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STUDY OF LOW-PRESSURE DISCHARGE BY OPTICAL EMISSION SPECTROSCOPY

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The axial distribution of excited argon and tungsten atoms in plasma of direct current magnetron discharge in crossed fields have been analyzed by optical emission spectroscopy. The influence of discharge parameters (discharge current I_d, buffer gas pressure p_{Ar} and zone of discharge glow Δl) and excited states energy E^* of studied particles on the axial distribution Ar and W atoms have been observed. The assumption about the excitation processes in the magnetron plasma is given.

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INTRODUCTION

The magnetron sputtering devices (MSD) are widely used in the microelectronics industry for plasma etching and deposition of thin film with unique physical and chemical characteristics [1-3]. Due to the importance to create high quality of thin films it is necessary to carry out systematic experimental investigations of the fundamental plasma parameters for the kinetics of the particles formation understand [4]. The most common plasma parameters diagnostic methods are Langmuir probe [5] and Optical Emission Spectroscopy (OES) [6, 7]. The use of the Langmuir probe method is complicated due to a possible plasma perturbation. OES is non-invasive. In addition, the experimental set-up is very simple: only diagnostic ports are necessary which provide a line-of-sight through the plasma. Although the optical emission spectra of plasma can be easily obtained, interpretation can be complex as should be done careful analysis of kinetic processes that contribute to populating and depopulating excited states of the species in the plasma. The optical emission spectra of plasma provide the information on the chemical composition of plasma particles and the population of its excited states by electron impact processes. The spatial distribution of the excited particles can provide information about the kinetics of particle formation to determine such fundamental plasma parameters as the electron density and the electron temperature. The optical radiation from plasma usually is studied using OES measurements performed through a window with an optical fiber. For spatial measurements an optical fiber is pointed to the plasma bulk perpendicularly to the window [2, 8] and is moved along the discharge axis as well as MSD is moved along the discharge axis [9].

In the present study the emission spectra of glow of planar magnetron discharge in crossed field in the visible spectral range have been obtained. The analysis of plasma optical radiation was carried out using software developed by our research group. The influence of the main discharge parameters (gas density, discharge current and the glow plasma domain) and the energy of the excited state of particles on the axial distribution the spectral line intensity have been studied. A discussion on the main kinetic processes by electron excitation in the magnetized plasma is given.

EXPERIMENTAL SET-UP

The experimental set-up consists of a MSD with optical arrangement for OES measurements (Fig. 1). A detailed description of the MSD is given in [10]. Typical experimental conditions of magnetron discharge (MD) for a tungsten target are as follows: buffer gas (Ar) pressure $p_{Ar} = 8...15$ Pa, discharge voltage $V_a = 350$ V, discharge currents $I_d = 10...160$ mA, magnetic induction $B = 0.05$ T. The choice of cathode material is due to the fact that tungsten has a low sputtering coefficient, a high melting point and rich atomic spectrum in visible spectral region.

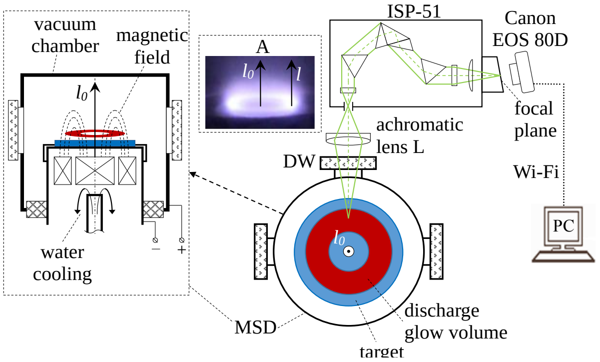

Fig. 1. Experimental set-up

The MD region can be conventionally divided into three main parts [4, 11]: the cathode sheath, the dense plasma magnetized region (bright glow space, ~ 10 mm) and then the space down to the anode that the MSD camera serves. The optical radiation from the bright glow region exit through the diagnostic window (DW) of the MSD camera, and then is focused using an achromatic lens (L) on the entrance slit of the ISP-51 spectrometer, in which the radiation is dispersed using three-prism system. After dispersion, the spectrum in the wavelength range of 390...650 nm is focused on the focal plane of the spectrometer output collimator. Inset A in Fig.1 shows the MD glow and the direction l of

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extent of the discharge glow parallel to the discharge axis l_0. With working geometry the optical radiation of the MD glow extent Δl distributed along of the spectrum lines high (Δh) with decrease 0.25 (Δh=0.25Δl). That image of spectrum was photographed using a Canon EOS 80D digital camera with a matrix size of 7000/5000 pixels. The photons of MD radiation incident on the sensor of the charge-coupled device (CCD) and converted into the electronic signal. After digitization the output signal (D) transferred to a computer for display image and storage.

