Effect of working gas pressure on mass-to-charge composition of plasma ions in high-current planar magnetron discharge

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Abstract. The mass-to-charge ion composition of a planar magnetron discharge plasma has been investigated. The measurements used a modified quadrupole mass-spectrometer and a time-of-flight spectrometer. The experiments were carried out on a copper magnetron target. Argon was used as a working gas. The operating pressure was 0.15÷1.3 Pa. The discharge current was 1÷20 A with a pulse duration of 30÷50 µs. The influence of main operating parameters (discharge current and working gas pressure) on mass-to-charge composition of plasma ions was measured. The mass-to-charge composition of plasma ions in the axial direction was measured as a function of working pressure. Plasma electron temperature was measured and its effect on the mass-to-charge composition of magnetron plasma ions was estimated.

1. Introduction
A pulsed magnetron discharge is of primary interest due to the high degree of ionization of the target material, which leads to a more intense energy action on the substrate, resulting in improved properties and characteristics of deposited films [1, 2]. The discharge current and the operating gas pressure are the main parameters of a magnetron discharge. It is known that an increase in the discharge current leads to an increase in the ratio of the target material in the plasma. In addition, the mass-to-charge ratio of the magnetron plasma ions is changing during the pulse duration [3]. The first several microseconds from the pulse beginning the plasma consists mostly of working gas ions. The ions of the target material begin to dominate 10÷20 microseconds after pulse beginning, depending on the discharge current. With increasing amplitude of the discharge current and increasing pulse duration, the fraction of target material ions in plasma can be as high as 90 percent [4]. Despite the fact that during most of the pulse duration, the discharge can operate predominantly on the atoms of the target material (the so-called “self-sputtering mode”), in order to initiate the next discharge pulse, it is necessary to provide a certain minimal level of the working gas pressure: typically, not lower than 0.2÷0.4 Pa. Thus, even in a high-current pulsed magnetron discharge, including the self-sputtering mode, the working gas pressure remains one of the key operating parameters.

As known, the working gas pressure has an essential effect on the properties and structure of films formed during the magnetron deposition in both DC and pulsed modes [5, 6]. The main effects are related to the deposition of gas atoms on the substrate, an increase in the cross-section of the transport.
scattering, and the loss of energy of the atomized atoms in the drift space. This, in turn, leads to a deterioration in the adhesive properties of the formed film, a decrease in the deposition rate and specific adatom energy. Additionally, however, varying the working gas pressure in the magnetron discharge changes the mass-to-charge ion composition of the plasma, from which the particles are subsequently deposited on the substrate [7]. Thus, the study of the effect of the working gas pressure on the magnetron discharge parameters is of key importance for understanding the physics of the deposition processes and properties of deposited films.

The aim of this work is investigating the effects of the working gas pressure on the mass-to-charge composition of plasma ions in a planar magnetron operating in a pulsed mode with a discharge current of 1÷20 A with use of two independent diagnostic methods.

2. Experimental method and technique

We used a planar magnetron sputtering system with a target diameter of 50 mm. The target was made of copper. The target thickness was 3 mm. The working gas (argon) flow rate varied in the range 5÷20 cm³/min and was monitored by an RRG-3.6 electronic gas flow meter (Eltochpribor, Russia). The magnetic field was produced by two NdFeB magnets: a central cylindrical magnet and a ring one at the periphery. The magnetic induction on the target surface at the center of the magnetic arc was 100 mT. All electrodes of the discharge system were cooled by running water. The magnetron discharge was electrically fed from a stabilized power source with an arc suspension function and stabilization modes for current, voltage, and power.

![Diagram of the experimental setup for the time-of-flight diagnostics of plasma ions](image)

**Figure 1.** Diagram of the experimental setup for the time-of-flight diagnostics of plasma ions: 1 – magnetron target; 2 – anode; 3 – high-voltage insulator; 4 – expander; 5 – suppressor; 6 – vacuum chamber; 7 – spectrometer; 8 – gate; 9 – electron multiplier; 10 – central plate; 11 – gas supply tube.

