Investigation of the emission spectra of a high-power impulse gas discharge of a magnetron during reactive sputtering

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Abstract. This paper presents the results of a study of the properties of a gas discharge of a magnetron during sputtering at a direct current (DCMS) and by high-power impulse method (HiPIMS). During the study, the copper target was sputtered in a mixture of argon and oxygen. It was found that, depending on the discharge parameters, sputtering is possible in two modes: oxide, during which deposition of oxide films occurs, and metal, in which deposition of metal structures is possible. As a result of the study, current-voltage characteristics, emission spectra of the gas discharge, as well as the dependence of the pressure in the vacuum chamber on the discharge current were obtained.

1. Introduction
Thin films are of great scientific interest and are used in many industrial areas. They can be used as protective coatings, metal tracks in microelectronic devices, solar batteries, photovoltaic cells and many others. The deposition of thin metal and oxide structures with high density, adhesion to the substrate, crystallinity and low roughness is an important task in the production of complex high-tech equipment [1–2]. One of the most promising methods for deposition of coatings, which makes it possible to achieve all of the above properties, is the high-power pulsed magnetron sputtering method (HiPIMS). Further study of this method will make it possible to achieve a significant increase in the quality of thin-film coatings relative to traditional methods for their deposition (direct current spraying (DCMS), cathode sputtering, arc deposition), and also contribute to accelerating its full-scale introduction into industrial production [3–5].

A feature of HiPIMS technology is the use of short current pulses, due to which a short-term highest power density is achieved without significant heating of the target. Due to this, in the sputtering process, up to 80% of the atomized atoms of the target can be ionized, which has a significant effect on the properties of the deposited coatings [6–7]. In the course of this work, a series of experiments was carried out, the pulse repetition rate in which was 200, 400 and 600 Hz, and their durations were 50, 150 and 250 μs. The study of high-power pulsed gas discharge requires an integrated approach, therefore, during the work, the influence of the parameters set by the power supply unit on the composition of the gas discharge and the pressure in the vacuum chamber was investigated. In order to most accurately determine these parameters for a gas discharge, the following
dependencies were obtained: current-voltage characteristics, emission spectra, and the dependence of pressure in a vacuum chamber on the average discharge current.

2. Experimental equipment
In the study a modernized vacuum unit UVN-71 (M) with diffusion steam oil pumping was used. The volume of the vacuum chamber was 0.15 m³. The pressure of the gas in the vacuum chamber during the experiments did not exceed 10⁻⁵ Torr. The purity of the gases used (argon and oxygen) was at the level of 99.99%. The copper target had a purity of 99.98%. The base pressure for argon was 1 mTorr. The area of the sputtered area was 31 cm². APEL-M-5HPP-1200 power supply system was used, due to which sputtering was possible both in HiPIMS and DCMS modes.

3. Optical emission spectroscopy of a gas discharge
Determining the composition of the gas discharge during sputtering in a high-power impulse mode is an extremely important task. As already mentioned above, a distinctive feature of this sputtering method is the presence in the gas discharge of a high proportion of ionized material of the sputtered target due to the application of short pulses with high voltage and current amplitudes to the cathode. Optical emission spectroscopy of the discharge makes it possible to obtain data on the ratio of the various components of a gas discharge using the contactless method, based on the intensities of the spectral lines corresponding to the ions and atoms of the substances in the discharge. In addition, the method of optical emission spectroscopy allows to obtain data in real time because of the high working rate of the measuring instrument. At the same time, taking into account the repetition rate of high-power pulses, it is necessary to understand that the spectral pattern is integral. With the use of emission spectroscopy, it is possible to accurately track the moment of transition of the discharge into a high-power mode, at which the ionized atoms of the target acquire the greatest intensity. This method also allows to detect the transition from the oxide sputtering mode to the metal one.

During the study, the copper target was sputtered in the mixture of argon and oxygen. The flow of oxygen into the chamber was 15 and 30 ml/min. Figure 1 shows the dependence of the intensities of the spectral lines on the discharge current. The flow of oxygen into the chamber was 15 ml/min (figure 1(a)) and 30 ml/min (figure 1(b)). In both cases, the pulse repetition frequency was 600 Hz, and their duration was 250 μs.

**Figure 1.** The intensity dependences of the spectral lines of copper atoms and ions and oxygen ions on the average discharge current for oxygen flows of 15 (a) and 30 (b) ml/min.

