

EUV Emission from a Regenerative Liquid Target Laser-Produced Plasma^{*)}

Tatsuya SORAMOTO¹⁾, Takeru NIINUMA¹⁾, Hiroki MORITA¹⁾, Shinichi NAMBA²⁾
and Takeshi HIGASHIGUCHI¹⁾

¹⁾*Department of Electrical and Electronic Engineering, Utsunomiya University, Utsunomiya 321-8585, Japan*

²⁾*Department of Advanced Science and Engineering, Hiroshima University, Higashihiroshima 739-8527, Japan*

(Received 15 July 2024 / Accepted 23 October 2024)

We demonstrated the efficient water-window extreme ultraviolet (EUV) source using a regenerative liquid metal target irradiated by a 10-Hz, 150-ps, solid-state laser at a wavelength of 1.064 μm . A regenerative liquid bismuth (Bi) target with a diameter of 30 μm was continuously injected into a vacuum. The number of photons was about 1×10^{13} photons/(sr · shot) in the spectral region from 2.3 to 4.4 nm, which was attributed to be $n = 4 - n = 4$ ($\Delta n = 0$) and $n = 4 - n = 5$ ($\Delta n = 1$) transitions. The fast ion was also observed with a maximum energy of 140 keV from a hot, dense Bi plasma.

© 2025 The Japan Society of Plasma Science and Nuclear Fusion Research

Keywords: laser-produced plasma, soft x-ray, water window, highly charged ion (HCI), bismuth

DOI: 10.1585/pfr.20.2406013

1. Introduction

Water window (WW) extreme ultraviolet (EUV) emission, ranging from the oxygen K-edge (O_K -edge) at 2.3 nm to the carbon K-edge (C_K -edge) at 4.4 nm, is of great interest due to its importance for high resolution *in vivo* microscopic biological structure imaging of samples such as cells and macromolecules [1]. A WW-EUV microscope has been demonstrated with high resolution [2] with a coherent beam [3]. In addition, a compact, laboratory-scale microscope has been presented, coupling with a laser-produced low atomic number (low-Z) plasma WW-EUV source [4]. Another field, EUV lithography, has recently been worked. One of the activities is the proposal of the shorter wavelengths as the next feasibility, such as beyond EUV (B-EUV) at a wavelength of 6.x nm and WW-EUV, so-called “Blue-X” [5]. The WW-EUV source is also helpful for bio-imaging and next-generation shorter wavelength EUV lithography.

A laser-produced plasma is a compact, high-brightness source on a laboratory scale. However, operating long-term for various applications, such as spectroscopy, imaging, and lithography, is challenging. One of the effective targets is the use of a regenerative liquid target, such as a liquid jet and a droplet train. Low-Z elements, such as ethanol [6] and liquid nitrogen [7], are practical for the WW-EUV source, coupling with a microscope and reflectometry. The regenerative liquid metal tin (Sn) target [8], on the other hand, is a promising technology for EUV lithography at a wavelength of 13.5 nm, which is matched with a maximum reflective coefficient of 70%

of the Mo/Si multilayer mirror [9]. Note that the melting point of Sn is 505 K.

A laser-produced low-Z plasma is applied to the microscope with continuous multi-shot operation, because the shot-by-shot output flux is low due to the narrowband feature of the line emission. To overcome the low shot-by-shot output flux, we have demonstrated a WW-EUV source using unresolved transition array (UTA) spectra [10]. One of the candidate high-Z elements is bismuth (Bi), because its UTA emission spectrum consists of many resonant lines that give rise to a feature greater than 0.5 nm in extent with a peak wavelength of 4 nm [11]. The use of the planar Bi target, on the other hand, is limited in the number of shots due to the surface damage caused by the laser ablation. A regenerative liquid target provides flesh-dense target material continuously. We propose the use of a regenerative liquid Bi target, which has a lower melting point of 545 K, with a diameter of 30 μm continuously injected into a vacuum. Since there is no report of the regenerative liquid Bi target plasma WW-EUV source, we focus on the feature of the EUV emission with fast ion as debris from a laser-produced Bi plasma at a repetition rate of 10 Hz for future WW-EUV microscope and shorter wavelength “Blue-X” short wavelength EUV lithography.

