https://doi.org/10.46813/2021-134-099 THERMALIZATION AND IONIZATION OF METAL ATOMS OF OPERATING ENVIRONMENT AT A METAL ION-SPATTERING SOURCE

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Fast atoms sputtered on a hot metal surface were suggested for thermalization at a metal ion-sputtering source and for further desorption into a discharge with temperature, which is equal to the temperature of the surface. In the suggested construction of the source, atom thermalization process is realized on an interior surface of an anode of the Penning discharge cell in an oscillation area of ionizing electrons. It has been experimentally shown that the proposed method of thermalization of Fe atoms increases the fraction of Fe⁺ ions in the extracted ion beam by three times.

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INTRODUCTION

A metal ion source is the key element of technological accelerating facilities applied in ion-beam surface modification, in radiation material science, ion-plasma coating and other technologies. Two initial work stages should be effectively organized when constructing metal ion sources: generation of sufficient concentration of a certain material atom in a gas-discharge chamber and ionization of received operation environment. The most universal and reliable mechanism of creation of atom operation environment from variety of metals is cathode sputtering in a glow discharge of plasma, because the sputtering is a result of atom collisions in surface layers of a solid body without striking thermal balance between them. The mechanism is particularly effective for generation of metal ions for which vapour pressure ~ 1Pa is attained at temperature over 1000° C.

The disadvantage of this mechanism is high energy E_v and velocity V_v of atom escape from the sputtered surface that decreases chance to ionize the fast atoms with an electron impact W ($W \propto 1/V_v$). Energy of the sputtered atoms may be lowered by their collision with atoms of a plasma gas up to the state of thermal balance [1]. In a small discharge chamber, effective thermalization requires increase of gas pressure so that gas atom concentration in a discharge could significantly exceed concentration of the sputtered atoms. For gas ion sources and metal ion sources, however, pressure of operating gas should be released [2]. In addition, there is a possibility to switch the sputtering source into the self-sputtering mode, thus impurity gas can be used only for a discharge ignition.

In connection with this, other mechanism for relaxation of fast atom energy is required. This paper experimentally proves the idea of thermalization of sputtered Fe atoms on the surface, described in [3], and evaluates the effectiveness of its contribution to the generation of Fe^+ ions.

EXPERIMENTAL FACILITIES AND RESULTS

Structural design of the considered source and distribution of longitudinal component of magnetic field B_z on an axis of discharge chamber are given in Fig. 1.



Fig. 1. Design of the sputtering type metal ion source and distribution of magnetic field B,

Sputtered insertions are made of the metal which ions should be generated (here it is iron) and are placed at face sides of a cylinder 4 (solid hatching in the figure). In this source, an interior surface of the cylinder anode 4 serves as a convertor of fast atoms into slow ones. From this surface, the thermalized atoms are desorbed into the discharge area where oscillation of fast electrons takes place. Thus, two fractions of atom concentration – the fast n_v and the thermalized n_s at one time will be would exist in an area of metal ions generation. Operating principle of the source and main physical processes taking place in it are described in details in work [3]. A brief material is presented in this article to the extent required for the given task consideration.

Gas discharge in the source is based on two consecutive glow discharges – the supporting magnetron discharge M and the main Penning discharge P. An inverse magnetron comprises a cold molybdenum cathode I and shortened anode 2 through which plasma-forming gas is supplied (Ar). The cathode of the Penning discharge is the plasma that is generated in the magnetron, the anticathode is electrode 3, and the anode of the Penning cell is the heat-insulated electrode 4. The required temperature mode of the anode 4 is achieved by distributing the power of the discharge itself without using external heaters. There are sputtering inserts at the axial area of the anticathode and at the gas magnetron outlet where the first (geometric) plasma contraction occurs. The inserts designed as pole tips of the cooled magnetic circuit cause increase of ion current density from plasma onto the sputtered surface. Such geometry of the discharge chamber allows formation of metal atoms directly in the area where ionizing electrons oscillate. Length of the discharge chamber may vary depending on whether the source was adjusted to generate Fe^+ or of Fe²⁺ ions. The magnetic system is based on permanent magnets. A magnetic path is marked with a STEEL hatching in the figure. The main magnets 5 (Nd-Fe-B) are mounted at the rear side of the discharge chamber. The compound correcting magnet 6 (SmCo₅), which generates a radial magnetic field, is fixed to a cylindrical part of the source magnetic path. Along with an additional magnetic pole 7, it allows correction of maximum of induction B_z on the Penning discharge axis. Such design of the magnetic system of the source allows formation of distribution of longitudinal component of a magnetic field on its axis, which is required for two successive glow discharges. Dependence graph for B_z = f(Z) is shown in the top of the figure. Induction of the longitudinal component magnetic field on the anode axis is 249 mT. Ions were extracted through a hole in the anticathode with a diameter of 1.5 mm at an output voltage of 18 kV. Length of an extraction interval was 5 mm, diameter of the aperture in the extraction electrode 8 was 3 mm. The source was operated mainly under a discharge current 2...4.5 A. Drop in voltage on a discharge (cathode 1 – anode 4) was varied in range 350...390 V depending on gas flow rate, which was 0.11...0.10 MPa×cm³×min⁻¹, respectively. Pressure in a body of the anode cylinder 4, which was measured without a discharge in the source at gas flow rate Q =0.11 MPa×cm³×min⁻¹, was $P \sim 2.5$ Pa. Distilled water was used to extract heat from the source electrodes.

