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Cite as: J. Appl. Phys. 126, 113103 (2019); https://doi.org/10.1063/1.5113617
Submitted: 05 June 2019. Accepted: 04 September 2019. Published Online: 20 September 2019


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Characterization of plasma emission in the 1-6 nm band from laser-irradiated cryogenic xenon targets

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ABSTRACT
We present measurements from laser-produced plasmas generated using cryogenic Xe targets and quantify the emission characteristics in the soft x-ray region (1 to 6 nm). The system is based on a LN2-cooled rotating drum, which allows for a high repetition rate, and Nd:YAG laser systems with energies up to 325 mJ on-target with pulse lengths of 130 ps, 600 ps, or 6.5 ns. High resolution spectra are measured using a grazing incidence spectrometer, and we present the first quantitative conversion efficiency (CE) measurements for Xe in this range. Data show CE values up to \( \sim 1\% \) at 6 nm and \( \sim 0.08\% \) at 1.5 nm (for 2% bandwidth and 2\( \pi \) solid angle), and there are lower limits on the required laser intensities and energies on target to achieve these efficiencies. In addition, the emission spot size is directly measured at 2 nm (620 eV) using a point-projection slit imaging method, with optimized emission spot sizes of \( \sim 20\mu m \).

I. INTRODUCTION
The development of sources for Extreme Ultraviolet (EUV) lithography and inspection is at the forefront of technological research. The drive toward viable shorter wavelength sources to enable ever more compact component design or metrology capabilities continues to enable innovation. There are stringent requirements on such sources; high repetition rate and availability are needed for commercial wafer metrology systems, and high conversion efficiency (CE) at the desired wavelength reduces the cost of development. Low debris buildup and simple maintenance make systems more user-friendly and less expensive to operate. The basis of these systems is the EUV source, and the form of this dictates many of the operational characteristics of a system.

While discharge-based and related systems are viable, laser-produced plasma (LPP) systems have dominated developments in recent years given their flexibility, ease of implementation, and relatively low debris issues. The most successful method to date uses Sn droplets heated using a laser beam to give high laser-to-EUV conversion efficiencies in the 13.5 nm band (e.g., Refs. 1–4). Tin is heated to a liquid and a droplet system introduces molten droplets into a vacuum chamber at a rate typically in the tens of kilohertz range. A synchronized laser beam Irradiates each droplet, rapidly converting it into a plasma of sufficient temperature to emit in the EUV range. This radiation is collected by a multilayer mirror (MLM) for use in lithographic processing. Peak CE values of \( \sim 6\% \) have been achieved with so-called advanced targets, involving the use of lasers with a prepulse to optimize target properties on the arrival of the main pulse.6 While efficient, issues of molten Sn debris must be addressed with such systems to limit impacts on the operational downtime. In addition to EUV emission, there is considerable interest in the possibility of developing efficient sources at shorter wavelengths, referred to as BEUV, Blue-X, or 6.x nm emission for lithography.

Xenon targets are attractive in that reasonably high conversion efficiencies can be achieved in the EUV, with the advantage of chemical inertness limiting the damage to surrounding optics. Several approaches have been investigated including Xe gas puff targets,9 Xe cluster jets,10 Xe liquid jets and droplet,11,12 and solid Xe pellets.11 A solid is desirable given the higher density available to generate emitters in the EUV and the ability to clearly define a focal surface for the incident laser. To this end, Fukugaki et al.12 developed a cryogenic drum system as a high average power solid Xe-based EUV source designed to operate at 10 kHz. The growth rate of known ice thickness on the drum surface was examined in detail. Subsequently, Amano et al.13–15 examined the crater formation on the ice surface and conversion efficiency using an Nd-YAG...
laser operating at \( \sim 10^{18} \) W/cm\(^2\). The maximum achieved CE is 0.9\% at 13.5 nm with 2\% bandwidth into 2\% solid angle for optimal conditions. The same team also investigated the emission characteristic in the 5–17 nm regime with an emphasis on the conversion efficiencies at 13.5 nm, examined the Xe ion debris, and assessed the expected damage to collecting Mo/Si mirrors.

