Modeling of the Atomic Processes and Photo Emission of the Plasmas for the EUV Source

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A study of the coefficient of radiative transfer of Sn plasmas for the extreme ultra-violet light source is presented. The atomic data of near 10 times ionized Sn ions are calculated using the Hullac code, and the emissivity and opacity are calculated using a collisional radiative model, which is used in the radiation hydrodynamics code to calculate the emission spectrum and conversion efficiency. The accuracy of the coefficients of radiative transfer are improved by considering the detailed spectral structure of strong 4d-4f resonance lines, as well as including a large number satellite lines, along with correction of transition wavelength after comparison with spectroscopic measurement.

Key Words: Laser produced plasmas, EUV, Atomic process, Spectroscopy.

1. Introduction

Laser produced plasma (LPP) EUV sources are being intensively investigated for realization of the next generation microlithography1,2. In order to obtain the required EUV power of 180W at λ=13.5nm within 2% bandwidth, we need to study the emission mechanism of the EUV radiation of the plasma in order to improve conversion efficiency (CE) > 1%, which is required to reduce the power as well as cost of the pumping laser system. Source power only within the etendue limit of 3mm² str is useful, so that the size of the plasma is also limited.

The radiation hydrodynamics simulation is expected to be useful for optimizing target and pumping conditions. However, the simulation is required to have high accuracy, for each of the physical components, as the emission intensity should be reproduced with an uncertainty of within few percent.

The simulation model of an LPP EUV source consists of several parts. In this paper, we present the model of the atomic processes, which is used to calculate coefficient of radiative transfer of the Sn plasmas.

Sn and Xe plasmas have both been used as EUV sources, which exhibit broad emission in EUV wavelength region, with a predominant peak at 11nm (Xe) and 13.5nm (Sn). Although, debris from solid targets requires mitigation techniques to extend the lifetime of the source for industrial applications, Sn targets are preferred because of higher conversion efficiency.

At a temperature of around 30eV, near 10 times ionized Sn and Xe ions are populated in the plasma. As those ions have a ground configuration of 4d, strong emission occurs through resonant 4d-4f, 4d-5p, and 4d-5f transitions. The wavelengths of 4d-4f transitions are almost constant over Pd-like to Rb-like ions. Sn plasmas are particularly interesting, because the wavelengths of many 4d-4f transitions lie within the 13.5nm band as shown in Figure 1. Furthermore, satellite lines from multiply excited states contribute to the broad emission spectrum3.

We investigate atomic processes in Sn and Xe plasmas based on theoretical atomic data calculated using the Hullac code4. Recent development of these computer codes enables us to model complex ions, for which the atomic data were unavailable experimentally. However, calculated atomic data may have errors. The collisional radiative model, which is used to determine population depends on the atomic model, and the choice of atomic levels included in the rate equations. We improve the accuracy of the coefficients of radiative transfer, through detailed calculation of atomic structure including the effect of configuration interaction (CI), as well as comparisons with a variety of experiments.

2. Model

We developed a collisional radiative model for Sn ions, to calculate tables of emissivity and opacity of a plasma, over temperature and density ranging from 5 to 250eV, and 10¹⁷ to 10²⁷/cm³, respectively.

Firstly, we calculated atomic data such as energy levels, radiative transition probabilities and autoionization rates using the Hullac code, based on the configuration averaged atomic model.

Secondly, we developed a collisional radiative model to calculate level populations. Figure 2 shows the level diagram
of Xe$^{10+}$ as a typical emitting ion. Strong emission through resonance lines such as,

$$4d^{-1}(4f \mid 5p \mid 5f) \rightarrow 4d'$$

$$4p'4d'^{-1} \rightarrow 4p'4d$$

appears from one electron excited configuration from the ground state, $4d^{-1}nl$. We also take $4d^{-2}4nl$, $4d^{-2}5nl$, $4d^{-2}5pnl$, and $4p'4dnl$ configurations into account, so that we include satellite lines,

$$4d^{-1}(4f \mid 5p \mid 5f)nl \rightarrow 4d'^{-1}nl$$

$$4p'4d'^{-1}nl \rightarrow 4p'4d'^{-1}nl$$

These multiply excited configurations have large statistical weight and have level energies below the ionization limit. Therefore, a large population into these levels has considerable effects on the ion abundance, as well as the emission spectrum.