A measurement design of a mass-charge spectrum of an ion beam is mentioned in work [3]. Surface temperature of the cylinder anode 4 was measured with a thermocouple (ion beam was not extracted from the source during the measurements).

Escape energy from the sputtered atoms surface in plasma of the glow discharge is usually no more than $E_{\nu} = 10...15 \text{ eV}$ [4] and will later relax on collision with plasma atoms. Different calculating models are used to describe the process of particle transportation, the simplest is the model of binary collisions in approximation of rigid spheres [1]. Using this model, the length of thermalization zone of the sputtered Fe atoms which collide with *Ar* atoms is estimated. From this model the length of thermalization zone is defined as

$$R_{\tau} = \ln \frac{E_{\tau}}{E_{V}} [\ln(1 - \beta)]^{-1} \cdot / N_{q}, \qquad (1)$$

where E_v is an average energy of sputtered atoms; E_T is energy of thermal motion of buffer gas atoms; $\beta = 2m_v m_g /(m_v + m_g)^2$, is an average relative portion of energy lost at elastic collision of particles with masses mv and mg; N = P/kT, is concentration of gas particles; $q = \pi (R_v + R_g)^2$ is full microscopic cross-section of elastic scattering of particles with radius R_v and R_g . At P = 2.5 Pa, $E_T = 0.013$ eV, $E_v = 10$ eV, $R_v = 156 \times 10^{-10}$ cm and $R_g = 71 \times 10^{-10}$ cm the length of thermalization zone is $R_T \sim 10$ cm. Length of an interval between the farthest surface area of the sputtered insert and the anode 4 is 1.6 cm that is considerably lower than the calculated length of thermalization zone of the sputtered atoms. Therefore, it is reasonable to suppose that the main part of the sputtered Fe atoms would move in the source plasma in a transit mode with energy kT_v until the atoms reach a surface of the cylinder anode 4 which has an operating temperature $T_s \sim 1800$ K ($T_s \ll T_v$). After the atoms collide with the surface, they exchange their energies with the surface atoms. Heat exchange efficiency is evaluated with thermal accommodation coefficient defined as

$$\alpha = \frac{(T_v - T_r)}{(T_v - T_s)},\tag{2}$$

where T_r is temperature of reflected atoms before equilibrium with a surface.

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At $\alpha = 1$, full thermal accommodation of the atom occurs due to excitement of phonons of the anode material. At $\alpha < 1$, the atom would transmit its excess energy only partially and having been reflected with energy $E_r = kT_r$, it would collide with another part of a cylindrical surface of the anode. After some collisions with the wall and the energy exchange between them, the atom, finally, comes to thermodynamic equilibrium with the wall ($\alpha = 1$) and oscillates in a potential well with a depth of equal of adsorption energy. Kinetic energy related to these oscillations corresponds to the surface temperature.

When the concentration of metal atoms thermalized on the surface is formed in the Penning discharge chamber, there is a question to be asked: what time interval would the absorbed atom lie on hot surface until it is further absorbed? On the assumption that the adsorbed atoms do not interact with each other, the average adatom lifetime may be expressed as follows [5]

$$\tau_{\alpha} = \frac{1}{v_0} e^{\frac{L_{ad}}{kT_s}},\tag{3}$$

where v_0 is frequency of adatom thermal vibrations in a crystal lattice, E_{ad} is an adsorbtion energy.

As is seen, average adatom lifetime is mainly defined by surface temperature and is shortened as the temperature rises. Real values of adsorbtion energy $E_{ad} = 0.25$ eV, surface temperature $T_s = 1800$ K and $v_0 = 10^{13}$ s⁻¹ substituted into the expression (3) give average adatom lifetime prior to him desorbtion, that is $\tau_a \sim 5 \times 10^{-13}$ s. In other words, in conditions where there is no nucleation, the thermalized atoms should be immediately desorbed at this temperature.

At high flow of sputtered atoms, the adsorbed particles on a surface can form a solid monoatomic layer and even cause a multilayer adsorbtion. Surface density of the adsorbed particles depends on the material of the surface, degree of its ideality and temperature, kind and current density of the sputtered atoms. If iron vapours are applied as a working agent, a growing metal film eventually causes magnetic field distortion in the Penning discharge cell. In this source, deceleration of nucleation rate is achieved first due to heating of the anode to high temperature that damages connection between the adsorbed atoms and the surface; and second due to reduction of specific area of the anode, i.e. its roughness.

