Comparison of plasma parameters and optical emission in DC, HIPIMS and hybrid DC+HIPIMS modes of magnetron sputtering

V A Semenov, A S Grenadyorov, V O Oskirko, A N Zakharov, S V Rabotkin, I V Ionov and A A Solovyev
Institute of High Current Electronics SB RAS, 2/3 Akademichesky Ave., Tomsk, 634055, Russia
E-mail: semenofvjacheslav@gmail.com

Abstract. Experimental studies of the parameters of the plasma of a magnetron discharge and optical radiation from plasma in the regime of direct current (DC), high-current mode (HIPIMS), and hybrid (DC+HIPIMS) mode for a copper cathode 100 mm in diameter were carried out. The hybrid mode of operation of the magnetron sputtering system was carried out by means of a matching unit of two power supplies APEL-M-5HIPIMS and APEL-M-5PDC. The results of the experiments showed that the plasma concentration at a distance of 120 mm from the cathode has maximum values ($5.6 \times 10^{11}$ cm$^{-3}$) in the HIPIMS regime at a frequency of 500 Hz. Measurement of the optical emission spectra for a copper cathode in an argon plasma showed, that at the same average discharge power (500 W), the intensity of the copper ion lines in the high-current and hybrid modes is 3 and 1.5 times, respectively, more than in the DC regime.

1. Introduction
The high-current mode of magnetron discharge (HIPIMS) is realized in the case of application to the cathode of a magnetron sputtering system of high-current pulses with duration of tens or hundreds of μs with a frequency from several Hz to kHz. In this discharge, the maximum power density during the pulse can reach 3 kW·cm$^{-2}$ with low average power. The HIPIMS mode allows generate plasma in which the degree of ionization of the atomized substance can reach 90% [1]. For the first time this mode magnetron sputtering has been described by Kuznetsov and others in 1999 [2]. The plasma density at HIPIMS can exceed the values typical for a magnetron discharge at a direct current (DC) by three orders of magnitude and reach $10^{13}$ cm$^{-3}$[3]. High plasma density leads to ionization of the atomized material, which leads to a higher ion flux density on the substrate. This allows obtain coatings with unique structural, optical and other properties. The disadvantage of this method is the reduction in the deposition rate of coatings compared to DC-mode. This is due to the attraction of the ionized atomized material back to the cathode [4]. The greater the degree of ionization of the sprayed material, the slower the deposition rate of the coatings. The quality of the formed coatings which have insufficient density due to the low plasma concentration ($10^9$–$10^{10}$ cm$^{-3}$) is a disadvantage of spraying in DC mode.

A combination of DC-mode and pulse high-current mode (DC+HIPIMS) is proposed for the formation of high-quality coatings without reducing the deposition rate [5]. The resulting properties of
films deposited by this method are a combination of the properties of films deposited in DC and HIPIMS modes [6].

In this work, the parameters of plasma and optical radiation from plasma in the hybrid mode DC+HIPIMS were studied. The parameters of plasma and optical emission in different modes of magnetron sputtering are investigated.

2. Methodology and materials
Experiments were carried out on the installation for magnetron deposition of NNV-6 coatings. The scheme of the experimental setup is shown in figure 1. The structure of the installation includes an unbalanced magnetron sputtering system (MSS) APEL-MRE-100 with a round flat Cu cathode with a diameter of 100 mm. The inner walls of the chamber serve as an anode in the gas-discharge system. Argon was used as the working gas, the working pressure in the chamber was maintained at 0.3 Pa during the experiment. To implement HIPIMS-mode, a switching power supply source APEL-M-5HIPIMS was used, which provides an average output power of up to 5 kW and a voltage in the range from 300 to 1000 V. To excite the magnetron discharge in DC mode, a power supply source APEL-M-5PDC was used, providing an average output power of up to 5 kW, a voltage in the range from 300 to 650 V. Hybrid mode of the discharge (DC+HIPIMS) was provided by the communication unit.

