4 where \zeta is the ionic charge, its importance is expected to increase with Z since the average ionic charge increases as the ionisation potentials are on average lower for a given \zeta with increasing Z and the ions spend more time in the interaction region because of their increased mass. However such behaviour cannot explain the absence of lines over extensive energy ranges in the heavier lanthanides since the extent of the continuum actually decreases with Z. Furthermore past Z = 74 lines are again present throughout the XUV in plasmas of all elements investigated up to and including uranium. It was shown that, because of the effects of 4f orbital collapse, 4f electrons are present in the ground configurations of all elements in the range 62 ≤ Z ≤ 74 in the ion stages produced in a typical Q-switched laser plasma where the emission is in general dominated by species with charge states ranging from 4+ to 16+ [17]. At the lower Z end of this group of elements the 4f binding energy in the neutral atom is lower than either the 5s or 5p but, because of the tendency of levels to regroup according to principal quantum numbers with increasing ionisation, the 4f binding energy is greater than either the 5s or 5p at the fifteenth or sixteenth ion stage [18]. At intermediate ion stages there is a near degeneracy first of 4f and 5p and later of 4f and 5s energies and as a result the ground configuration in most ion stages contains an open 4f subshell. These near degeneracies produce a vast array of low lying excited states which greatly increases the number of possible transitions. It was also shown that the statistics of the levels follow the same behaviour as compound nuclear states and may form a quantum chaotic system [19].
lines which appear in the spectra of higher Z elements of this group only do so because the 4f subshell is filled or contains at most one vacancy, and ground configurations of many stages involve only open 5s or 5p subshells which give rise to relatively simple spectra. From these considerations it was deduced that the transitions blended to form weak bands of quasi-continua which blended with and enhanced the recombination continuum. These plasmas were shown to provide a stable, compact and bright alternative source of XUV radiation to the synchrotron in experiments where a knowledge of absolute XUV flux or polarisation was not required. In this mode they have been used for applications that include photoabsorption studies using the dual laser plasma (DLP) technique.

The basis of the DLP technique is that a Q-switched laser (1 Hz, 0.8J in 45 ns) may be focused using a cylindrical or spherical lens onto the element of interest at a relatively low power density so that the maximum ion stage obtained does not radiate itself in the EUV while a second laser pulse is focused tightly onto a W or Sm target to yield a backlighting continuum. By varying the interplasma time delay or probing different plasma regions, the absorption of particular ion stages may be differentiated and optimized [20].

A typical result of an experiment on iodine photoabsorption at two different time delays is shown below [21] where they are compared with calculations using the Cowan code [22]. In obtaining the theoretical spectra, cross-sections were obtained for each term of the open 4d subshell calculations which were then weighted and summed assuming a Boltzmann distribution for a particular plasma temperature as described elsewhere [23]. This process was repeated for each ion stage and the contribution of each to the absorption profile then summed again assuming the same constant temperature. For all but closed shell species it is difficult to isolate a spectrum free of excited state absorption. The situation is particularly complex for open 4d and 4f ions and it was recently found to
be impossible to observe a simple spectrum in higher ions of the Xe isolectronic sequence again reflecting the effects of 4f and 5p level crossing.

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