Characteristics of extreme ultraviolet emission from high-Z plasmas

H. Ohashi, T. Higashiguchi, Y. Suzuki, M. Kawasaki, C. Suzuki, K. Tomita, M. Nishikino, S. Fujioka, A. Endo, B. Li, T. Otsuka, P. Dunne, and G. O’Sullivan

1Utsunomiya University, Department of Advanced Interdisciplinary Sciences, Yoto 7-1-2, Utsunomiya, Japan
2National Institute for Fusion Science, 322-6 Oroshi-cho, Toki, Japan
3Interdisciplinary Graduate School of Engineering and Sciences, Kyushu University, 6-1, Kasugakoen, Kasuga, Fukuoka, Japan
4Quantum Beam Science Directorate, Japan Atomic Energy Agency, 8-1-7 Umemi-dai, Kizugawa, Kyoto, Japan
5Institute of Laser Engineering, Osaka University, 2-6 Yamada-Oka, Suita, Japan
6HiLASE Project, Institute of Physics AS, CR, Na Slovance 2, 18221 Prague 8, Czech Republic
7School of Physics, University College Dublin, Belfield, Dublin 4, Ireland

E-mail: higashi@cc.utsunomiya-u.ac.jp

Abstract. We demonstrate the extreme ultraviolet (EUV) and soft x-ray sources in the 2 to 7 nm spectral region related to the beyond EUV (BEUV) question at 6.2 nm and the water window source based on laser-produced high-Z plasmas. Resonance emission from multiply charged ions merges to produce intense unresolved transition arrays (UTAs), extending below the carbon K edge (4.37 nm). An outline of a microscope design for single-shot live cell imaging is proposed based on high-Z plasma UTA source, coupled to multilayer mirror optics.

1. Introduction

Extreme ultraviolet (EUV) lithography by use of a 13.5-nm sources [1] is focused on high-volume production of semiconductor devices. In fact, the fabrication tools for integrated circuits with 0.33NA (numerical aperture), to attain a half-pitch (HP) of 22 nm, have already been developed and are capable of reaching a HP of 16 nm with off-axis illumination techniques [2]. In order to achieve very high resolution HP, a shorter wavelength, which couples to highly reflective multilayer mirrors (MLMs) with a theoretical reflective coefficient of ~ 70%, is a more attractive proposition and explains the present trend in lithography source development [3]. Wavelengths around 6.2 nm are especially useful for the final stage beyond the 13.5-nm EUV source [4]. In fact, the 6.2-nm beyond EUV (BEUV) emission can be coupled with a Mo/B₄C or La/B₄C multilayer mirror whose reflectivity is currently 50% at 6.2 nm, (theoretical maximum > 70%). The unresolved transition array (UTA) emission exploited in Sn is scalable to shorter wavelengths, and Gd has a similar conversion efficiency to Sn, though at a higher plasma temperature, within a narrow spectral range centered near 6.7 nm.

Before discussing the high-power high-Z plasma source, it is important to summarize the characteristics of efficient UTA light sources used in the 5 to 15 nm region. All are based on
\( n = 4 - n = 4 \) \( (4d - 4f \text{ and } 4p - 4d) \) transitions that overlap to generate an intense UTA. For efficient 13.5-nm operation, which corresponds to a photon energy \( h\nu \approx 92 \) eV, it is important to produce an optimum plasma electron temperature of 30 – 50 eV. The rare-earth elements of gadolinium (Gd, \( Z = 64 \)) and terbium (Tb, \( Z = 65 \)) produce strong emission near \( \lambda = 6.7 \) nm \( (h\nu \approx 183 \) eV) which is maximized at electron temperatures in the 100 – 120 eV range depending on initial focusing conditions \([5, 6]\). The spectral behavior of Gd and Tb plasmas is expected to be largely similar to that of Sn plasmas, because in the temperature range of interest, both are dominated by 4d open-shell ions. Although the conversion efficiency from the input laser energy to the output in-band EUV emission energy depends on the bandwidth (BW) of the reflection coefficient of the MLM, the maximum CE's have been observed to be higher than 1%. Because it moves to shorter wavelength with increasing atomic number, \( Z \), the \( n = 4 - n = 4 \) UTA is expected to lie in the water window if higher \( Z \) elements from \( Z = 79 \) (Au) to \( Z = 83 \) (Bi) are used \([7]\). Higher \( Z \) elements such as uranium also emit in the water window but their radioactivity prohibits their use. Much of the previous work on high-\( Z \) plasmas has concentrated on the production of quasicontinuum spectra at moderate laser intensities, employing electron temperatures below 100 eV \([8]\).

