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Novel methods for synthesizing high-quality thin films through short and ultrashort high-power pulsed magnetron sputtering

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Abstract. The present study focuses on investigating the increase in average ion current density to the substrate in short and ultra-short high-power impulse magnetron sputtering (HiPIMS). Theoretical and experimental evidence demonstrates that, while maintaining the average power level of HiPIMS, the ultra-short mode enables a more than threefold increase in the ion current density of the target material onto the substrate. These findings hold promise for enhancing the quality of HiPIMS ion-plasma vapor deposition (IPVD) coatings.

Keywords: high-power impulse magnetron sputtering, plasma vapor deposition, thin films

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Материалы конференции
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Новые методы синтеза высококачественных тонких пленок с помощью короткого и ультракороткого мощного импульсного магнетронного распыления

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Аннотация. Данная статья посвящена исследованию повышения средней плотности тока ионов на подложку при коротком и сверхкоротком импульсном магнетронном распылении с высокой мощностью (HiPIMS). Теоретические и экспериментальные данные показывают, что при сохранении среднего значения мощности HiPIMS сверхкороткий режим обеспечивает более чем трёхкратное увеличение плотности тока ионов материала мишени на подложку. Полученные результаты обладают перспективами для повышения качества покрытий HiPIMS IPVD методом осаждения.

Ключевые слова: мощное импульсное магнетронное напыление, плазменное напыление, тонкие пленки

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Introduction

High-power impulse magnetron sputtering (HiPIMS) represents a contemporary and technologically advanced physical vapor deposition technique [1]. It is known as pulsed-periodic modification of a direct current magnetron sputtering (dcMS). Characteristic features of HiPIMS are low duty cycle and high peak discharge current. These electrophysical HiPIMS properties greatly improve the ionization of the target material and the ion deposition contribution to the coating growth process (IPVD). The latter improves the formation of high-density coatings with good adhesive quality. It also allows the coating crystallization control, phase composition, and microstructure enhancement. However, HiPIMS is accompanied by a substantial reduction (up to 75%) in the deposition rate, which serves as a trade-off for achieving superior coating quality [1].

In [2], by reducing the duration of HiPIMS pulses from 100 to 10 μs, a twofold increase in the average ion current density to the substrate was achieved. Furthermore, in [3], reducing the pulse duration to 3–5 μs led to a threefold increase in the average ion current density on the substrate at a constant current amplitude and average discharge power. Theoretical estimations been identified the main reason for the average substrate ion current density growth, which lies in the dissipation of plasma ionization region (IR) after the discharge pulse (afterglow phase). As a result of the elevated discharge current during the HiPIMS pulse, a substantial accumulation of metal and gas ions occurs, effectively confined within an electrically-induced “recirculation trap”. After the pulse ends and the target potential reaches zero, the ions leave the trap, transport toward the substrate, and provide a higher average ion current density. In the case of pulse duration reduction, the pulse repetition frequency significantly increases at a constant average discharge power. Along with the pulse frequency, the number of ion accumulation acts in the “recirculation trap” and their release/transportation to the substrate also increases.

The present investigation encompasses both theoretical and experimental results about to the augmentation of the average ion current density on the substrate. This augmentation occurs during the transition from the upper threshold of pulse durations in the voltage regime ~ 40 μs (s HiPIMS) to significantly shorter pulse durations ~ 4 μs (us-HiPIMS), while maintaining a constant average discharge power level of $P_{d.avg.} = 1$ kW.

Materials and Methods

To theoretically model the processes leading to an increase in ion current to the substrate in pulsed magnetron discharge under fixed average discharge power, the Ionization Region Model (IRM) of HiPIMS was employed. The IRM is a comprehensive plasma-chemical model that describes the averaged values of electron, ion, and neutral particle concentrations, as well as the temperature of “cold” electrons, i.e., electrons resulting from electron collision processes, within the ionization region (IR). The temporal evolution of the concentration and temperature parameters is determined by a set of ordinary first-order differential equations, which represent the time derivatives of electron energy and the densities of all considered particles within the IR [1].

The afterglow phase represents a crucial stage in the discharge process, whose significance increases as the pulse duration decreases. After the voltage pulse ends, the system assumes a floating potential, resulting in the disappearance of the electric field that predominantly returns ions from the ionization region (IR) back to the target. This leads to a redistribution of ion and neutral particle fluxes, directed towards both the target and the substrate. The IRM incorporates the attraction of ionized sputtered particles with a reverse attraction probability denoted as $\beta \sim 0.9$. During the pulse, the ion flux into the diffusion region (DR) is calculated based on the flux towards the racetrack. It is assumed that sputtered particles possess a directed velocity away from the target. After the voltage pulse is switched off, ions acquire velocities close to those of the sputtered metal particles. Therefore, in our modeling approach, we assume that β during the afterglow phase is close to zero [4].

In this study, the IRM employs an advanced plasma-chemical model [5] where the plasma-chemical reaction constants for argon-copper plasma are determined through the electron temperature, which is consistently calculated as a solution to the system of equations governing mass and energy balance [1].