Sn and Xe ions have a large number of multiply excited configurations. In order to decide the most appropriate set of levels to be included for modeling radiative coefficients, we repeated calculations by increasing the size of the collisional radiative model, until the mean charge converged. Energy levels, which consist of a common core configuration with one excited electron, are defined as a group, and the number of groups in the model is increased according to the ascending order of the energy of the level, which corresponds to the core configuration. The principal and orbital quantum number of an excited electron are limited to $n \leq 8$ and $l \leq 3$, respectively. This procedure allows us to construct the atomic model automatically.

We found that the mean charge depends on the number of groups of levels in the model until 5, and it no longer changes significantly if we included further groups beyond 5. On the other hand, in the temperature range of the interest, the mean charge varies from less than 5 to more than 20. At each temperature, the ion abundance of any charge state decreases rapidly below and above the most abundant ion. Therefore, the level population was calculated with a model, which includes only 3 charge states of each side of the most abundant one.

The level population was calculated assuming an optically thin, collisional radiative equilibrium (CRE) state of the plasma. Rates of spontaneous emission and autoionization are calculated using the Hullac code, whereas rates of collisional excitation and ionization, and radiative recombination were inferred from hydrogenic empirical formulas as functions of transition energy and oscillator strength.$^5$

Thirdly, we calculated the emissivity and opacity, using the population of each configuration averaged level, and the transition probabilities and widths of the transition arrays. Each transition array is assumed to have a Gaussian profile, except for strong 4d-4f, 4d-5p, 4d-5f and 4p-4d transitions, for which wavelengths and spectral profile were corrected after detailed theoretical calculations and comparison with experiment.

It is known that the wavelengths and spectral distribution of 4d-4f and 4p-4d transition arrays change significantly due to the effect of configuration interaction (CI)$^6$. Calculations with CI show that the wavelength becomes considerably shorter and almost constant over several charge states, and the spectral width becomes narrower. However, it is known that convergence of a CI calculation in terms of number of configurations is slow, and the calculated wavelength sometimes has errors.

Therefore, we determined the wavelength of the most important 4d-4f transitions by independent measurement. Using the charge exchange spectroscopy (CXS), each single charge state of Sn ions was selected from those produced in the ECR ion source, and the EUV emission spectrum after the charge exchange collision with He or Xe gas is observed.$^7$

Experiments were repeated from Pd-like to Kr-like ions, and broad unresolved transition arrays (UTA's) were observed, which were identified as 4d-4f, 4d-5p and 4d-5f transitions. It was found that the calculated wavelengths of the 4d-4f transitions are systematically shorter by approximately 0.3nm with respect to the experimental results as shown in Fig.1. In the case of 4d-5p and 4d-5f transitions good agreement between calculated and measurement is obtained. Furthermore, the calculated profile of the UTA is very similar to the experimental one. Therefore, we shifted the wavelength of 4d-4f transitions in further calculations of the emissivity and opacity.

Fig.1 Averaged wavelength of 4d-4f, 4d-5p, and 4d-5f transitions from Sn$^{3+}$ to Sn$^{14+}$.

Fig.2 Energy level diagram of Ru-like Xe (Xe$^{10+}$).
3. Results

Figure 3 shows the calculated emissivity of the Sn plasma at a typical ion density (=10^{19}/cm^3) of laser produced plasmas. The graph shows a peak of the emissivity in the 13.5 nm band at a temperature of around 30 eV. The peak wavelength decreases as temperature increases at low temperature, and in contrast increases toward longer wavelength beyond the optimal condition. This behavior arises from the dependence of the wavelength of 4d-4f transitions as shown in Fig. 1. It is shown that ions with higher charge than Kr-like also emit EUV radiation through 4p-4d and 4d-4f resonance and satellite lines, with emission wavelength increasing from 14 to 16 nm as the ion charge increases. It is shown that the width of the peak is less than 1 nm. It implies that the emissivity as well as the resultant conversion efficiency from the radiation hydrodynamics simulation also depends critically on the accuracy of the wavelength of the emission lines.

