Dynamic transition in the discharge current between gas-dominant discharge and self-sputtering in high-power impulse magnetron sputtering

Zhongzhen Wu a,⁎, Shu Xiao a, Zhengyong Ma a, Suihan Cui a, Feng Pan a, Xiubo Tian a,b, Ricky K.Y. Fu c,⁎, Paul K. Chu c

a School of Advanced Materials, Peking University Shenzhen Graduate School, Shenzhen 518055, China
b State Key Laboratory of Advanced Welding and Joining, Harbin Institute of Technology, Harbin 150001, China
c Department of Physics and Materials Science, City University of Hong Kong, Tat Chee Avenue, Kowloon, Hong Kong, China

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1. Introduction

In high-power impulse magnetron sputtering (HiPIMS), unipolar pulses with a low duty cycle and up to 10 kW/cm² momentary power density [1,2] produce a larger plasma density [3] to boost the ionization efficiency of the sputtered species [4]. By controlling the energy and impact angle of the ionized materials to the substrate, HiPIMS provides greater flexibility in microstructure engineering and better film quality than conventional direct-current (DC) magnetron sputtering (MS) [5–7]. During deposition, a stable discharge current is a key factor. The typical current waveform during pulsing in HiPIMS shows three distinct regimes: (I) Plasma initiation and a current maximum, (II) a transition in which the current decays to a lower-current steady-state regime, and (III) a regime in which the discharge voltage is stable. With regard to materials such as Cr [9], the HiPIMS curve is characterized by two power law fits, \(I_D = U_D^n[10]\), in \(I_D\) and \(U_D\) are the time-dependent discharge current and voltage measured at the cathode, and simultaneously measured at the peak current [11]. The two fits intersect at a kink for a certain current density marking the transition from the DC-like discharge to a self-sputtering dominant regime with higher discharge impedance [12,13].

However, they have been inferred from stable waveform data which are usually averaged on the oscilloscope. In fact, careful examination of the instantaneous curves reveals an unstable transition between the two stages and in this work, the unstable discharge phenomenon and formation mechanism are investigated.

2. Experimental

The experiments were performed in a turbo-molecular pumped vacuum chamber with a diameter of 40 cm and height of 40 cm and the base pressure is \(3 \times 10^{-3}\) Pa. The working gas (99.9997% pure Ar) is introduced through a leak valve. The magnetron cathode was a Cr target (ϕ50 mm × 6 mm) and powered by a hybrid pulsed power supply developed in our laboratory. The discharge pulse width and frequency were 200 μs and 100 Hz, respectively and the working pressure was kept at 0.5 Pa. A digital oscilloscope was used to record all the instantaneous curves without any fitting and the average currents were derived by the integration division by the pulse width.
3. Results and discussion

Fig. 1 shows the three stages in the instantaneous discharge current curves as the discharge voltage increases. When the discharge voltage is < 740 V, the discharge is weak and the current increases with discharge voltage. The current curves of each pulse overlap when the voltage is fixed, as shown in the Fig. 1a). They are stable and show gas dominant discharge characteristics [14]. When the voltage is between 740 V and 800 V, the current curves at different pulses begin to change. The instantaneous current curves acquired at four random pulses with discharge voltages of 770 V and 780 V are depicted in (Fig. 1b and c) and significant differences can be observed. The current curves exhibit a self-sputtering dominance in and out of this discharge stage [14]. When the voltage is over 800 V, the current curves are stable again. The current curves obtained for different pulses are consistent with each other and the discharge is dominated by self-sputtering [14, 15].

It has been documented that the conventional magnetron $I-U$ characteristics follow a power law: $I = kU^n$. The exponent, $n$, is typically in the range 5–15 [16] and small when the discharge occurs at high current density, such as that in HiPIMS [12]. The average values of the discharge currents are presented in Fig. 2. The pulsed discharge exhibits two slopes depending on the target voltage. At low discharge voltage (< 740 V), the exponent, $n$, is approximately 6 indicating a gas-dominant discharge. As the discharge voltage is increased to over 780 V, the discharge becomes self-sputtering and the exponent, $n$, decreased to about 3. When the discharge voltage is between 740 V and 780 V, the current fluctuates revealing a dynamic transition from the gas-dominant discharge to self-sputtering one.

In the gas-dominant discharge, the plasma density is small and the amount of metallic atoms is much less than that of gas atoms. In the dynamic transition, the discharge is unstable in that both of them are not suitable for deposition. Owing to the high density discharge and large metallic atom contents, only the self-sputtering dominant regime is useful to the deposition and the threshold voltages of the stable self-sputtering regime is important. Fig. 3 shows the threshold voltages versus pressure and hybrid DC current. When the pressure is low, the threshold voltage of the stable self-sputtering discharge is large and it decreases monotonically with increasing pressure reaching a stable stage at a pressure above 1.0 Pa. When a hybrid DC current is applied, the threshold voltage of the stable self-sputtering discharge is smaller and decreases linearly with increasing current. Compared to pressure, the influence of hybrid DC current on the threshold voltage is smaller. The results suggest that both the pressure and hybrid DC current affect the stable discharge threshold and consequently deposition effectiveness.

