Influence of Nonequilibrium Ionization Process on the Efficiency of a Plasma Discharge Xenon Source

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(Received 14 October 2003 / Accepted 21 October 2003)

Abstract
In connection with fast plasma heating in capillary discharges, the transient ionization effect on the plasma conversion efficiency of 4d–4d transitions ($\lambda \approx 13.5$ nm) of xenon has been theoretically investigated. Results indicate that ion abundance is a function of the history of plasma evolution, and the fast heating process produces higher conversion efficiency values compared with the steady-state plasma.

Keywords: extreme-ultraviolet lithography, EUV source, conversion efficiency, capillary discharge, ionization dynamics

We were motivated to carry out the present work by our interest in the possible use of pulsed-power plasma sources of the Z-pinch type in light source experiments. Extreme-ultraviolet (EUV) sources in ionic systems using laser-produced plasmas and discharge-produced pinch plasmas have been investigated both theoretically [1-3], and experimentally [4-8]. The major efforts in EUV source development are now focused on xenon as the plasma emitter. In xenon, the transitions responsible for a wavelength of $\lambda \approx 13.5$ nm are in the Xe$^{10}$ ionization state. In plasmas created by capillary discharge, ionization dynamics are extremely important due to the very fast timescales of changing plasma parameters at pinching time, and ionization state distributions are considered to be far from their steady-state values. The goal of the present letter is to investigate the effects of nonequilibrium ionization (NEI) on the conversion efficiency of a Xe-EUV source based on fast heating in capillary discharge plasmas. We have presumed that the excited states are in local thermodynamic equilibrium. Therefore, in order to evaluate the NEI effects on conversion efficiency, only the time evolution of ground states is traced using a set of coupled differential equations following the suggested idea of describing a non-LTE transient plasma by a few dominant ion states [9,10] as

$$\frac{dn_k}{dt} = n_e[T_{k-1}n_{k-1} + S_k - n_{k-1} - (T_k + S_k)n_k],$$

$$k = 1, ..., Z_a + 1,$$

where $n_k$ is the ground state number density of ion $k$, $T_k = R_k + n_eC_k + D_k$ the total recombination term, and $Z_a$ the atomic number. Here, $S_k$, $R_k$, $C_k$, and $D_k$ are respectively, the electron collisional ionization, radiative, three-body, and dielectronic recombination (DR) rate coefficients of ion $k$ with condition $T_1 = S_0 = T_{Z_a+2} = S_{Z_a+1} = 0$. The treatment of rate coefficients has been described in Refs. [11,12]. In this approach, the ionization state distributions are practically identified by two parameters, namely, the electron temperature and the time integral of the electron density ($\int n_e dt$, in units of cm$^{-3}$ s), often denoted by $n_e t$.
The \( n_e \tau_R \) (where \( \tau_R \) is the effective relaxation time) values required for the partially ionized xenon to reach a steady-state is shown in Fig. 1 (a). In order to calculate the relaxation time, we followed the time evolution of ionization state distributions until the average ionic charge state at a given electron density and electron temperature \( kT_e \) is close to 99% of its steady-state value [11]. In this letter, the time evolution of electron density at a given electron temperature and time is computed using ion density and average ionic charge state. When an envelope is taken from the ionization state distributions at time \( t \geq \tau_R \) as shown in Fig. 1 (b), a correlation is seen between different degrees of ionizations and the \( n_e \tau_R \) diagram. Namely, the small value of \( n_e \tau_R \) is a result of the closed shell structure of the Pd-like (Xe+8) ion. A comparison of Figs. 1 (a) and 1 (b) show that the relaxation time for the Xe+10 ionization state can be approximately scaled with electron density as

\[
\tau_R \approx 5 \times 10^{10} \frac{n_e}{(\text{cm}^{-3})} \text{(s)}
\]

The average ionic charge state against electron temperature is shown in Fig. 1 (c), for different values of \( n_e \tau_R \). The solid line \( n_e \tau_R = 3 \times 10^{11} \text{ cm}^{-3} \text{ s} \) almost corresponds to the result of the steady-state ionization model. An inspection of Figs. 1 (b) and 1 (c) indicates that plateau regions are due to the closed shell ionization state distribution, i.e., Xe+8 degree of ionization. As the value of \( n_e \tau_R \) decreases, the average ionic charge state gets smaller at a given electron temperature due to the finite time required for the ionization to occur.

