Deposition and sputtering yields on EUV collector mirror from Laser Plasma Extreme Ultraviolet Sources

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Abstract. Based on the self-similar solution of gas dynamic equations, spherical expansion of the highly ionized plasma with limited mass into a vacuum is investigated for the droplet target laser-produced plasma extreme ultraviolet (LPP-EUV) sources. Using partially numerical and partially analytical technology, the velocity, the temperature and the density profiles in the plume versus ionization degree, adiabatic index and initial conditions are presented. Furthermore, the spatial thickness variations of the deposited substrate witness and ion sputtering yields for Ru, Mo, and Si under Sn ion bombardment are theoretically calculated, which can be useful to enable LPP-EUV sources suppliers to estimate collector lifetime and improve debris mitigation systems.

1. Introduction

Extreme ultraviolet lithography (EUVL) is currently a leading candidate for the manufacture of integrated circuits at the 32 nm node and below [1]. Recent theoretical and experimental data clearly demonstrate the advantage of the combination of a CO₂ laser with a Sn for the high conversion efficiency [2]. However, generation of debris in EUVL sources is an inherent and real threat to the lifetime of collection optics [3]. In order to mitigate the problem of debris from the Sn plasma, a mass-limited Sn-based target such as Sn droplet target with a diameter in the range of tens of µm, which has a minimum mass that can still deliver the sufficient EUV power, is effective [4,5]. Much theoretical work, using self-similar theory, has been done to understand the many complex physical processes in the plume dynamics and to reproduce the temporal and spatial evolution of the profiles in the pulsed laser deposition (PLD) expansion [6,7,8]. Nevertheless, Most of such previous works are described in planar geometry and the relevant analytical work on the droplet target LPP-EUV source plume expansion is rare. Moreover, most of existing self-similar models describing the behavior of expanding laser produced plasma plume are based on the neutral gases assumption. Recently, X.Y. Tan et.al. [9] and D.Bennaceur-Doumaz et.al. [10], using the Saha equation to estimate the ionization fraction, studied the single ionization effect in the expanding plume. However, the species emitting extreme ultraviolet light are highly ionized ions. In the present work, we deal with a theoretical investigation of highly ionized plasma expansion, limited to very small temporal and spatial domains.
just after the shot pulse, valid in vacuum, especially for LPP-EUV micro-droplet target sources. A simple analytical model is presented for hydrodynamic expansion of laser-produced plasma with a limited mass, which expands isothermally during laser irradiation and adiabatically after turning off the laser under the spherical geometrical conditions. Based on the gas dynamic equations, using the self-similar solution assumption and taking into account the high ionization degree of the plume, the ionization degree, adiabatic index and initial conditions are investigated to find their influence on the expansion features. The following spatial thickness variations and energetic ion-induced normal incidence sputtering yields of the Mo/Si multilayer materials are calculated.

2. Description of the model
Hydrodynamic spherical expansion of plasma plume into vacuum from a droplet target produced by pulsed laser irradiation can be divided into two stages before particles reached the mirror surface. The first stage is isothermal expansion during laser irradiation. After the cessation of the pulsed laser radiation, there is no additional input in the number of particles injected into the plasma from the target, nor is there any absorption of laser energy. Thus an adiabatic expansion occurs where the temperature can be related to the dimension of the plasma by a well-known thermodynamic relation given by

\[ T \left( \frac{4}{3} \pi R^3 \right)^{-\gamma^{-1}} = \text{const}. \quad (1) \]

In this equation, \( \gamma = c_p/c_v \) is the adiabatic exponent and \( R \) refer to a time-dependent spherical radius of the leading edge of the plasma. The two successive expansions are found to be connected smoothly with each other in time and space and each of the two stages can be considered separately [11]. The later stage leads to the characteristic nature for the laser deposition and sputtering yields on collector mirror process.

2.1 Plasma expansion dynamics
During the initial stage of plasma expansion, the particle density is sufficiently high and the mean free path of the particles is short allowing the plasma to behave as a continuum fluid. Many collisions take place between the various particles so the plasma can be considered in local thermal equilibrium (LTE) [10]. This means that in a sufficiently small region, the electrons, ions and neutrals can be characterized with a common temperature. The plasma plume motion dynamics were considered as a compressible, viscous ideal gas. Therefore, the plume expansion is described by solution of gas-dynamic Euler equations in spherically symmetric case \((r \text{ is the radial coordinate})\) and they are [12]

\[ \frac{\partial \rho}{\partial t} + \frac{1}{r^2} \frac{\partial}{\partial r} (r^2 \rho u) = 0 \quad (2) \]

\[ \frac{\partial u}{\partial t} + u \frac{\partial u}{\partial r} + \frac{1}{\rho} \frac{\partial p}{\partial r} = 0 \quad (3) \]

\[ \frac{\partial T}{\partial t} + u \frac{\partial T}{\partial r} + (\gamma - 1) \frac{T}{r^2} \frac{\partial}{\partial r} (ru^2) = 0 \quad (4) \]

where \( \rho, u, P \) and \( T \) are the plasma vapor density, mass velocity, pressure and temperature, respectively.

