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Ionic composition of the planar magnetron plasma in gas and vacuum operation mode

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The results of experimental study and numerical simulation of the fractional content of argon and copper ions in plasma generated in the direct current planar magnetron with the copper target in gas mode (at the argon pressure of 0.1 Pa) and vacuum mode (at the residual gas pressure of 0.04 Pa) are provided. It is demonstrated that the copper ions fractions in gas and vacuum modes at the discharge current sufficient to maintain the self-sputtering mode (10 A) are quite similar and amount to 97 % and 100 % respectively. The results of experiments and numerical evaluations evidence the possibility to perform stable operation of continuous discharge and obtain the metal ions flow in high vacuum of the planar magnetron without effects of thermal evaporation or sublimation of the copper target.

Keywords: planar magnetron; self-sputtering mode; vacuum operation mode.

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Introduction

Today magnetron sputtering is the most common method of physical deposition of protective and functional coatings. Meanwhile the free length of sputtered atoms is compatible or even lower than the relevant distances from the sputtered magnetron target to the substratum under standard values of the operating pressure during magnetron sputtering (0.5–10 Pa) [1, 2].

Collisions with the operating gas particles in the space between the target and the substratum result in the sputtered atoms power dissipation and reduction, which affect the created coating quality and properties [3–5]. With the increasing pressure of the operating gas, the created coating becomes porous and has the prevailing column-type structure [6, 7].

Elimination of the above-mentioned deficiencies is possible in so-called self-sputtering mode, in which the discharge is maintained by virtue of ionization of the sputtered material of the target, but not the

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one of the operating gas [8, 9]. Under sufficient discharge energy and therefore the amount of the sputtered metal atoms in the ionization area, such discharge is able to transfer to a continuous form and operate without the operating gas supply at the operating pressure determined by the vacuum exhaust system only [10–12].

Regardless of the fact that this type of discharge has been previously implemented in the experiment, today there is a lack of works related to simulation of the processes taking place in it, including the mass-to-charge plasma composition.

This work provides the results of experimental study and numerical simulation of the fractional content of argon and copper ions in plasma generated in the direct current planar magnetron with the copper target, under gas mode (at the argon pressure of 0.1 Pa) and vacuum mode (at the residual gas pressure of 0.004 Pa).

Experimental procedure and method

The planar magnetron with the directly cooled copper target 100 mm in diameter was used in the experiments. The initial target thickness was 4 mm. The intensive water-cooling system (20 l/min as a minimum) ensured the possibility of the magnetron discharge operation in continuous mode, with the power output up to 8 kW. The magnetic field of 100 mT on the target surface in the intensive sputtering area was ensured by NdFeB magnets of N35 grade placed in the water jacket at the maximum operating temperature of + 80 °C. The circular electrode (for creation of the round cross section of the particles flow) with the diameter of 100 mm that had the potential of the grounded vacuum chamber walls and was located 8 cm away from the cathode target edge, was used as the magnetron anode. The discharge voltage in the vacuum mode was 590–610 V at 8–12 A magnetron discharge current. The voltage was approximately 50–60 V lower in the gas mode, with the same discharge current (probably due to more efficient sputtering of the copper target with argon ions as compared to copper ions [8]).

By virtue of a large internal volume of the vacuum chamber (about 65 liters) and water cooling of the target, the temperature of the external target surface (from the discharge side) did not exceed 200 °C during experiment with the magnetron operating at 8 kW. The surface of the heated target can have the higher gas temperature but the lower concentration, though it is true generally for all types of particles (neutrals and ions of both argon and copper), and therefore it hardly affects the trends of the curves. Thus, it was considered that the target was not the source of the operating gas heating, and the temperature thereof was assumed as the room temperature in the assessment. Pressure in the chamber was maintained constant, with the equal processes of argon supply close to the target and exhaust thereof with the pump through an aperture located in the outermost area of the vacuum chamber.