The mass-to-charge measurements of the ion composition of the magnetron discharge plasma were carried out using two independent methods. The first involved the use of time-of-flight spectrometry. A diagram of the experimental setup is shown in figure 1. The discharge system was insulated from the “ground” potential by insulator 3. Magnetron anode 2 was electrically connected with expander 4, the end of which was closed with a fine-grain grid, 0.7 mm × 0.7 mm, that formed the plasma emission surface. The expander, the grid and the discharge anodes were held at a steady high accelerating potential of up to +20 kV with respect to the ground potential. To suppress the reverse flux of secondary electrons, a small negative potential of –300 V was applied to suppressor electrode
5. Ions were focused in the region of gate 8 (gate) and directed to current collector 9. The pulse 2 kV for 100 ns was applied between the TOF gate rings, allowing the different mass-to-charge ion fractions, accelerated by the same voltage, to reach the Faraday cup at different times. In all experiments, the deflecting voltage pulse was applied at the end of the current pulse. In the absence of a deflecting pulse, ions were prevented from hitting the input of the current collector by central plate 10. A VEU-6 (Russia) channel electron multiplier was used as a current collector. The spectrometer base was 1.2 m. To maintain a stable ignition of the high-current magnetron discharge at a low working pressure, a low-current (10 mA) high-voltage (1.5 kV) power source was connected in parallel to the main source in continuous mode to create a “background” plasma. Gas was feeding into the expander cavity through special tube 11 near the magnetron anode. The pressure drop across the electrodes of the extraction system supported, on the one hand, high gas pressure in the expander region for the sable operation of the magnetron discharge and, on the other hand, low gas pressure in the ion flight region in the spectrometer drift space. The vacuum chamber was evacuated using a turbomolecular pump at a pump rate of 900 l/s. The residual gas pressure was \(3\times10^{-3}\) Pa. In what follows, all working pressure values are given for the vacuum chamber region, not accounting for the pressure drop across the extraction system electrodes.

![Diagram of the experimental setup](image)

Figure 2. Diagram of the experimental setup for the quadrupole diagnostics of plasma ions: 1 – magnetron target; 2 – anode; 3 – Wilson vacuum feed-through; 4 – window; 5 – quadrupole rods; 6 – Faraday cup; 7 – computer; 8 – gas supply tube.

The second method of measuring the mass-to-charge composition of plasma ions in the magnetron discharge employed a retrofitted quadrupole mass-spectrometer RGA-100 (SRS, USA) (figure 2). The standard gas ionizer unit was replaced by an input diaphragm with an aperture diameter of 1.5 mm. The quadrupole mass-spectrometer was mounted on the butt of the vacuum chamber, opposite to the magnetron and coaxial with it at a distance of 10÷30 cm. The drift space in the quadrupole region was evacuated using a separate turbomolecular pump at a rate of 110 l/s, providing pressure not worse than \(3\times10^{-3}\) Pa. The pressure drop across the input aperture of the mass-spectrometer supported, on the one hand, sufficient pressure for stable operation of the magnetron sputtering system and a low degree of dusting of the quadrupole rods by the ion sputtering products of the target and, on the other hand, a low pressure in the quadrupole region, reducing the effect of the resonant ion charge exchange. Increasing the input diaphragm diameter allowed us to increase the amplitude of the recorded signal at the expense of the spectrometer resolution which deteriorated significantly. Since the spectrometer current collector is designed to be at the potential of the chamber body (“ground”), in order to accelerate positive ions, the plasma potential in the spectrometer was positively biased with respect to the grounded walls of the chamber. To this end, the magnetron anode was connected to the positive
lead of an auxiliary power supply source. The plasma bias potential was experimentally found to be \(+40\pm50\) V. The main vacuum chamber, made of stainless steel, was evacuated using a turbomolecular pump at a rate of 500 l/min, providing the residual gas pressure not worse than \(3\times10^{-3}\) Pa. During experiments with the quadrupole mass-spectrometer, the working pressure in the vacuum chamber was measured in direct vicinity of the magnetron discharge system.