![Figure 1. Scheme of experimental unit for measurement of plasma parameters in DC, HIPIMS and DC+HIPIMS modes.](image)

A Langmuir probe made of nichrome wire with a diameter of 0.7 mm and a working surface length of 5 mm was used to determine the plasma parameters. The probe was located at a distance of 120 mm from the cathode surface. To remove the probe characteristics, a laboratory constant voltage source B5-50 was used, which provided the electric potential of the probe displacement from -100 to +100 V.

Optical emission parameters were obtained using Avantes AvaSpec-2048 spectrophotometer with a wave measurement range from 200 to 750 nm.

The surface morphology of the films was investigated by a QUANTA 200 scanning electron microscope.
Table 1. Characteristics of magnetron sputtering modes.

| Sputtering mode                          | Maximum discharge voltage and current |
|------------------------------------------|---------------------------------------|
|                                           | $U_{\text{HIPIMS}}$ (V) | $I_{\text{HIPIMS}}$ (A) | $U_{\text{DC}}$ (V) | $I_{\text{DC}}$ (A) |
| DC 500 W                                 | -                          | -                      | 332                | 1.55               |
| HIPIMS 500 W, 500 Hz                     | 460                        | 40                     | -                  | -                  |
| DC 250 W+HIPIMS 250 W, 500 Hz            | 423                        | 20                     | 304                | 0.83               |

3. Results and Discussion

3.1. Process characteristics

At the first stage, the current-voltage characteristics of the magnetron discharge were measured in different modes of operation of the magnetron sputtering system at different pressures of the working gas Ar in the vacuum chamber. Figure 2 shows the current-voltage characteristics obtained in DC, HIPIMS and hybrid mode DC+HIPIMS. The magnetron sputtering system operated stably in DC mode with a minimum working gas pressure of 0.12 Pa (figure 2a).

With an increase in the working gas pressure to 0.4 Pa, the ignition voltage of the magnetron discharge decreases from 350 to 270 V. the discharge Power in DC mode at a working gas pressure of
0.3 Pa was 3.3 kW. This corresponds to the power density at the cathode 42 W·cm⁻². The maximum DC discharge current at a working gas pressure of 0.3 Pa was 7.2 A (figure 2a).

Figure 2b shows the current-voltage characteristics obtained in the high-current mode of magnetron sputtering with a pulse repetition rate of 500 Hz and a pulse duration of 100 µs. To construct the current-voltage characteristics in HIPIMS mode, the maximum values of the pulse current were used. At a pressure of 0.3 Pa, the value of the pulse discharge current was 270 A. This corresponds to the average discharge power of 3 kW and the pulse power density of 2 kW·cm⁻². For a pressure of 0.12 Pa, the maximum pulse discharge current was 300 A. For the reduced pressure range, the maximum pulse current was limited by the transition of the discharge to the arc stage. For volt-ampere characteristics of high-current discharge (figure 2b) it is possible to allocate two plots with different growth rate of the current. In the first section (from 0 to 50 A), the current increase is due to the ionization of the working gas (Ar). The second section (from 50 A and above) characterizes the so-called self-spraying mode [7], where with a slight increase in voltage, the current changes by tens of amperes. In this type of discharge, the atoms of the cathode material are ionized. Current-voltage characteristics for the hybrid mode sputtering was obtained at a fixed power DC-discharge, and voltage change of combustion of high-current discharge. Figure 2c shows the current-voltage characteristics for the HIPIMS, 500 Hz, 100 µs + DC 250 W. As can be seen from the current-voltage characteristics, at a working gas pressures of 0.12 and 0.2 Pa there is no transition to the self-spray mode. This is due to the fact that at a low pressure of the working gas, with an increase in the discharge voltage, breakdowns appear at the cathode and the discharge passes into an arc. The maximum pulse discharge current in the hybrid mode at a working gas pressure of 0.3 Pa is 250 A. The Maximum pulse power density on the cathode surface was 1.8 kW·cm⁻². Further studies of the plasma parameters were carried out at a pressure of Ar of 0.3 Pa, since at this pressure in all the studied modes stable operation of the magnetron sputtering system is observed. At the same time, at a pressure of 0.4 Pa, a larger current is observed at a lower voltage for all the studied modes, but at this pressure the vacuum pumping system undergoes large loads.