The transient effects become of increasing importance if the time scales for atomic processes are large in comparison to the time scales for the change of the plasma parameters. For example, if the plasma suffers a fast heating process such as shock wave heating (i.e., \( S_{\text{heating}} \leq (n_e \tau_{\text{heating}})^{-1} \)), where \( S_{\text{heating}} \) is the ionization rate coefficient, and \( \tau_{\text{heating}} \) the heating time), or in the case of fast cooling (i.e., \( T_{\text{cooling}} \leq (n_e \tau_{\text{cooling}})^{-1} \)), where \( T_{\text{cooling}} \) is the recombination rate coefficient, and

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**Fig. 1** (a) Diagram of \( n_e \tau_R \) (cm\(^{-3}\) s) value versus electron temperature, where \( \tau_R \) is the relaxation time. (b) The xenon ionization state distributions at time \( t \geq \tau_R \). (c) The average ionic charge state for different values of \( n_e \tau_R \) (cm\(^{-3}\) s) against electron temperature; \( n_e \tau_R = 3 \times 10^4 \) (solid), \( n_e \tau_R = 3 \times 10^6 \) (dotted line), \( n_e \tau_R = 3 \times 10^8 \) (dashed line), and \( n_e \tau_R = 3 \times 10^9 \) (dot-dashed line). Note that the scale of the abscissa is not same.

**Fig. 2** Time evolution of some ionization states (ionization stage of Xe+10 in NEI model: solid line) as a function of \( n_e \tau_R \) (cm\(^{-3}\) s) when electron temperature increases linearly versus time from 0.1 to 150 eV during 15 ns (thick dot-dashed line). The initial ion density is assumed \( N_T = 5 \times 10^{17} \text{ cm}^{-3} \), and the time evolution of the electron density is calculated using the average ionic charge state. For comparison, the Xe+8 ionization state (dashed line) calculated using the steady-state ionization model is shown.
to achieve the maximum Xe$^{+10}$ ionization state due to the finite relaxation time than in the case of steady-state plasma.

In the capillary discharges, the high-energy-density plasma is made by an electromagnetic implosion accompanied shock wave [13]. The well-known Rankine-Hugoniot relations predict that for very high Mach numbers the temperature for each particle species is proportional to the particle mass. Therefore, rapid collisional shock heating initially heats the ions. Also, at the final phase of the implosion, the plasma is heated under a condition of almost constant ion density. In order to understand the time-dependent behavior of the conversion efficiency of a xenon source excited in capillary discharges, we have performed calculations for a transient case (i.e., at regime of $n_{e,f} \leq 10^{11}$ cm$^{-3}$ s) using shock wave equations [14].

We have estimated the conversion efficiency using the ratio of radiation energy to plasma energy as described in [15-17]

$$\eta = \frac{L_\lambda \pi r_p^2 \Delta \lambda \Delta \lambda}{E_{\text{plasma}}}.$$  (2)

where $L_\lambda$ is spectral radiance, $\Delta \lambda$ the line profile, $r_p$ the plasma radius, and $\Delta \lambda$ emission duration. The plasma energy is calculated based on the ionization energy for generating the emitting ion ground state and the excitation energy for generating at least one photon for each ion. In a thermal plasma there is an additional need to heat both ion and electron gas as: $E_{\text{plasma}} = E_{\text{i-ion}} \pi r_p^2 l_{\text{plasma}} n_i$, and $E_{\text{im}} = E_{\text{exc}} + \sum_{i=1}^{Z_{\text{ave}}} I p_{\text{ion}} + \frac{3}{2} k T_e + \frac{1}{2} Z_{\text{ave}} k T_e$. Here, $I p_{\text{ion}}$ is the ionization potential of the $i$th ionization state, $l_{\text{plasma}}$ plasma length, $n_i$ ion density, $Z$, and $Z_{\text{ave}}$, respectively, the mean ionic and the average ionic charge states [18]. Calculations of the ionization balance and the opacity in the line center of a Doppler-broadened line in a stationary plasma [19] show that for temperatures ranging $\approx 20 - 100$ eV and an electron density of $n_e = 5 \times 10^{18}$ cm$^{-3}$, the optical depth of the $\lambda = 13.5$ nm transition of Xe$^{+10}$ (oscillator strengths $= 0.18$ [5]) changes from $= 5$ across the plasma (radius = 0.02 cm) to $= 25$ along the plasma column (length = 0.1 cm). Hence, in order to estimate spectral radiance, Planck’s law is assumed as an approximation (which means the excited states are in local thermodynamic equilibrium) as