To display the measured emission spectra and determine the qualitative and quantitative characteristics of MD plasma, a multifunctional interactive GUI application OSA (Optical Spectrum Analyzed) is used [12, 13]. The OSA application was created in the Python programming language, using the Tkinter graphics library, and uses a set of additional modules: PIL, SciPY, NumPy, and Matplotlib. Mathematical algorithms and procedures have been developed that allow to processing the numerical matrix corresponding to the selected digital image and visualize the results. For conversion the digitization signal (D) into photonic signal (I) the response function (RF) was obtained with using nine-stage attenuator.

EXPERIMENTAL RESULTS AND DISCUSSION

The spectra of the MD radiation were recorded in a wide wavelength range of 400...650 nm and cover all experimental conditions explored in the study (p_{Ar} = 8, 11, and 15 Pa; I_d = 10, 20, 30, 40, 50, 60, 70, 100, 120, 140 and 160 mA). Spectra in the wavelength 400...650 nm mainly composed of tungsten atom lines that belong to the transition from states with electron configuration $5d^4 6s6p$ to grand state with electron configuration $5d^4 6s^2$. The spectra also contained weak lines of argon atoms (the transition from the states with electron configuration $3s^2 3p^5 5p$ and $3s^2 3p^5 5d$ to states with electron configuration $3s^2 3p^5 4s$ and $3s^2 3p^5 4p$) and weak lines from singly ionized Ar^+ ions. As was pointed out in [14] the pronounced argon lines are observed in the red region (690...900 nm). The emission spectrum of the MD glow in the wavelength range of 400.0...650 nm is given in Fig. 2. Discharge external parameters: I_d = 70 mA, U_d = 350 V, p_{Ar} = 10 Pa. In the emission spectrum are predominantly present W I lines and a series of Ar I and Ar II lines. There is a significant difference in the intensity (the digitization signal D) distribution of spectral lines emitted by excited W and Ar particles along their height (h).

The spectral line intensity (I) depends on the population density of the excited level (n^*) as:

$$I = n^* \cdot A \cdot (h \cdot c/\lambda), \qquad (1)$$

where A is the Einstein transition probability, h – Planck constant, c – speed of light and λ – wavelength of line. Therefore a change in the line intensity (I) along its height (h) reflects change in the population density of excited particles (n^*) along direction (l). In order to study the spatial distribution of excited particles along the discharge axis, some lines were selected, the parameters of which are given in the Table. The choice of lines was determined by the following: the excitation energies of the observed tungsten lines are in the range from 2.48 to 3.24 eV, therefore three lines with the lowest, highest, and intermediate excitation energy were chosen for analysis. Since the Ar lines observed in the spectrum has the excitation energy of ≈ 15 eV, for the study the line with high intensity was chosen for the study.

Measurements of $I(h)$ of the selected lines were performed for various parameters (p_{Ar}, I_d) of the MD. The results are shown in Figs. 3, 4.

