Atomic processes and equation of state of high Z plasmas for EUV sources and their effects on the spatial and temporal evolution of the plasmas

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Abstract. Laser-produced plasma (LPP) extreme ultraviolet (EUV) light sources have been intensively investigated due to potential application to next-generation semiconductor technology. Current studies focus on the atomic processes and hydrodynamics of plasmas to develop shorter wavelength sources at $\lambda = 6.\times$ nm as well as to improve the conversion efficiency (CE) of $\lambda = 13.5$ nm sources. This paper examines the atomic processes of mid-Z elements, which are potential candidates for $\lambda = 6.\times$ nm source using $n=3-3$ transitions. Furthermore, a method to calculate the hydrodynamics of the plasmas in terms of the initial interaction between a relatively weak prepulse laser is presented.

1. Atomic processes of mid-Z elements for shorter wavelength sources

Extreme ultraviolet (EUV) light sources at $\lambda = 6.\times$ nm have attracted attention as a mean to further reduce the process rule of lithographic technology at wavelengths where multilayer optics with a high reflectivity is available. The 4d-4f transition from near Pd-like ions, which is used for $\lambda = 13.5$ nm sources with Sn plasmas, is scalable to shorter wavelengths by increasing the atomic number of the target material. Gd and Tb plasmas show strong emission in the $\lambda = 6.\times$ nm band [1]–[3].

The 4d-4f transition exhibits an unresolved transition array (UTA) with a broad structure [4], from which strong emission can be obtained. However, for sources in the $\lambda = 6.\times$ nm band, the spectral width is too broad compared to the 0.5% bandwidth of the multilayer optics. Moreover, Gd and Tb sources should require a high temperature plasma of $T_e \approx 100$ eV, a pumping laser with a one order of magnitude greater intensity than those used for Sn sources may be necessary. Thus, an alternative source material may be better suited to realize efficient sources.

In this study, we investigate the atomic energy levels and transition probabilities using the HULLAC code [5] for a variety of atomic elements over a wide range of charge states. Figure
1 shows the wavelengths of various transitions from Mo ions, where the numbers indicate the principal quantum numbers of the lower and upper states of the transition. It is shown that Mo can be used as a source for different wavelengths as the ionization proceeds. The figure also suggests that the n=3-3 transition from ions with Z ≈ 20 of Mo may be a potential source at λ = 6.8 nm.

Figure 2 shows the average wavelength of 3d-4f and 3p-3d transitions. The wavelength of the 3d-4f transition depends strongly on the ion charge. In the case of Kr, the emission near the λ = 6.8 nm band can only be obtained from Co-like ion. On the other hand, the wavelength of the 3p-3d transition is almost constant from a Co-like to K-like ions, exhibiting a trend similar to those of the 4d-4f and 4p-4d transitions of Sn.

Figure 3 shows the calculated emission spectrum from the Mo plasma. The emission from a plasma sphere with a radius of 0.3 mm is calculated assuming that the plasma is in a local thermal equilibrium (LTE) state [7]. The strong peak at λ ≈ 6.9 nm arises from the 3p-3d (j=3/2-1/2) transition of Co-like Mo. The estimated spectral efficiency, which is defined by the ratio of the emission in 0.5% bandwidth relative to the total emission, is 1%. It has been shown that the calculated wavelengths have an uncertainty due to the effect of the configuration interaction. For this line, the calculated wavelength is 6.8711 nm, which is slightly shorter than the value in the NIST database (6.9596 nm) [6]. The emission line should be chosen using its accurate wavelength and then the co-optimization with the multilayer should be carried out.

**Figure 1.** Average wavelengths of various transitions of Mo ions.

**Figure 2.** Average wavelengths of (a) 3d-4f transition of Cu-like to K-like ion of Ge to Sr, and (b) 3p-3d transition of Co-like to K-like ion of Sr to Cd.

**Figure 3.** Calculated emission spectrum from Mo plasma at $T_e = 43$ eV and $n_i = 10^{19}$ cm$^{-3}$. Arrow denotes the 3p-3d (j=3/2-1/2) transition of Co-like Mo.
2. Modeling the development of the spatial structure of a laser irradiated Sn droplet

During the development of EUV sources at $\lambda = 13.5$ nm, a mass-limited target such as a small droplet of Sn with a radius of few 10 $\mu$m has been found to be useful to minimize the emission of debris. However, such a droplet is smaller than the spot size of the laser, resulting in an inefficient absorption of the laser energy. To increase the efficiency and EUV power from a plasma, a longer scale length of the plasma is useful. Therefore, double-pulse irradiation is employed, where the first laser pulse produces a plasma with a scale length of $\approx 100$ $\mu$m and the second laser pulse heats the plasma to a temperature sufficient for the EUV emission, as illustrated in fig. 4 [8]. Because the average density of the mist is reduced three orders of magnitude compared to the solid density, the power balance model suggests that a CE of more than 4% is feasible [9]. To further optimize of EUV sources, the initial interaction between the relatively weak laser pulse and the solid target as well as subsequent fragmentation of the target material are important.

Anisimov and Khokhlov have suggested mechanisms for the instabilities associated with the melting, evaporation, and condensation processes during the interaction between a relatively weak laser pulse and the target material [10]. We have investigated modeling methods of such phase transition processes and their effects on the structure of the plasma. Figure 5 shows the Van-der-Waals equation of state of Sn [11]. Sn has a two-phase region below the critical temperature of $T_c = 7297$ K in which the liquid and gas phases of Sn coexist with an arbitrary arrangement.

Because the material in a cell is assumed to be in a single state in conventional hydrodynamics simulations, the average state is considered even for a two-phase region. In this study, as shown in fig. 6, the cell is divided to two once the temperature and volume of the cell reach the liquid-gas two-phase region. The volume fraction and mass of each cell, which correspond to the liquid and gas phases, is determined according to the Maxwell construction assuming that the total volume is conserved, but the arrangement of each region is determined using a probability. This method allows the spontaneous formation of the plasma structure due to phase transition processes to be simulated. As an example, fig. 6 also shows the result of a test calculation for the fragmentation of a liquid droplet during isothermal expansion.

3. Summary

We show the results of the recent studies on the atomic processes and hydrodynamics of plasmas used as EUV sources. We propose a method to simulate the hydrodynamics of the initial interaction between a laser and the matter as well as subsequent particle formation. In the future, we intend to theoretically and experimentally validate our method and apply it to the optimization of EUV sources.
Figure 5. Van-der-Waals equation of state for Sn, and schematic diagram of the possible arrangements in the liquid and gas phase regions.

Figure 6. Modeling of the liquid-gas phase transition using cell division. When the density and temperature of the cell reach those of the two phase region, the cell is divided into two cells, where one is in the liquid phase and the other is in the gas phase.

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