

PROPERTIES OF THE IONIZATION REGION OF A MAGNETRON DISCHARGE IN ARGON WITH COPPER VAPOR ANALYZED USING A GLOBAL MODEL

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Using a global model, the ionization region (IR) of a magnetron discharge in argon with copper target is studied for the conditions corresponding to carried out experiments (the neutral gas pressure is in the range 0.01...0.1 Torr and the magnetron current is between 10 and 300 mA). It is found how the densities of thermal and hot electrons (emitted secondary electrons at ion bombardment of the target), copper and argon ions, copper atoms and metastable argon atoms in the IR depend on the magnetron current and gas pressure. The current dependences for the voltage drop across the sheath near the target, the sheath size and the energies of ions with which they bombard the target are also determined for different pressures. It is found that the temperature of thermal electrons decreases with increasing the magnetron current and pressure, in agreement with experimental data on the temperature of energy level population (so called excitation temperature) of argon atoms determined by Boltzmann plot technique on the basis of argon spectral lines.

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INTRODUCTION

Magnetron sputtering has been widely applied for the deposition of thin films [1, 2] and the production of nanoparticles [3]. The technology is effectively used in many industrial applications [1]. Therefore, magnetron plasmas have been intensively studied in experiments and theoretically [1-3]. In particular, to analyse properties of magnetron sputtering discharges, analytical, hybrid Monte Carlo (MC)-fluid, MC and particle-in-cell-Monte Carlo collision models have been developed by different scientific groups [2]. For the analysis of the ionization region (IR) near the target, global models have been also used [4-8].

However, most of the global models were developed for magnetron discharges operating in the low-pressure regime [5-8] (pressure P is less than 1 Pa). For this regime, the mean free paths of ions λ_i are usually larger than the IR dimensions and the size of the sheath (s) near the target. Global models for the IR at $P > 20$ Pa were developed only in a few works [4, 9]. In [9], the study of properties of the IR at $20 \text{ Pa} < P < 210 \text{ Pa}$ was carried out assuming that $\lambda_i < s$.

Meanwhile, to the best of our knowledge, magnetron sputtering discharges have not yet been analyzed for the intermediate pressure regime ($1 \text{ Pa} < P < 20 \text{ Pa}$) using global models. In this pressure regime, the both cases, $\lambda_i < s$ and $\lambda_i > s$, may take place (it depends on external conditions). Therefore, for these pressures, the sheath properties should be analyzed taking into account that the ratio of λ_i to s is unknown.

In this paper, the IR of a magnetron plasma in argon gas with copper target is studied for the pressure range 1.33 Pa (0.01 Torr) $< P < 13.33$ Pa (0.1 Torr), using the global model developed in [9]. The model in [9] is slightly modified to take into account that the sheath near the target may be collisionless. Using this model,

the IR properties (the densities of ions and electrons, copper atoms and metastable argon atoms and the electron temperature) and the sheath properties (the sheath size and the ion energies with which they bombard the target) are analyzed under conditions corresponding to the experiments in a magnetron argon plasma with copper vapor.

1. EXPERIMENTAL STUDY AND SIMULATION

1.1. EXPERIMENTAL TECHNIQUE

Experimental studies were carried out on a magnetron sputtering system based on a planar cathode of 72 mm diameter. A copper tube as an anode was located at a distance of 15 mm from the cathode. As a source of material for sputtering, a metal target made of copper was placed on the surface of the cathode. The detailed description of the magnetron discharge device can be found in [10].

In the experiment, the parameters of the magnetron discharge in argon (the voltage drop between the anode and cathode and the temperatures of energy level population of atoms) were analyzed at neutral gas pressure in the range of 0.01...0.1 Torr and the magnetron current from 10 to 300 mA. The emission spectra of copper-argon plasma were studied by the Solar LS SDH-IV spectrometer. The temperatures of energy level population of argon atoms were obtained by Boltzmann plot technique on the base of spectral lines of this element.

