Research Repository UCD

Title	Spectral and temporal behavior of an alkali metal plasma extreme ultraviolet source for surface morphology applications
Authors(s)	Higashiguchi, Takeshi, Yamaguchi, Mami, Otsuka, Takamitsu, Terauchi, Hiromitsu, Yugami, Noboru, Yatagai, Toyohiko, D'Arcy, Rebekah, Dunne, Padraig, O'Sullivan, Gerry
Publication date	2011-02-28
Publication information	Higashiguchi, Takeshi, Mami Yamaguchi, Takamitsu Otsuka, Hiromitsu Terauchi, Noboru Yugami, Toyohiko Yatagai, Rebekah D'Arcy, Padraig Dunne, and Gerry O'Sullivan. "Spectral and Temporal Behavior of an Alkali Metal Plasma Extreme Ultraviolet Source for Surface Morphology Applications" 98, no. 9 (February 28, 2011).
Publisher	American Institute of Physics
Item record/more information	http://hdl.handle.net/10197/3586
Publisher's statement	The following article appeared in Applied Physics Letters 98, 091503 (2011), DOI:10.1063/1.3560304 and may be found at http://dx.doi.org/10.1063/1.3560304 . The article may be downloaded for personal use only. Any other use requires prior permission of the author and the American Institute of Physics.
Publisher's version (DOI)	10.1063/1.3560304

Downloaded 2023-04-07T10:52:29Z

The UCD community has made this article openly available. Please share how this access benefits you. Your story matters! (@ucd_oa)

© Some rights reserved. For more information

Spectral and temporal behavior of an alkali metal plasma extreme ultraviolet source for surface morphology applications

Takeshi Higashiguchi^{1,2}, Mami Yamaguchi¹, Takamitsu Otsuka¹, Hiromitsu Terauchi¹, Noboru Yugami^{1,2}, Toyohiko Yatagai¹, Rebekah D'Arcy³, Padraig Dunne³, and Gerry O'Sullivan³

¹Department of Advanced Interdisciplinary Sciences, and Center for Optical Research & Education (CORE), Utsunomiya University, Yoto 7-1-2, Utsunomiya, Tochigi 321-8585 Japan

²Japan Science and Technology Agency, CREST, 4-1-8 Honcho, Kanagawa, Saitama 332-0012, Japan

³School of Physics, University College Dublin, Belfield, Dublin 4, Ireland

Abstract

We have characterized the emission spectrum and temporal history of a pure potassium plasma in a capillary discharge. Strong broadband emission was observed around 40 nm due to 3s–3p, 3p–3d, and 3d–4f transitions in ions ranging from K^{2+} to K^{4+} at a time-integrated electron temperature of about 12 eV. The temporal behavior of this emission strongly follows the recombination phase in the plasma and it was successfully reproduced by a hydrodynamic simulation of the potassium plasma which accounted for atomic processes.

Short wavelength radiation in the vacuum ultraviolet (VUV) and extreme ultraviolet (XUV or EUV) spectral regions has sufficient photon energy to induce photochemical reactions in almost all organic molecules, which consist of hydrogen, carbon, and oxygen [1,2]. A compact short wavelength light source is useful to provide surface interaction between photons and materials. VUV and XUV radiation are thus applied in promising technologies, such as EUV semiconductor lithography (EUVL) [3-7], material processing, photochemistry, and biological imaging. A photo-induced desorption spectrometer [8] employing a compact, efficient XUV source has been proposed for surface morphology applications, as operation at these wavelengths has the advantage of not damaging the surface during irradiation [9]. The photo-induced desorption spectrometer consists of a light source in the XUV spectral region, a mechanical stage for selection of the material surface to be irradiated, and a mass spectrometer coupled to the vacuum chamber. For surface analysis, a low kinetic energy of the particles, which originate from the irradiated test material surface, is required for the mass spectrometer. The photons emitted from the light source, on the other hand, must have an energy higher than the binding energy. These requirements, therefore, must be traded off. To reduce the kinetic energy and to induce a photochemical reaction, wavelengths of around 40–50 nm, which correspond to photon energies of 25–31 eV, are required for the light source. A 40-nm XUV source based on potassium vapor was produced in a capillary with a 500- μm diameter at an electron temperature of about 12 eV and a discharge current of about 200 A [10]. In the present paper, the spectrum of the discharge-produced plasma is compared with that of a laser-produced plasma, because the polytetrafluoroethylene (PTFE) used in the capillary-based discharge plasma, combined with the oxide film on the potassium target surface produces oxygen, hydrogen and carbon line emission, together with the potassium