When matter, especially medium and/or high \( Z \) element, is heated to high temperature, it will be ionized many times. The degree of ionization has a strong effect on many plasma properties. In LTE case, an approximate method to calculate mean ionization degree which is fitted to the scaling law and numerical results of the ionization potential of Thomas-Fermi statistical model is given in the form [13]

\[ \eta = Z \left[ 1 - \exp \left\{ -4.115 \left( \frac{T^{3/2}}{Z^{4/3}} \ln \frac{A \cdot T^{3/2}}{\eta \cdot n_i} - 1.16 \times 10^{-4} \right) \right\} \right] - 0.5 \quad (5) \]
where $A=4.8\times10^{24} \text{cm}^3 \text{\(10^9\)K}^{-3/2}$, is the mean ionization degree of the plume, $n_i$ is the ion number density, $Z=50$ for Sn element. Eq. (5) can be iterative calculated to determine $\eta$ for the particular ion number density and temperature. Under the condition of quasi-neutrality assumption $n_i=\eta n_e$, the governing equations (2)-(4) are closed by an ideal gas equation.

Equations (2)-(4) can be constructed for a self-similar isentropic solution in the following form:

$$u = \frac{r}{R} \frac{dR}{dt}$$  \hspace{1cm} (6)

$$\rho = \frac{M}{J_1 R^2} \left(1 - \frac{r^2}{R^2}\right)^{\gamma/2}$$  \hspace{1cm} (7)

$$P = (1+\eta) \frac{E}{J_1 R^3} \left(\frac{R_0}{R}\right)^{3(\gamma-1)} \left(1 - \frac{r^2}{R^2}\right)^{\gamma/2}$$  \hspace{1cm} (8)

$$T = \frac{\mu E}{R} J_1 \left(\frac{R_0}{R}\right)^{3(\gamma-1)} \left(1 - \frac{r^2}{R^2}\right)^{\gamma/2}$$  \hspace{1cm} (9)

where $R_0$ is the initial size of the plume, $r$ is the radial coordinate, $R^*$ is the universal gas constant, $\mu$ is the molar mass of the fuel material. The values of the total mass $M$ and the initial energy of the plasma $E$ in expressions (7)-(9) are defined as the integrals over the volume of the plume. $J_1$ and $J_2$ are the normalization constants determined from the conditions of mass and energy conservation.

Substituting self-similar solutions (6)-(9) into the set of gas dynamic equations, one can easily find that the continuity Eq. (2), as well as the Eq. (4), are fulfilled identically. The Euler’s Eq. (3) is transformed into an ordinary differential equation which describes the motion law for expanding plume boundary. The derived equation is similar to the formula in Ref. [9], which can be reduced to the normalized form as follows:

$$\frac{d^2 R^*}{dt^2} = (5\gamma - 3)(1+\eta) \left(\frac{1}{R^*}\right)^{3(\gamma-1)}$$  \hspace{1cm} (10)

$$R^* \bigg|_{t^*=0} = 1, \frac{dR^*}{dt} \bigg|_{t^*=0} = u_0^*.$$  

Here, we used the dimensionless variables $t^* = R_0 \sqrt{M/E}$, $t^* = t/t_0$, $R^* = R/R_0$, $u_0^* = u_0 J_1 / R_0$.

The Eq. (10) has the solution in the form of inverse function through further integration

$$t^* = \left[u^2_0 + \frac{2}{3} (\gamma-1) \right] \left[\frac{1}{2} (a,b; c; -(3+5\gamma)(1+\eta)) \frac{1}{1+\frac{3}{2} (\gamma-1)u_0^2} \right] - \frac{1}{2} (a,b; c; -(3+5\gamma)(1+\eta)) \frac{1}{1+\frac{3}{2} (\gamma-1)u_0^2}.$$  \hspace{1cm} (11)

where $F(a,b;c; z)$ is the hypergeometric function with parameters $a=1/2$, $b=-1/3$, $c=1+b$.