1.2. SIMULATION MODEL

The global model assumes that the ionization region near the target can be described as a homogeneous half torus of mean radius R_m and of toroidal radius R_{tor} confined above the cathode racetrack [4, 8]. The IR

plasma volume is $V_{IR} = \pi^2 R_m R_{tor}^2$, the area of its surface near the target is $S_{RT} = \pi[(R_m + R_{tor})^2 - (R_m - R_{tor})^2]$, while the area of half of torus surface is $S_{tor} = 2\pi^2 R_m R_{tor}$. Here, it is assumed that $R_m = 2.5$ cm and $R_{tor} = 0.75$ cm. The dimensions of the IR correspond to the 7.2 cm cathode used in [10].

The dominant neutral species in the magnetron discharge are argon atoms in the ground state (Ar_0) at density $n_{Ar} \approx P/(k_B T_g)$, where k_B is the Boltzmann constant and T_g ($= 400$ K) is the neutral gas temperature. It is assumed that the discharge contains also copper atoms with density n_{Cu} which appear in the IR due to target sputtering by ion fluxes. We assume that there are thermal electrons (e^t) with density n_e and temperature T_e (in eV), singly ionized argon (Ar^+) and copper (Cu^+) ions with densities n_{Ar+} and n_{Cu+} , respectively, and hot electrons (e^H) (emitted secondary electrons at ion bombardment of the target) with density n_{eh} and temperature T_{eh} (in eV). The magnetron discharge also contains metastable argon atoms (Ar_m) with density n_m , argon atoms in the resonance 4s states (3P_1 and 1P_1) (Ar_r) with density n_r , as well as argon atoms in 4p states (Ar_{4p}) with density n_{4p} .

The energy distributions for ions and electrons are assumed to be Maxwellian, and the temperatures of all ions and neutral species are assumed to be equal to T_g . We assume that the temperature of hot electrons T_{eh} is $(2/3)(U_{sh} + U_{IR}/2)$, where U_{sh} and U_{IR} are the potential drops across the sheath near the target and the ionization region, respectively. $U_{sh} + U_{IR} = U_D$ where U_D is the voltage applied to the target.

It is assumed that the IR plasma is quasineutral, i.e.

$$n_{Ar+} + n_{Cu+} = n_e + n_{eh}. \quad (1)$$

The fluxes of argon and copper ions to the target are calculated as $\Gamma_{Ar+} = h_{LAr} u_{BAr} n_{Ar+}$ and $\Gamma_{Cu+} = h_{LCu} u_{BCu} n_{Cu+}$, where h_{LAr} , h_{LCu} , u_{BAr} and u_{BCu} are the ratios of the ion density at the sheath edge to that in the IR, and the ion (Bohm) velocities at the sheath edge with the IR plasma for Ar^+ and Cu^+ , respectively. The expressions for h_{LAr} , h_{LCu} , and u_{BAr} and u_{BCu} are presented in [9].

The fluxes of ions are linked to the magnetron current I_D via the relation

$$I_D/(e S_{RT}) = (1 + \gamma_{Ar+})\Gamma_{Ar+} + \Gamma_{Cu+}, \quad (2)$$

where e is the absolute value of electron charge, $\gamma_{Ar+} = 0.112$ is the secondary electron emission (SEE) coefficient for collisions of argon ions with the copper target. Here, it is assumed that the SEE coefficient for collisions of copper ions with the target is zero [7].

Using Eqs. (1) and (2) and the expressions for Γ_{Ar+} and Γ_{Cu+} , one gets the following expression for the electron density

$$n_e = I_n + (1 - k_n) n_{Ar+} - n_{eh}, \quad (3)$$

where $I_n = I_D/(e S_{RT} h_{LCu} u_{BCu})$ and $k_n = (1 + \gamma_{Ar+}) h_{LAr} u_{BAr} / (h_{LCu} u_{BCu})$.