In this paper, we report the EUV and soft x-ray sources in the 2 to 7 nm spectral region related to the BEUV and the water window source based on laser-produced high-\( Z \) plasmas. Resonance emission from multiply charged ions merges to produce intense UTAs, extending below the carbon K edge \( (4.37 \) nm). An outline of a microscope design for single-shot live cell imaging is proposed based on high-\( Z \) plasma UTA source, coupled to multilayer mirror optics.

### 2. Atomic number dependence on peak wavelengths of UTA emission

Because it moves to shorter wavelength with increasing \( Z \), the \( n = 4 - n = 4 \) \( (\Delta n = 0) \) UTA emission can be used for BEUV source and transmission x-ray microscopy for biological imaging in the water window (see Fig. 1). We have made preliminary studies of the potential of Gd and Tb for BEUV source \([5, 6]\) and Bi as the “water window” soft x-ray source \([7]\).

![Figure 1. Calculated position of \( n = 4 - n = 4 \) transitions in key ions in elements from indium \((Z = 49)\) to uranium \((Z = 92)\).](image-url)
3. Results and discussion

The EUV spectra around 3 and 6 nm from high-Z plasmas are shown. Figures 2(a)–2(c) show time-integrated spectra from Au, Pb, and Bi plasmas at a laser intensity of $1 \times 10^{14}$ W/cm$^2$ with a 150-ps pulse duration. Time-integrated EUV spectra between 1 and 6 nm from each element display strong broadband emission near 4 nm, which is mainly due to $n = 4 - n = 4$ transitions from ions with an open 4$f$ or 4$d$ outermost subshell, together with broadband emission around 2–4 nm due to $n = 4 - n = 5$ transitions from multi-charged state ions with an outermost 4$f$ subshell. The latter merge to form a structured feature from which the contributing ion stages may be readily inferred. The intensity of the $n = 4 - n = 4$ UTA emission is higher than that of the $n = 4 - n = 5$ emission. The atomic number spectral dependence is summarized in Fig. 2(d). The predicted photon energy of each experimental peak wavelength was shifted to higher photon energy with increasing atomic number. Neither the emission spectra nor the plasma electron temperatures, however, have been optimized, as shown below. The emission intensity of the $n = 4 - n = 5$ transitions, however, was compared with that of the $n = 4 - n = 4$ UTAs.

One efficient EUV source at 6.x nm is provided by laser-produced plasmas (LPPs) of Gd and Tb. These elements produce strong narrowband emission, which is attributed to a $n = 4 - n = 4$ ($\Delta n = 0$) UTA, at 6.x nm. The spectral behavior of Gd and Tb plasmas is expected to be similar to that of Sn plasmas. Figure 3 shows time-integrated EUV emission spectra from the Nd:YAG LPPs at different laser intensities ranging from $9.7 \times 10^{11}$ to $6.6 \times 10^{12}$ W/cm$^2$. The peak wavelength shifts from 6.7 to 6.8 nm, and is mainly due to $n = 4 - n = 4$ ($\Delta n = 0$) transitions in ions with an open 4$f$ or 4$d$ outermost subshell. Emission at wavelengths less than 6 nm, increases with increasing laser intensity and according to numerical evaluation, lines in the $\lambda = 2.5 - 6$ nm ($h\nu = 207 - 496$ eV) spectral region originate from Gd ionic charge states between Gd$^{19+}$ and Gd$^{27+}$, and arise from $n = 4 - n = 5$ ($\Delta n = 1$) transitions.

![Figure 2](image1.png)

**Figure 2.** Time-integrated spectra from the picosecond-laser-produced high-Z plasmas by use of Au (a), Pb (b), and Bi (c), respectively, and the atomic number dependence on the photon energies of peak emission of the $n = 4 - n = 4$ transition (circles) and the $n = 4 - n = 5$ transition (squares).