The electron temperature was measured using a double probe made of stainless steel. Each plate had dimensions of \(1\times1\) cm\(^2\). The probe was placed co-axially at a distance of 7 cm from the magnetron target. For this case, the magnetron anode was grounded.

### 3. Experimental results

Figure 3 illustrates a typical time-of-flight spectrum of plasma ions in the planar magnetron with a pulsed discharge current of 1 A. The plasma consists of mostly singly charged ions \(\text{Cu}^+\) of the target materials and of the singly charged working gas ions \(\text{Ar}^+\). The fraction of doubly charged copper and argon ions is significantly lower; the presence of doubly charged copper ions becomes noticeable only at a discharge current greater than 1 A. Background components, as a rule, are ions of the residual gas and water vapor (\(\text{H}_2\text{O}^+, \text{O}^+, \text{H}^+, \text{N}_2^+\)).

![Figure 3](image1)

**Figure 3.** Time-of-flight mass-to-charge spectrum of magnetron discharge plasma ions. \(I = 1.5\) A, \(L = 6\) cm, \(p = 0.15\) Pa.

![Figure 4](image2)

**Figure 4.** Ion fractions in the magnetron discharge plasma obtained using the time-of-flight method (a) and the method of quadrupole spectrometry (b). a – \(\tau = 30\) µs, \(f = 10\) Hz; b – \(\tau = 50\) µs, \(f = 300\) Hz.
Increasing the discharge current at a fixed working pressure leads to a monotonous growth of the fraction of the target material in plasma (figure 4 (a)). At a discharge current of about 10 A, the fraction of metal ions in plasma exceeds 70 percent, i.e., the discharge operates predominantly on the atoms of the target material. It may be said that the discharge operates in the so-called self-sputtering mode. Figure 4 (b) shows a similar dependence of the composition of plasma ions on the discharge current when using quadrupole mass-spectrometry. Both techniques qualitatively agree with regard to the mass-to-charge plasma composition. The dependences obtained demonstrate that in a planar magnetron with a target diameter of 50 mm the excess of the fraction of the target material ions over the working gas ions occur at a discharge current of 1.5÷2 A.

Figure 5 shows the argon and copper ion fraction dependencies in plasma at a distance of 9 and 14 cm from the magnetron target. The results were obtained by the time-of-flight method at a discharge current amplitude of 10 A. Measurements of the argon and copper ion fractions in axial direction using the quadrupole spectrometer are shown in figure 6.

The conducted investigations unambiguously indicate that a decrease in the working pressure results in an increase of the argon fraction in plasma ions. Similar characteristic dependences were obtained previously for a planar magnetron discharge with a pure boron target in argon [8] and krypton [9] atmospheres using the time-of-flight plasma diagnostics. The effect of increasing the argon fraction over the copper fraction is observed up to the limiting low pressure at which a high-current discharge becomes unstable.

It should be noted that at a pressure level of 1 Pa, the registered fraction of working gas ions does not exceed 5÷10%. In the absence of a complete picture of the axial distribution of the mass-to-charge composition, the data obtained for considerable distances from the target at high working pressure may be misinterpreted as relating to a magnetron discharge operating in the self-sputtering mode. This is especially clearly manifested for increased distances in the axial direction from the magnetron target and is associated, first of all, with the fact that the energy of gas ions in a magnetron discharge is approximately twice as low as that of the target material ions. With increasing distance from the target at high working pressure, the overall ion signal attenuates as a result of a decrease in the free path length; however, the fraction of argon ions decreases much faster than that of copper ions. The second possible cause for the decrease in the fraction of argon ions with increasing distance is the resonant charge exchange of argon. At a working pressure of 0.03 Pa and an argon ion energy of 1 eV, the...
mean ion free path length before the resonant charge exchange is several centimeters [10]. The resonant charge exchange of copper ions is also possible, however, in a lesser degree, since the concentration of copper atoms is at least an order of magnitude lower than that of argon atoms in the drift space. An increase in the fraction of doubly charged argon ions, as compared to singly charged ions, with increasing distance from the magnetron target is due to a decrease in the cross-section of the resonant charge exchange with increasing primary ion energy [11].