The spectral efficiency and mean charge for the Sn plasma is shown in figs 4 and 5, respectively. The spectral efficiency is defined by the fraction of in-band emission to the total emission. The dashed line in Fig. 5 corresponds to the temperature where the spectral efficiency has a peak value for each ion density. It is shown that high efficiency can be obtained in a relatively narrow temperature region, where the mean charge of the plasma becomes 10, except for low density (n_\text{i}<10^{19}/cm^3) conditions. The peak spectral efficiency increases up to more than 40% as the ion density decreases. This trend is due to less contributions from satellite lines as shown in the emission spectrum in fig. 6. It is seen that in the case of n_i=10^{19}/cm^3, 2/3 of total emission may be attributed to the satellite lines. Unfortunately, most of the satellite line emission appears outside the EUV band (\lambda=13.5 nm, 2% BW).

The satellite lines, which gives rise to broad spectra, arise from multiply and inner-shell excited configurations. The spectator satellites usually appear on the longer wavelength side of the resonance line, and the difference in energy decreases as the principal quantum number of the spectator electron increases. In the case of higher charge states, transitions between inner-shell excited configurations also contribute. As the optical depth of the plasma increases, the emission spectrum approaches a Planckian distribution. Therefore, the spectral efficiency becomes low at high density.

4. Discussion

We have calculated the emissivity and opacity of Sn plasmas. We have also performed a hydrodynamics calculation using present radiative coefficient and see that the simulation reproduced the experimental emission spectrum as well as the conversion efficiency for various laser and target conditions, implying that the present simulation is accurate enough for the optimization of an EUV source.

On the other hand, the result of the radiation hydrodynamics simulation differs slightly from experiment in the case where the optical depth of the plasma is large, where the photo excitation and ionization have considerable effects on the dynamics of the plasma. The experimental spectrum shows a broad background over EUV region, which is not present in the calculated result. More satellite channels may be included to reproduce the background structure. In particular, highly charged ions have a large number of inner-shell satellite lines, and the effect of CI is also significant for these transitions. Comparison with the measured opacity of the plasma may be useful for improvement of atomic model and atomic data.

In the present radiation hydrodynamics simulation, the effect of radiation transfer is taken into account by solving the
transfer equation using multi-group diffusion approximation. In the present model, the effects of radiation on the atomic process is taken into account using the interpolation method by taking the average of the coefficient of radiative transfer at CRE and LTE, as given by Novikov\(^{10}\). A calculation using the direct coupling of atomic processes and radiation hydrodynamics is in preparation to verify the model of calculation.

It is seen from the density dependence of the emissivity, that the conversion efficiency of the EUV source should improve if it were operated at low density, which implies an advantage for using a CO\(_2\) laser as a pumping source, because the density of the plasma is determined by the critical density, and depends on the wavelength of the pumping laser. On the other hand, the optical depth of the plasma should be more than 1, in the case of a CO\(_2\) laser pumped source, a combination of the prepulse and main pulse laser irradiation with longer pulse duration will be essential to have the longer scale length of the plasma needed for efficient coupling between laser power and extraction of EUV emission. By optimizing the pumping condition, a sufficient optical depth of the plasma will be maintained, keeping the source size below the etendue limit.

The coefficient of radiative transfer calculated using the present model can be applied also to the modelling of discharge pumped plasma (DPP) EUV sources as well as more advanced laser assisted discharge sources, for the calculation of emission spectra as well as the dynamics including the effect of radiation transport. Moreover, the present method of calculation can be applied to other atomic elements with which plasma light sources over the energy range of few 10eV to 10keV can be obtained.

References