![Figure 3](image2.png)

**Figure 3.** Time-integrated EUV emission spectra from the Nd:YAG LPPs at different laser intensities of $9.7 \times 10^{11}$ (a), $2.2 \times 10^{12}$ (b), and $6.6 \times 10^{12}$ W/cm$^2$ (c), respectively. The peak wavelength shifts from 6.7 to 6.8 nm with increasing the laser intensity.

The spatial and temporal evolution of the Gd plasma for 6.x-nm emission were studied using the modified 1-dimensional Lagrangian, laser-plasma hydrodynamic simulation code MED103 [9] for a cylindrical solid Gd target. Simulations were performed for a Nd:YAG LPP for laser
parameters: $\lambda_L = 1064$ nm, pulse width, $\tau_L = 8.5$ ns (FWHM) and peak intensity $2 \times 10^{12}$ W/cm$^2$ incident on a target with a radius of 10 $\mu$m. The electron temperature and density obtained as a function of time and space are presented in Fig. 4. A higher temperature plasma, $T_e$ up to 150 eV, was obtained. Based on a collisional-radiative simulation [10], ionic populations of Rh- to Ag-like are maximized at an electron temperature of 80 $-$ 130 eV. The plasma will thus have strong emission at 6.76 nm when $T_e$ is close to 80 eV [6].

**Figure 4.** Electron temperature, $T_e$ (a) and electron density, $\log_{10} n_e$ (b) distributions as a function of time and space using 1.06-µm Nd:YAG laser simulated by the modified hydrodynamic code: MED103.

### 4. Summary

We characterized the EUV and soft x-ray emission in the 2 to 7 nm spectral region related to the BEUV at 6.37 nm and the water window source based on laser-produced high-Z ($Z = 64$ – 83) plasmas. Resonance emission from multiply charged ions merges to produce intense UTAs, extending below the carbon K edge ($\lambda = 4.37$ nm).

### Acknowledgement

A part of this work was performed under the auspices of MEXT (Ministry of Education, Culture, Science and Technology, Japan), and “UU Interdisciplinary Project for in vivo Bioimaging and Sensing” from MEXT. One of the authors (T.H) also acknowledges support from The Canon Foundation, Research Grant (Basic Research) on TEPCO Memorial Foundation, and Gigaphoton Inc.

### References

[1] George S A, Hou K-C, Takenoshita K, Galvanauskas A and Richardson M C 2007 Opt. Exp. 15 16348
[2] Meiling H, Boeij W, Bornebroek F, Harned N, Jong I, Meijer H, Ockwell D, Peeters R, Setten E, Stoeldraijer J, Wagner C, Young S, Kool R, Kürz P and Lowisch M 2012 Proc. SPIE 8322 83221G
[3] Platonov Y, Rodriguez J, Kries M, Louis E, Feigl T and Yulin S 2011 Proc. 2011 International Workshop on EUV Lithography (Maui, Hawaii, 13–17 June)
[4] Banine V Y, Koshelev K N and Swinkels G H P M 2011 J. Phys. D 44 253001
[5] Otsuka T, Kilbane D, White J, Higashiguchi T, Yugami N, Yatagai T, Jiang W, Endo A, Dunne P and O’Sullivan G 2010 Appl. Phys. Lett. 97 111503
[6] Li B, Dunne P, Higashiguchi T, Otsuka T, Yugami N, Jiang W, Endo A and O’Sullivan G 2011 Appl. Phys. Lett. 99 231502
[7] Higashiguchi T, Otsuka T, Yugami N, Jiang W, Endo A, Li B, Dunne P and O’Sullivan G 2012 Appl. Phys. Lett. 100 014103
[8] Zeng G-M, Takahashi M, Daido H, Kanabe T, Aritome H, Nakatsuka M and Nakai S 1990 J. Appl. Phys. 67 3597
[9] Christiansen J P, Ashby D E T F and Roberts K V 1974 Comput. Phys. Commun. 7 271
[10] Colombant D and Tonon G F 1973 J. Appl. Phys. 44 3524