Fig. 1 depicts a schematic representation of the experimental configuration showcasing the short and ultra-short pulse HiPIMS system. The system comprises a vacuum chamber equipped with a gas pumping system, a magnetron sputtering system, a magnetron power supply, and a measurement system for capturing the electrical parameters of the plasma and its radiation.

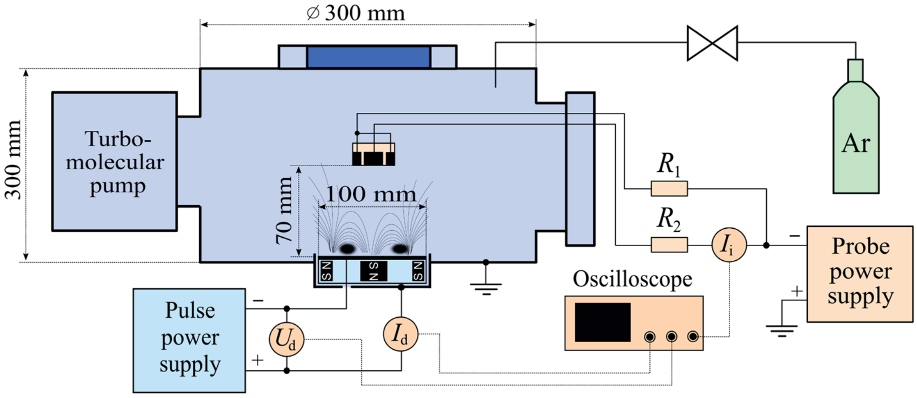

Fig. 1. Schematic of s/us-HiPIMS experimental setup

The magnetron employed in this study features an unbalanced magnetic field configuration and utilizes a copper target with a diameter of 100 mm. The magnetic field strength on the cathode surface measures B = 730 G, while the geometrical unbalance coefficient, K_G = 1.2. The inner walls of the vacuum chamber serve as the anode for the discharge system. The pulsed power supply for the magnetron was chosen to be APEL-M-5HPP-1500U (Applied Electronics Ltd, Tomsk, Russia) providing rectangular voltage and triangular current pulsed with sharp edges. The experiments have been held for the argon pressure of 0.15 Pa. To sustain the average power at 1 kW keeping also 150 A current peak while reducing the voltage pulse t_i from 40 μs to 3.5 μs we have adjusted the operating frequency as well as voltage peak amplitude (Fig. 2). In the IRM calculations the current and voltage pulse waveforms were closely alighted to the experimental data.

To measure the total ion current density, a probe with a protective ring has been used in experiments, which was maintained at a constant negative bias voltage of -100 V. Within the framework of the IRM, the current densities of three types of ions, namely Ar^+, Cu^+, and Cu^{++}, were computed in the direct calculations. Both in theory and in experiments, the oscillograms of the total ion current density were averaged over the power supply voltage period.

Results and Discussion

Fig. 3 shows key experimental and theoretical results of this paper. In Fig. 3, *a* the temporal profiles of the electric charge delivered by the ion current to the probe are depicted. It illustrates the transfer of the electric charge to the probe during the discharge current pulse (0–50 μs) and the subsequent afterglow stage (50–140 μs). At t_i = 39 μs, approximately one third of the total charge is transferred to the probe during the discharge current pulse, while the remaining charge is transferred during the afterglow stage. When the pulse duration is reduced to 3.5 μs, the total charge transfer during the pulse period decreases by approximately 2.5 times. Consequently, the entire charge is transferred to the probe during the afterglow stage, as the charge flowing during the discharge current pulse diminishes to zero.

In Fig. 3, *b*, the temporal profiles of the average ion current density on the probe during the discharge current pulse and afterglow stage are shown. The average ion current density, $j_{sb.avg}$,

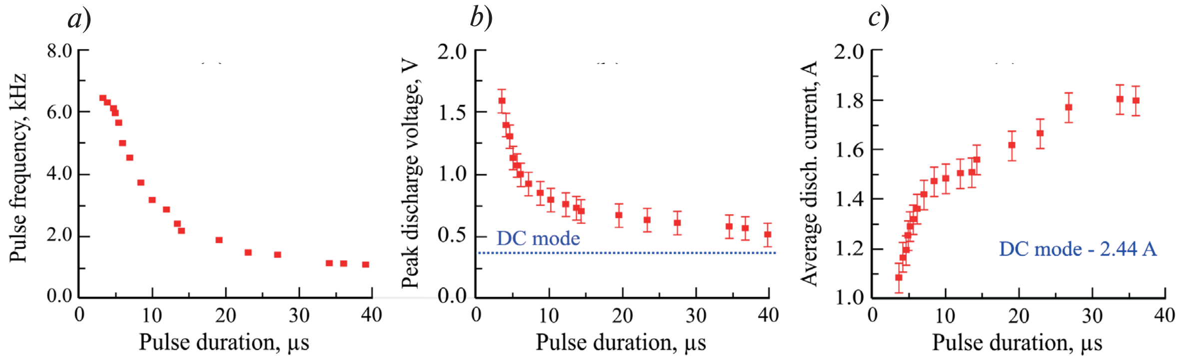

Fig. 2. Time-dependencies of pulse frequency (*a*), peak voltage (*b*), and average discharge current (*c*) provided by pulsed power voltage supply

