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Measurements of hydrogen gas stopping efficiency for tin ions from laser-produced plasma

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Experimental studies of stopping of ion fluxes from laser-produced plasma by a low-pressure gas atmosphere are presented. A modification of the time-of-flight spectroscopy technique is proposed for the stopping cross-sectional measurements in the ion energy range of 0.1–10 keV. The application of the proposed technique is demonstrated for Sn ion stopping by H₂ gas. This combination of elements is of particular importance for the development of plasma-based sources of extreme ultraviolet radiation for lithographic applications. *Published by AIP Publishing.*

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Hot dense plasma is very attractive as an efficient source of short-wavelength radiation, including soft X-rays and extreme ultraviolet (EUV) radiation.¹ This can be laser-produced plasma, discharge-produced plasma, or their combination—laser-assisted discharge-produced plasma. The range of applications of plasma-based radiation sources includes water-window microscopy,^{2,3} photoelectron spectroscopy,⁴ optical metrology,^{5–8} and photolithography.^{9,10} Photolithography in the EUV range or EUV lithography is the key driver of the plasma-based radiation source technologies nowadays. Radiation sources for EUV lithography make use of hot-dense tin (Sn) plasmas to produce photons at the wavelength of 13.5 nm. However, besides the production of the useful radiation, plasma-based sources also generate fluxes of corpuscular debris as a side product. These can be Sn micro-particles, ions and neutral atoms.^{11–14} All these are harmful for optical elements of the source. But the ions are considered to be the main concern for the optics lifetime since the optimal operation of the EUV source assumes nearly complete conversion of the target material to the plasma state.⁹

In order to protect the source optics from the plasma debris, various approaches for the debris mitigation are being developed. These are deflection of the ion and plasma fluxes with a strong magnetic field,^{15–17} stopping of these corpuscular fluxes with a background gas,^{18,19} and combination of the two approaches.^{9,20} The application of the background gas is important because it also allows us to mitigate the neutral atom flux (if such is present). The choice of the background gas elemental composition and its pressure level in the source chamber is governed by the EUV transmission of the gas and its effectiveness for debris mitigation. Harilal *et al.* have pointed out that the most transparent gases for EUV

radiation with a wavelength of 13.5 nm are argon (Ar), helium (He), and hydrogen (H₂).²⁰ The interaction of Sn plasma plume with background gases has been studied in various experimental conditions.^{18–22} However, direct measurements of Sn ion stopping in the above-mentioned gases are still lacking. This paper reports on the experimental studies of Sn⁺ and Sn²⁺ ion stopping by hydrogen for ion energies in the range of ~0.1–10 keV.

Energy losses dE of ions moving through a gas layer of thickness dx and particle concentration n are quantified by the stopping cross-section which is defined as

$$S = -\frac{1}{n} \frac{dE}{dx}. \quad (1)$$

Note that the value of dE/dx in Eq. (1) is usually referred to as the stopping power. The stopping cross-section, as defined by Eq. (1), cannot be directly measured. Characteristics of ion fluxes that can be straightforwardly measured are the ion energy, ion current, and ion time of flight. The classical works on the ion stopping measurements used the pre-conditioned mono-energetic ion beams.^{23–27} In such a case, the time of flight for ions with a given energy flying through a gas-filled tube can be reliably measured. Using the measured time of flight, one can determine the ion energy losses over its pass length in the tube and thus determine S . Here, we describe a method of the stopping cross-sectional measurements for plasma sources of ions with a broad energy spectrum. The proposed method also makes use of the time-of-flight measurements, but the ion energy selection is carried out at the end of the tube using an electrostatic ion spectrometer.

To explain the proposed method, let us first consider ions flying through a tube of length L filled with a gas. The mass of each ion is denoted by m . The concentration of the

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gas varies along the tube, i.e., $n(x) \neq \text{const}$. Kinetic energy of the ions is assumed to be much higher than the thermal kinetic energy of the gas particles. We are interested only in ions that arrive to the end of the tube with a certain energy E . For any given gas concentration profile $n(x)$, the end energy E corresponds to a certain value of the initial ion energy E_0 , i.e., the ion energy at the entrance of the tube. The loss of the kinetic energy of such ion species at a distance x from the entrance of the tube can be calculated using the definition of the stopping cross-section as

$$\Delta E(x) = E_0 - E(x) = \int_0^x n(x') S(E(x')) dx'. \quad (2)$$

According to Eq. (2), $E_0 - \Delta E(x)$ corresponds to the ion energy at the distance x from the entrance of the tube. Then, $E_0 - \Delta E(L) \equiv E$. Since the experimental studies deal with gas pressure p rather than gas concentration n , it is convenient to rewrite Eq. (2) using the ideal gas law ($p = nkT$) as

$$\Delta E(x) = \frac{1}{kT} \int_0^x p(x') S(E(x')) dx', \quad (3)$$

where k is the Boltzmann constant and T is the gas temperature. Let us introduce notation $\Delta E'(x) = \Delta E(L) - \Delta E(x)$ that will be used below.