In this work, we use the same drum system to investigate the emission characteristics of the laser-produced plasma from a solid Xe target at wavelengths down to 1 nm, into the soft x-ray (SXR) regime, for the single shot operation. We examine the dependence of the conversion efficiency on the laser energy and pulse length in a fixed optical configuration designed to produce a suitable small laser focal spot. We present SXR spectral measurements, including conversion efficiency determination, followed by direct imaging measurements of the source size.

II. EXPERIMENTAL SETUP AND CRYOGENIC XE TARGET SYSTEM

A solid Xe target was generated using a rotating cryogenic drum system developed by Fukugaki et al. This comprises a liquid nitrogen cooled cylindrical drum maintained at 77 K onto which Xe is introduced at a fixed flow rate typically between 50 sccm and 150 sccm to allow solidification onto the drum surface. The thickness of the Xe layer is controlled by a series of wipers set at ~1 nm from the drum surface, and the drum was rotated at 100 rpm so that a fresh xenon surface is presented to each laser shot.

The primary laser used in this study is an Ekspla SL335 with a 1064 nm stimulated Brillouin scattering (SBS) compressed pulse of variable length (FWHM) between 130 ps and 600 ps. The measured maximum energy available at the target position is 325 mJ with a variable length (FWHM) between 130 ps and 600 ps. The measured spectral measurements of the source size.

III. YIELD AND SPECTROSCOPIC MEASUREMENTS

Spectroscopy and energy yield are recovered from the experiments simultaneously to allow the determination of the absolute energy emitted in each energy band. Along with the measured laser incidence energy, this allows the determination of the conversion efficiency at each wavelength over a given bandwidth, here taken as 2\% of the center wavelength studied. The emitted energy in the SXR range is measured by a calibrated AXUV100G diode (OptoDiode Inc) with various metallic filters (Lebow Company) which can be exchanged in vacuum using a filter set mounted on a translation slide. Transmission profiles for the filters used here are shown in Fig. 2(a). The filter slide is suitably baffled to ensure that the photon emission from the target does not circumvent the filter to impinge on the diode through small angle scattering. The diodes are fielded unbiased, and the recovered traces are integrated to determine the total energy emitted on each laser shot. Figure 2(b) shows a typical diode signal and integrated traces using an Fe filter.

Figure 2(c) shows the measured integrated diode signal using the Fe filter as the lens is scanned through the best focus position for experiments on 3 different days. The integrated signal is proportional to the total energy emitted in the region around 2 nm determined by the filter transmission [Fig. 2(a)]. The traces show the central dip at 0.0 nm in energy emitted, as the laser spot diameter is reduced and the laser intensity reaches its maximum. This dip is indicative of the best focal position of the lens in the presently measured spectral bands. Higher energy ion species are generated, and emission is reduced in the 2 nm waveband as emission increases at shorter wavelengths (<1 nm). These processes are discussed in detail in Refs. 13–17. The three repeated lens scans in Fig. 2(c) show the excellent repeatability and stability of the system over long-term operation. Error bars in the plot indicate the typical shot-to-shot variability of \( \sim 5\% \).

Spectral measurements are recovered using a McPherson 251MX spectrometer with a flat-field aberration corrected 2400 lines/mm grating. This gives a range of 1–6 nm with a 0.01 nm resolution using an angle-of-incidence of 87\°. The spectrometer slit is fixed at 20 \( \mu \)m, and the spectrum is imaged onto an Andor Newton CCD (2048 \( \times \) 512, 13 \( \mu \)m pixel size). A typical spectrum is given in Fig. 3.

Emission in the 1–2 nm region is dominated by Xe XXVII lines due to 3d \( \rightarrow \) 4s, 4p, 4f transitions, with contributions from
Xe XXVI \(4s \rightarrow 4p\) transitions. Xe XXVI \(4p \rightarrow 5f\) to, \(4f \rightarrow 6g\), and \(4d \rightarrow 5f\) transitions dominate in the 3–4 nm range, and Xe XXVI \(4f \rightarrow 5g\) and \(4d \rightarrow 5p\) lines dominate in the 5–6 nm range. There also appears to be some contribution from Xe XXIX lines due to transitions within the 3d shell in the 4–6 nm range. The sharp drop in emission at \(\sim 4.3\) nm is likely due to either some small carbon contamination in the system, which has an absorption edge at this wavelength, or self-absorption in the plasma itself. Due to this, the analysis presented below avoids using this small spectral band for comparisons.