The density of α ions (α is Ar^+ or Cu^+) is described by the following balance equation:

$$\frac{\partial}{\partial t} n_\alpha = \sum_i R_i^\alpha - \frac{\Gamma_\alpha S_{RT}}{V_{IR} f_t}, \quad (4)$$

where R_i^α is the collisional rate for the i -th reaction accompanied by generation or loss of α ions. Here, it is assumed that the fraction of ions leaving the IR plasma towards the target, f_t , is the same for argon and copper ions. Detail information on the processes describing the generation and loss of ions in different collisions and on the corresponding rate coefficients is presented in [9].

For the steady-state case ($\partial/\partial t = 0$) considered here, it follows from Eq. (4) that

$$f_t = \frac{(\Gamma_{Ar+} + \Gamma_{Cu+}) S_{RT}}{(\sum_i R_i^{Ar+} + \sum_i R_i^{Cu+}) V_{IR}}. \quad (5)$$

To calculate the ion densities, one also needs to know the density of Cu atoms which can be found from the following equation

$$\begin{aligned} \frac{\partial}{\partial t} n_{Cu} = & \frac{(\Gamma_{Ar+} Y_{Ari} + \Gamma_{Cu+} Y_{Cui}) S_{RT}}{V_{IR}} - K^{Ar+Cu} n_{Ar+} n_{Cu} \\ & - K^{eCu} n_e n_{Cu} - K^{ehCu} n_{eh} n_{Cu} - \\ & K^{mCu} (n_m + n_r + n_{4p}) n_{Cu} - \frac{n_{Cu} D_{Cu}}{R_{tor}^2}, \end{aligned} \quad (6)$$

where Y_{Ari} and Y_{Cui} are the sputtering yields describing generation of Cu atoms in collisions of Ar^+ and Cu^+ ions with the target, respectively. The yields as functions of the ion energies E_α with which they bombard the target are taken from [11]. K^{Ar+Cu} , K^{eCu} , K^{ehCu} , and K^{mCu} are the rate constants for the reactions $Ar^+ + Cu \rightarrow Ar_0 + Cu^+$, $e^t + Cu \rightarrow Cu^+ + 2e^t$, $e^H + Cu \rightarrow Cu^+ + e^t + e^H$, and $Ar^* + Cu \rightarrow Ar_0 + Cu^+ + e^t$, respectively, where Ar^* is Ar_m , Ar_r or Ar_{4p} . The rate constant values and their dependencies on T_e and T_{eh} are presented in [9].

The density of hot electrons is described by the following expression [9]

$$\partial n_{eh}/\partial t = \gamma_{Ar+} \Gamma_{Ar+} S_{RT}/V_{IR} - \nu_h n_{eh}, \quad (7)$$

where $\nu_h = 1/\tau_{diff} + Q_h/\varepsilon_h$ is the loss frequency of hot electrons; $\varepsilon_h = U_{sh} + U_{IR}/2$ is the average energy of hot electrons; τ_{diff} is the time for hot electrons to exit from the ionization region due to diffusion and Q_h is the power loss by hot electrons in different collisions (the expression for Q_h is presented in the supplementary material for [9]). $\tau_{diff} = R_{tor}^2/(2D_h)$, where $D_h = e T_{eh}/[m_e(\nu_{eah} + \nu_{iah})] \times [1 + \omega_c^2/(\nu_{eah} + \nu_{iah})^2]^{-1}$ [12]; m_e is the electron mass; ν_{eah} is the frequency for collisions of hot electrons with atoms; ν_{iah} is the ionization frequency of discharge atoms by hot electrons and ω_c is the electron gyro-frequency. The studies are carried out for the magnetic field strength of 0.03 T.