To study the mass-to-charge composition of the plasma ions, the modified quadrupole mass-spectrometer RGA-200 (Stanford Research Systems) was used, the input aperture of which was located 25 cm apart of the magnetron target. The spectrometer collector was located under the “ground” potential, like the grounded walls of the vacuum chamber. The magnetron discharge plasma was diffused into the cylindrical expander, the end of which was cut-off with the fine-structure non-magnetic stainless-steel mesh. The potential gradient for ions acceleration into the spectrometer

was ensured by an individual power source between the magnetron anode and the grounded walls of the vacuum chamber. The optimum value of the bias potential was 30–50 V. The detailed description of the discharge cell of the planar magnetron and the equipment for measurement of the mass-to-charge composition of ions used in the experiment can be found in [12]. Experimental peaks of the ionic mass spectrum were integrated for more specific determination of the fractional ratio of ionic components in plasma.

Numerical model

The discharge processes in the previously described magnetron were simulated based on the following provisions. It was assumed that the flat cathode target of the magnetron had the disk shape with the diameter D, the discharge plasma occupied the internal volume between the target and the annular anode with the height h. Opposite to the model described in [13], the above-mentioned model did not account for the electron beam injection into the discharge area, there was no additional reflecting electrode, and the discharge was initiated and maintained with the voltage U_d between cathode and anode. The operating gas in the model was argon at p_{Ar} pressure and permanent $T_{\mathrm{Ar}} = 300$ K temperature; weight of the atom thereof, ionization potential and ionization cross section within the energy range of 0 to 3.000 eV (that reaches the maximum of $\sim 4\times10^{-16}$ cm^2 at ~31 eV) [14, 15] were considered. Under the ion impact of the copper target surface, its sputtering took place which was determined by sputtering coefficient γ_{sp} depending on the ion energy [8, 16], and copper atoms entered the argon plasma, which were further ionized in it with the energized electrons, taking into account the potential and ionization cross section for copper [14], without consideration (for simplification) of any target alloying by the incoming ions of any type. The sputtering coefficient was approximated with the following formula:

$$\gamma_{sp}\left(\varepsilon_{i\mathrm{Ar}}\right) = \frac{0.06}{\varepsilon_t}\sqrt{Z_x}\left(\sqrt{\varepsilon_{i\mathrm{Ar}}} - \sqrt{\varepsilon_{th}}\right)$$

where 0.06 is the constant with the dimension of $(\mathrm{eV})^{-1/2}$ [17], $\varepsilon_{i\mathrm{Ar}}$ is the energy of argon ions accelerated in the cathode potential fall, ε_t is the target material bond energy, and ε_{th} is the threshold ion energy from which sputtering is started, $Z_x = 2Z_{\mathrm{Cu}}\Big/\left(\left(Z_{\mathrm{Ar}}/Z_{\mathrm{Cu}}\right)^{2/3} + \left(Z_{\mathrm{Cu}}/Z_{\mathrm{Ar}}\right)^{2/3}\right)$ is the mentioned atomic number determined by atomic numbers of the operating gas Z_{Ar} (argon) and the target Z_{Cu} (copper).

The fraction of copper $n_{i\mathrm{Cu}}$ and argon $n_{i\mathrm{Ar}}$ ions in the plasma was determined based on the ratio of concentrations thereof to the total concentration of ions in the plasma $n_i = n_{i\mathrm{Ar}} + n_{i\mathrm{Cu}}$. The above-mentioned values were determined as a result of the following equations solution:

$$n_{i\mathrm{Ar}} = I_d\left(n_{\mathrm{Ar}}\sigma_{i\mathrm{Ar}}\left(U_d\right)\times\left(U_d/W_{i\mathrm{Ar}}\right)h\right)\Big/ \Big/\left[0.4\left(1+\gamma_{ie}\right)e\sqrt{2kT_e/M_{\mathrm{Ar}}}S_c\right], \quad (1)$$

$$n_{i\mathrm{Cu}} = I_d\left[n_{\mathrm{Cu}}\left(\gamma_{sp}\right)\sigma_{i\mathrm{Cu}}\left(U_d\right)\times\left(U_d/W_{i\mathrm{Cu}}\right)h\right]\Big/ \Big/\left[0.4\left(1+\gamma_{ie}\right)e\sqrt{2kT_e/M_{\mathrm{Cu}}}S_c\right] \quad (2)$$