In this paper, we report on the continuous efficient WW-EUV source using a 30- μm diameter regenerative liquid Bi target irradiated by a solid-state laser pulse at a wavelength of 1.064 μm . The energy spectrum of the fast ion is also observed.

author's e-mail: higashi@cc.utsunomiya-u.ac.jp

^{*)} This article is based on the presentation at the 26th International Conference on Spectral Line Shapes (ICSLS2024).

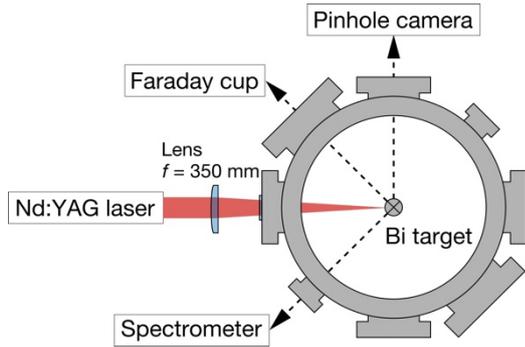


Fig. 1 Schematic diagram of the experimental setup.

2. Experimental Setup

A schematic diagram of the experimental setup for the WW-EUV source is shown in Fig. 1. The liquid Bi target was formed inside a vacuum chamber. A Q -switched Nd:YAG laser at a wavelength of $1.064\ \mu\text{m}$ produced a maximum pulse energy of 200 mJ with a pulse width of 150 ps [full width at half-maximum (FWHM)]. After expanding the beam diameter, the laser pulse was focused on a liquid Bi target using a lens with a focal length of 35 cm. The laser pulse was focused on about 1.5 cm from the nozzle exit. The diameter of the liquid Bi target was $30\ \mu\text{m}$ at the laser-irradiation point. The focused spot size was evaluated to be $35\ \mu\text{m}$. The laser intensity was varied by adjusting the laser pulse energy with a fixed laser spot diameter. The maximum laser intensity was about $1 \times 10^{14}\ \text{W}/\text{cm}^2$.

We observed the spectra using a flat-field grazing incidence spectrometer with an unequally ruled 2400 grooves/mm grating. The spectrometer and a Faraday cup were positioned at 45° with respect to the incident laser axis. Time-integrated EUV spectra were recorded by a thermoelectrically cooled back-illuminated x-ray charge-coupled device (CCD) camera (Andor Technology). The typical spectral resolution was better than 0.01 nm. The spectrometer has been calibrated [12]. The source image was observed using an EUV pinhole camera with a 500-nm Ti filter, which was positioned at 90° with respect to the incident laser axis.

3. Results and Discussion

We show the time-integrated EUV spectra at four different laser intensities, as shown in Fig. 2. The number of photons was observed to be about 1×10^{11} photons/(0.01 nm · sr · pulse) at the peak wavelengths of 3.9 and 4.2 nm, and arises from $n = 4 - n = 4$ ($\Delta n = 0$) transitions, and about 0.4×10^{11} photons/(0.01 nm · sr · pulse) at a peak wavelength of 2.4 nm, which can be attributed to $n = 4 - n = 5$ ($\Delta n = 1$) transitions at a laser intensity of about $1 \times 10^{14}\ \text{W}/\text{cm}^2$. The spectrum consists of strong UTA emission around 4 nm, which is mainly due to $n = 4 - n = 4$ transitions from ions with an open 4f or 4d outermost subshell and many small peaks between 2

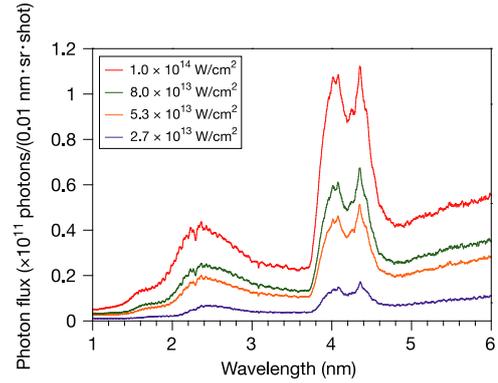


Fig. 2 Laser intensity dependence of the time-integrated EUV spectra.