emission. To isolate the pure potassium emission from the compact, discharge-produced plasma XUV source, we used a laser-produced potassium plasma, which has very similar characteristics to the discharge-produced plasma [10]. The use of a laser-produced plasma enables one to control the plasma parameters by changing the laser intensity and wavelength, as the electron temperature and the critical density depend on the laser intensity and wavelength, respectively. As a comparison with a laser-produced pure potassium plasma and a numerical analysis are essential for understanding the behavior of the hollow cathode discharge-produced plasma XUV source, we characterized the emission spectra from the laser-produced pure potassium plasma at a time-averaged electron temperature of 12 eV and an electron density of $1 \times 10^{20} \text{ cm}^{-3}$. In addition, we used the collisional-radiative equilibrium (CRE) model for plasma processes [11-13] and the multi-configuration Hartree-Fock atomic structure code of Cowan [14] to analyze the results.

In this paper, we characterize a laser-produced pure potassium plasma to optimize spectral and temporal behavior of the XUV emission. Potassium ions produce strong broadband emission around 40 nm at a time-averaged electron temperature of approximately 12 eV. The temporal behavior of this emission was measured and numerically analyzed.

Calculations were performed for transitions between the configurations $3s^23p^n$ and $3s^23p^{n-1}3d^1$, ($1 \leq n \leq 6$) in one to six times ionized potassium. The corresponding oscillator strength (gf) distributions calculated with the Cowan code are presented in Fig. 1 to show the resonant emission in ions ranging from K^+ to K^{6+} . In particular, emission from K^{2+} to K^{4+} combines to produce a strong emission band near 40 nm. The ion population of the potassium plasma as a function of electron temperature was calculated using a time-

dependent CRE model at an electron density of $1 \times 10^{20} \text{ cm}^{-3}$ in a plasma that was assumed to be optically thin for EUV radiation [11]. A range of ion stages is present in a plasma at any given electron temperature, where higher temperatures produce higher ion stages. The 3s–3p, 3p–3d, and 3d–4f transitions in the ions K^{2+} and K^{4+} dominate the spectrum around 40 nm for electron temperatures of 5.5 and 11 eV, respectively. In this work, we focus on the wavelength region 39–40 nm, where emission is predicted to be maximized at an electron temperature of 12 eV.

Fig 1: The weighted oscillator strength spectra of the resonant lines for ion stages whose resonance transition.

A Q-switched Nd:YAG (Nd:yttrium-aluminum-garnet) laser operating at 1064 nm with a maximum pulse energy of 2 J and a duration of 10 ns (FWHM) was focused, using a 12-cm focal length lens, onto a planar potassium target of thickness 1 mm inside a vacuum chamber. The focused spot size was monitored by use of a telescope and a charge coupled device (CCD) camera placed on the laser axis. The spot size was measured to be 500 μm (FWHM). This spot size was chosen to minimize the plasma hydrodynamic expansion loss [15]. Adjusting the pulse energy at the fixed spot diameter of 500 μm varied the laser intensity. The emission from the plasma in the XUV spectral region was analyzed with a normal incidence vacuum spectrograph using an iridium coated grating of 1200 lines/mm (Acton VM502), and was viewed at 45° with respect to the incident laser axis. Time-integrated spectra were obtained by a thermoelectrically-cooled back-illuminated x-ray CCD camera. The typical spectral resolution was better than 0.2 nm. In addition, a higher resolution spectrometer (a 2-m grazing incidence design with a 300-groove/mm platinum coated grating) with wavelength resolution better than 0.02 nm was also positioned at 90° with respect to the incident laser axis. The temporal behavior of the 39-nm XUV emission was measured by a fast photo-multiplier tube connected to a sampling digital photon counter, which was coupled to the first spectrometer. In the present case, we set the laser intensity at $2 \times 10^{10} \text{ W/cm}^2$ to produce a time-averaged electron temperature of 12 eV. The temporally and spatially integrated electron temperature, which has been evaluated by use of the intensity ratios of the line emissions of oxygen ions in the time-integrated XUV spectra, increased with increasing laser intensity (not shown). This laser intensity dependence of the electron temperature was also reproduced by the hydrodynamic simulation.

Fig 2: (a) Temporal behavior of the emission at 39 nm at a laser intensity of $2 \times 10^{10} \text{ W/cm}^2$. The dots represent the raw, experimentally measured data and the solid line (blue) is a smoothing of the experimental result. The dashed line (red) is the calculated temporal waveform of the 39-nm emission. (b) Numerical calculation of the temporal change of the ionic population based on a time-dependent CRE model.

