Detailed atomic modeling of Sn plasmas for the EUV source

A. Sasaki¹, A. Sunahara², K. Nishihawra³, T. Nishikawa¹, F. Koike⁵, and H. Tanuma⁶

¹Quantum Beam Science Directorate, Japan Atomic Energy Agency, 8-1 Umemidai, Kizugawa-shi, Kyoto, Japan, 610-0215
²Institute for Laser Technology, 1-8-4, Utsubohonmachi, Nishi-ku, Osaka, 550-0004
³Institute for Laser Engineering, Osaka University, 2-6 Yamadaoka, Suita, Osaka, 565-0871
⁴Okayama University, 1-1-1 Tsushima-naka, Okayama, 700-8530
⁵Kitasato University, 1-15-1 Kitasato, Sagamihara, Kanagawa, 228-8555
⁶Tokyo Metropolitan University, 1-1 Minamiosawa, Hachioji, Tokyo, 192-0397

E-mail: sasaki.akira@jaea.go.jp

Abstract: An atomic model of Sn plasmas is developed to calculate coefficients of radiative transfer, based on the calculated atomic data using the Hullac code. We find that the emission spectrum and conversion efficiency depend critically on the wavelength and spectral structure of the 4d–4f transition arrays. Satellite lines, which have a significant contribution to the emission, are determined after iterative calculations by changing the number of levels in the atomic model. We also correct transition wavelengths through comparison with experiments. Using the present emissivity and opacity, the radiation hydrodynamics simulation will be carried out toward the optimization of the EUV source.

1. Introduction

Sn plasmas, which have a strong emission in the EUV wavelength, are considered as a candidate for the light source for the next generation microlithography [1]. Near 10 times ionized Sn is produced in the laser or discharge pumped plasma at the electron temperature of 20-50eV. These ions have a ground configuration of 4di, where i is the occupation number, so that strong emission is obtained through 4d–4f, 4d–5p, and 4d–5f resonance lines. The wavelength of 4d–5p and 4d–5f transitions decreases as the ion charge, whereas the wavelengths of 4d–4f transitions are almost constant [2]. Although several atomic species with the atomic number around 50 have similar dependence of the wavelengths of 4d–4f, only Sn has the characteristic emission wavelength at 13.5nm, which is especially interested for the lithographic EUV source.

The plasma EUV sources are being studied theoretically and experimentally. We present an atomic model of Sn ions mainly for the laser produced plasma (LPP) EUV source, which is used to calculate the coefficients of radiative transfer. Firstly, we calculate atomic data such as energy levels and radiative transition probabilities using the Hullac code [3]. Secondly, we develop a collisional radiative model of Sn ions by choosing energy levels, which are expected to have a significant population in the plasma. Thirdly, the emissivity and opacity of the plasma are calculated from the level population. Finally, detailed spectral profile in the EUV wavelength region as well as wavelength correction is applied to the resonance lines and other strong transitions. The theoretical simulation is useful for finding the optimum condition of the EUV source to obtain high conversion
efficiency and output power.

2. The atomic model

We develop an atomic model of Sn including neutral Sn atom to Ar-like ions. The coefficients of radiative transfer should be accurate over a wide range of the density and temperature, because the laser produced plasma has a spatial non-uniformity. Moreover, the density and temperature change significantly depending on the pumping laser intensity and wavelength. We use the detailed configuration accounting (DCA) model, and the atomic data are calculated by the Hullac code, which provides the configuration averaged energy levels, radiative rates, and autoionization rates. On the other hand, the rates of electron collisional excitation and ionization are calculated using the empirical formulas.

Sn ions have a large number of inner-shell and multiply-excited states. We find energy levels, which have significant population by iterative calculations. A group of levels is determined, which have a same core configuration with one excited electron such written as 4d\(^{i}\)nl. Calculation of population is repeated for all groups of configurations. We see the mean charge converges including 5 groups of levels, in the case of near 10 times ionized state, such as 4d\(^{1}\)nl, 4d\(^{2}\)4fnl, 4d\(^{2}\)5sln, 4d\(^{2}\)5pnl, and 4p\(^{4}\)dl\(^{n}\)l configurations [4]. For each group, excited levels up to n=8 and l=3 are taken into account. Similar iterative calculation is carried out in order to find satellite lines, which have significant contribution to the spectrum.