The ion energy E_α depends on the ratio of the sheath size s to the value of the ion mean-free path λ_α . If $\lambda_\alpha \geq s$, we assume that $E_\alpha = U_{sh} = U_D(1 - f_{IR})$, where $f_{IR} = U_{IR}/U_D$. For the case, when ion-neutral collisions in the sheath are intensive ($\lambda_\alpha < s$), the energy with which Ar^+ and Cu^+ ions bombard the target can be found using equation (6.5.2) in [13]

$$E_\alpha = e \lambda_\alpha E/\pi, \quad (8)$$

where $E = \left[\frac{3e(\Gamma_{Ar+} + \Gamma_{Cu+})}{2\varepsilon_0\sqrt{2e\lambda_{iav}/\pi m_{iav}}} \right]^{2/3} s^{2/3}$ is the electric field strength in the collisional sheath near the target [13], ε_0 is the vacuum permittivity; $\lambda_{iav} = (\lambda_{Ar+} n_{Ar+} + \lambda_{Cu+} n_{Cu+}) / (n_{Ar+} + n_{Cu+})$ is the average ion mean free path; $m_{iav} = (m_{Ar} n_{Ar+} + m_{Cu} n_{Cu+}) / (n_{Ar+} + n_{Cu+})$ is the average ion mass, m_{Ar} and m_{Cu} are the masses of argon and copper atoms, respectively.

In the case when $\lambda_{iav} \geq s$, the sheath size is found using Eq. (6.3.14) in [13]

$$s = \frac{\sqrt{2}}{3} \lambda_{Des} \left(\frac{2U_{sh}}{T_e} \right)^{3/4}, \quad (9)$$

where λ_{Des} is the Debye length at the boundary of the sheath near the target with the IR plasma.

For $\lambda_{iav} < s$, the sheath size is calculated using equation (6.5.7) in [13]

$$s = \left(\frac{2U_{sh}^{3/2}}{3e(\Gamma_{Ar+} + \Gamma_{Cu+})} \left(\frac{5}{3} \right)^{\frac{3}{2}} \varepsilon_0 \left(\frac{2e\lambda_{iav}}{\pi m_{iav}} \right)^{\frac{1}{2}} \right)^{2/5}. \quad (10)$$

The model also includes the balance equations for argon atoms in the metastable, resonance 4s states and 4p states and the power balance equation for thermal electrons. The detail information about these equations and the model used here can be found in [9].

A steady-state ($\partial/\partial t = 0$) solution of the time-dependent balance equations of the IR model was found by using the DVODE package [14]. In all considered cases, the calculations were performed for the case $f_t = 0.9$, as in [9].

2. RESULTS OF NUMERICAL CALCULATIONS

The model presented in previous section was used to analyze properties of the IR and the sheath near the target of a magnetron discharge for the conditions of experiments in [10]. The model equations were solved for P , I_D , and U_D shown in Table. In Table, there are also the temperatures of energy level population of argon atoms which were determined in [10] by Boltzmann plot technique on the basis of argon spectral lines. We assume that the temperature of thermal electrons is proportional to the temperature of energy level population of argon atoms.

In Fig. 1,a, the densities of electrons and ions and the copper atom density as functions of I_D are presented for $P = 0.05$ Torr. Fig. 1,a shows that the densities of ions, electrons and copper atoms increase when the magnetron current becomes larger because of increasing the input power $I_D U_D$ (see Table) and due to intensification of the sputtering process (see Eq. (6)).

For the conditions considered here, the density of Ar^+ ions is larger than the Cu^+ density because the magnetron currents are not very high and, as a result, $n_{Ar} \gg n_{Cu}$ (see Fig. 1,a). The densities of argon atoms are $2.414 \cdot 10^{14}$, $1.207 \cdot 10^{15}$, and $2.414 \cdot 10^{15} \text{ cm}^{-3}$ for $P = 0.01$; 0.05 and 0.1 Torr, respectively.

With increasing pressure, the temperature of thermal electrons decreases (see Fig.1,b), in agreement with the experimental results presented in Table. T_e also

decreases with increasing the magnetron current. In our opinion, the decrease of T_e is mainly because the density of Cu atoms becomes larger (the threshold energy for ionization of Cu atoms is smaller than that for ionization of Ar atoms) and due to increasing the density of hot electrons (see Fig. 1,a).