The wavelength-integrated energy between 25 and 60 nm was previously maximized in a discharge-produced plasma [10]. Figure 2 shows a time-integrated spectrum between 20 and 60 nm from the laser-produced plasma at a flux of $2 \times 10^{10} \text{ W/cm}^2$. The spectral bandwidth of the potassium plasma emission was approximately 8 nm (FWHM) at 39 nm. The insert in Fig. 2 shows the time-integrated spectrum of the potassium laser-produced plasma emission in the narrow band spectral region recorded in the higher resolution measurement, together with the stronger emission between 40 and 42 nm. The difference between these spectra is due to the different measurement angles and self-absorption (opacity) effects in the potassium plasma. The origin of the emission lines may be identified with the aid of

calculations using the Cowan multi-configuration Hartree-Fock code [14] which provides theoretical data for transition energies and oscillator strengths as shown in Fig. 1.

Fig 3: Normalized time-averaged XUV spectra at laser wavelengths of 1064 (red) and 532 nm (green), respectively.

Figure 3(a) shows the temporal behavior of the emission at 39 nm. The dots represent the measured emission, and the solid line is a smoothing of the experimental result. It should be noted that the time at $t = 0$ ns corresponds to the laser intensity peak with the pulse duration of 10 ns (FWHM). The duration of the emission was 50 ns (FWHM). The dashed line is a numerical simulation of the 39-nm emission. The calculation is based on a time-dependent CRE model, as shown in Fig. 3(b), which includes plasma processes such as ionization, recombination, emission, collisional excitation, and de-excitation between several levels of the potassium ions. The electron temperature, determined experimentally by the

laser pulse profile, was calculated in the hydrodynamic simulation and determines the ion population and therefore the 39-nm emission. The peak and time-averaged electron temperatures were calculated to be 26 and 12 eV, respectively, at the chosen laser intensity of $2 \times 10^{10} \text{ W/cm}^2$. The dominant 39-nm emission is shown both spatially and temporally to be generated when the electron temperature is close to 15 eV. The highest ion stages attained initially are K^{5+} to K^{7+} ; thereafter the plasma recombines to lower ionization states, and starts emitting the 36–42 nm band in this recombining phase (see Fig. 3(b)).

Fig 4: The numerical calculated-spectrum comparison without (a) and with (b) self-absorption (opacity) effects in the potassium plasma at an electron temperature of 12 eV based on the time-dependent CRE model combined with a Cowan code simulation. The evaluated transmission coefficient at an effective plasma thickness of $5 \mu\text{m}$ (c).

To explore self-absorption effects in the K plasma, we also observed spectra at different laser wavelengths to evaluate the effects of self-absorption, because different color irradiation produces plasmas with different densities [4,6,7,15-18]. The spectra were seen to be almost identical at laser wavelengths of 1064 and 532 nm at the same laser intensity of $2 \times 10^{10} \text{ W/cm}^2$, as shown in Fig. 4. At first sight, this result indicates that the potassium plasma may be only weakly self-absorbing. However, to explore self-absorption effects further, an atomic level numerical calculation was also used. We obtained theoretical spectra of potassium plasma emission at an electron temperature of 12 eV, as shown in Figs. 5(a) and 5(b); the first spectrum is due to the emission only while the second includes opacity effects. The differences between these spectra are due to absorption broadening around 39 nm, which is also included in the numerically calculated transmission coefficient for an effective ionic plasma thickness of 5 μm in Fig. 5(c). As a result, the 39-nm emission is seen to originate from strong resonant line emission and this emission is also strongly self-absorbed. Figure 5(b) represents the numerically calculated spectrum at a time-integrated electron temperature of 12 eV and an electron density of $1 \times 10^{20} \text{ cm}^{-3}$. The experimentally observed spectrum in Fig. 2 is in reasonably good agreement with the calculated one. The numerical calculation, therefore, provides a clear insight to the optimization of XUV emission. As this spectrum is similar to that of the hollow cathode-mode discharge plasma spectrum in Ref. [10], it indicates that the emission from the discharge-produced plasma occurs in a region of high electron density close to 10^{20} cm^{-3} .

Fig 4: The numerical calculated-spectrum comparison without (a) and with (b) self-absorption (opacity) effects in the potassium plasma at an electron temperature of 12 eV based on the time-dependent CRE model combined with a Cowan code simulation. The evaluated transmission coefficient at an effective plasma thickness of 5 μm (c).

