



Theoretical investigation on impact ionization of argon gas-filled cavities

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Dedicated to Prof B N Basu

In this study, a theoretical analysis of electron beam impact ionization of argon-filled cavities has been carried out. A kinetic model has been developed to investigate the evolution of the different species of argon atoms and ions generated due to the energy deposition of the electron beam. The model includes a set of equations for neutral and charged species. Numerical analysis has been performed for the electron beam of energy 10 keV having different current densities. The typical evolution of densities of Ar, excited Ar, Ar⁺, excited Ar⁺, and Ar²⁺ has been presented at different gas pressures. It is found that most of the electron beam energy is deposited in the generation of Ar⁺ ions. Excitation states 4s(⁴P_{3/2}) and 4s(⁴P_{1/2}) of Ar⁺ ions are populated dominantly which can give extreme ultraviolet (EUV) radiation in the range of 40-110 nm. © Anita Publications. All rights reserved.

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1 Introduction

Extreme ultraviolet (EUV) radiation, due to its shorter wavelength and higher energy compared to the visible spectrum of electromagnetic radiation, finds its use for various applications like photolithography, surface modification at the nanoscale, medical imaging and microscopy, metrology and material science, and scientific research to name a few [1-3]. Dedicated research and development activities have been carried out in the area of EUV radiation generation [4]. These radiations were generated using sources like synchrotron radiation facilities and laser-produced plasmas (LPPs) [5-7]. High-cost investment, ownership, and high maintenance are required for a synchrotron radiation facility. On the other hand, the operation of LPPs require powerful and bulky lasers and also have the problem of the formation of debris inside the chamber. The debris formation can damage the optical arrangement inside the operating chamber [8]. Generation of EUV radiation through a gas discharge is another alternative scheme. Some of the methods are capillary discharge and gas puff Z-pinch [9-11]. These sources, however, have several limitations, like, short lifetime due to erosion of electrode systems by high current generation (~ 10 kA), gas heating due to pinching effect, and large cooling facility for operating in the kHz range [12].

Pseudospark discharge (PD) based EUV sources have received much attention for the generation of EUV radiation due to their unique characteristics [13]. PD is well known for the generation of high current density (>108 A/m²) along with high brightness (up to 10¹² Am⁻² rad⁻²) short pulse energetic electron beam [14]. The fast current rise (up to 10¹² A/s) and high power density (~109 W/cm²) of such beams make them suitable for various potential growing applications, such as microwave and THz radiation generation, soft and intense X-ray sources, electron beam lithography, surface modification, and EUV generation [15-18].

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The electron beam interacts with the neutral gas atoms, depositing its energy into the gas medium through elastic and inelastic collisions. Excitations and de-excitations during collisions result in the generation of different radiation that is mainly dependent on the electron beam characteristics and medium in which the electron beam propagates. In this study, in order to optimize the EUV component of radiation, a theoretical analysis of electron beam impact ionization of argon-filled cavities has been carried out.

Several electron beam deposition models have been developed earlier to calculate the population of different states resulting from the excitation of the gas atoms by the degradation of electron beam energy. Peterson and Allen employed the model for the electron beam energy deposition in argon gas based on the continuous slowing down approximation (CSDA) [19]. Bretagne *et al* used the same approximation to calculate the beam deposition efficiencies for the excitation of various states of argon (Ar) [20]. Petrov *et al* used the Boltzmann equation to solve this problem [21]. Mcgarahh *et al* also employed the CSDA method to calculate the excitation and ionization rates which were further used in the kinetic model to determine the intensities of the lasing lines of argon ions [22]. A non-equilibrium collisional model was developed to compute the population of Ar excited states and line intensities in Ar/N₂ plasmas [23]. Most of these previously developed kinetic models of electron beam energy deposition are aimed at investigating the emission of VUV/Visible radiations or for the application of material processing. The present kinetic model is developed to investigate the evolution of the Ar, Ar⁺, and Ar²⁺ species in the ground state and different excited states as their corresponding transitions range from 40-110 nm [24]. This will enable us to calculate the density of the generated EUV photons in the range of 40-110 nm.

The study involves the calculation of time-dependent rate equations for various species viz., neutralized atoms, excited atoms, ions, and excited ions in a particular energy state. The formulation includes electron impact ionization, electron impact excitation/de-excitation, radiative decay, radiative recombination, and three-body recombination processes. The effect of the pressure variation and the electron beam current density on the different species' production rates has also been investigated.

2 Model Description

The model includes the coupled set of rate equations for different species in the form of

$$\frac{dn_i}{dt} = S_i - L_i \quad (1)$$

Here n_i denotes the density of the i^{th} species which is evolved in time due to the corresponding source and loss creation mechanisms S_i and L_i , respectively [22,25]. The model follows the main species which are ground state argon atom Ar, excited argon atom Ar*, singly ionized argon Ar⁺, excited argon-ion Ar^{+*}, and doubly ionized argon Ar²⁺ that are produced due to the electron impact collision. Higher ionized species are not included in the model due to low probability of these ionization states.

