The atomic model of the Sn plasmas for the EUV source

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Abstract. An atomic model of Sn to study the level population and radiative properties of near 10 times ionized states in laser produced plasmas is presented. The extreme ultra-violet (EUV) spectrum is investigated in conjunction with the effect of the detailed structure of resonance lines as well as satellite lines from multiply- and inner-shell excited states. The coefficients of radiative transfer are calculated with present model, which used in the radiation hydrodynamics simulation for the optimization of the EUV light source.

1. Introduction

Sn plasmas are being intensively studied as a candidate of the extreme ultra-violet (EUV) light source for the next generation microlithography. More than 180W of EUV output power at a wavelength of $\lambda=13.5\text{nm}$ within the 2% bandwidth is expected using laser produced plasmas (LPP) or discharge pumped plasma (DPP) sources [1].

Laser produced plasmas have been interested for their capacity to produce incoherent and coherent emission up to x-ray wavelength region. However, those sources required extreme conditions to work, therefore industrial application has not been demonstrated. Recently, it is found that plasmas of mid-z ions ($z\approx50$) excited by laser irradiation with a moderate intensity ($\approx10^{10}\text{W/cm}^2$) or discharge can be applicable to high power EUV sources.

The highly charged ions of mid- to high-z elements have characteristic emission spectrum in several different UV to x-ray bands depending on the plasma temperature and corresponding ion charge. In contrast to the wavelength of $\Delta n\neq0$ transitions, which shifts according to the ion charge, the
wavelength of \( \Delta n=0 \) transitions are nearly constant over several charge states, which is useful for application to light sources, as shown in Figure 1.

Sn plasma, heated to electron temperatures of 20-50eV, is ionized up to a state with a mean charge of \( \approx 10 \), which have a ground configuration of \( 4d^{10}nl \), where \( i \) is the occupation number. In the case of these 4d ions, strong configuration mixing takes place between \( 4d^{1-1}4f \) and \( 4p^34d^{i+1} \) configurations, so that predominant emission is obtained at \( \lambda=13.5\text{nm} \), because a large number of 4d-4f and 4p-4d fine structure transitions overlap each other in a same wavelength region [2]. Ions beyond Kr-like state still have intense emission near \( \lambda=13.5\text{nm} \) through 4p-4d transitions, therefore Sn is an efficient emitter over a wide range of plasma temperature.

In addition to the 4d-4f resonance lines, satellite lines from multiply- and inner-shell excited states contribute to the EUV emission. As the optical depth of the plasma increases, the emission intensity from resonance lines at \( \lambda=13.5\text{nm} \) saturates, whereas relative intensity of satellite lines in the longer wavelength increases [3]. Then the spectral width increases resulting in more than 50% of pumping energy is converted to the total EUV emission. However, the conversion efficiency to the 2% bandwidth at \( \lambda=13.5\text{nm} \) should rather increase by decreasing the optical depth of the plasma, by reducing the spectral width and excess emission outside the required emission band, therefore the pumping condition requires optimization [4].

We develop an atomic model of Sn ions and calculate the coefficients of radiative transfer of Sn plasmas, in order to investigate the emission spectrum and conversion efficiency of the EUV sources through the radiation hydrodynamics simulation.

### 2. The atomic model

We develop a collisional radiative model of Sn ions, and the level population is calculated assuming collisional radiative equilibrium (CRE) as a function of electron temperature and ion density. In order to investigate the emission over a wide temperature range, the neutral to Ar-like Sn ions are taking into account.

The atomic states are defined in terms of their non-relativistic electron configuration. A set of rate equations corresponding to each state is solved including ionization and excitation processes in plasmas through radiation and electron collisions. The energy of each state, radiative transition probability and autoionization rates are calculated using the HULLAC code [5]. Rates of collisional excitation and ionization are calculated using empirical formulas as a function of the transition energy, as well as the oscillator strength in the case of excitation [6]. Rates of radiative recombination are also calculated using an empirical formula. Rates of inverse processes such as collisional deexcitation and three-body recombination are determined from the detailed balance.