All the voltage pulses are the same during the discharge process and hence, the discharge current curves should be the same in principle. However, considering the real situation, the discharge voltage consists of a series of pulses and the discharge triggered by the later pulses is influenced by the previous one resulting in the different discharge current curves. A theoretical expression of the discharge current slope and the six modes in the HiPIMS current curves have been proposed [17]. The current curves in the gas dominant and self-sputtering regimes belong to mode V and mode III, respectively. The average secondary electrons emission coefficient $\gamma$ and ratio of the ions lost by diffusion and
recombination with ions not returning to the target $\varepsilon$ determine the current waveforms[17]. In HiPIMS, the frequency and duration of the pulse are small and the off-time between two consecutive pulses is sustained for many milliseconds or even dozens of milliseconds, which are long enough for diffusion and recombination in the discharge plasma resulting in completely disappearance before the next pulse[18,19]. Hence, $\gamma$ does not change in the discharge of each voltage pulse. However, $\varepsilon$, the determining factor (temperature) is a slowly changing entity and it can’t cool down completely in the off-time between pulses and the temperature will rise producing different “initial” conditions of the discharge during each voltage pulse. That is, the previous pulse does affect the discharge of the later pulse due to the temperature thereby producing the dynamic transition in the discharge current curves between mode V (gas dominating) and mode III (self-sputtering dominating).

When the voltage is small, the discharge is weak and the temperature increase near the target is insufficient to alter the discharge of the next pulse. As a result, the current curves in the gas-dominant stage are stable and repeatable for each pulse indicative of mode V. As the voltage is increased, the discharge becomes more intense and self-sputtering begins to dominate. The plasma composition balance is reached before the current slope $\Phi = 0$ and the current curves represent Mode III[17] as shown in Fig. 4. Here, the discharge power rises sharply together with the temperature to improve rarefaction. The high temperature resulting from the previous pulse or last several pulses decreases the initial pressure near the target of the discharge prior to the new voltage pulse. The increase of $\varepsilon$ leads to $\Phi = 0$ ahead of the balance of the plasma composition and the current curve recovers to the configuration of Mode V. After the transition, the discharge becomes weak and the temperature goes down gradually. The initial pressure recovers to the original condition and the discharge reverts back to Mode III. The repetitive dynamic transition between Mode V and Mode III produces the oscillating as shown in Fig. 1. When the voltage is high enough, the discharge is intense and self-sputtering becomes dominant. Although the temperature still affects the initial pressure of the next pulse, the plasma composition balance is reached before the current slope $\Phi = 0$ and the temperature does not increase further since the system reaches a thermal balance. Hence, the current curves stabilize again and exhibit the configuration of Mode III.

Formation of the self-sputtering dominating regime is determined by whether the plasma composition reaches equilibrium before $\Phi = 0$ and it depends on the sputtering process $S(t)$ which is also related to the sputtering yield $Y$. The relationship can be expressed as follows:[20]

$$S(t) = n'(t) \cdot \beta \cdot Y$$

$$Y = 0.042 \frac{\alpha M_2/M_1 S_0(E)}{U_s} \left[1 - \left(\frac{E_{th}}{E}\right)^{1/2}\right]$$

where $\alpha(M_2/M_1)$ is a constant determined by the ratio of the incident ion mass $M_1$, and target atom mass $M_2$, $S_0(E)$ is the cross-section of the elastic collision, $E$ is the kinetic energy of the incident ions, $U_s$ is surface bonding energy, and $E_{th}$ is the energy threshold of the target atoms. All
the quantities are constant except the kinetic energy of the incident ions. Hence,

\[ Y \propto E^{1/2} \]

\[ S(t) \propto n_i(t) \cdot \beta \cdot E^{1/2} \]

The sputtering process is determined by the amount and energy of the incident ions as well as sputtering yield of the target materials. There are two pressure effects. The first one is to enhance the plasma density to reach equilibrium rapidly and the second one is to reduce diffusion and recombination \( \varepsilon \) thus extending the time to reach \( \Phi = 0 \). Both effects improve the stability of the discharge and the voltage threshold of the self-sputtering dominant regime diminishes. The main effect of hybrid DC current is to raise the sputtering power to increase the plasma density and sputtering rate. In this way, the discharge will be more stable and the voltage threshold in the self-sputtering dominant regime decreases as well.

4. Conclusions

In summary, there is a dynamic transition between the gas-dominant discharge regime and self-sputtering one as manifested by the unstable current in the HiPIMS discharge current curves. The formation mechanism and time dependence of the transition process between the two stable regions are studied. The temperature increase from previous pulses plays an important role in the transition and the results are important to future work pertaining to enhancement of the efficiency of HiPIMS as well as microstructure of the deposition films.

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