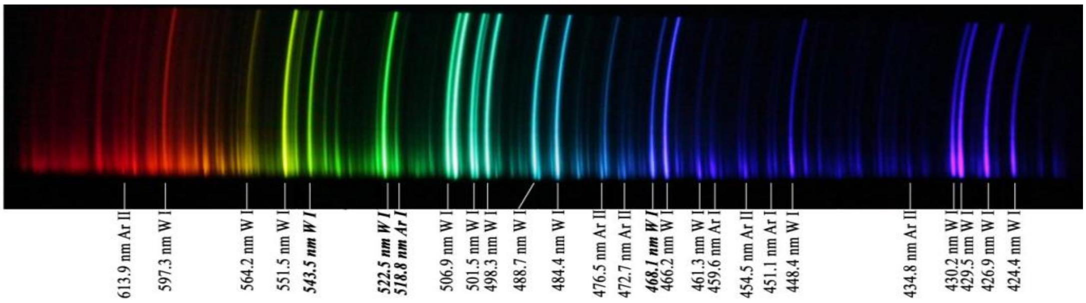

Fig. 2. The emission spectrum of the magnetron discharge glow

The general behavior is as follows: as h increases the I rises sharply to a peak and then slowly falls. However, the shape of I(h) is determined by the discharge parameters (Id, pAr) as well as excitation energy (E*) of the emitting particle. The intensity of line is directly proportional to the number of excited particles emitting this line therefore I(h) displays the distribution of excited particles along the discharge glow extent (Δl). For the λλ 468.1 nm W I, 518.8 nm Ar I lines emitted by the atoms excited in states with high excitation energy E* I(h), a pronounced peak is determined, and then a sharp decrease. That means, that the highly excited particles are formed in the nearest to cathode part of glow. At the same time, for the λλ 543.5 nm, 522.5 nm W I lines emitted by the atoms excited in the states with low excitation energy E* I(h) changes insignificantly. Therefore, particles excited to levels with low E* are formed in the entire registration region of glow with almost identical probabilities. Such a difference in I(h) for the lines with E*<3 eV and E*>3 eV is probably due to the presence of two group of electrons in MD plasma [17]. i) The energetic electrons which results from the injection of electrons accelerated in the cathode sheath with subsequent energy degradation in the glow. These energetic electrons can allow the direct formation of excited atoms in states with high excitation energy. ii) The thermal electrons which are located in the magnetized zone of the discharge. These electrons are formed atoms in the states with low excitation energy.

The parameters of the spectral lines studied in the work

Wavelength, nm λ_{exp}, / λ_{air} [16]	Interpretation	Excitation energy E*, eV [15]	Spectral transition [16]	
			Upper state Configuration State	Lower state Configuration State
543.5 / 543.5032	W I	2.48	$5d^46s(^6D)6p \quad ^7F_1^0$	$5d^46s^2 \quad ^5D_1$
522.5 / 522.4675	W I	2.97	$5d^46s(^6D)6p \quad ^7D^{\circ}_2$	$5d^46s^2 \quad ^5D_3$
468.1 / 468.0509	W I	3.24	$5d^46s(^6D)6p \quad ^7D^{\circ}_3$	$5d^46s^2 \quad ^5D_3$
518.8 / 518.7746	Ar I	15.30	$3s^23p^5(^2P^o{}_{1/2})5d \quad ^2[1\,½]^o{}_2$	$3s^23p^5(^2P^0{}_{3/2})4p\ ^2[½]_1$

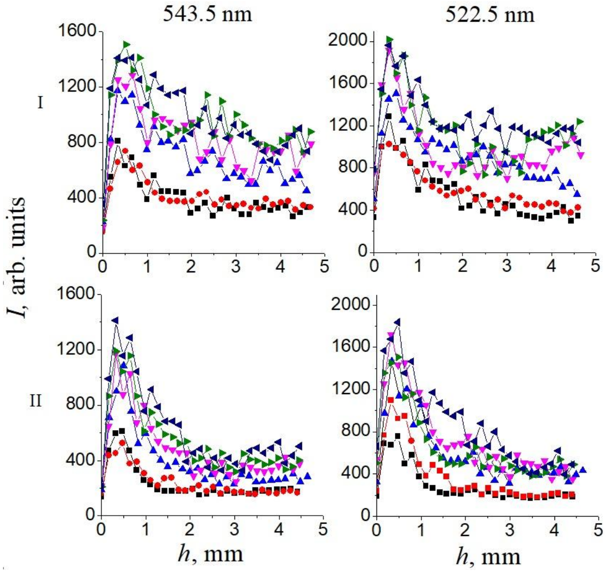

Fig. 3. Variation of intensity (I) of denoted lines on it height (h). I – p_{Ar} =15 Pa, II – p_{Ar} =8 Pa;
I_d = 60 mA (■), 70 mA (●), 100 mA (▲), 120 mA (▼), 140 mA (▶), 160 mA (◀)

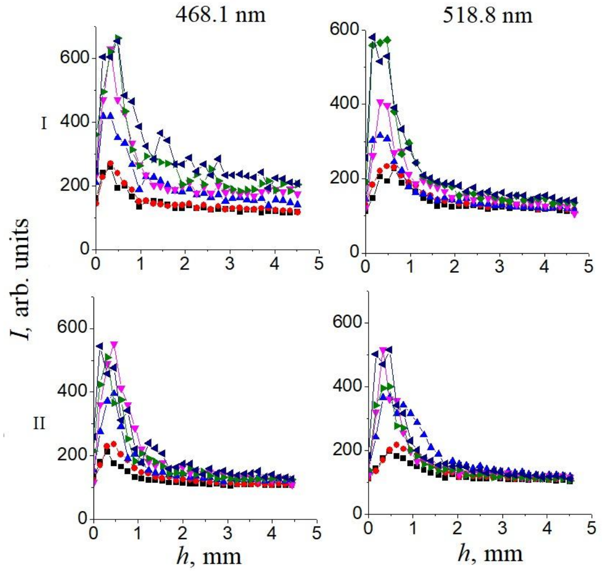

Fig. 4. Variation of intensity (I) of denoted lines on it height (h). I – p_{Ar} =15 Pa, II – p_{Ar} =8 Pa;
I_d = 60 mA (■), 70 mA (●), 100 mA (▲), 120 mA (▼), 140 mA (▶), 160 mA (◀)

