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Brilliance improvement of laser-produced extreme ultraviolet and soft x-ray plasmas based on pulsed gas jets

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Two methods improving the brilliance of laser-induced plasmas emitting in the extreme UV (EUV) and soft x-ray (SXR) regions were investigated, using three different gases (nitrogen, krypton, and xenon) from a pulsed gas jet. Utilizing a newly designed piezoelectric valve, up to almost ten times higher gas pressures were applied, resulting in increased target densities and thus, higher conversion efficiencies of laser energy into EUV and SXR radiation. Secondly, geometrically reducing the angle between the incoming laser beam and the observed plasma emission minimizes reabsorption of the emitted short wavelength radiation. Combining both methods, the source brilliance is increased by a factor of 5 for nitrogen. Furthermore, a compact EUV focusing system for metrological applications is presented utilizing the optimized plasma source. An energy density of 1 mJ/cm² at wavelength λ = 13.5 nm in the focal spot of an ellipsoidal mirror is achieved with xenon as the target gas being sufficient for material removal of PMMA samples with an ablation rate of 0.05 nm/pulse. © 2019 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).

I. INTRODUCTION

Extreme ultraviolet (EUV) sources based on laser-produced plasmas (LPP) emitting at a wavelength of 13.5 nm are currently developed with tremendous effort for the next generation of semiconductor microlithography.¹,² Besides high power sources for high volume manufacturing, compact EUV sources of lower average power are also needed for metrology purposes, e.g., for mask inspection, actinic material testing, or optics and sensor characterization. Moreover, tabletop LPP sources are also employed to generate soft x-ray (SXR) radiation for microscopy³,⁴ or absorption spectroscopy⁵,⁶ (NEXAFS) in the water window spectral range (2.2–4.4 nm). A variety of different types of targets are used for LPP sources, as there are solids,⁷ including cryotargets, liquid jets,⁸,⁹ and mass limited droplets,¹⁰ as well as pulsed gas jets.¹¹,¹² Although plasmas generated from gas jets are generally less brilliant due to their lower particle density as compared to liquid or solid targets, such sources are of particular interest for metrological applications due to their simplicity and compactness. Moreover, although the laser plasma is ignited very close to the nozzle, almost no debris is generated since the gas flow protects the nozzle against erosion from the plasma, accomplishing a clean and long-term stable operation. For these reasons, several efforts have been undertaken to enhance the target particle density and therefore the brilliance of gas jet based LPPs without waiving its inherent advantages. Fiedorowicz et al.¹³ developed a double-stream nozzle where an outer helium jet confines the main target gas jet. Mey et al.¹⁴ investigated the expansion of a gas jet into a moderate helium atmosphere, creating a barrel shock with an increased particle density.

Also, experiments with a prepulse-induced shock wave to enhance the target gas density have been performed.¹⁵ However, there is definitely the need for further improvements to increase the conversion efficiency from laser to EUV or soft x-ray radiation. Additionally, the photon yield of gas jet based laser plasma sources is reduced, as the short wavelength radiation is absorbed by the gas itself.

Thus, in this paper, two different approaches are pursued to enhance the brilliance of a gas jet based laser plasma. At first, the target particle density is increased by raising the applied gas pressure at the nozzle. Secondly, reabsorption in the flowing target gas jet is minimized geometrically by reducing the angle between the incoming laser beam and the direction under which the generated short wavelength radiation is utilized. The optimized EUV laser plasma was focused with the help of an ellipsoidal mirror, generating a focal spot suitable for surface modification of a PMMA sample at a wavelength of 13.5 nm.

II. ENHANCEMENT OF SOURCE BRILLIANCE

A. Experimental setup

Short wavelength radiation in the extreme UV or soft x-ray spectral region is emitted from a laser-produced plasma source [see Fig. 3(a)], utilizing a Nd:YAG laser (Innolas, wavelength 1064 nm, pulse energy 600 mJ, pulse duration 6 ns) being focused onto a pulsed gas jet target by a planoconvex lens (focal length 100 mm). The setup has been described in detail elsewhere.¹⁶ The spectral emission properties of the plasma depend on the target gas: SXR radiation between 1 and 5 nm [see Fig. 1(a)] can be obtained by using either nitrogen (narrowband) or krypton (broadband); for broadband EUV radiation [5–20 nm, see Fig. 1(b)], xenon is chosen.¹⁷

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The pulsed gas jet is provided by a fast valve with a conically diverging nozzle (300–550 μm opening diameter, thickness 1 mm). In contrast to previously used piezodisc translators (Physik-Instrumente, P-286.72), the newly developed valve operates with a stronger and faster piezoelectric rod actuator (P-842.60), allowing for almost tenfold higher gas pressures (p_g up to 200 bar) at a two times shorter opening time (t_open = 450 μs).

