Spectral measurements in the plasma of microwave and magnetron discharges

O I Shipilova, A A Chernikh and V L Paperny
Irktusk State University, 1 Karl Marx Str., Irktusk, 664003, Russia
E-mail: paperny@math.isu.runnet.ru

Abstract. Spectroscopic methods were used to study the parameters of a low-power (≤ 100 W) magnetron discharge plasma, as well as a low-pressure microwave discharge (6–40 Pa) in argon. It is shown that in the magnetron discharge the concentration of atoms and ions of the buffer gas (Ar) and the target material (Cu) decreases exponentially with distance from the target cathode and in the first case it occurs much more sharply. The temperature of electrons in a plasma stream, which is emitted by a discharge burning near the surface of the cathode, was estimated from the relative intensity of the spectral lines according to the LTE model and the coronal model. The values obtained in both cases were close, lying near 1 eV, and the temperature remained almost constant with the distance from the cathode. The electron temperature in the microwave discharge turned out to be almost constant over the cross section of the plasma column, while the concentration near the microwave emitter was 2–3 times higher than in the rest of the column.

1. Introduction
Low-temperature plasma is widely used to create modified layers in dielectrics. In particular, using a combination of a magnetron discharge emitting a stream of metal atoms (Au, Ag, or Cu) and a thermal evaporator emitting a stream of atoms of a dielectric matrix (LiF), composite dielectric films containing nanoscale metallic clusters were synthesized which possessed, for example, the non-linear optic (in case of Au) [1] or memristor (in the cases of Ag and Cu) [2] characteristics that are promising for applications. The parameters of the film, in particular, the content of metal nanoclusters, their size, the degree of defectiveness of the film, etc. are determined by the parameters of the plasma flux of a magnetron source, therefore the measurement of these parameters is important to justify the method of film synthesis. Taking into account that the degree of ionization of a low-power magnetron discharge plasma does not exceed 10^-5, probe diagnostic technique for this plasma is ineffective, and spectroscopic methods should be used.

Such measurements of the dependence of plasma flow parameters on the distance to the cathode are very few [3, 4], and in these works, the LTE model was used to estimate the electron temperature $T_e$, the applicability of which under given experimental conditions seems to be unjustified. Therefore, conducting such measurements and analyzing the applicability of various radiative models for estimating $T_e$ seems an important problem.

Microwave discharges of different types are used to create in the dielectric matrix modified layers containing radiation-induced defects (color centers), which have numerous applications (see, for example, review [5] and references there). In particular, the microwave discharge of reduced pressure...
we used earlier to create these layers in LiF crystals [6]. To optimize this method, given the high energy density in such a discharge and the thermal instability of the induced centers, the actual problem is to study the distribution of plasma parameters in the working chamber.

In the present work, spectroscopic measurements of the spatial distribution of plasma parameters in the types of discharges above and the analysis of the obtained results were carried out in order to substantiate the methods used.

2. Experimental details and discussion

2.1. Measurements in a magnetron discharge

The emission spectrum of a magnetron discharge plasma was measured with an AvaSpek 2048 fiber spectrometer in the wavelength range of 200–1100 nm with a spectral resolution of about 1 nm. The input end of the movable optical fiber was oriented perpendicular to the axis of the plasma jet emitted by the discharge and moved along this axis, registering radiation from the cross section of the plasma column, which was located at a distance $h$ from the surface of the target-cathode. The spectra were measured at typical current and voltage of the discharge of 240 mA and 420 V, respectively, and a buffer gas pressure (Ar) of $1.7 \times 10^{-2}$ Torr.

![Figure 1](image)

Figure 1. The dependence of the intensity of the brightest lines of copper and argon atoms, as well as the lines of argon ions on the distance to the cathode target in a magnetron discharge. The vertical line denotes the conditional boundary of a discharge burning near the cathode surface.

Figure 1 shows that in the discharge area, the intensity of the lines, i.e. the concentration of emitting plasma atoms is relatively high; when we leave the discharge area, it drops sharply; with a subsequent distance from the discharge, line intensities in the ejected plasma jet drops exponentially. Note that the intensity of both components of the argon lines, therefore, the concentration of the emitting argon atoms, falls much more sharply than the concentration of the emitting copper atoms. The observed drop of the concentration of fast atoms of the plasma jet ejected by the discharge is due to their scattering by thermal atoms of the background buffer gas, as a result of which the atoms of the jet lose directional velocity, leave the jet, and their radiation is not recorded further. The characteristic free path lengths obtained from the data in figure 1 for copper atoms were 2 cm, and for argon atoms, 1 cm, which with satisfactory accuracy corresponds to the estimates obtained for these experimental conditions.
Let us now consider the dependence of the concentration of plasma components on the discharge current that was measured near the discharge boundary, depicted in figure 2. From the figure, it can be seen that the concentration grows exponentially with increasing current, and the exponent for copper atoms is greater than for argon (0.016 and 0.01, respectively). This indicates a significantly more rapid increase in the concentration of sputtered atoms of the cathode material that is naturally associated with an increase in the concentration of argon ions in the discharge plasma accelerated by the electric field of the cathode and providing atomization of the target material. Thus, we can conclude that with increasing voltage and, accordingly, the discharge current, the concentration of the discharge plasma increases exponentially.