The normalized density, pressure and temperature profiles within the plume are given by

$$\rho^* = \frac{\rho}{\rho_0} = \left(\frac{1}{1+\xi^2}\right)^{\gamma/2}$$  \hspace{1cm} (12)

$$P^* = \frac{P}{P_0} = (1+\eta) \left(\frac{1}{R^*}\right)^{\gamma/2} \left(1 - \xi^2\right)$$  \hspace{1cm} (13)

$$T^* = \frac{T}{T_0} = \left(\frac{1}{R^*}\right)^{3(\gamma-1)} \left(1 - \xi^2\right)$$  \hspace{1cm} (14)

where $\rho_0 = \frac{M}{J_1 R_0^3}$, $P_0 = \frac{1}{J_2 R_0^3}$, $T_0 = \frac{J_1 \mu E}{J_2 R_0^3 \eta}$ and $\xi = \frac{r}{R} (0 \leq \xi \leq 1)$ is the Lagrangian coordinate.

2.2 Deposition and sputtering EUV mirror
In the case of Sn sources, mirror lifetime presents great challenge. Apart from erosion due to high energetic ions, Sn also condenses and deposits on mirror surfaces. The EUV reflectivity in this case is affected by erosion as well as roughness caused by deposition. A Mo/Si witness sample is exposed to a Sn EUV sources (see Fig.1). The plasma plume material contaminates the sample and causes non-uniform thickness profiles \( h(\theta) \). The mass flux to the substrate surface is as follows [6]:

\[
j(r, R_s, t) = j(r, R_s, t) = \begin{cases} \frac{M R_s}{J_s R^3} \frac{dR}{R^2} \left[1 - \frac{r^2}{R^2}\right]^{1/2}, & t \geq t_s(r) \\ 0, & t \leq t_s(r) \end{cases}
\]

Figure 1. Schematic diagram of the spherical expansion plume deposition on witness sample.

Here \( t_s(r) \) is the time when the outer boundary of the expanding cloud reaches the witness surface \( r = R_s \ sec = R(t) \), where \( r \) is the radial coordinate and \( \theta \) is the radial angle. Integrating the mass flux (15) over the time from \( t_s(r) \) to infinity and dividing the result by the density of the deposited material \( \rho_s \), we obtain the thickness profile:

\[
h(\theta) = \frac{M R_s}{\rho_j J_j} \int_{r_s}^\infty \frac{1}{R^3} \left[1 - \frac{R^2}{R^2 \cos^2 \theta}\right]^{1/2} dt
\]

Sputtering is the erosion of a solid surface by energetic particles bombardment. Sputtering yield is defined as average number of ejected target atoms per incident ion. For a simple physical sputtering system, the yield is described in terms of \( Y(E, \theta, M_1, M_2, Z_1, Z_2) \), which is dependent on the incident ion energy \( E \), incident angle \( \theta \) relative to the target surface normal, \( Z_1 \) and \( Z_2 \) are the atomic numbers, \( M_1 \) and \( M_2 \) are the masses of the projectile and the target atom, respectively. Based on the nuclear stopping power for the Thomas-Fermi potential \( S_{TF} \), Yamamura and Tawara derived a new universal equation as follows [14]:

\[
Y(E) = 0.042 \frac{Q(Z_2) \alpha}{U_s} \frac{S_{TF}(E)}{1 + \Gamma k_s e^{0.3}} \left(1 - \frac{E_{th}}{E}\right)^3
\]

where * and \( Q(Z) \) are the parameters fitting to the data available, \( E_{th} \) is the sputtering threshold energy, \( k_s \) is the Lindhard electronic stopping coefficient, is the reduced energy, \( U_s \) is the surface binding energy of the target solid and \( \Gamma = W(Z_2)/[1+(M_1/7)^3] \) is a dimensionless factor. The detailed calculation method is given by Ref.[14].

3. Results and discussions

3.1 The calculation of the velocity evolution of the plume edge

With such a time-dependent homogeneous ionization degree assumption, equation (10) can be numerically solved coupling with equation (5). Fig.2 shows the evolution of ionization degree of monatomic gas ( =1.67) versus normalized expanding plume length under the initial conditions given in table 1. We have noticed the ionization degree decreasing very fast with the expansion as a consequence of recombination processes and decrease of the temperature. The further integration of Eq. (10) yields \( u_s = u_0 + 2(5\gamma - 3)(1 + \eta)E / 3(\gamma - 1)M \), which is consistent to the results from Fig.3. It is
easy to see directly from the derived equation above that the velocity of expansion tends to constant \( \frac{dR}{dt} \rightarrow u_f \) for sufficiently extended time, i.e. inertial stage of expansion. The results of calculations show that the maximum expansion velocity of the ionized plasma is increasing with the ionization degree. This shows that ionization strongly affects the dynamics evolution of plasma, which is also verified by the Ref. [9-10]. In summary, the self-similar behavior of all the expansion profiles when ionization effect is taken into account, are more extended than that of a neutral gas vapor. We can see from Fig.4, the lower value of \( \gamma \) would result in higher terminal velocities, but at the early stage of adiabatic expansion vice versa. For laser-generated ionized species, the value of \( \gamma \) is much lower than monatomic gases, probably in the range of 1.1-1.3 when the degrees of freedom due to excitation and ionization are taken into account. We assume that the external source of energy is removed, so that the sum of the internal energy and the kinetic energy is conserved. This behavior is due to the adiabatic expansion where thermal energy is converted into directed kinetic energy. The plume dynamic behavior is also affected by different initial conditions, including initial plasma edge velocity and radius, i.e. the absorbed CO\(_2\) pulsed laser energy and the entire disintegrated mass of the droplet target by the end of isothermal expansion stage.