where I_d is discharge current; $n_{\mathrm{Ar}} = p_{\mathrm{Ar}}/kT_{\mathrm{Ar}}$ is the concentration of neutral argon atoms; $n_{\mathrm{Cu}}(\gamma_{sp})$ is the concentration of neutral copper atoms knocked out from the copper atoms target depending on γ_{sp} under established U_d

and I_d; $\sigma_{i\mathrm{Ar}}(U_d)$, $\sigma_{i\mathrm{Cu}}(U_d)$ is the cross section of argon and copper atoms ionization with discharge electrons; $W_{i\mathrm{Ar}} \approx 2\varphi_{i\mathrm{Ar}} \approx 30$ eV, $W_{i\mathrm{Cu}} \approx 2\varphi_{i\mathrm{Cu}} \approx 15$ eV is the potential of ion-electron pair formation for argon and copper ($\varphi_{i\mathrm{Ar}}$, $\varphi_{i\mathrm{Cu}}$ are their ionization potentials) [18]; $\gamma_{ie} = 0.1$ is the ion-electron emission coefficient (for the energies below 1 keV) [19]), M_{Ar}, M_{Cu} is the mass of argon and copper atom, $S_c = \pi D^2/4$ is the cathode target area. To simplify the calculations, it was assumed that the electrons temperature in the plasma T_e has adopted the typical value of 5 eV. The rest of the model equations are provided in details in [20]. The multipliers $(U_d/W_{i\mathrm{Ar}})h$ of (1) and (2) expressions have the meaning of the number of ionization processes per the discharge span length, without consideration of the electron multiplication avalanche in the plasma volume (due to rarity of such events and the need of more sensitive additional experiments to confirm significance of the above-mentioned process under the reviewed experimental conditions).

Equations (1) and (2) have been recorded under approximation of the absence of electron multiplication in the layer, as it is well known that there is no electron multiplication in the cathode layer of the low-pressure magnetron discharge (especially in its high current form because the layer is collision-free) [21–23].

The concentration of copper and argon ions was calculated based on the balance that ions generation in the discharge determined by the numerators of expressions (1) and (2) has been compensated by Bohm flow of ions away from the plasma, which is described by the denominator of these expressions. Applicability of the Bohm equation, that describes that the density of the ionic flow from the plasma border is proportional to the ions concentration and does not depend on their temperature and the applied voltage, is justified by the fact that the directed velocity of the magnetron discharge plasma ions is less than the Bohm velocity (that depends on electrons temperature in the plasma).

The degree of the plasma ionization was calculated based on two approximations: (1) given all plasma components – both neutrals and ions of copper and argon (α_{tot}), and (2) taking into account the metal (copper) ions and neutrals (α_m) only, according to the following expressions:

$$\alpha_{tot} = 100\ \% \times \left[\left(n_{i\mathrm{Ar}} + n_{i\mathrm{Cu}} \right) / \left(n_{\mathrm{Ar}} + n_{\mathrm{Cu}} \right) \right], \quad (3)$$

$$\alpha_m = 100\ \% \times \left[\left(n_{i\mathrm{Cu}} \right) / \left(n_{\mathrm{Cu}} \right) \right] \quad (4)$$

The degree of ionization determined based on equation (3) considers all plasma components (residual atmosphere, operating gas and sputtered target atoms), and therefore allows for characterizing the plasma to the fullest extent possible, while the direct experimental determination thereof can be complicated by the presence of multiple components in the plasma which are able to condensate on the measuring equipment elements and effect on the measurements accuracy.

Ionization degree (4) that considers the metal component of the plasma only can be generally measured directly with the use of electric [24] or magnetic [25] condensation probes and therefore comparison of the two methods of ionization degree assessments is of the large significance for the analysis of experimental measurements correctness.

The calculations results and comparison thereof with the experiment and the analysis are provided below.

Results and their discussion

Comparison of experimental and calculated fractions of argon and copper ions in the plasma is shown in Fig. 1*a*.