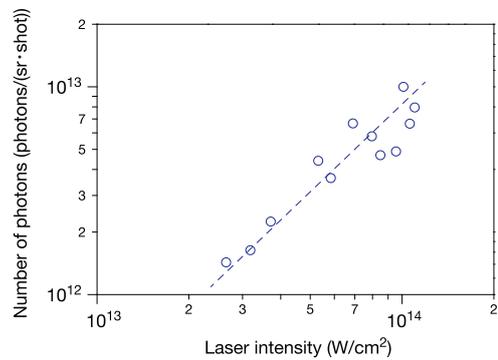


Fig. 3 Laser intensity dependence of the WW-EUV spectrum-integrated photon numbers.

and 3.5 nm, due to $n = 4 - n = 4$ transitions from multi-charged state ions with an outermost 4f subshell [13]. The position of the $n = 4 - n = 4$ transition peak is observed to be essentially unchanged with increasing the ion stage; however, the mean position of the $n = 4 - n = 5$ peaks shift towards a shorter wavelength with increasing laser intensity because of the increase of electron temperature and ionic charge state. The emission in 2 – 3.5 nm originates primarily from 4f – 5g transitions in ions with an open 4f subshell. The strong UTA emission around 4 nm comes mainly from 4p – 4d and 4d – 4f transitions in ions with an open 4d subshell. The emission at wavelengths longer than 4.2 nm includes a contribution from 4d – 4f emission from lower stages with an open 4f outermost subshell. This emission may be associated with the recombining phase of the expanding plasma.

The laser intensity dependence of the wavelength-integrated number of photons is plotted in Fig. 3. The number of photons increases with increasing the laser intensity. In the present case, the average number of photons produced by a single pulse was evaluated to be about 1×10^{13} photons/(sr · shot) in the range of 2.3 – 4.4 nm from a quasi-mass-limited metal target plasma, resulting in one-order lower output photon flux than from a planar

target (non-mass-limited) in single-shot operation. Note that a decrease in the number of photons is expected in the plasma volume due to the quasi-volume limited target at a laser irradiation point. Here, the average number of photons at a repetition rate of 10 Hz was comparable to that from the previous single-shot planar target source. The continuous EUV source can be extended with current thin-disk and thin-rod laser technologies for high repetition rate operation [14, 15]. The energy conversion efficiency (CE) of the integrated emission in the WW-EUV wavelength (2.3 – 4.4 nm) is estimated to be 0.3%/(sr · shot), corresponding to a CE of 1.9%/pulse in a solid angle of 2π sr for lithography evaluation. The exposure time in the x-ray CCD camera in the spectrometer was 1 s at the laser repetition rate of 10 Hz. Therefore, Figs. 2 and 3 show the averaged number of photons in 10 shots. The present source size is measured to be 30 μm (FWHM) (vertical) and 25 μm (FWHM) (horizontal) by the EUV pinhole camera, as shown in Fig. 4. The source image in Fig. 4 was time-integrated in the exposure time of 1 s.

Figure 5 shows the energy spectrum of the fast ions emitted from the plasma obtained using a Faraday cup at the maximum laser intensity of about 1×10^{14} W/cm². This evaluation provides collector mirror damage information in microscopy and lithography applications. The spectral shape is essentially thermal, and this spectrum includes the

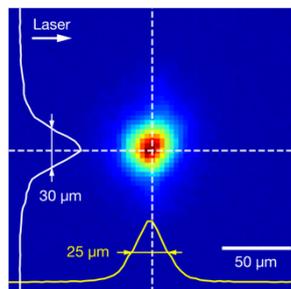


Fig. 4 Source image recorded by an x-ray pinhole camera at the laser intensity of about 1×10^{14} W/cm².