The dependences presented in figure 1 show that at low current values the intensity of the lines of neutral copper atoms exceeds the intensity of its ion lines, which indicates that sputtering is conducted in a mode similar in its properties to sputtering at a direct current. With a further increase in the average discharge current, the intensity of the lines is redistributed in favor of the ions of the target material, which is a sign of the transition of the deposition system to the HiPIMS mode.
From the shown above dependences of the intensities of the spectral lines, it is also possible to determine the transition point from the oxide to the metal sputtering mode. It can be seen that initially, despite the measurements were taken in the cathode region, oxygen lines have the highest intensity, which indicates that the target surface is covered with an oxide layer. With a further increase in the discharge current, a decrease in the intensity of oxygen lines and the beginning of active growth of copper lines occurs, from which it can be concluded that the system is in the metal sputtering mode. It can also be seen that with a stream of oxygen of 30 ml/min (figure 1(b)), this transition occurs a little later than with a stream of 15 ml/min.

Figure 2 shows the emission spectra of a gas discharge during sputtering in a high-power impulse mode and at a constant current. The data of the spectral characteristics clearly show that during sputtering in the HiPIMS mode, the lines of metal ions are most intense, while in the DCMS mode, the lines of neutral atoms prevail. This clearly demonstrates the difference in the composition of the gas discharge for HiPIMS and DCMS modes.

Figure 2. Spectral characteristic of the gas discharge during HiPIMS (a) and DCMS (b).

4. The analysis of a discharge through gases on a current–voltage characteristic

Current-voltage characteristic is extremely important when studying a gas discharge. It establishes a connection between the two main characteristics that influence the entire process of magnetron sputtering. In addition, it allows you to accurately determine the boundaries between the oxide, transition and metal modes during sputtering in a reactive gas environment.

Figure 3 shows the current-voltage characteristics of a gas discharge during high-power impulse sputtering in the mixture of argon and oxygen (a) and the dependence of the total pressure of oxygen and argon in the vacuum chamber on the discharge current (b). Sputtering is carried out at a frequency of 600 Hz, pulse duration of 250 μs and oxygen flow into the chamber of 15 ml/min. Two modes of sputtering can be clearly seen from the discharge current-voltage characteristic (figure 3(a)): oxide (the target surface is coated with oxide film and copper oxide is deposited on the substrate surface) and metal (pure target material sputtering). The transition between these modes occurs at ~ 1 A and is accompanied by a noticeable change in the course of the current-voltage characteristic. To confirm the data obtained on the basis of the current-voltage characteristics of the discharge in figure 3(b), the dependence of the gas pressure in the vacuum chamber on the discharge current is also presented. It can be seen that as the discharge current increases, a gradual decrease in pressure occurs with further stabilization of this parameter (on the level of base pressure of argon of 1 mTorr) at a current of ~ 1.2 A. This is due to the fact that with an increase in discharge current, an increasing number of oxygen particles turns out to be sorbed by atoms and ions of the target material, and the establishment of a constant pressure at a discharge current of ~ 1 A mentions transition of the sputtering system to a fully metallic operation mode.
Figure 3. Current-voltage characteristic of HiPIMS discharge during deposition the mixture of argon and oxygen (a); dependence of the total pressure in the vacuum chamber on the average discharge current (b).

The mentioned above findings are also fully confirmed by the results of optical emission spectroscopy. Figure 4 shows the dependence of the intensities of the lines of oxygen atoms on the current. The intensity of the oxygen line slowly decreases up to discharge current of ~1 A, and then a more abrupt decrease begins. This indicates the transition of the system from the oxide sputtering regime to the metallic one.

Figure 4. The dependence of the radiation intensity on the average discharge current for the spectral lines of oxygen.

5. Conclusion
In the course of the work, a comprehensive study of the properties of the gas discharge of a magnetron during high-power impulse sputtering in a mixture of argon and oxygen gases was made. A number of results was obtained such as current-voltage characteristics of the gas discharge, its emission spectra, as well as the dependence of pressure in the vacuum chamber on the average discharge current.

Based on the dependencies obtained, the capabilities of the sputtering system in the metal and oxide modes were determined depending on the parameters of the power supply unit and the gas pressure in the vacuum chamber. It was also found that even at a high-power impulse mode at low values of the average current, the power density is insufficient for an effective ionization process of target atoms, and the operation of the magnetron system is similar in its properties to sputtering at a direct current. This suggests that when spraying in a high-power pulsed mode, it is necessary to strictly monitor the parameters of a gas discharge in order to achieve the maximum possible ion concentration of the target material in the gas discharge.
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