In order to study the influence of the discharge parameters (discharge current, zone of the extent of MD glow and gas pressure) on the excited particles formation, the intensities of the selected lines (I_λ) against the discharge current (I_d) at the Ar pressures p_{Ar} = 11 Pa and two zones of discharge present in Fig. 5. Logarithmic scale have been chosen to interpret the results as $I_\lambda = k\cdot I_d^{\alpha}$.

At the bright glow region of MD (see max *I(h)*, Figs. 3, 4) the alike behavior of the intensity of all the denoted lines on the discharge current was observed: as I_d increases the I_λ rises too up to saturation at high currents. The saturation at low currents was observed too, but it was conditioned by the background noise of the digital signal at weak lines intensity. The insignificant increase in the line intensity with $p_{\rm Ar}$ = 15 Pa was observed. Although the intensity of selected lines behaves like $I_\lambda = k\cdot I_d^{\alpha}$, the slopes in log-log plot are very different: α=1.4 for λ543.5 nm W I, α=1.6 for λ522.5 nm W I and α~1 for lines λ486.1 nm W I and λ518.8 nm Ar I.

As it was pointed out earlier (1), the line intensity correlates with the population of the excited level of studying transition. From the experimental data it follows that the excited states population of plasma species strongly depends on the discharge current and different processes are determine the population of studied levels. The excitation energy of the tungsten atoms studied in the work is in the range 2.5…3.5 eV. The argon atoms excitation energy of all excited states is above 11 eV [18]. So the direct electron impact excitation processes are the most important ones for W. Therefore, the direct electron impact excitation processes are more significant for W than for Ar.

A simple approach to the population density in electron-atom collisions is presented by the corona model [6], where it is assumed that the upward transition occurs only due to electron collisions, while the downward transitions only due to radiative decay. In the simplest case, the population of an excited state (n_i) is balanced by electron impact excitation from the ground state (n_0) and decays by spontaneous emission (optically allowed transitions to lower level (k)):

$$n_0\cdot n_e\cdot X_{0i}^{exc} = n_i\cdot\Sigma A_{ik}, \qquad (2)$$

where X_{0i}^{exc} is excitation rate coefficient which can be obtained from the convolution of the cross section with the corresponding energy distribution function (EEDF) of the impact particle; n_e is the electron density. As following from (1) the spectral line intensity (I_{ik}) can be written as:

$$I_{ik} = (A_{ik}/\Sigma A_{ik})\cdot n_0\cdot n_e\cdot X_{0i}^{exc}. \qquad (3)$$

In MD the density of sputtered metal atoms is proportional to the discharge current and the Ar^+-W sputtering coefficient (Y); the electron density is proportional to the discharge current and the W coefficient of ion-electron emission (γ). So (3) can be written as:

$$I_{ik} = K_{ik}\cdot(I_d)^2, \qquad (4)$$

where $K_{ik} = Y\gamma\cdot(A_{ik}/\Sigma A_{ik})$. Therefore, it can be concluded that the increasing part of $I(I_d)^\alpha$ of lines W I in Fig. 5 reflects the change in the population of particles excited in identified state on I_d. As it is seen α is not exactly equal to 2 as X_{0i}^{exc} is depends on such plasma parameters as n_e, n_0 etc. [6].

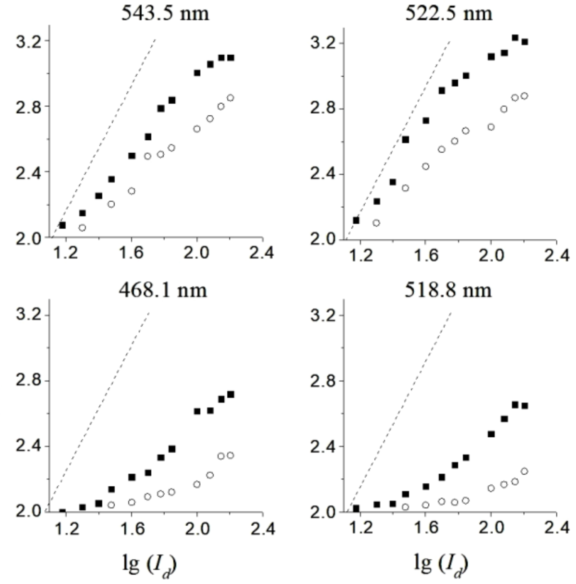

Fig. 5. Variation of the Ar I and W I lines intensity (I) on discharge current (I_d) in lg-lg plot. ■ – the peak of I(h); ○ – at h =1.5 mm from the peak I(h); p_{Ar} = 11 Pa. The dotted line denotes $I \sim I_d^2$