The calculated curves of ion fractions demonstrate satisfactory consistence with the experiment, which may be an indirect evidence of correctly calculated absolute values of the plasma ionic components concentrations and ionization thereof. According to the numerical simulation results, increase of the copper ions fraction (up to 97% in gas mode and up to 100% in vacuum mode) with the increasing discharge current is related to intensification of sputtering which is justified both by increase of the number of ions in the flow, and by the increase of the sputtering coefficient γ_{sp} of the copper target with the ions accelerated by the cathode voltage drop U_d.

Figure 1*b* shows the calculated values of ionization degree α_{tot}, α_m of the plasma in gas and vacuum modes. It shows that the ionization degree increases in line with the discharge current (from ~ 2 % at 3 A current in gas mode, up to ~ 15 % at 12 A current in vacuum mode), which is consistent with the experiment [26]. Some difference of about 1 % is observed between α_{tot} and α_m in gas mode only, when the argon ions and neutrals fraction are notable. In vacuum mode, when the fraction of these particles becomes negligible, the values of ionization degree α_{tot}, α_m are nearly the same, and can be equally used to assess the plasma ionization degree.

Figure 2 shows the calculated concentrations of argon and copper neutrals and ions in gas and vacuum modes.

From among all calculated concentrations in both modes and at the maximum values of the magnetron current, the maximum value is reached by the copper ions concentration due to intensive copper sputtering. The minimum value under the same conditions is taken by the argon ions concentration due to its lower ionization cross section and ionization potential exceeding the copper one. The relatively rapid increase of both ion types concentration with the increasing discharge current is notable in gas mode and at small currents (up to 3–4 A). With further increase of the discharge current, dependency of both ionic and neutral components of the plasma transfers to saturation in both gas and vacuum modes.

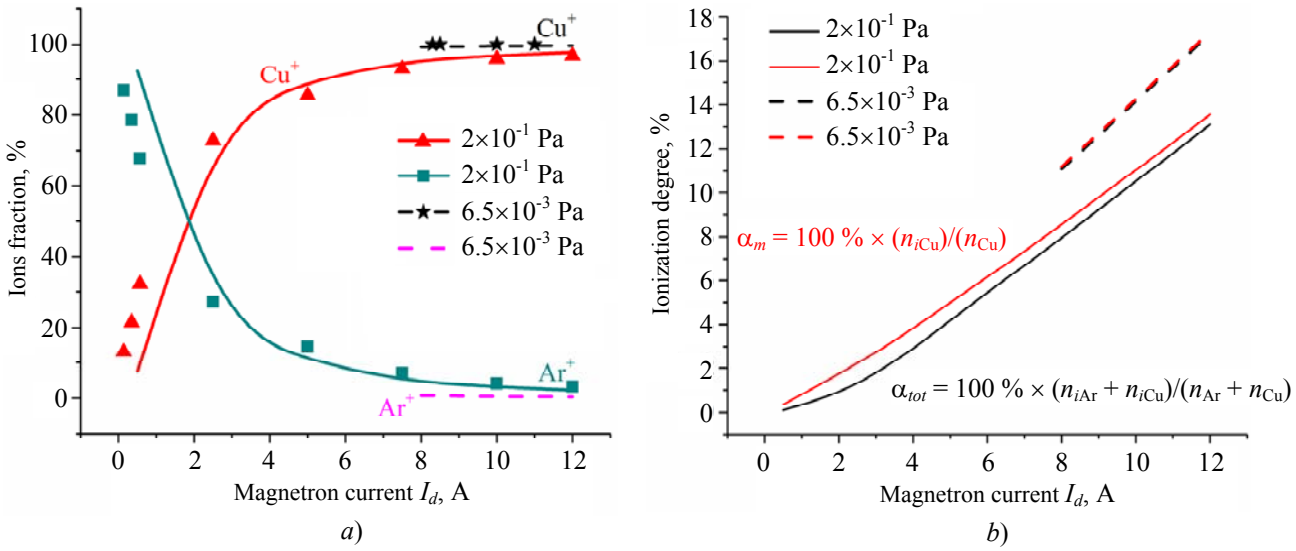

Fig. 1. a) – Calculated and experimental fractions of copper and argon ions in plasma versus the magnetron current, and b) – calculated ionization degrees α_{tot} and α_m of the plasma in gas (at argon pressure of 0.2 Pa) and vacuum (at argon pressure of 0.0065 Pa) modes