Then, for each plasma component, from the intensity ratio of different pairs of spectral lines of this component, the temperature was estimated for the distribution of emitting atoms among the excited states of $T_{\text{exc}}$.

Assuming also that the gas of the excited atoms is in thermodynamic equilibrium with the gas of the exciting electrons, we will assume that the temperatures of their equilibrium distributions are equal, i.e. $T_{\text{exc}} = T_e$. This approximation is commonly referred to as the local thermodynamic equilibrium (LTE) model. The temperature was estimated by the known formula by a large number (100-150) line pairs:

$$\ln\left(\frac{I_{n_2}A_{n_2}}{I_{n_1}A_{n_1}}\frac{g_{k_n}}{g_{k_j}}\right) = -\frac{\Delta E_k}{kT}$$

where $I_n$, $\lambda_n$, $g_k$, $A_{k_n}$ ($n = 1,2$ are indexes of lines of the considered pair) are, respectively: intensity, wavelength, statistical weight and probability of a spontaneous radiative transition from the $k$-th, top level, to the lower, $i$-th level and $\Delta E_k$ is the difference of the upper energy levels of this pair of lines.

The result of estimating the temperature at different distances from the cathode is shown in figure 3 from which it follows that the obtained values lie in the range of 0.6–1.1 eV, and the scatter of values obtained over different pairs of lines is quite significant. However, the values are distributed in a rather chaotic way, so this variation should be attributed to the errors of the technique. In this case, it is possible to speak of a certain the same average temperature for the excitation of all plasma components, which it is natural to accept as the equilibrium electron temperature $T_e$. From figure 3 it can be seen, also, that with the distance from the cathode, a slight decrease in temperature is observed in the vicinity of the discharge, and with the subsequent movement of the plasma jet emitted by the discharge, the temperatures of all the components remain practically unchanged.
However, estimates show that for given experimental conditions, the conditions of applicability of the LTE model are not satisfied, and the coronal model is more appropriate. Within the framework of this model, according to the experimental data obtained, an estimate of the electron temperature was obtained according to the formula

\[ \ln \left( \frac{I_{k_i} A_{k_i}}{A_{k_i} a_{k_i}} \right) = \frac{E_{k_i}}{k T_e} + \text{const} \]

Here, \( a_{k_i} \) is the proportionality coefficient for approximating the velocity coefficient of collisional excitation at temperature \( T_e \) from the ground state to the excited state \( \hat{k} \), and the summation goes over all underlying transitions. These coefficients, as well as the transition probabilities \( A_{k_i} \) for the Ar lines obtained from the experiment, are taken from [8].

The result depicted in figure 4, shows that the temperature remains almost constant with distance from the cathode and averages \( 1.0 \pm 0.3 \) eV. This value is quite close to the temperatures obtained above using the LTE method.

![Figure 4.](image)

**Figure 4.** T. The dependence on the distance to the cathode of the electron temperature, calculated according to the coronal model.

### 2.2. Measurements in a microwave discharge

The discharge is based on a household microwave oven, into the chamber of which the working volume is placed (see figure 5). The volume was pumped off to a pressure of 4 Pa, and then the working gas (Ar) was injected, the pressure of which was adjusted in the range of 6–60 Pa. The discharge was excited by the 800 W magnetron radiation with a frequency of 2.45 GHz (\( \lambda = 12.5 \) cm).

![Figure 5.](image)

**Figure 5.** The scheme of the experiment with a microwave discharge: (1) is a microwave oven, (2) is working chamber, (3) is input for the working gas, (4) is exhaust pipe, (5) is the AvaSpec 2048 spectrometer, 6 is a laptop.

![Figure 6.](image)

**Figure 6.** The dependence on the gas pressure of the intensity of lines of argon atoms and ions in a microwave discharge.
The dependence of the intensity of spectral lines on the pressure of the working gas was also investigated. The result, shown in figure 6, shows that the intensity increases rapidly with increasing pressure and after reaching a pressure of about 10 Pa, the intensity of the lines (i.e., plasma concentration) remains almost constant over the entire pressure range studied.

Figure 7. The intensity of the brightest spectral lines of argon in different sections of the working volume (a) the temperature of the electrons in these sections, estimated from the ratio of the intensities of the spectral lines (b). The coordinates $x_i$ correspond to the positions of the fiber shown in figure 5. $P = 30$ Pa.

The emission spectrum of argon plasma was recorded in sections of the plasma column located at different distances from the microwave emitter. The measurement results shown in figure 7 show that the intensity of the brightest argon lines near the all of the working chamber closest to the microwave emitter (the position of the optical fiber entrance $x_1$ in figure 5) is approximately 1.5 times the intensity of the lines near the far wall (the position $x_3$) and more than 3 times the intensity of the lines in the center of the camera (the position $x_2$). From this it follows that for more efficient creation of modified layers in dielectric samples this process should be carried out at maximum pressure of the working gas.

3. Conclusions
- The results obtained show that emission spectroscopy allows one to obtain the spatial distribution of the most important parameters, namely, the concentration and electron temperature, of a low-temperature plasma with a small degree of ionization. These data make it possible to optimize methods for the synthesis of composite layers consisting of a dielectric matrix and metal nanoclusters or radiation-stimulated defects.
- It can be concluded that, in the given experimental conditions, the methods for estimating the temperature of excitation of plasma components by the relative intensity of spectral lines in the framework of the LTE model and the coronal model give close results.

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