Another important assumption we make is that the overall energy loss on the pass length L is much smaller than the initial energy, i.e., $\Delta E(L) \ll E_0$. Hence, $S(E)$ can be treated as constant. Then, one can write

$$\Delta E'(x) \approx \frac{S(E)}{kT} \left(\bar{p}L - \int_0^x p(x') dx' \right), \quad (4)$$

where $\bar{p}$ denotes the average pressure of the gas in the tube

$$\bar{p} = \frac{1}{L} \int_0^L p(x) dx. \quad (5)$$

The total time of flight of the considered particle can be calculated via its velocity $v(x)$ as

$$t = \int_0^L \frac{dx}{v(x)} = \int_0^L \frac{dx}{\sqrt{2(E + \Delta E'(x))/m}}. \quad (6)$$

Applying the Taylor series expansion for the denominator of the integrand in Eq. (5), one can derive

$$t \approx t_{vac} \left(1 - \frac{1}{2EL} \int_0^L \Delta E'(x) dx \right), \quad (7)$$

where $t_{vac} = L/\sqrt{2E/m}$ corresponds to the time of flight of ions with energy E through the tube at zero gas pressure. Using Eqs. (3) and (6), one can calculate the difference between t and t_{vac} as

$$\Delta t = t - t_{vac} \approx \frac{t_{vac} S(E) \bar{p} L}{4E} \eta, \quad (8)$$

where

$$\eta = \frac{\bar{p}L^2 - \int_0^L \int_0^x p(x') dx' dx}{\bar{p}L^2/2}. \quad (9)$$

η is a form-factor that characterizes the non-uniformity of gas distribution in the tube. For uniform distribution, $\eta = 1$. From Eq. (10), it follows that the stopping cross-section for the given ion energy E can be calculated as

$$S(E) = -\frac{4EkT}{p_{eff}L} \frac{\Delta t}{t_{vac}}, \quad (10)$$

where $p_{eff} = \bar{p}\eta$. Note that the product $p_{eff}L$ characterizes the total number of collisions of the considered ion over the path length L for a given gas pressure distribution $p(x)$.

Our experiments on the measurements of the hydrogen gas stopping for Sn ions have been designed according to the above described approach. The experimental setup is schematically shown in Fig. 1. For producing tin plasma, a pulsed CO₂ laser system operating at the wavelength of 10.6 μm was used. The laser system was assembled from two Transversely Excited Atmospheric (TEA) laser units Infralight SP-10 by Optosystems. One was used as a master oscillator, and the other was used as an amplified oscillator. The CO₂:He:N₂ gas mixture with a 300:650:40 stoichiometric composition was used as the active medium. An Ophir PE-50 IR detector was used to measure the laser pulse energy. The maximum pulse energy in the experiments was 553 mJ (energy delivered to the target surface with account for all reflection losses). The laser beam was focused on the target surface using a ZnSe lens with a focal length of 20 cm. The target in the form of a tin plate attached to a rotating stage was placed in the center of a stainless steel vacuum chamber. Ion flux from plasma was analyzed using a custom designed cylindrical electrostatic ion spectrometer by ISTEQ B.V. The spectrometer was attached to the vacuum chamber through a long time-of-flight (TOF) tube. The overall distance between the target surface and the entrance of the spectrometer was 156 cm. Hydrogen was supplied into the chamber through the inlet located approximately in the middle of the TOF tube. The gas pressure was measured using PMI sensors at three different points as indicated in Fig. 1.

The experiments were performed for the maximal pressure of hydrogen in the chamber varying in the range of 10^{-2} Pa to 1 Pa. For each experimental point, we used data from all pressure sensors to calculate the pressure profile in the tube. For that, we used linear interpolation between the points (x_i, p_i) . Using these pressure profiles, the value of p_{eff} [involved in Eq. (10) for S] was calculated. Obviously, the base pressure in the chamber is caused by the air leakage and outgassing, but not by hydrogen. The base pressure also provides stopping of the ions. For that reason, when calculating

FIG. 1. Scheme of the experimental setup.

the value of p_{eff} for a given pressure profile of hydrogen, we subtracted the value of p_{eff} corresponding to the base pressure.