Table 1. Initial parameters of the plume which have been used in calculations.

| Initial parameters of the plume | value |
|--------------------------------|-------|
| Initial size, \( R_0 [\mu m] \) | 300   |
| Plume mass, \( M [\mu g] \)    | 2     |
| Plume total energy, \( E [mJ] \) | 50     |
| Initial velocity of expansion, \( u_0 [km/s] \) | 6     |
| Initial temperature, \( T_0 [eV] \) | 33     |

Figure 2. The averaged ion charge variation with the plume size under the assumption of homogeneous ionization degree. The initial parameters are listed in table 1.

Figure 3. The time history of the normalized velocity at the edge of the plume for \( \eta > 0 \) and \( \eta = 0 \).
3.2 Density, temperature and pressure profiles as a function of time and space
The normalized plasma plume density, temperature and pressure distributions versus time and space are given by the Fig. (5) and Fig. (6). In this free-expansion regime, we see that the density, temperature and pressure decrease with time and space. The results are just compatible with the analytical expressions mentioned above. Note that the density, temperature and pressure profiles describe a plume with a sharp external edge. This is the consequence of adiabatic assumption, which leads to the relation (1). Thus, the density, temperature and pressure approach zero at the front, in contrast to an isothermal expansion, where the velocity of sound at the outer edge of the plume remains finite and generates density and pressure tails.
3.3 Plasma deposition thickness profiles and sputtering yields on the EUV mirror
EUVL technology, which is based on reflective optics for 13.5nm, by using Mo/Si reflective coatings with protecting capping layers of Ru, it is possible to achieve reflectivity of as high as 70% [15-16]. However, reflectivity degradation of the collector optics is an effect of debris interaction with the collector optics. For nearly normal incidence mirrors, sputter-induced erosion, in addition to roughness and contamination, plays a critical role in reflectivity degradation. A simple analytical expression for $h(\theta)\cos^{3}\theta$ can be obtained from equation (16) for the case of spherical expansion, which is satisfied in many experimental situations. The thickness variation controlled by adiabatic index is shown in Fig.7. It can be observed that the deposition thickness $h(\gamma)$ increases as adiabatic index and/or substrate-target distance decreases under the same conditions.

As mentioned above, in spite of mirror surface contamination, the energetic species bombardment may induce sputtering damage to the topmost surface. The sputtering yields of Ru, Mo and Si are theoretically calculated and plotted in Fig.8. It can be shown that the lightest element Si has the highest sputter threshold $E_{th}$. When the incident energy is in the range of 10keV, the sputter yields approach an asymptotic maximum value more than 10. Since energetic ion sputtering is found to be responsible for the degradation of reflectivity of the multilayer (ML), it is critical for longer lifetime to reduce energetic species flux to the EUV mirror surfaces.

![Figure 7](image1.png)

Figure 7. The thickness $h(\gamma)$ normalized by that of monatomic gas under the initial conditions listed in Table 1.

![Figure 8](image2.png)

Figure 8. Energy dependence of the sputtering yields of Si, Mo and Ru with Sn using Yamamura and Tawara’s formula.

4. Conclusion
Mathematically, the present self-similar solution under the physics of the one-fluid plasma model exists only for certain specific values of the adiabatic index depending on the spherical geometry in a very small temporal and spatial domain. The initial conditions required for the calculation above are chosen so as to be relevant to the EUV plasma. In addition to tin deposition, a major factor that determines the lifetime is ion sputtering of the material at the collector surface. Controlling or
mitigating the debris has been attempted by various groups using different methods, including cavity confined plasma [17], mass limited droplet targets, foil trap [18], the application of ambient gas, magnetic fields and combined effects [19].

In summary, based on the consideration of ionization degree and conservations of mass, momentum and energy of the plasma, plasma spherical expansion into vacuum has been studied, in which the plume evolvement profiles are obtained after the laser radiation. The influences of ionization degree, adiabatic index and initial conditions on plume expansion are discussed. In order to estimate the collector mirror lifetime and investigate the ML damage mechanism for EUV light source development, the non-uniform collector surface contamination and the sputtering yields of Si, Mo and Ru under the energetic Sn debris bombardment are also calculated theoretically.

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