The emissivity and opacity are calculated from the level population considering free-free, bound-free, and bound-bound transitions, among which the last component dominates over the EUV wavelength region. The emission lines consist of a large number of fine structure transitions. In order to reproduce the emission spectrum of Sn, we include the spectral structure of resonance lines and the satellite lines from Sn ions. 4d-4f transitions of Sn\(^{10+}\) ion have a peak around 13.5nm with a width of 1nm. The wavelength of 4d-4f transition is calculated taking the effect of CI (configuration interaction) into account, because due to the strong interaction between 4d\(^{1}\)4f and 4p5d\(^{1}\) configurations, the 4d-4f and 4p-4d transition arrays shift towards shorter wavelength, and the width of the arrays also becomes narrower. We find the accurate wavelength is essential to determine the conversion efficiency, therefore, we observe the emission spectrum from each ion using charge exchange spectroscopy (CXS). According to the measurement, we shift the wavelength of 0.3nm used in the model to obtain an agreement with the experimental spectrum [5].

A considerable amount of emission from Sn ions arises from satellite lines. Satellite lines from multiply excited states with a spectator electron in the Rydberg state sometimes appear in the longer wavelength side of the resonance lines. Especially, in the case of near 10 times ionized state, we typically take emission from 4d\(^{1}\)nl, 4d\(^{2}\)4fnl, 4d\(^{2}\)5sln, 4d\(^{2}\)5pnl, and 4p\(^{4}\)dl\(^{n}\)l configurations into account. Averaged wavelength and width of each satellite channel is determined, after taking into the effect of CI into account, and adjusting the line position to have the wavelength asymptotically approach the resonance line at the limit of n=\(\infty\).

Although, efficient EUV emission are mainly expected from near 10 times ionized Sn ions, Sn ions may be ionized above Kr-like charge state in the case of higher plasma temperature caused by higher pumping laser intensity and lower density. 4p-4d and 4d-4f transitions from Kr- to Cu-like ions may also contribute to the EUV emission. The wavelength are within EUV region and slightly increases from 13 to 16nm as the ion charge increases. The effect of CI changes calculated wavelength and spectral structure of these lines considerably. Preliminary calculation suggests emission from more inner-shell excited configurations should be included. Measurement of line wavelength using CXS has
also been carried out, and wavelength in the model will be corrected.

3. Result and Discussion

Figure 1 shows the mean charge and spectral efficiency, which is defined from the ratio of emission into 2% band width at $\lambda=13.5$nm to the total emission, calculated by present collisional radiative model. The plasma temperature to have a mean charge=10 is slightly different with respect to the electron density, which corresponds to the temperature for which the maximal spectral efficiency is obtained. The maximal spectral efficiency increases up to more than 40% as the electron density decreases.

![Figure 1](image1.png)

**Figure 1.** Mean charge (a) and spectral efficiency (b) of the Sn plasma calculated by the collisional radiative model.

Figure 2 shows the spectral emissivity for the electron density of the plasma of $10^{17}$ and $10^{19}$/cm$^3$. It is shown that the peak photon energy is increased with increase in the plasma temperature as expected from the z dependence of the energy of the 4d-4f transition. Spectrums from the low density plasma ($n_i=10^{17}$/cm$^3$) are much narrower than those from the higher density, due to the smaller contribution from satellite line emission, resulting in the higher spectral efficiency. In the case of $n_i=10^{19}$/cm$^3$, 2/3 of the total emission is originates from the satellite lines. These results support the use of the CO$_2$ laser as a pumping source to achieve higher conversion efficiency, because the density of the emission region of the plasma is expected to be lower due to the low critical density. On the other hand, the lowest density will be decided by the source size limited by the etendu. In order to extract the pumping energy stored in the plasma through radiation efficiently, the optical depth of the plasma should be $>1$, which indicates that the plasma size will increase as the density decreases. An optimization of laser and target parameters will be investigated using radiation hydrodynamics simulation.

In conclusion, we show the atomic model of Sn and the calculated coefficient of radiative transfer using the present model. We show the usefulness of calculated atomic data for the modeling complex multiple charged ions. The accuracy of the atomic data and atomic model is improved by detailed comparisons with experimental spectrum, and comparisons between calculated results using different atomic models.
Figure 2. Calculated spectral emissivity of Sn plasma for low (n_i=10^{17}/cm^3) (a-d) and high (n_i=10^{19}/cm^3) (e-h) density. Numbers in each graph correspond to the plasma electron temperature and calculated mean charge.

References
[1] Stamm U, et al., 2006 Proceedings of SPIE vol.6151, “Emerging Lithographic Technologies X”, 61510O.
[2] O’Sullivan G, and Caroli P.K 1981 J. Opt. Soc. Am. 71, 227.
[3] Bar-Shalom A, Oreg J, and Klapisch M 1997 Phys. Rev. E56, R70.
[4] Sasaki A, Sunahara A, Nishihara K, Nishikawa T, Fujima K, Koike F, Kagawa T and Tanuma H 2007 HEDP, 3, 250-255.
[5] Tanuma H, Ohashi H, Shibuya E, Kobayashi N, Okuno T, Fujioka S, Nishimura H, and Nishihara K 2005 Nucl. Inst. Meth. Phys. Res. B235, 331.