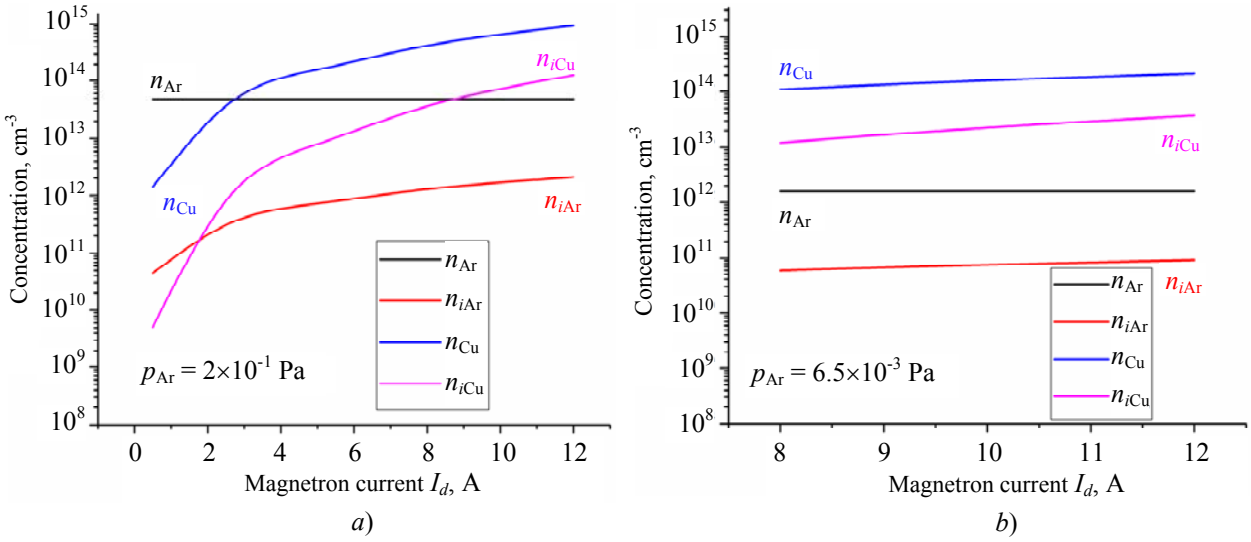

Fig. 2. Calculated concentrations of neutral and ionic components of argon and copper in gas (a) and vacuum (b) modes

Given the cathode layer is collision-free in the low-pressure (up to 0.004 Pa in this case) discharge, and its width in the intensive sputtering area on the target surface does not exceed 1 mm at the discharge current of 10 A, and that the ionic compositions of the plasma at such current value in gas and vacuum modes is nearly the same (97 and 100 %, respectively), it can be suggested that the differences of the potential distribution (due to different burning voltage in gas and vacuum modes) would not significantly affect the concentration growth mechanism.

Conclusion

The results of experimental study and numerical simulation of ionization processes in plasma generated in the direct current

planar magnetron with the copper target in gas and vacuum modes allow for explaining the mass-to-charge composition of the plasma ions in different operation modes of the magnetron discharge. The obtained assessments of the sputtered target material ionization degree have insignificant difference for gas and vacuum modes, which is explained by the prevailing metal ion component in plasma at relatively high values of the discharge current independently of its operation mode.

The numerical simulation allows for calculation of the numerical values of neutrals and ions concentrations of operating gas, target material and the degree of ionization of the discharged gas metal plasma. As it comes from the simulation results, the process of ionic spurring is sufficient to operate the magnetron discharge in the vacuum self-spurring mode without involving any auxiliary effects of thermal evaporation and sublimation of the magnetron target.

One of the main conclusions obtained in this work is that the proposed model allows for assessing the degree of the sputtered material ionization consistently with the experiment (the parameter which is one of the key parameters in sputtering the coatings, as it justifies the level of ionic effect on the electrical bias surface). The obtained results are of scientific and practical interest in terms of comparison of the vacuum mode with any other modes of magnetron sputtering (DC, HIPIMS).

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