Spatial distribution and intensity of the plasma are monitored by an EUV and SXR sensitive pinhole camera (pinhole diameter 50 μm, phosphor-coated CCD chip, Sony ICX285, pixel size 6.45 μm, 1280×1024 pixels), placed under an angle of either 90° or 20° referred to the incoming laser beam, respectively [see Fig. 3(a)]. The pinhole is equipped with either a zirconium (EUV) or titanium (SXR) foil (thickness 200 nm) to block out-of-band radiation. Brightness measurements are conducted with a calibrated photodiode (IRD, SXUV 100). The particle density distribution of the gas jet is quantified with the help of a Hartmann–Shack sensor, measuring the wavefront deformation of a collimated test beam after transmission through the jet. From this, the refraction index distribution can be calculated, yielding the spatially resolved particle density by using the Lorentz–Lorenz equation [see Fig. 2(a)].

B. Results and discussion

The results of particle density and brightness measurements for a nitrogen plasma (λ = 2.88 nm) at different gas pressures of up to 175 bar are compiled in Fig. 2, indicating a linear increase of particle density with pressure. In contrast, the enhancement of the plasma intensity saturates at gas pressures >75 bar. This can be attributed to two different effects:

1. Reabsorption of the emitted short wavelength radiation within the surrounding gas jet.
2. Reflection of laser radiation at the over-dense plasma.

With higher-Z gases like krypton or xenon as the laser target, the plasma intensity already stagnates at around 30 bar due to an even stronger reabsorption and higher electron densities, resulting in an enhanced back-reflection of laser radiation. Thus, to further increase the source brilliance, reabsorption is reduced by minimizing the optical path length through the gas jet, utilizing the radiation emitted in the direction of the incoming laser beam. Corresponding pinhole camera images of a xenon plasma recorded at 90° and 20° are shown in Figs. 3(b) and 3(c). Lateral and axial positions of the laser beam were optimized for both angles to achieve maximum plasma intensity.

Fig. 1. Integrated emission spectra (p_g = 10 bar) over 100 pulses of (a) nitrogen and krypton and (b) xenon.

Fig. 2. (a) Density distributions of the nitrogen jet at gas pressures of 25 and 75 bar, respectively. (b) Pressure dependence of particle density (nitrogen, 500 μm below the nozzle) and plasma brightness at λ = 2.88 nm.

Fig. 3. (a) Schematic drawing of the laser-produced plasma source and pinhole camera images of a xenon plasma (distance to nozzle 1 mm, gas pressure 20 bar, Ti-filtered, averaged 10 pulses) taken under an angle of (b) 20° and (c) 90° referred to the incoming laser beam. (d) Centroid positions of 500 xenon plasma pulses recorded for both angles.
For all three investigated gases, emission in the backward direction is strongly favored: The effective source area of the plasma becomes significantly smaller; intensity and positional stability [see Fig. 3(d)] are increased (see Table I). In total, the brilliance is enhanced by a factor of 2–4 depending on the target gas. By combining the geometrical improvement with a higher gas pressure, the brilliance is increased even up to a factor of 5 for nitrogen. These results are in qualitative agreement with earlier investigations by Kranzusch et al. 12 at about ten times lower xenon gas pressures.

### III. EUV APPLICATION

With the new setup, ablation and irradiation experiments at a wavelength of 13.5 nm are conducted with xenon as the target gas. The experimental arrangement is illustrated in Fig. 4(a). It consists basically of the plasma source described above utilizing an emission angle of 20° and an ellipsoidal mirror for focusing the EUV light into the sample plane.

Using a differential pumping system, the background pressure is reduced by two orders of magnitude from approximately 10\(^{-3}\) mbar in the source chamber during operation (gas pressure \(p_G = 20\) bar, repetition rate 5 Hz) to approximately 10\(^{-5}\) mbar within the optics and sample chamber. Narrowband radiation at \(\lambda = (13.55 \pm 0.26)\) nm is obtained by a plane Mo/Si multilayer mirror [optiX fab, AOI 25°, \(R = 58\%\), bandwidth (BW) = 3.8\%] and a 150 nm Zr-filter \([T = 59\% \text{ at } \lambda = 13.5\text{ nm} (\text{Ref. 19})]\). An axisymmetrical ellipsoidal mirror (Rigaku, Ni-coated, \(\text{NA}_{in} = 0.044, M = 0.62\)) on a 5-axes mount, adjustable from outside the vacuum, collects and focuses the radiation into the sample plane \([z = 0 \text{ mm}, \text{see Fig. 5(a)}]\) where a target (e.g., polymers, metals, etc.) can be placed. To adjust the ellipsoid, intensity profiles were measured along the optical axis (\(z\)-axis) around the focal plane [see Fig. 4(b)] utilizing a phosphor-coated CCD chip (as in pinhole camera). To avoid overexposure of the camera, the EUV radiation was sufficiently attenuated by replacing the Zr-filter with a Ti-filter [thickness 400 nm, \(T = 0.05\% \text{ at } \lambda = 13.5\text{ nm} (\text{Ref. 19})\)]. Source energy is measured with a calibrated photodiode (IRD, SXUV100) behind the EUV mirror. The pulse-to-pulse stability amounts to approximately 2\% (standard deviation). Parameters of the source and focusing system are listed in Table II.