The processes followed in the model for the population of Ar, Ar⁺, Ar²⁺ are shown below:

Electron impact excitation	$e + \text{Ar} \rightarrow e + \text{Ar}^*$,
Electron impact ionization	$e + \text{Ar} \rightarrow 2e + \text{Ar}^+$,
Electron impact excitation of Ar ⁺	$e + \text{Ar}^+ \rightarrow e + \text{Ar}^{+*}$
Electron impact ionization of Ar ⁺	$e + \text{Ar}^+ \rightarrow 2e + \text{Ar}^{2+}$,
Spontaneous emission	$\text{Ar}^* \rightarrow \text{Ar} + h\nu, \text{Ar}^{+*} \rightarrow \text{Ar}^+ + h\nu,$
Radiative recombination	$e + \text{Ar}^+ \rightarrow \text{Ar} + h\nu, e + \text{Ar}^{+*} \rightarrow \text{Ar} + h\nu,$
Three body recombination	$e + \text{Ar} + \text{Ar}^+ \rightarrow \text{Ar} + \text{Ar}, \text{ and } e + \text{Ar} + \text{Ar}^{+*} \rightarrow \text{Ar} + \text{Ar}$

The production rate of the electron beam and generated secondary particles are calculated using the continuous slowing down approximation (CSDA) formalism. This approach comprises an integral equation

for the total number of k^{th} excited states produced in the course of complete degradation of electron energy. The number of excitations $N_{ik}(E)$ are:

$$N_{ik}(E) = N_{ik}^0(E) + \sum \int_{E_{ik}}^{(E-I_{ij})/2} N_{ik}(X)n_{ij}(X)dX \quad (2)$$

$$\text{where } N_{ik}^0(E) = \int_{E_{ik}}^E \frac{\sigma_{ik}(X)dX}{L_i(X)} \quad (3)$$

$$\text{and } N_{ij}(E, E_s) = \int_{2E_s + I_{ij}}^E \frac{S_{ij}(X, E_s)dX}{L_i(X)}. \quad (4)$$

$N_{ik}^0(E)$ is the number of excitations directly due to the primary electrons of the electron beam as their energy reduces from E to E_k . Here, $\sigma_{ik}(E)$, $S_{ij}(E, E_s)$, and $L_i(E)$ are respectively, the excitation cross-section of the k^{th} state, differential ionization cross-section of the j^{th} shell and total loss function which is calculated using the formula given in [20,26]. Here, $n_{ij}(E, E_s)$ is the number of secondary electrons generated due to the ionization of primary electrons having a kinetic energy in the range of E_s and $E_s + dE_s$.

The calculated number of excitations during the energy degradation from E to E_{ik} is further used to calculate the deposition efficiency which represents the fraction of energy going into the production of k^{th} species

$$\eta_{ik}(E) = \frac{N_{ik}(E)E_{ik}}{E} \quad (5)$$

The production rates $K_{ik}(E)$ of the different excited and ionized species produced during the propagation of the electron beam in the gas-filled cavity obtained from these efficiencies are:

$$K_{ik}(E) = \frac{\eta_{ik}(E)L_i(E)J_b}{E_{ik}e} \quad (6)$$

The calculated production rates are used in the analysis of source and loss creation mechanisms S_i and L_i . The approach is straightforward as given in Ref [25]. The set of differential equations given by Eqs (1) is then solved by using the RK4 method to determine the density of charged and neutral species of argon at different instants of time.

3 Result and Discussion

In this section, we present the numerical results obtained by solving the set of Equations (1) for the species considered. In the computation, we have taken the following set of parameters for the electron beam: beam energy 10 keV, pulse duration 50 ns, and current density 10^4 A cm^{-2} . The background argon gas pressure is 10 Pa. Impact excitation by the primary and the generated secondary electrons results in the enhancement in the number density of the Ar atoms in the different exciting states as shown in Fig 1. It is clearly observed that the maximum energy is deposited in the excitation state 3d (1/2) of Ar. It is also found that the excitation of Ar in the states 3d (1/2), 4s (1/2), and 6s (3/2) is much more as compared to the other states.

One of the major channels for the energy deposition of the electron beam in the argon gas is the formation of the ions. The electron beam interacts with the atoms of the gas and ionizes them to form ions. Figure 2 shows the number density of the singly charged and the doubly charged ions produced by the electron beam due to the interaction with the argon gas of pressure 10 Pa. The density of Ar^+ at the end of the beam pulse is $7.27 \times 10^{13} \text{ cm}^{-3}$, while the density of Ar^{2+} is $4.5 \times 10^{11} \text{ cm}^{-3}$. The density of the different species clearly shows that the energy of the electron beam deposited in the ionization is more as compared to the excitation. This implies that the dominance of electron impact ionization process degrades the electron beam energy.