To determine the absolute conversion efficiency of laser energy to SXR emission in a given waveband, the spectra are cross-calibrated with the diode energy yield measurements. Diode signals and spectra are taken on the same shots using nominally identical filters, which is repeated for all four of the filters presented in Fig. 2(a). The integrated diode signal is combined with the diode spectral response curve (e.g., Refs. 18–20) to give the total energy emitted for each shot. This is used to calibrate the filtered spectrum, giving values in terms of energy per count recorded on the spectrometer charged coupled device (CCD). The calibration was carried out for four fixed bands in the range from 1 to \(\sim 4\) nm. For coverage of the whole spectral range of the spectrometer (from 1 to 6 nm), we used spectral curves of the grating efficiency and spectrometer CCD for this band. This is repeated over several shots with varying energy on the target for each of the four filters to give an average calibration value. The energy yield in any waveband can then be divided by the incident laser energy to determine the absolute conversion efficiency as a function of wavelength. Emission is assumed to be isotropic into \(2\pi\) solid angle. Here, we use our most conservative calculations of the calibration factors and so present minimum values for the CE in each case. We can then examine the change of the CE with the laser parameters and lens position to determine the peak possible efficiencies as well as the dominant requirements for achieving them for a given waveband.
An extensive set of spectra was obtained using a range of laser pulse lengths, on-target energies, and at different lens focal positions. Figure 4 shows CE plots for pulse lengths of 130 ps, 600 ps, and 6.5 ns as the lens position is scanned, using a fixed laser on-target energy of ~200 mJ. Note that the best CE values for long wavelengths are found slightly on either side of the best focus position, consistent with Fig. 2(c).

The maximum CE at 6 nm (~200 eV) is about 1%, and for 1.4 nm (870 eV), the best observed CE is ~0.08%. The variation with wavelength is approximately the same for both the 130 ps and 600 ps data, with no notable differences in the spectra, despite the 5x increase in the laser intensity. The 6.5 ns data are significantly different, however. At 6 nm, the CE is reduced by a factor of 10%–0.1%, and at 1.4 nm, the CE is very close to zero. The high ionization states emitting in the 1–2 nm region for the 130 ps and 600 ps data are not present in the plasma generated with the 6.5 ns laser, even with 200 mJ of energy on-target.

We can also examine the variation of the conversion efficiency with laser energy at a fixed, optimal lens position. This is shown for the 130 ps and 600 ps pulse length shots in Fig. 5. Data for the 6.5 ns pulse lengths are not included here due to the low conversion efficiencies at all lower wavelengths.

Trends for both the 130 ps and 600 ps are again similar, but differences are noted for different wavelengths as the laser energy is scanned. For the 5.8 nm band, the CE increases as the laser energy rises. After ~100 mJ, the CE remains approximately constant close to the peak value of ~1%. For the 1.42 nm waveband, the CE continues to rise until at least 200 mJ and only begins to tend to a constant value above this energy. It is also interesting to note that the curve for the 1.42 nm and 2.5 nm wavebands cross at ~125 mJ for both pulse lengths.

To reach the peak CE values observed here, there are clear requirements for the laser system, at least using the optical deliver chain implemented here. The longer pulse length reduces the laser intensity to peak values of $6 \times 10^{12}$ W/cm$^2$ (assuming a 25 μm spot size—see Sec. IV), and CE values are strongly reduced. The 130 ps and 600 ps pulse lengths produce peak intensities up to $3 \times 10^{13}$ W/cm$^2$ and $7 \times 10^{13}$ W/cm$^2$, respectively. The CE of the shorter pulses drops rapidly as the on-target energy is reduced, which reduces the on-target intensity. We can suggest here that intensities at the target must be $>10^{13}$ W/cm$^2$ to produce emission in the 1–2 nm region with the maximum efficiency. We can also note that significantly larger energy on the target is required to generate strong emission (i.e., highest CE values) in the 1–2 nm range than in the 5–6 nm range independent of the pulse length. This again sets requirements for the laser system, which depends on which emission wavelength is desired for an application.