So only two processes are important: the electron-impact excitation process from the ground state W atoms and spontaneous radiation from the excited atoms. Thus the population of W atoms in excited states can be described by corona model which is valid for excited plasma species in low-temperature plasma with low pressure (<10 Pa) [7]. A more general approach to population is to set up rate equations for each state of the particles together with the coupling to other particles. Such model, which balances the collisional and radiative processes, is called the collisional radiative model (CRM) [6]. So, the saturation I_λ depending on I_d at high I_d (Fig. 5, W I) can be associated with non-radiative electron or tungsten atom impacts with excited tungsten atoms when the n_0 of tungsten atoms rises with growth of I_d.

Dependence the Ar I line intensity on discharge current as $I_\lambda = k\cdot I_d^\alpha$ cannot be described by corona model because in Ar plasma at $p_{Ar} \approx 10$ Pa the tail (E>10 eV) of the low-energy part of the EEDF is decreased as compared with Maxwellian distribution [19]. The formation of Ar atoms in the 5d state with the excitation energy of 15.34 eV [18] in the direct electron-Ar atom in ground state impact is unlikely. But the Ar atom has two metastable states with the excitation energies 11.548 and 11.723 eV [18], therefore, it is most likely that the excited in 5d state Ar atom is formed in two-step processes: e+Ar(0)→Ar(m) (1); e+Ar (m)→Ar (5d) (2). In zone of the extent of MD glow at h=1.5 mm (see *I(h)*, Figs. 3, 4) the dependence of the intensity of all the denoted lines on the discharge current significantly changes, as a result of change in density distribution of electrons in that zone [20] which affects the behavior of $I_\lambda(I_d)$.

CONCLUSIONS

In the work represented the experimental set-up is made of MSD with an optical arrangement for OES measurements. The significant feature of the optical arrangement is the relationship between the change in the spectral line intensity along its height and the change in the excited particles population in the glow part of MD along the axial direction. The optical radiation from the bright glow region was analyzed by a multifunctional interactive GUI application Optical Spectrum Analyzed at different discharge parameters. From the obtained data it follows that the spatial distribution of excited particles varies along the axis and strongly depends on the MD parameters and the type of discharge species. A change in the line intensities in correlation with the discharge current indicates that particles of the cathode material (W) and the buffer gas (Ar) get excited in different collision processes. Excitation of W atoms occurs in the direct electron-impact process and can be described by corona model. An excited Ar atom in 5d state with a high excitation energy due to the presence of two metastable states is formed in two-step processes within the collisional radiative model.

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ИССЛЕДОВАНИЕ РАЗРЯДА НИЗКОГО ДАВЛЕНИЯ МЕТОДОМ ОПТИЧЕСКОЙ ЭМИССИОННОЙ СПЕКТРОСКОПИИ

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Методом оптической эмиссионной спектроскопии проанализировано аксиальное распределение возбужденных атомов аргона и вольфрама в плазме постоянного магнетронного разряда в скрещенных E×H-полях. Обнаружено влияние параметров разряда (ток разряда I_d, давление буферного газа p_{Ar} и область свечения разряда Δl) и энергии возбужденных состояний E^* исследуемых частиц на распределение атомов Ar и W вдоль оси разряда. Сделано предположение о процессах возбуждения в магнетронной плазме.

ДОСЛІДЖЕННЯ РОЗРЯДУ НИЗЬКОГО ТИСКУ МЕТОДОМ ОПТИЧНОЇ ЕМІСІЙНОЇ СПЕКТРОСКОПІЇ

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Методом оптичної емісійної спектроскопії проаналізовано аксіальний розподіл збуджених атомів аргону і вольфраму в плазмі постійного магнетронного розряду в схрещених E×H-полях. Виявлено вплив параметрів розряду (струм розряду I_d, тиск буферного газу p_{Ar} і область світіння розряду Δl) та енергії збуджених станів E^* досліджуваних частинок на розподіл атомів Ar і W уздовж осі розряду. Зроблено припущення про процеси збудження в магнетронній плазмі.