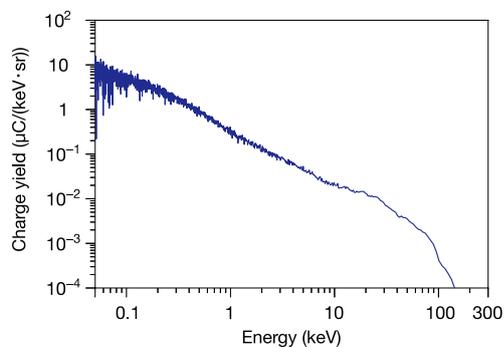


Fig. 5 A charge-state-integrated spectrum of the fast ion from a laser-produced Bi plasma WW-EUV source at the laser intensity of about 1×10^{14} W/cm².

highly charged ions. Charge-separated spectra, however, were not measured in the current work. The Faraday cup can detect the charge-integrated pulse current. The maximum energy observed is 140 keV. In our previous report, the maximum ionic charge state was observed to be Bi¹⁷⁺, possessing a maximum energy of about 200 keV at a laser intensity of about 1×10^{14} W/cm² [16]. We observed similar emission spectra in Fig. 2 and fast ion energy spectra in Fig. 5. The highly charged ion distributions in the plasma determined the emission spectra. The charge states of the highly charged ion distributions depend on the plasma parameters. In addition, we also observed a similar fast ion energy spectrum from the plasma under similar irradiated laser parameters for the previous planar Bi target experiments. Therefore, we evaluated the production of identical plasma parameters.

4. Summary

In summary, we have demonstrated an efficient WW-EUV source using a regenerative metal Bi liquid target with a diameter of 30 μm . The number of photons was observed to be about 1×10^{11} photons/(0.01 nm · sr · pulse) at peak wavelengths of 3.9 and 4.2 nm due to $n = 4 - n = 4$ ($\Delta n = 0$) transitions, and 0.4×10^{11} photons/(0.01 nm · sr · pulse) at a peak wavelength of 2.4 nm, which can be attributed $n = 4 - n = 5$ ($\Delta n = 1$) transitions. The number of photons was about 1×10^{13} photons/(sr · shot) in the 2.3 – 4.4 nm wavelength range, corresponding to the CE of 0.3%/(sr · shot). Fast ion was also observed with a maximum energy of 140 keV. This evaluation provides collector mirror damage information in microscopy and short wavelength EUV lithography applications. For the “Blue-X” proposal [5], we need to further investigate the short wavelength UTA light source in continuous operation for the lithography applications.

Acknowledgments

The authors are indebted to Masaki Kume and Takeru Niinuma (Utsunomiya University) for useful technical support and discussion. T.H. acknowledges the support from the Japan Society for the Promotion of Science (JSPS) / INTERNATIONAL JOINT RESEARCH PROGRAM and The Sumitomo Foundation.

- [1] J.C. Solem and G.C. Baldwin, *Science* **218**, 229 (1982).
- [2] H. Legall *et al.*, *Opt. Express* **20**, 18362 (2012).
- [3] T. Gorniak *et al.*, *Opt. Express* **19**, 11059 (2011).
- [4] M. Kördel *et al.*, *Optica* **7**, 658 (2020).
- [5] V. Bakshi, *EUV Roadmap Needs Extension*, *EE Times* (2018); <https://www.eetimes.com/euv-roadmap-needs-extension/>
- [6] L. Rymell and H.M. Hertz, *Opt. Commun.* **103**, 105 (1993).
- [7] M. Berglund *et al.*, *Rev. Sci. Instrum.* **69**, 2361 (1998).
- [8] S. Bajt *et al.*, *Opt. Eng.* **41**, 1797 (2002).
- [9] V. Bakshi, *EUV Sources for Lithography* (SPIE, Washington, 2006), Chap. 11.

- [10] T. Higashiguchi *et al.*, Appl. Phys. Lett. **100**, 014103 (2012).
- [11] G. Arai *et al.*, Opt. Express **26**, 27748 (2018).
- [12] T.-H. Dinh *et al.*, Rev. Sci. Instrum. **87**, 123106 (2016).
- [13] T. Wu *et al.*, J. Phys. B **49**, 035001 (2016).
- [14] A. Giesen *et al.*, Appl. Phys. B **58**, 365 (1994).
- [15] I. Kuznetsov *et al.*, Opt. Lett. **43**, 3941 (2018).
- [16] H. Kawasaki *et al.*, Rev. Sci. Instrum. **91**, 086103 (2020).