In summary, we have shown the emission spectra of a K plasma and its temporal behavior around 40 nm. Potassium ions produced strong broadband emission around 40 nm ranging from K^{2+} to K^{4+} ions at a time-integrated electron temperature of about 12 eV. Emission at 39 nm commences during the recombining phase. In the near future, it is planned to optimize a compact, hollow cathode discharge-produced potassium XUV source, coupled with a Sc/Si multilayer mirror for use in a photo-induced desorption spectrometer.

The authors are also grateful to Keisuke Kikuchi and Takafumi Oba for their unparalleled technical support. Part of this work was performed under the auspices of MEXT (Ministry of Education, Culture, Science and Technology, Japan) and “Utsunomiya University Distinguished Research Projects.” One of the authors (T.H.) also acknowledges support from Research Foundation for Opto-Science and Technology. The UCD group acknowledges support from Science Foundation Ireland under Principal Investigator research grant 07/IN.1/I1771.

References

- [1] T. Ohtsubo, T. Azuma, M. Takaura, T. Higashiguchi, S. Kubodera, and W. Sasaki, Appl. Phys. A **76**, 139 (2003).
- [2] Y. Maezono, K. Toshikawa, K. Kurosawa, K. Amari, S. Ishimura, M. Katto, and A. Yokotani, Jpn. J. Appl. Phys. **46**, 3534 (2007).
- [3] C. W. Gwyn, R. Stulen, D. Sweeney, and D. Attwood, J. Vac. Sci. Technol. B **16**, 3142 (1998).
- [4] H. Tanaka, A. Matsumoto, K. Akinaga, A. Takahashi, and T. Okada, Appl. Phys. Lett. **87**, 041503 (2005).
- [5] Y. Ueno, G. Soumagne, A. Sumitani, A. Endo, and T. Higashiguchi, Appl. Phys. Lett. **91**, 231501 (2007).
- [6] T. Otsuka, D. Kilbane, J. White, T. Higashiguchi, N. Yugami, T. Yatagai, W. Jiang, A. Endo, P. Dunne, and G. O'Sullivan, Appl. Phys. Lett. **97**, 111503 (2010).
- [7] T. Otsuka, D. Kilbane, T. Higashiguchi, N. Yugami, T. Yatagai, W. Jiang, A. Endo, P. Dunne, and G. O'Sullivan, Appl. Phys. Lett. **(to appear in 2010)**.
- [8] N. Hirashita, M. Kinoshita, I. Aikawa, and T. Ajioka, Appl. Phys. Lett. **56**, 451 (1990).
- [9] M. Wasamoto, M. Katto, M. Kaku, S. Kubodera, and A. Yokotanim, Appl. Surf. Sci. **255**, 9861 (2009).
- [10] T. Higashiguchi, H. Terauchi, N. Yugami, T. Yatagai, W. Sasaki, R. D'Arcy, P. Dunne, and G. O'Sullivan, Appl. Phys. Lett. **96**, 131505 (2010).
- [11] D. Colombant and G. F. Tonon, J. Appl. Phys. **44**, 3524 (1973).
- [12] T. Fujimoto, J. Phys. Soc. Jpn, **47**, 265 (1979); *ibid* **47**, 273 (1979).

- [13]H. Griem, *Principle of Plasma Spectroscopy* (Cambridge Monographs on Plasma Physics, 2005).
- [14]R. D. Cowan, *The Theory of Atomic Structure and Spectra*, University of California Press, Berkeley (1981).
- [15]R. C. Spitzer, T. J. Orzechowski, D. W. Phillion, R. L. Kauffman, and C. Cerjan, J. Appl. Phys. **79**, 2251 (1996).
- [16]M. Yamaura, S. Uchida, A. Sunahara, Y. Shimada, H. Nishimura, S. Fujioka, T. Okuno, K. Hashimoto, K. Nagai, T. Norimatsu, K. Nishihara, N. Miyanaga, Y. Izawa, and C. Yamanaka, Appl. Phys. Lett. **86**, 181107 (2005).
- [17]S. Miyamoto, A. Shimoura, S. Amano, K. Fukugaki, H. Kinugasa, T. Inoue, and T. Mochizuki, Appl. Phys. Lett. **86**, 261502 (2005).
- [18]J. White, P. Dunne, P. Hayden, F. O'Reilly, and G. O'Sullivan, Appl. Phys. Lett. **90**, 181502 (2007).