The conversion efficiencies measured at 6 nm are comparable to those achieved at 13.5 nm by Amano et al.21 who obtained 0.9% using a 10 ns, 1 J, 1064 nm laser pulse with the same cryogenic Xe system. We have shown similar CE values at 6 nm using only 200 mJ with both short pulse beams. Since our data using a long pulse showed poor conversion efficiency at 6 nm (0.1%), this suggests that a shorter pulse length is beneficial for the SXR generation. It is also interesting to note that Amano et al. in Ref. 21 also
suggest a minimum energy requirement of 300 mJ for the peak conversion efficiency at 13.5 nm using a 24 ns laser pulse length.

At present, several methods are being examined for use in the region around 6.7 nm (often termed as 6x nm). These are based on the molten metallic droplet method developed for Sn but using higher atomic number materials such as gadolinium and terbium amongst others (e.g., Refs. 22 and 23). Simulation results presented in Ref. 23 show that for laser intensities up to $5 \times 10^{12}$ W/cm$^2$ the peak CE for both Tb and Gd is $\sim 0.9\%$. While experimental data have been presented for these materials, CE has not been directly calculated. Additionally, at present, no quantitative experimental data are available for these systems at wavelengths down to 1 nm to compare to our Xe results in the present work.

**IV. SOURCE SIZE MEASUREMENTS**

Typically, the source size in the XUV or SXR region is determined from direct imaging. Here, we use a slit imaging method,$^{24-27}$ which uses the generated emission spot to radiograph a target of known dimensions at high magnification. The advantage is that the slit method allows the direct determination of the spot imaging capabilities in the spectral region of interest using a suitable filter arrangement. This avoids the use of imaging components in which optic component aberrations limit resolution, and multiple reflection (e.g., from an MLM) are not required, giving a high contrast image for analysis. Additionally, the spatial scale in both the horizontal and vertical directions can be determined simultaneously directly from the spot radiation, avoiding the possible asymmetric distortion of the relay element in imaging. As with direct imaging, the largest error is the determination of the system magnification and the resolution of the detector at the desired magnification.

The diagnostic comprises a rectangular aperture across which a 100 $\mu$m diameter wire was mounted vertically and a 200 $\mu$m diameter wire mounted horizontally (see Fig. 6). This arrangement was 11 cm from the Xe surface with the camera a further 57 cm from the Xe surface and filtered with a 1 $\mu$m Fe foil. The illumination generated as the laser is incident on the Xe ice backlights the imaging target, casting a shadow onto the CCD (Andor Newton) as a point projected image in a radiation band around 2 nm. For this point-projection arrangement, the magnification is equal to $M = (p + q)/p$, where $p$ is the distance from the Xe target to the imaging object ($p = 11$ cm), and $q$ is the distance from the imaging object to the CCD ($q = 57$ cm) [Fig. 6(a)]. The magnification is $\times 6.2$ for the present system.

The source size is expected to be of the order of tens of micrometers, and so diffraction effects at the imaging object edges are not expected. In the point projection, the diffraction limit is equal to $\Delta \lambda = f(\pi p/(p+q))^{1/2}$, which is typically simplified to $\Delta \lambda = f(\lambda p)^{1/2}$ for large magnifications (i.e., $q > p$). From the experimental setup, this gives $\Delta \lambda \sim 14$ $\mu$m, where the wavelength $\lambda$ is taken as 1.8 nm, which is the peak transmission wavelength through the Fe filter used in the imaging [Fig. 2(a)]. The imaging objects are several times the expected source size and are opaque to the imaging radiation. As the laser focusing lens is scanned toward the best focus position [at 0.0 mm in Fig. 6(b)], it is clear that the transmission profile across the edges in the image can be used to determine the size of the source producing the image. There are multiple positions at which this can be carried out for the images, and one lineout position across the 100 $\mu$m wire is shown in Fig. 6(c). The spatial separation on the detector of the 10% and 90% transmission point multiplied by the magnification is approximately equal to the source size. Strictly, the source size $d = \Delta x \times p/q$, where $\Delta x$ is the 10%–90% transmission point dimension. $^{24-27}$ It can be seen that the transmission profile for the 0.6 mm image lineout is significantly different than that for the 0.0 mm image. Measurements show that the source size has reduced from 77 $\mu$m to 24 $\mu$m as the lens is brought toward the best focus. Note that for an approximately Gaussian beam profile, taking the 10%–90% transmission points recovers a source size...
which approximately represents the 1/e diameter (or ~0.75 times the 1/e^2 diameter) of the emitting region. Often, it is useful to quantify the emission spot dimensions in both the horizontal and vertical directions, to examine whether the incident laser produced a suitably round spot. From the images in Fig. 6, this is readily achieved by taking multiple lineout analyses both horizontally and vertically. The results of this process are shown for a 300 mJ, 130 ps experiment where the lens is scanned through the best focus region [Fig. 7(a)]. Data are the result of an averaging of 4 lineouts in each direction. Note that far from focus, the emission spot is larger in the horizontal direction than the vertical, due to a slight beam asymmetry. Close to focus, both dimensions are similar and give a minimum spot size of ~20 μm diameter round spot. The error in the measurement is due to the determination of the magnification, the pixel resolution across the image used to describe the profiles analyzed, and the scatter in data.