Our spectrometer allowed us to explore the ion energy range of 100 eV to 5 keV in the experiments. For each ion energy, signals from the spectrometer were recorded for different pressure values in the tube. Figure 2 shows the example of ion signals recorded for $E/Z = 720$ eV at $p_{eff}L = 0$ and $p_{eff}L = 0.081$ mBar cm of the hydrogen pressure. At zero pressure, one can see four peaks corresponding to Sn^+ , Sn^{2+} , Sn^{3+} , and Sn^{4+} ions and peaks corresponding to impurities. At higher pressures only Sn^+ and Sn^{2+} were observed—higher ionization degrees vanish due to electron exchange reaction that occurs at tin-hydrogen collisions.

In order to determine S for a given ion energy E , we measured the dependence of time-of-flight t of the detected ions with this energy on the gas pressure, more precisely on the value $p_{eff}L$. Note that the experimental time of flight is defined as the center of mass of a considered ion peak from the spectrometer signal. Figure 3 exemplifies a plot of $t - t_{vac}$ versus $p_{eff}L$ for Sn^+ ions with $E = 720$ eV. Obviously, the slope of this curve gives the value of $\Delta t/p_{eff}L$, which is needed to determine S according to Eq. (10). Thus, from the measured data, we obtain $\Delta t/p_{eff}L = -5.51 \mu\text{s}/(\text{mBar cm})$ and $t_{vac} = 49.4 \mu\text{s}$ which result in $S = 1.4 \times 10^{-14} \text{ eV cm}^2$ for Sn^+ ions with $E = 720$ eV. The main cause for the measurement errors is the variation of pressure in the system during the measurements. Acquisition of each ion spectrum takes 5–10 min. It was found that the pressure value can vary by $\pm 10\%$ during this time.

The described algorithm of S measurements was applied over the entire aforementioned energy ranges for Sn^+ and Sn^{2+} ions. The resulting plot of S on E is given in Fig. 4. It is important to note that the measured stopping cross-section can be approximated with the power law $S = cE^\alpha$ for the ion energies up to approximately 5 keV. The least squares fitting gives the best approximations $S = 1.59E^{0.7}$ and $S = 1.46E^{0.54}$ for Sn^+ and Sn^{2+} ions, respectively. The best fit for Sn^{2+} ions is close to the root dependence of S on E , i.e., the stopping cross-section is nearly proportional to the ion velocity. This might be an indication for the physical mechanism responsible for the energy loss of the Sn ions in collisions with the H_2 molecules: such scaling arrears in the Firsov²⁸

FIG. 2. Signals from the electrostatic ion spectrometer obtained when its settings tuned for registration of Sn^+ ions with N energy of 720 eV. The red line corresponds to $p_{eff}L = 0$, and the purple line corresponds to $p_{eff}L = 0.081$ mBar cm.

FIG. 3. Measured time-of-flight for Sn^+ ions with an energy of 720 eV vs $p_{eff}L$.

and Lindhard and Scharff²⁹ models describing the electronic mechanisms of the stopping of slow heavy ions in matter. It is also remarkable that the measured stopping cross-section for Sn^{2+} ions is lower than that for Sn^+ ions for energies above 1 eV. This can be interpreted as another indication for the electronic mechanism of ion stopping. The prime parameter in both the aforementioned models is the radius of electron density or atomic radius a . More precisely, the Firsov model suggests that the stopping is proportional to a .³⁰ The electron configurations of Sn^+ and Sn^{2+} ions are $[\text{Kr}]4d^{10}5s^25p^1$ and $[\text{Kr}]4d^{10}5s^2$, respectively. The Sn^{2+} ion has a closed shell electron configuration, which results in a smaller ionic radius. Hence, according to the Firsov model, stopping for Sn^{2+} ions should be lower than that for Sn^+ ions, which agrees with the experimental results.

To summarize, we experimentally investigated stopping of ion fluxes from laser-produced plasma by a low-pressure gas atmosphere. For that, we proposed a modification of the time-of-flight spectroscopy technique. The application of the proposed technique was demonstrated for Sn ion stopping by H_2 gas. This combination of elements is of particular

FIG. 4. Stopping cross-section of H_2 gas for Sn ions: red dots correspond to the measured data for Sn^+ ions; green dots correspond to the measured data Sn^{2+} ions; Solid lines correspond to the model fits of the experimental data with the power law.

importance for the development of plasma-based sources of EUV radiation for lithographic applications. As a result of the measurements, we obtained the data for the H_2 gas stopping cross-section for Sn^+ and Sn^{2+} ions in the energy range of ~ 0.1 – 10 keV. The data can be used to optimize operation of the industrial EUV sources that use H_2 gas for Sn ion mitigation.

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