The ion abundance and level population usually depend on the set of atomic states included in the model. In particular, atomic states with low excitation energies should be included, because these states have a large population. However, Sn ions have a really large number of atomic states. We propose a method to choose an appropriate set of states.
We define a set of excited states of each ion, from low excited states of the next charge state. For example, from the ground and excited states of Mo-like Sn such as 4d⁶, 4d⁵5s, 4d⁵5p, 4d⁴f, 4p⁴d⁷, according to the order of excitation energy, groups of excited states of Tc-like Sn such as 4dⁿˡnl, 4d⁵5snl, 4d⁵5nl, 4d⁴4nl, 4p⁴dⁿl can be generated. The atomic model for all charge states can be determined recursively. Then we perform iterative calculations of level population increasing the number of groups of excited states of each ion, until the ion abundance and radiation loss converge. Typically, we take 5 groups of states into account, for example in the cases of Pd-like to Rb-like ions, we typically include 4dⁿ⁻¹nl, 4dⁿ⁻²5nl, 4dⁿ⁻²5nl, 4dⁿ⁻²4nl, and 4p⁴dⁿnl configurations with maximum principal and orbital quantum number of n≥8 and l≤3. Figure 2 shows the temperature dependence of the mean charge, which converges including more than 5 groups of states [7].

Using the calculated level population, we calculated the emissivity and opacity of the plasma. It is shown that each emission line consists of a large number of fine structure transitions called unresolved transition array (UTA) [8]. Although, as the spectral width of 4d-4f transition arrays are 0.3-0.5 nm, the accurate central wavelength and structure should be determined, in order to apply calculated radiative coefficients to optimization of the efficiency.

We calculate the wavelength and detailed spectral structure of 4d-4f and 4p-4d transitions using the Hullac code considering the effect of configuration interaction (CI). We see energy levels converge after including sufficient CI, and spectral profile agrees well with those observed in the charge exchange spectroscopy (CXS) [9]. Therefore, we used calculated level energies and radiative rates in the collisional radiative model, however transition wavelengths are corrected according to the observed shift between Hullac code and CXS.

Calculated spectrum also depends on how many satellite channels are included in the model, as well as their spectral structure. We calculated the detailed structure of satellite lines, which have considerable contribution to the spectrum. We also corrected the wavelength of 4d-4f, and 4p-4d satellite lines so as their wavelength converge to those of the resonance line of the next charge state, at the limit of the principal quantum number of the spectator electron of n=∞. The same iterative method, which is used to determine level population, is applied to find dominant satellite channels. As shown in Figure 3, the emissivity in the EUV wavelength region increases as the number of groups of levels included in the model increases. Although, the peak emissivity at λ=13.5 nm is almost the same, the spectral width of the emissivity increases considerably by increases number of atomic states in the model. The emissivity also converges if the number of groups of states is increased above 5, where we include 4dⁿnl, 4dⁿ⁻²5nl, 4dⁿ⁻²5nl, 4dⁿ⁻²4nl, and 4p⁴dⁿnl configurations to include resonance and satellite lines of 4d-4f, 4d-5p, and 4p-4d transitions with one spectator electron in an outer orbital.

Figure 2. Electron temperature dependence of the mean charge of Sn plasmas and its atomic model dependence.

Figure 3. Calculated emissivity of Sn plasmas in the EUV wavelength region.
3. Result and discussion

Figure 4 shows calculated specific spectral emissivity and its temperature dependence for the ion density ranging from $10^{17}$ to $10^{21}$/cm$^3$. It is shown that the spectral width increases as the ion density increases, due to increased contribution of the satellite lines.

It is found that the spectral efficiency, which is defined by the ratio of inband emissivity to the total, has a maximum value where the mean charge of the plasma becomes $\approx 10$, which increases as density decreases up to 40% at $n_i=10^{17}$/cm$^3$.

Peak wavelength decreases as temperature increases until it reaches the lowest value of 13nm, and then increases up to 16nm at the electron temperature of 100eV. This arises from the $z$-dependence of the wavelength of the 4d-4f transitions.

Finally, we carry out radiation hydrodynamics simulation with present atomic data. We compared calculated and measured spectrum as well as conversion efficiency to see reasonable agreement [4]. We also find improvement of conversion efficiency using low density plasmas excited by CO$_2$ lasers, which is implied in the dependence of the spectral efficiency on ion density shown in Figure 4.

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