3.2. Plasma parameters

A Langmuir probe made of nichrome wire with a diameter of 0.7 mm and a working surface length of 5 mm was used to determine the plasma parameters. The probe was located at a distance of 120 mm from the surface of the magnetron target. Figure 3 shows the probe characteristics for different modes of operation of the magnetron. For all high-current modes, the probe characteristics were measured at the maximum current per probe.
As can be seen from figure 3, the highest electron current per probe was obtained in HIPIMS mode, 500 W at a frequency of 100 Hz. The values of floating potential, plasma potential, ion and electron saturation currents, and electron temperature were obtained from the analysis of probe characteristics (table 2).

Table 2. Characteristics of plasma parameters in different magnetron sputtering modes.

| Sputtering mode          | Floating potential $U_f$ (V) | Plasma potential $U_p$ (V) | Electron temperature $T_e$ (eV) | Plasma concentration $n_0$ (cm$^{-3}$) |
|-------------------------|------------------------------|---------------------------|-------------------------------|---------------------------------------|
| DC 500 W                | -6.9                         | 2.7                       | 0.7                           | $6.7\times10^{10}$                    |
| HIPIMS 500 W, 500 Hz, 100 µs | -9.1                        | 6.4                       | 2.4                           | $7.5\times10^{11}$                    |
| DC 250 W+HIPIMS 250 W, 500 Hz, 100 µs | -14.7                       | 6.7                       | 1.8                           | $6.5\times10^{11}$                    |

Figure 4 shows the radial distribution of the plasma concentration at a distance of 120 mm from the target. When measuring the probe characteristics, the probe was moved from the center of the target to its edge with a step of 5 mm. The maximum plasma concentration was observed opposite the erosion groove of the target, the center of which was located at a distance of 30 mm from the axis of the magnetron sputtering system.

3.3. Optical emission
The optical emission parameters were obtained using the Avantes AvaSpec-2048 spectrometer with a wave measurement range from 200 to 750 nm. The optical sensor was installed in a vacuum chamber (figure 1) in place of the Langmuir probe. Figure 5 shows typical emission spectra of a magnetron discharge obtained in different modes of magnetron sputtering. Figure 5 shows that at the same average power the intensity of the spectral lines of copper atoms in the mode of HIPIMS 500 W, 500 Hz is about 3 times higher than in the DC mode. In a hybrid HIPIMS mode 250 W,
500 Hz+DC 250 W this intensity increases two times compared to the DC mode. Table 3 lists the wavelengths of high and low intensity spectral lines observed in the plasma generated in the argon atmosphere by a magnetron sputtering system with a copper cathode in the range of 200–750 nm. Table 3 also shows the excited neutral and ionized particles are identified using the database Nist Atomic Spectra Database [5].

![Radiation spectra from plasma obtained in different modes of magnetron sputtering.](image)

**Figure 5.** Radiation spectra from plasma obtained in different modes of magnetron sputtering.

In the wavelength range 505–535 nm in high-current and hybrid modes, the appearance of single-charge copper ions, which are absent in DC modes, is observed. Spectral lines of excited argon atoms are observed in the wavelength range 745–770 nm. At the same time, in the high-current mode, the intensity of argon lines is minimal, which is due to the self-spraying mode. In hybrid mode, the intensity practically coincides with the intensity in DC modes.
Table 3. Types of ions and spectral lines observed in a plasma generated in the argon atmosphere by a magnetron sputtering system with a copper cathode.