Difference between probability of ionization of the iron atoms desorbed after thermalization on surface W_s and moving in transit mode under other equal conditions is evaluated below. Probability of atom ionization by an electron impact is [6]

$$W = \frac{\sigma_i j_e L}{V},\tag{4}$$

where σ_i is ionization cross section, which depends on electron energy, j_e is current density of ionizing electrons, *L* is path length where the atoms interact with the ionizing electrons, *V* is velocity of the neutral atoms. Suggest that $T_s = 1800$ K, which corresponds to $E_s = 0.158$ eV, and energy of the sputtered atoms is $E_v = 15$ eV, then velocity of the desorbed and sputtered iron atoms will be $V_s = (2 \times 0.155/\text{m})^{1/2}$ and $V_v = (2 \times 15/\text{m})^{1/2}$, respectively. Here of it follows

$$W_{\rm s} \approx 9.85 \, W_{\rm v} \,, \tag{5}$$

i.e. probability of atoms ionization by an electron impact is orders of magnitude higher. The ion source was tested in two operating modes at the same j_e , L, σ_i and two different temperatures of the convertor T_s , in order to evaluate the efficiency of contribution of the thermalized iron atoms into Fe⁺ ions generation. For these experiments, the length of the gas discharge gap (from the exit aperture of the gas magnetron to the emission discharge) and the discharge operating modes were adjusted to generate single charged ions.

Mass spectrum of the ion beam was measured by the technique described in the work [3]. Here two collimating slits cut out a paraxial ribbon beam, which gets to a deflecting magnetic field of an analyzer. Under the magnetic field, the ions are deflected from the analyzer axis by a value proportional to ratio of ion charge to ion mass and are analyzed by a movable wire collector with an increment of 0.1 mm. The results are displayed on the monitor. Ion current of each mass in the ribbon beam is proportional to square of a corresponding peak. Percentage of each ion mass of the ribbon beam was calculated from sum of currents of all its fractions. In impulse mode of the source ($t_{puls} = 1 \text{ ms}, f = 1 \text{ Hz}$), value of a signal of the wire collector was recorded after each step with a digital oscillography GDS-2064 and was averaged over four measurements.



Temperature Penning's anode is 450 K

Fig. 2 shows M/Z spectrum of a ion beam extracted from the source, which operates in the impulse mode. The surface temperature here for the sputtered atoms thermalization was $T_s = 450$ K.



Fig. 3. Mass and charge spectrum of the ion beam. Temperature Penning's anode is 1800 K

Fig. 3 shows a M/Z spectrum of a beam extracted from the source, which operates in the stationary mode at the convertor temperature $T_s = 1800$ K.



Fig. 4. Comparison of the intensities of different fractions of the ion beams

Fig. 4 shows ratio of currents of different fractions of the ion beam to a total beam current for two different temperatures of the convertor at the same parameters of the discharge. As is seen from the diagram, Fe⁺ fraction was ~ 12% of total beam current at low temperature of the convertor when contribution of the thermalized concentration ns was low. If temperature is high, fraction of the thermalized atoms at the zone of ionizing electrons oscillation increases. It results in more efficient generation of Fe⁺ in the discharge and in increase of Fe⁺ fraction in the extracted beam up to ~ 36%.

CONCLUSIONS

The results of the tests performed for the metal ion sputtering source showed that thermalization of the Fe atoms sputtered on the hot metal surface increases current of Fe⁺ fraction in the extracted beam by three. In any case, under optimal discharge current and plasma forming gas (Ar) pressure, total current of single and double charged iron ions in the ion beam is 46%. Further design of the source would be aimed at generation of refractory metal ions.

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ТЕРМАЛИЗАЦИЯ И ИОНИЗАЦИЯ АТОМОВ МЕТАЛЛОВ РАБОЧЕЙ СРЕДЫ В МЕТАЛЛИЧЕСКОМ ИОННО-РАСПЫЛИТЕЛЬНОМ ИСТОЧНИКЕ

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В источнике металлических ионов распылительного типа предложено производить процесс термализации быстрых распыленных атомов на горячей металлической поверхности с последующей их десорбцией в разряд с температурой, равной температуре этой поверхности. В рассматриваемой конструкции источника термализация атомов реализована на внутренней поверхности анода пеннинговской разрядной ячейки в области осцилляции ионизирующих электронов. Экспериментально показано, что термализованные таким образом распыленные в разрядной камере атомы Fe увеличивают фракцию Fe⁺ в экстрагируемом ионном пучке в три раза.

ТЕРМАЛІЗАЦІЯ І ІОНІЗАЦІЯ АТОМІВ МЕТАЛІВ РОБОЧОГО СЕРЕДОВИЩА В МЕТАЛІЧНОМУ ІОННО-РОЗПИЛЮВАЛЬНОМУ ДЖЕРЕЛІ

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У джерелі металевих іонів розпилювального типу запропоновано проводити процес термалізації швидких розпорошених атомів на гарячій металевій поверхні з наступною їх десорбцією в розряд з температурою, що дорівнює температурі цієї поверхні. У розглянутій конструкції джерела термалізація атомів реалізована на внутрішній поверхні анода пенінговської розрядної комірки в області осциляції іонізуючих електронів. Експериментально показано, що термалізовані таким чином розпорошені в розрядній камері атоми Fe збільшують фракцію Fe⁺ в екстрагованому іонному пучку в три рази.