Parameters of magnetron discharge measured in [10]

P , Torr	I_D , mA	U_D , V	T , K
0.01	10	255	19600
0.01	50	280	16750
0.01	100	300	14500
0.01	150	310	13600
0.01	300	320	11800
0.05	10	230	17200
0.05	50	260	14150
0.05	100	280	13300
0.05	150	290	13000
0.05	300	320	11700
0.1	10	220	15200
0.1	50	245	13400
0.1	100	275	13000
0.1	150	290	11900
0.1	300	320	10700

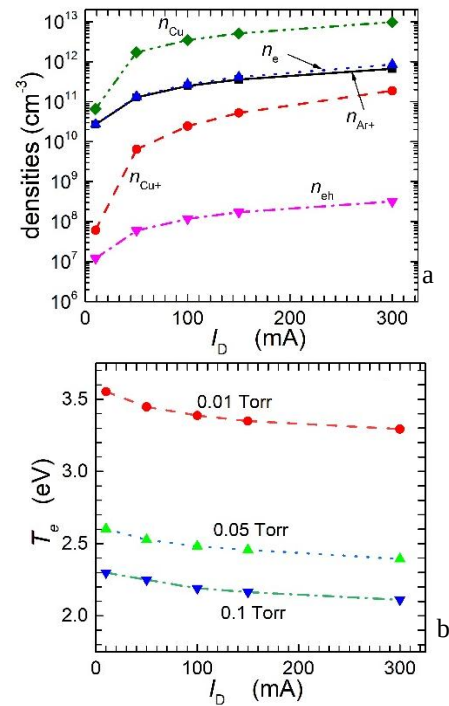


Fig. 1. The calculated electron and ion densities and Cu density for $P = 0.05$ Torr (a) and the temperature of thermal electrons (b) as functions of the magnetron current for different pressures

However, the calculated variation of the electron temperature with I_D is smaller than the variation of the temperature of energy level population of argon atoms (see Table). In our opinion, this difference is mainly due to simplifications used in this model which are discussed in next section.

The densities of most plasma species (thermal electrons, Ar^+ and Cu^+ ions (for $I_D \geq 100$ mA), metastable argon atoms and Cu atoms (for $I_D \geq 100$ mA)) become larger with increasing pressure (Figs. 2 and

Figs. 3,a) for most of the currents considered here because of increasing the Ar_0 density. Note that for the conditions considered in [9] ($P > 20$ Pa), the Cu and Cu^+ densities decreased at increasing P . The decrease observed in [9] was mainly due to essential decreasing the ion energies with which they bombard the target [9].

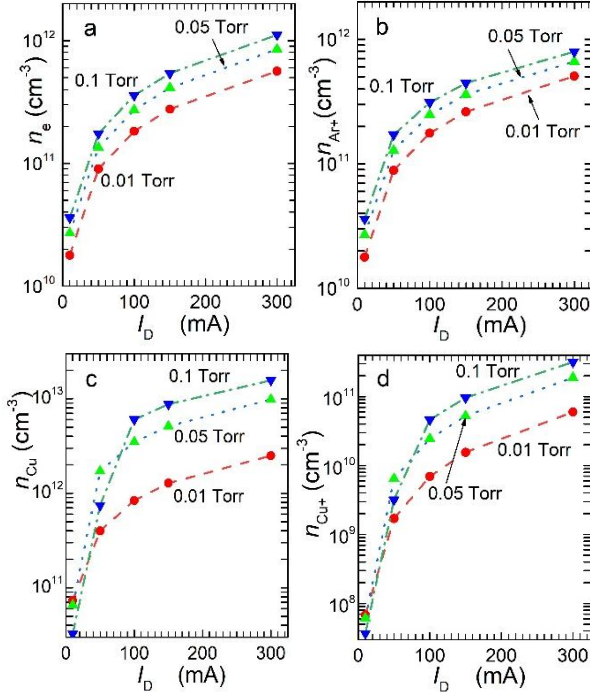


Fig. 2. The calculated densities of thermal electrons (a), argon ions (b), copper atoms (c) and copper ions (d) as functions of the magnetron current for different pressures