![Figure 6](image6.png)

**Figure 6.** Axial distribution of ion fractions in magnetron for a discharge current of 10 A (a) and 20 A (b). $\tau = 50 \mu s, f = 200$ Hz.

An increase in the fraction of working gas ions with decreasing working pressure can be explained by several reasons. Figure 7 shows the temperature dependence of plasma electrons in the magnetron discharge on the working pressure. The experiments demonstrate that a fourfold decrease in the working pressure leads to a twofold increase in the plasma electron temperature.

![Figure 7](image7.png)

**Figure 7.** Pressure dependences of the magnetron plasma electron temperature. $I = 4$ A, $\tau = 30 \mu s, f = 20$ Hz.

The main ionization region in a planar magnetron is located at a distance not exceeding a few cm from the target surface [12]. The electron energy axial distribution in a magnetron discharge at a distance of up to 10 cm from the target can be assumed Maxwellian [13, 14]. Ionization of working
gas atoms and sputtering of metal atoms is carried out mostly by high-energy electrons from the tail of the Maxwellian distribution. Figure 8 shows the electron energy distribution function (EEDF) for two boundary electron temperature values in the working pressure range in question (figure 7). The obtained distributions show that at high working pressure and characteristic electron temperature therein, at the level of 2 eV, the high-energy tail contains a small number of electrons with the energy sufficient for ionization of argon atoms (solid line), while the copper ionization potential is 7.7 eV. With decreasing working pressure and increasing the electron free path length, the energy of high-energy electrons becomes sufficient for ionization of both copper and argon atoms (dashed line).

The second factor that affects the fraction ratio of argon and copper in the magnetron discharge drift space is the so-called Penning ionization [15] occurring according to the following reaction:

\[ \text{Ar}^* + \text{Cu} \rightarrow \text{Ar} + \text{Cu}^+ + e. \]

The argon excitation potential to the metastable 4s3P1 level is 11.5 eV, while the copper ionization potential is only 7.7 eV [11]. The Penning ionization cross section is of the order of magnitude \(10^{-16}\) cm\(^2\) and is close to the copper ionization cross section by the electron impact [11]. The mechanism of Penning ionization is also facilitated by the fact that in the operating pressure range (0.2 Pa) under study, the density of Ar neutrals is an order of magnitude greater than that of Cu ions (about \(5 \times 10^{13}\) cm\(^{-3}\) and \(5 \times 10^{12}\) cm\(^{-3}\) at a discharge current of 10 A, respectively). A decrease in the working gas pressure in this case reduces the probability of the Penning ionization and of the number of copper ions.

4. Conclusion
The conducted experiments have shown that in a planar magnetron with a target diameter of 50 mm, the threshold current for transition to the self-sputtering mode \((\text{Cu}^+ > \text{Ar}^*)\) is \(1.5 \pm 2\) A at a distance from a target of approximately 10 cm. An increase in the fraction of working gas ions with decreasing working pressure is due to an increase of the plasma electron temperature and a sharp difference in the ionization potentials of the working gas and the target material. The obtained results are in good agreement with previous experiments for other working gas and target material pairs carried out in planar magnetrons: Ar–B and Kr–B. Investigations of dependencies of the mass-to-charge composition of magnetron plasma ions on the discharge current carried out using the time-to-flight and quadrupole techniques are in qualitative agreement. Both methods testify to a change in the fractional ratio of argon and copper ions in the axial direction, with a growing proportion of copper ions as the distance from the magnetron target increases.
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