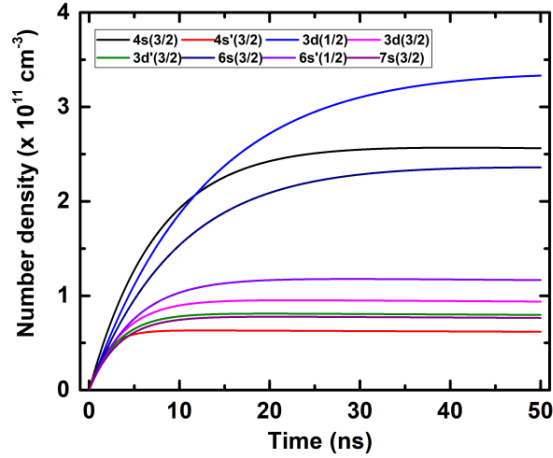


Fig 1. Number density of Argon atoms in different excited states produced by electron beam of energy 10 keV, current density 10^4 A cm^{-2} at an argon gas of pressure 10 Pa.

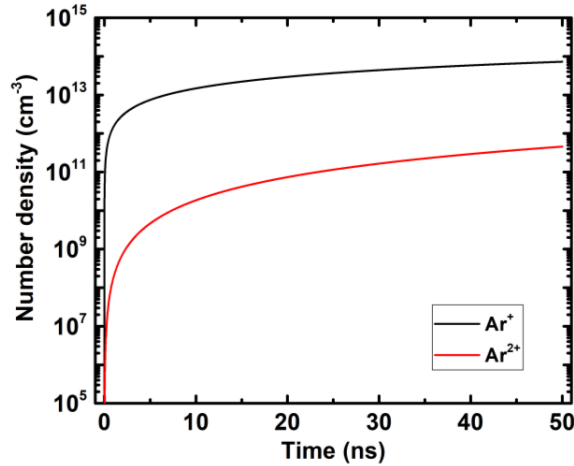


Fig 2. Number density of ions Ar^+ and Ar^{2+} produced by electron beam of energy 10 keV, current density 10^4 A cm^{-2} at an argon gas pressure of 10 Pa.

Another channel through which the electron beam degrades its energy while propagating in the argon gas medium is by exciting Ar^+ ions. The density of excited Ar^+ ions in various excited states is shown in Fig 3. The figure illustrates that the states $4s(^4\text{P}_{3/2})$ and $4s(^4\text{P}_{1/2})$ are more populated by the electron beam as compared to the states $4s(^2\text{P}_{3/2})$ and $4s(^2\text{P}_{1/2})$. For the chosen set of parameters, the density of the Ar^+ ion in the excited state $4s(^4\text{P}_{3/2})$ and $4s(^4\text{P}_{1/2})$ reaches a maximum value of $2.37 \times 10^8 \text{ cm}^{-3}$ and $2.28 \times 10^8 \text{ cm}^{-3}$, respectively.

4 Conclusion

A kinetic model is developed to investigate the evolution of the Ar, Ar^+ , and Ar^{2+} species in the ground state and different excited states as their corresponding transitions range from 40-110 nm. For the chosen set of parameters, it is found that most of the electron beam energy is deposited in the generation of Ar^+ ions. Excitation states $4s(^4\text{P}_{3/2})$ and $4s(^4\text{P}_{1/2})$ of Ar^+ ions are populated dominantly. This investigation is expected to be helpful in the modeling of electron beam-based EUV sources.

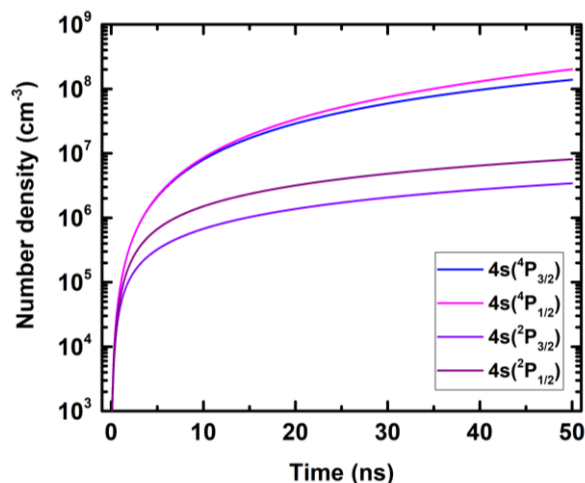


Fig 3. Number density of ion Ar^+ in the excited states produced by electron beam of energy 10 keV, current density 10^4 A cm^{-2} at an argon gas pressure of 10 Pa.

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