FIG. 6. (a) Setup for point projection imaging, (b) example images at different lens positions along with (c) lineouts showing the determination of the emission spot size for 300 mJ, 130 ps shots (28 November 2017). Plots are scaled by magnification.

FIG. 7. Plots of source size (1/e^2 diameter) as a function of lens position (both vertical and horizontal) and averaged source diameter with pulse length and energy.
from different lineouts on the same image. Typically, errors are no better than 5%. Note that the horizontal measurements are corrected for the 25° viewing angle of the diagnostic.

To compare across different laser parameters during lens position scans, we average the vertical and horizontal measurements into a single measurement for each scan. Given the differences in the spot dimensions far from focus, this gives larger errors here but still allows a meaningful comparison. In Fig. 7(b), we show emission spot measurements from a 130 ps scan at 300 mJ, a 130 ps scan at 50 mJ, and 600 ps scan at 300 mJ to examine the change in dimensions with laser energy and pulse length. In general, the energy on target does not affect the emission spot size between 50 mJ and 300 mJ, as data overlap inside error bars. There may be a very small dependence on pulse length, emission spot sizes may be slightly larger with 600 ps vs 130 ps pulses, but this is very close to error bar overlap throughout the plot. Generally, no strong dependences on laser parameters are observed. For an idealized input laser beam, the optical arrangement used (F# ~ 4.4), gives a Rayleigh range of ~100 μm. From the plots in Fig. 7, the observed average Rayleigh range is 280 μm, likely due to the poor beam symmetry. The direct imaging of the incident laser spot size also reports minimum 1/e² diameters of ~20 μm, and so the emission spot size appears approximately correlated to the incident laser spot at intensities at the incident laser intensities investigated here.

V. CONCLUSIONS

We have presented calibrated spectra and absolute conversion efficiencies for cryogenic Xe targets in the soft x-ray (1–6 nm) region for the first time. Conversion efficiency values range from ~1% at 6 nm (~200 eV) to ~0.08% at 1.4 nm (870 eV) for the rotating drum system employed here. Additionally, minimum laser energy was required to achieve these CE values for the measured minimum laser spot diameter (1/e²) of about 20 μm, which varied with the emission wavelength. Approximately, 100 mJ was required on target for peak CE at 6 nm, while ~200 mJ was required at 1.4 nm. Both the CE values and the required energy on target were consistent with pulse lengths of 130 ps and 600 ps, despite the 5x change in intensity at the target surface. This contrasts with data at the same energy but longer pulse length, which showed strongly reduced CE in all wavelengths. These observations suggest that a certain incident laser intensity is required in the SXR for a (~10¹³ W/cm²), and that above this minimum, the energy delivered to the target is a more important factor in generating the high ionization state required to provide efficient emission at wavelengths down to 1 nm. The emission spot size at 2 nm (620 eV) was directly determined by a point projection slit analysis method. The measurements report the observed asymmetry in the initial laser beam but show an approximately round focal spot with diameters down to ~20 μm at the best focus. The dimensions of the spot size showed little dependence on the laser pulse length or energy on target, which were the same within errors.

The data presented in this work suggest that solid xenon targets may provide a feasible alternative to high atomic number metallic droplet systems in the SXR region. Conversion efficiencies are already similar to those expected from Tb and Gd systems currently being investigated at 6 nm (called 6× nm sources) and values approach 0.1% for 1.4 nm. Future work will seek to optimize this system further and investigate the ion spectrum to assess debris issues.

ACKNOWLEDGMENTS

This work is funded by the KLA Corporation under Contract No. 209114251.

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