The density of hot electrons decreases with growth of P (see Fig. 3,b) due to increasing the Cu and Ar_0 densities. Because of the increase of the densities of neutral species, the loss of hot electrons in collisions with argon and copper atoms becomes more intensive.

The voltage drop in the sheath becomes larger when the magnetron current increases (Fig. 4,a), in agreement with [9]. $U_{sh} = U_D(1 - f_{IR})$ decreases with increasing P (see Fig. 4,a) because of an increase of the parameter f_{IR} (see Fig. 4,b), which determines the voltage drop in the IR. Here, where $f_{IR} = U_{IR}/U_D$ is rather small (≤ 0.26), in agreement with the results in [6] and [7].

The sheath size decreases with increasing I_D (Fig. 5,a). At large pressures ($\lambda_{iav} < s$), the decrease is because the total ion flux density to the target, $\Gamma_{Ar^+} + \Gamma_{Cu^+}$, becomes larger (see Eq. (10)). In the case $\lambda_a \geq s$, the sheath size decrease may be explained by increasing the Debye length λ_{Des} (see Eq. (9)).

Note that for the same pressure (at $P = 0.05$ and 0.1 Torr), the sheath size can be smaller than the average ion mean free path if I_D is rather large and can be larger than s for small magnetron currents (see Fig. 5,a). Thus, the transition from the collision sheath regime to the collisionless one is possible at variation of I_D .

When I_D becomes larger, the energies of ions with which Ar^+ and Cu^+ ions bombard the target increase (see Fig. 5,b).

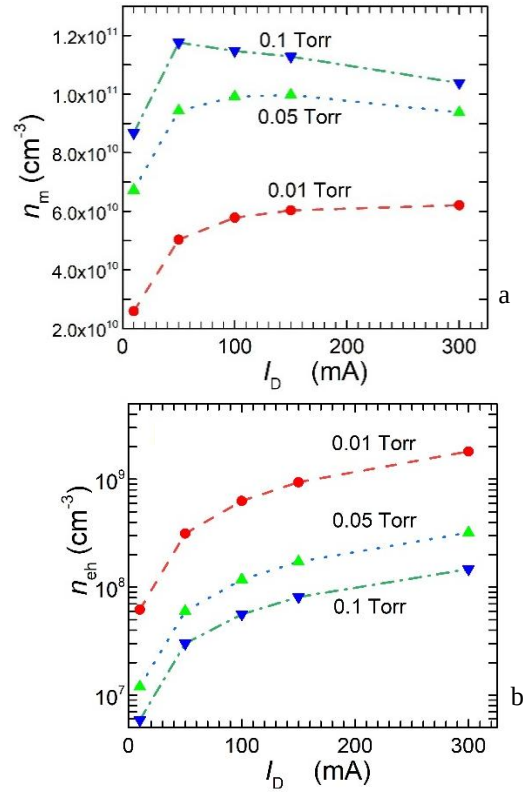


Fig. 3. The calculated n_m (a) and n_{eh} (b) as functions of the magnetron current for different pressures