exhibits an increase during the afterglow stage due to the current flow. Conversely, $j_{sb.avg}$ during the discharge current pulse is zero when the pulse duration is shortened. As a result, a larger fraction of current flows to the probe during the afterglow stage. Despite the decrease in charge during the pulse duration, the ion current density on the probe continues to rise. This is attributed to the fact that the average ion current density is determined by the product of the charge generated by each pulse and the pulse frequency. When the pulse duration is reduced, the pulse frequency increases by approximately sevenfold (as shown in Fig. 2, *a*), while the charge decreases by 2.5 times. Consequently, the increased frequency not only compensates for the lower charge generated by each pulse but also leads to a roughly threefold increase in the average ion current.

Finally, Fig. 3, *c* presents the theoretical and experimental relationships between the average ion flux to the substrate and the duration of the voltage pulse. The theoretical calculations exhibit good agreement with the experimental data, confirming the observed trend of increasing average ion current density during the transition from normal pulse durations (50–500 μs) to s-HiPIMS (< 50 μs) and us-HiPIMS (< 5 μs) pulses while maintaining a constant average discharge power. To maintain a stable discharge power as the voltage pulse duration decreases, a significant increase in pulse frequency is required, while the characteristic decay time of the plasma in the afterglow remains unchanged.

Fig. 3, *c* also shows the behavior of the average ion current density on the substrate in the us HiPiMS mode. It is evident that as the discharge voltage pulse time is progressively shortened, there is a marginal decrease in the average ion current density. Nevertheless, this decrease remains insignificant when compared to the maximum average ion current density achieved at t_i ~ 5 μs. This phenomenon can be attributed to the fact that, even at higher discharge voltages, the reduction in pulse duration ultimately diminishes the effective duration of high-density plasma generation. Although the decrease in plasma number density, particularly in the Cu$^+$ component,

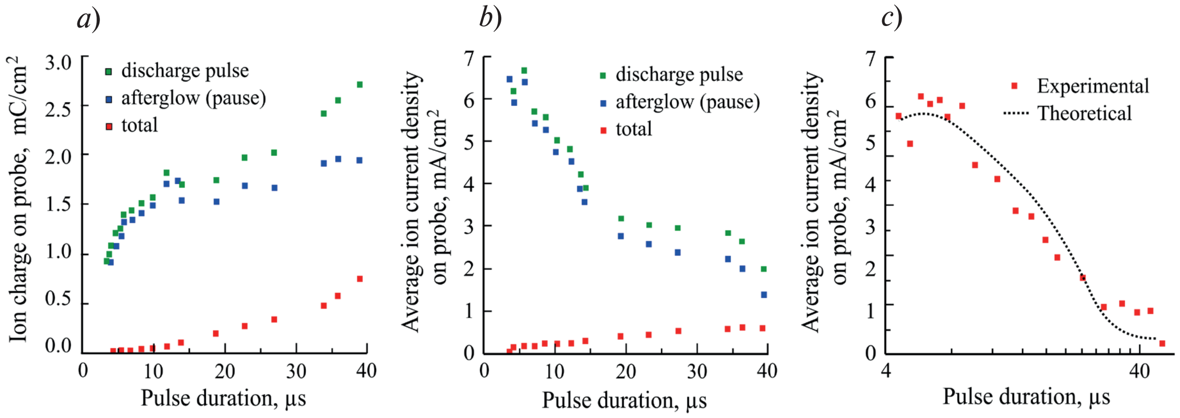

Fig. 3. Period-averaged s/us-HiPIMS characteristics from the pulse duration: charge transferred to the probe during the pulse interval and afterglow stage (experimental) (*a*), average ion current density on the probe during the discharge current pulse, afterglow stage, and their combined influence (experimental) (*b*), total average current density to the probe (experiment vs. theory) (*c*)

does not surpass an order of magnitude, it does contribute to the observed reduction in average ion current density on the substrate, as reported in several experimental studies.

Conclusions

This study has demonstrated unequivocally that reducing the pulse duration during Cu target magnetron sputtering, while maintaining a constant average discharge power and peak current, results in a substantial increase in ion flux onto the substrate. The findings are based on an experimental investigation of ion current density dynamics and theoretical calculations utilizing an ionization region model to analyze the behavior of neutral and charged plasma particles. The analysis reveals that, during the afterglow stage, the ion current density is enhanced as the ions generated in the circulation trap during the pulse are directed toward the substrate. By optimizing parameters such as pulse duration, peak current, and pulse frequency, a remarkable augmentation in ion current density on the substrate is achieved, surpassing that of MFMS, DCMS, and HiPIMS modes, the latter employing longer average pulses (> 50 μs).

In our assessment, the utilization of pulses ranging from 8 to 15 μs appears to be the most favorable approach. These pulses yield a substantial increase in ion current density, approximately 2 to 3 times higher, while concurrently maintaining a higher deposition rate and a discharge voltage below 1000 V. These criteria hold significant importance for system performance and equipment complexity, particularly concerning the power supply.

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