| Type   | Wavelength (nm)            |
|--------|----------------------------|
| Cu I   | 324.7; 327.4; 333.7; 353.0; 402.3; 427.5; 510.5; 515.3; 521.8; 570.0; 578.2 |
| Cu II  | 404.3; 416.1; 450.6; 455.6; 464.5; 470.2; 526.9; 529.2; 740.4 |
| Ar I   | 419.8; 603.2; 687.1; 696.5; 706.7; 738.4; 750.4; 751.5; 763.5; 772.4 |

* According to the accepted international spectral designations Cu I, Ar I correspond to the excited neutral atoms, Cu II – single-charge ions.

3.4. Surface morphology analysis

Scanning electron microscopy (SEM) analysis was performed in order to investigate the surface topography of 1-µm-thick Cu films deposited on Si substrates films in more details. The films surface morphology was in strong relationship with deposition mode. Film deposited in DC mode has the roughest surface as shown in figure 6a. Films deposited in hybrid mode show smoother, denser and less defective surface.

![Figure 6](image)

Figure 6. Surface SEM images of Cu films deposited in: (a) DC 500 W; (b) HIPIMS 500 W, 500 Hz; (c) DC 250 W+HIPIMS 250 W, 500 Hz sputtering modes.

In hybrid DC + HiPIMS mode plasma is more ionized. As a consequence, more ions bombard the surface of the growing films which enhances the mobility of adatoms. Therefore the surface morphology of the films deposited in hybrid mode is relatively denser than that of DC mode. Table 4 shows the deposition rates of copper films in different modes of magnetron sputtering.
Table 4. Deposition rate of copper films.

| Sputtering mode                        | Deposition rate (nm·min⁻¹) |
|----------------------------------------|----------------------------|
| DC 500 W                               | 130                        |
| HIPIMS 500 W, 500 Hz, 100 µs           | 75                         |
| DC 250 W+HIPIMS 250 W, 500 Hz, 100 µs | 110                        |

4. Conclusion
The density of electrons in HIPIMS and DC+HIPIMS modes is about 2 orders of magnitude higher than in magnetron sputtering at a constant current. According to optical measurements from plasma in DC mode and at the initial stages of high-current pulses in HIPIMS and DC+HIPIMS modes, excited atoms and Ar⁺ ions dominate in the discharge. During a high-current pulse in the plasma, the number of metal ions increases and the discharge enters the self-sputtering mode. The smoothest surface was observed in copper films obtained in the mode of HIPIMS 500 W, 500 Hz, 100 µs. Spraying speed in high-current mode is 2 times lower than in DC mode. The speed of spraying in hybrid mode is close to the speed of spraying in DC mode and was 110 nm·min⁻¹.

Acknowledgements
This work was supported by RFBR (grant No. 18-32-00179).

References
[1] Bugaev S P, Koval N N, Sochugov N S and Zarharov A N 1996 Proc. XVII Intern. Symp. on Discharges and Electrical Insulation in Vacuum (Berkeley, USA) 1074
[2] Kouznetsov V, Macák K, Schneider J M, Helmersson U and Petrov I 1999 Surf. Coat. Technol 122 290
[3] Odivanova A N, Sochugov N S, Oskomov K V and Podkovyrov V G 2011 Plasma Physics Reports 37 239
[4] Christie D J 2005 J. Vac. Sci. Technol. 23 330
[5] Bobzin K, Brogelmann T, Kruppe N C and Engels M 2016 Thin Solid Films 620 188
[6] Hashmi S 2014 Comprehensive Materials Processing.Elsevier 4 5634
[7] Huo C, Lundin D, Raadu M A, Anders A, Gudmundsson T and Brenning N 2014 Plasma Sources Science and Technology 23 0250175
[8] Kramida A, Ralchenko, Yu, Reader J and NIST ASD Team 2019 National Institute of Standards and Technology 5.6.1