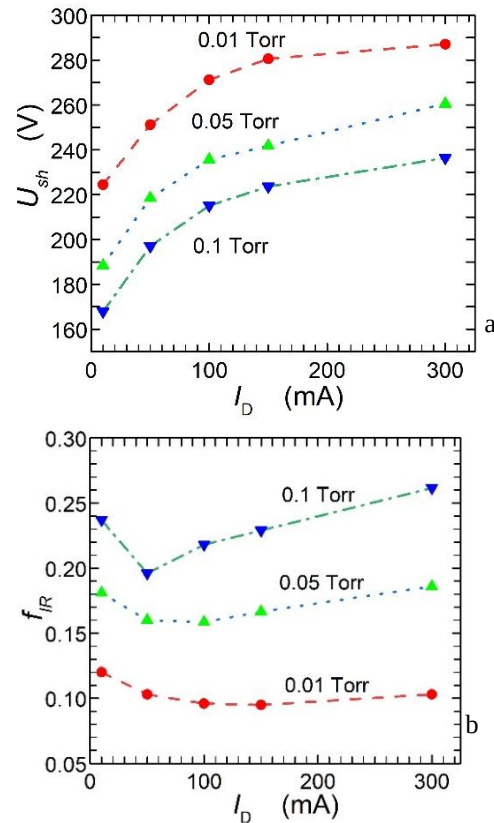


Fig. 4. The calculated U_{sh} (a) and $f_{IR} = U_{IR}/U_D$ (b) as functions of the magnetron current for different pressures

For $\lambda_{iav} < s$, this is due to increasing the total ion flux density to the target (see Eq. (8)). For $\lambda_{iav} > s$, the increase is due to the increase of the voltage drop in the sheath (see Fig. 4,a). E_{Ar+} and E_{Cu+} become larger with decreasing P . For small pressures ($\lambda_{iav} > s$), this is due to increasing U_{sh} (see Fig. 4,a). For $\lambda_{iav} < s$, the increase of E_{α} is mainly due to increasing the ion mean free paths (see Eq. (8)).

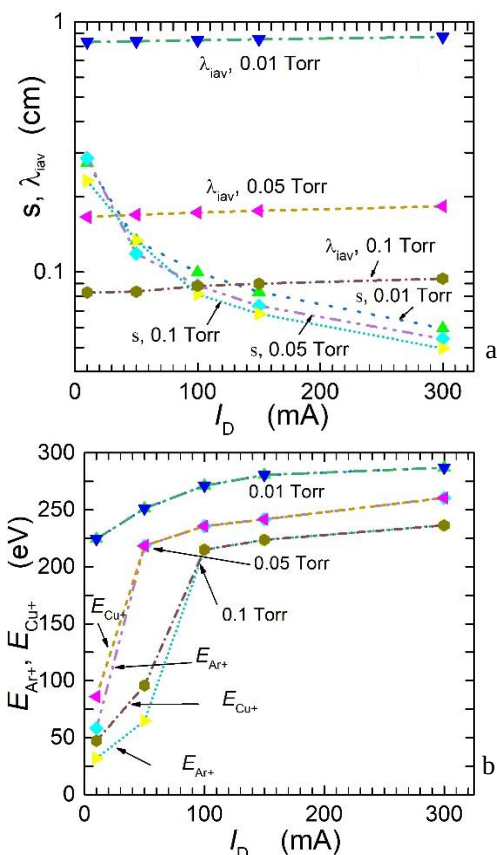


Fig. 5. λ_{iav} (a) and E_{Ar+} , E_{Cu+} (b) as functions of the magnetron current for different pressures

SUMMARY

Thus, the ionization region of a magnetron discharge with copper target has been investigated for the pressure range $0.01 \text{ Torr} \leq P \leq 0.1 \text{ Torr}$, using the global model developed in [9]. For different pressures, it has been analyzed how properties of the IR and the sheath near the target depend on the magnetron current. The studies have been carried out for the conditions corresponding to the experiment in [10]. It has been found that the densities of thermal and hot electrons, $Ar+$ and $Cu+$ ions and Cu atoms increase with increasing I_D (see Fig. 2). The densities of most discharge species also increase with increasing pressure because of a growth of the argon density.