$$L_\lambda = B_\lambda \left(1 - \exp (-\tau_\lambda)\right),$$

where $\tau_\lambda$ is the optical depth at the line center of the Doppler-broadened line, and $B_\lambda d\lambda$ is the energy emitted per unit time, surface area, and the solid angle in the spectral interval ($\lambda, \lambda + d\lambda$) given by

$$B_\lambda (k T) = \frac{2 h^2 c^2}{\lambda^5} \frac{1}{\left[g_i n_i \right]_{\text{LTE}} - 1}$$

$$= \frac{2 h^2 c^2}{\lambda^5} \frac{1}{\exp \left[\frac{hc}{\lambda k T}\right] - 1}.$$  (4)

Here, $h$ is Planck’s constant, $c$ the speed of light in a vacuum, $g_m$ the statistical weights of level $m$, and $n_m$ the population of level $m$.

Since the broadening of the optically thick line depends only weakly on the optical depth in the line center, we have assumed that the line profile is $\Delta \lambda \approx 2 \lambda_{\text{Doppler}}$ [7,15].

The effect of shock heating with different Mach numbers on the Xe$^{+10}$ ionization state is shown in Fig. 3 (a) at a constant total ion density of $N_T = 10^{18}$ cm$^{-3}$, and an initial condition for temperature as $k T_e = k T_i = 10$ eV. We have made the simplifying assumption that the plasma is in equilibrium at the initial time and temperature at which it starts, and the initial ionization state distributions are calculated using a steady-state ionization model. Following shock heating, the thermalization of electrons and ions to a common
temperature is calculated based on a rate equation using Coulomb collision equilibration time [20]. Therefore, the temperature profiles are defined by the strength of the shock wave. For comparison, Fig. 3 (a) also shows the Xe$^{+10}$ stage calculated using the steady-state ionization model (solid line). The change of the corresponding conversion efficiency of the heating plasma to the radiation with $\lambda \approx 13.5$ nm ($\Delta \lambda \approx 0.27$ nm around 13.5 nm) as a function of electron temperature is shown in Fig. 3 (b), and for comparison, its steady-state value. The conversion efficiencies were estimated for a xenon plasma with dimensions of 0.02 cm in radius, 0.1 cm in length, and an emission duration of 40 ns. In fact, the emission duration at pinching time depends on ionization time and also on recombination time due to the reduction of electron density owing to the plasma’s expansion. In such a case, the recombination processes freeze out, a phenomena which has been noted many times previously [21]. Since the spectral radiance was based on Planck’s law: $L_\lambda \propto \frac{1}{\exp\left(\frac{hc}{\lambda kT}\right) - 1}$, \(\lambda\) is proportional to the population of the ground state of interest ion (Xe$^{+10}$ ionization state), and in which the higher temperatures are favorable as long as a sufficient ground state of interest ion is existent to maximize conversion efficiency. Figure 3 reveals that in the presence of the process involving a rapid rise in electron temperature, the consequent lagging of ionization states behind the electron temperature due to the relaxation effects leads to an increase in the EUV output of a discharge-pumped xenon source.

More detailed estimation requires a calculation where magnetohydrodynamics, the atomic physics of ionization and excitation, and radiation transport are coupled self-consistently and advanced in time. However, calculated results regarding the population of excited states based on the quasi-steady-state hypothesis also revealed the same quantitative behavior of conversion efficiency as that described in present letter [12]. In fact, a reduction in the mismatch between the electron temperature that maximizes the Xe$^{+10}$ ionization stage in a steady-state situation, and optimizes the kinetics of the excited state owing to the relaxation effects, leads to an increase in the conversion efficiency of xenon in a band of $\lambda \approx 13.5$ nm. This mismatch arises because the excitation energy into the 4d5p levels ($E_{exc} \approx 92.4$ eV) is 0.4 times the ionization energy and thus a temperature which gives large ground state excitation rates will lead to large ionization rates if the electron density is sufficient.

Based on the calculation results, we can point out that for transiently heating plasma the optimum temperature moves to a higher value compared to that of the steady-state condition, and that temperature and density history as well as ionization dynamics effects must be carefully controlled for optimizing the conversion efficiency of plasma discharge EUV sources.

The authors wish to express their gratitude for the encouragement and support received from Prof. Eiki Hotta. Acknowledgment is also due to Dr. Akira Sasaki for his valuable discussions.

References