### Table I. Plasma characteristics for different target gases \((p_G = 20\) bar\) under emission angles of 20° and 90°, respectively, measured with a pinhole camera. The distance from the plasma to the nozzle is about 500 \(\mu\text{m}\) for nitrogen and 1 mm for krypton and xenon.

<table>
<thead>
<tr>
<th>Gas</th>
<th>(N_2)</th>
<th>Kr</th>
<th>Xe</th>
</tr>
</thead>
<tbody>
<tr>
<td>Angle (deg)</td>
<td>20</td>
<td>90</td>
<td>20</td>
</tr>
<tr>
<td>Total energy (MCnts)</td>
<td>8.1</td>
<td>8.9</td>
<td>2.5</td>
</tr>
<tr>
<td>Maximum energy (kCnts)</td>
<td>12.3</td>
<td>7.5</td>
<td>4.1</td>
</tr>
<tr>
<td>Plasma size x, FWHM ((\mu\text{m}))</td>
<td>240</td>
<td>510</td>
<td>255</td>
</tr>
<tr>
<td>Plasma size y, FWHM ((\mu\text{m}))</td>
<td>190</td>
<td>175</td>
<td>195</td>
</tr>
<tr>
<td>Area FWHM (Ellipse) ((\text{mm}^2))</td>
<td>0.036</td>
<td>0.07</td>
<td>0.039</td>
</tr>
<tr>
<td>Pointing stability x ((\mu\text{m}))</td>
<td>18.5</td>
<td>48.2</td>
<td>8.4</td>
</tr>
<tr>
<td>Pointing stability y ((\mu\text{m}))</td>
<td>10.1</td>
<td>13.8</td>
<td>3.4</td>
</tr>
<tr>
<td>Brilliance improvement factor</td>
<td>1.8</td>
<td>4.2</td>
<td>2.1</td>
</tr>
</tbody>
</table>

### Table II. EUV plasma source and focusing system parameters at \(p_G = 20\) bar.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength (\lambda)</td>
<td>13.5 nm</td>
</tr>
<tr>
<td>Plasma size x, y (FWHM)</td>
<td>350 (\mu\text{m}), 280 (\mu\text{m})</td>
</tr>
<tr>
<td>EUV source peak brilliance ((t = 5) ns) (a)</td>
<td>(1.2 \times 10^{13}) photons/s mm(^2) mrad(^2) 3.8% BW)</td>
</tr>
<tr>
<td>Focal spot size (FWHM)</td>
<td>245 (\mu\text{m})</td>
</tr>
<tr>
<td>Focal spot energy density</td>
<td>1 mJ/cm(^2)</td>
</tr>
</tbody>
</table>

\(a\)Reference 20.
To prove the suitability of the compact setup for EUV ablation experiments, a cleaned PMMA sample positioned in the focal plane was irradiated with 3000 EUV pulses at an energy density of 1 mJ/cm², causing a perceptible surface modification. The analysis by means of a white light interferometer shows a 145 nm deep crater of 230 μm width (FWHM). This shape fits well to the intensity distribution in the focal plane [see Figs. 5(b) and 5(c)]. As the induced photon energy of 92 eV is much higher than the binding energy of PMMA (3–7.7 eV), every photon is able to break multiple bonds. However, since the energy density is below the ablation threshold of 10 mJ/cm² for PMMA, photon-induced material removal is achieved by a statistical desorption process. Its yield is proportional to the EUV intensity, explaining the very good agreement between the crater shape and the EUV beam profile. The ablation rate of about 0.05 nm/pulse is slightly higher compared to values found in previous experiments by Barkusky et al., using a Schwarzschild focusing optic, which results in a different intensity distribution and a smaller focal spot size.

IV. SUMMARY AND CONCLUSIONS

We have presented a compact laser-induced EUV/SXR plasma source with improved brilliance, which can be applied for various metrological applications including material interaction studies and surface modification. Brilliance enhancement has been achieved by an increased target gas density using a newly designed valve. However, stagnation occurs at higher pressures due to a stronger reabsorption of EUV radiation by the plasma surrounding gas and reflection of incoming laser radiation at an over-dense plasma. Minimizing the angle between observation and incoming laser beam reduces reabsorption. Moreover, the plasma becomes smaller and its lateral pointing stability improves. Depending on the target gas, the brilliance is increased by a factor of 2–5. Based on these results, an EUV focusing setup has been developed, characterized, and successfully applied to the structuring of a PMMA sample. In summary, we have demonstrated an EUV and SXR laser plasma source of improved brilliance and stability, maintaining its inherent cleanliness and compactness.

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