However, the density of hot electrons decreases when pressure becomes larger (see Fig. 3,b) because of increasing the loss of hot electrons in collisions. At small I_D , the densities of Cu and $Cu+$ may become also smaller at larger pressure (see Fig. 2,c and d) because of decreasing the energies of ions with which they bombard the target (see Fig. 5,b) (the sputtering becomes less intensive). For $P \geq 0.05 \text{ Torr}$, the density

of metastable argon atoms increases with increasing I_D at small currents and decreases at large ones (see Fig. 3,a). In our opinion, the increase is due to increasing n_e , while the decrease is due to decreasing T_e and an enlargement of the density of copper atoms (increasing the loss of metastable atoms in collisions with Cu atoms).

The voltage drop in the sheath becomes larger with an increase of the magnetron current, while the ratio of the voltage drop in the IR to the voltage applied to the target decreases with a decrease of pressure (see Fig. 4,b) (in our opinion, the decrease is due to a decrease of the energy loss of thermal electrons in collisions).

The sheath size becomes smaller if I_D increases. As a result, s may become smaller than the average ion mean free path for large I_D and larger than λ_{iav} for small I_D (see Fig. 5,a).

The energies with which ions bombard the target increase with increasing I_D (see Fig. 5,b) because of increasing U_{sh} for the case $\lambda_{iav} > s$ (see Fig. 4,a) and due to the increase of the electric field strength in the sheath if $\lambda_{iav} < s$ (see Eq. (8)).

The global model shows that the temperature of thermal electrons becomes smaller with increasing I_D (see Fig. 1,b) because of increasing the densities of copper atoms and hot electrons. The temperature also decreases when P becomes larger. The results on T_e are in good agreement with the obtained experimental results on the temperature T of energy level population of argon atoms. However, the decrease of T_e with increasing I_D , which gives the model, is essentially smaller than the decrease of T in the experiment (see Fig. 1,b and Table). In our opinion, the differences between the model results and the experimental ones are mainly due to model simplifications. In particular, the model does not account that the electron energy distribution function (EEDF) in the experiment may differ essentially from the Maxwellian EEDF assumed in this model [13, 15]. It also does not account for formation of hot and warm argon atoms after interaction of argon ions with the target [6]. Moreover, the description of ion diffusion at $P \leq 0.01 \text{ Torr}$ should be improved because for these pressures, the ion mean free path is larger than the IR width ($\sim R_{tor}$) (see Fig. 5,a). Therefore, the model here can be used only for qualitative analysis of the IR and requires an essential improvement to describe properties of the IR plasma, especially at low pressures ($\leq 0.01 \text{ Torr}$). The model and results presented here can be useful for verifying experiments and numerical simulations, particularly concerning magnetron sputtering systems widely used in modern technologies.

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ВЛАСТИВОСТІ ІОНІЗАЦІЙНОЇ ОБЛАСТІ МАГНЕТРОННОГО РОЗРЯДУ В АРГОНІ З ПАРАМИ МІДІ, ПРОАНАЛІЗОВАНІ ЗА ДОПОМОГОЮ ГЛОБАЛЬНОЇ МОДЕЛІ

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Використовуючи глобальну модель, досліджено іонізаційну область (ІО) магнетронного розряду в аргоні з мідною мішенню за умов, що відповідають проведеним експериментам (тиск нейтрального газу в діапазоні 0.01...0.1 Торр, а струм магнетрона в межах 10 та 300 мА). Встановлено як густини теплових та гарячих електронів (вторинних електронів, що генеруються за іонного бомбардування мішені), іонів міді та аргону, атомів міді та метастабільних атомів аргону в ІО залежать від струму магнетрона та тиску газу. Для різних тисків також визначено залежності від струму для падіння напруги у шарі просторового заряду біля мішені, розміру шару просторового заряду та для енергій іонів, з якими вони бомбардують мішень. Встановлено, що температура теплових електронів зменшується зі збільшенням магнетронного струму та тиску, що узгоджується з експериментальними даними про температуру заселення енергетичних рівнів атомів аргону, визначеними методом діаграм Больцмана із залученням спектральних ліній аргону.