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Origin of Initial Current Peak in High Power Impulse Magnetron Sputtering and Verification by Non-Sputtering Discharge *

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A non-sputtering discharge is utilized to verify the effect of replacement of gas ion by metallic ions and consequent decrease in the secondary electron emission coefficient in the discharge current curves in high power impulse magnetron sputtering (HiPIMS). In the non-sputtering discharge involving hydrogen, replacement of ions is avoided while the rarefaction still contributes. The initial peak and ensuing decay disappear and all the discharge current curves show a similar feature as the HiPIMS discharge of materials with low sputtering yields such as carbon. The results demonstrate the key effect of ion replacement during sputtering.

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High power impulse magnetron sputtering (HiPIMS) is attractive to the fabrication of hard, decorative, biomedical, and photoelectric coatings due to the high ionization of the sputtered materials,[1−3] subsequent epitaxial growth, excellent film adhesion, and good coating density.[4−7] During deposition, a stable discharge current is crucial.[8,9] The typical current in HiPIMS exhibits an initial peak followed by a sharp decay to a stable value and it is generally considered to originate from the rarefaction effect.[10,11] As the local temperature increases during the process, the local gas density decreases and the mean free path increases correspondingly.[12] However, according to the discharge current slope relationship derived in our previous work,[13]

$$\varPhi = \gamma + \frac{\alpha}{\beta} - \Big[1 - \varepsilon\Big(1 - \frac{1}{\beta}\Big)\Big], \tag{1}$$

where $\varPhi$ is the instantaneous slope of the discharge current in the pulse duration, the average number of electrons produced during collisions α and percentage of ions attracted back to the target β have constant values, $\varPhi$ is determined by both the average secondary electrons emission coefficient γ and ratio of the ions lost by diffusion and recombination to the ions not returning to the target ε which is related to rarefaction.[11] The decrease in the secondary electron emission coefficient due to the replacement of gas ions by metallic ions during sputtering is also important to the current decay following the peak in addition to the rarefaction effect due to the fact that the secondary electron emission coefficients of metal ions are smaller than those of gas ions for the same charge.[14]

In this work, to verify the effects of the plasma composition evolution on the discharge current, a non-sputtering discharge with hydrogen instead of argon is implemented. Increased gas discharge and decreased metallic discharge are observed when hydrogen is introduced into the vacuum chamber. When ion replacement is avoided, the secondary electron emission coefficient no longer decreases in the discharge inducing continuously increase in the discharge current until 'arcing' or pulse end. The initial peak and following decay in the current curves disappear, suggesting that Eq. (1) is valid.

The experiments were performed in a vacuum chamber with a diameter of 40 cm and the height of 40 cm, at a base pressure 3×10^{-3} Pa, as shown in Fig. 1. Hydrogen gas mixed with Ar (99.9997% pure) was introduced through a leak valve and the magnetron cathode was driven by a custom hybrid pulsed power supply. According to the phasic discharge characteristics and target current modes of HiPIMS, the I–V characteristics are different for different target materials and voltages.[3,15] To validate the effects of the sputtering process in each discharge case, Cr (gas discharge dominant) and Cu (metallic discharge dominant) targets (ϕ50 mm × 6 mm) and different target voltages were studied. The discharge pulse width and frequency were 200 µs and 100 Hz, respectively, and the pressure was 0.5 Pa. A digital oscilloscope was employed to monitor the discharge current under dif-

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**Corresponding author. Email: wuzz@pkusz.edu.cn ferent discharge conditions via a $1\,\Omega$ resistor between the anode of the HiPIMS power supply and vacuum wall.

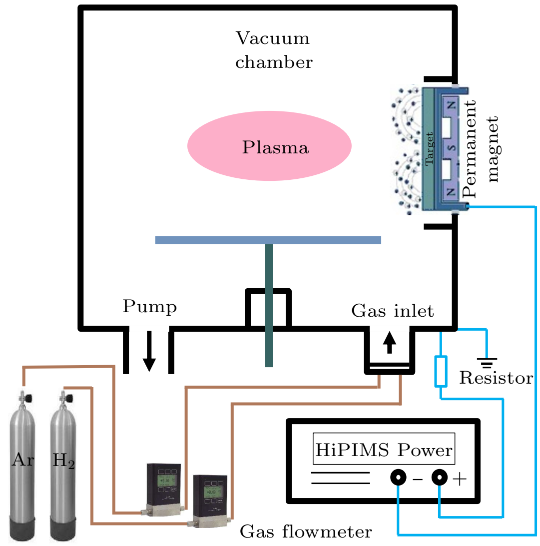

Fig. 1. Schematic diagram of the experimental setup.

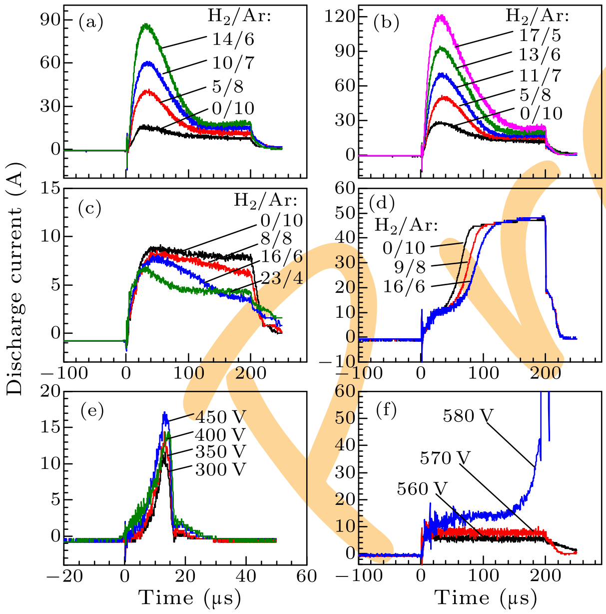

Fig. 2. Waveforms of the HiPIMS discharge currents: (a) Cr target at 500 V, (b) Cr target at 550 V, (c) Cu target at 560 V, (d) Cu target at 580 V under different H$_2$/Ar conditions; (e) Cr target and (f) Cu target at different voltages for pure H$_2$.

The discharge voltage is above 800 V when self-sputtering occurs on Cr. To avoid arcing, the waveforms of the HiPIMS discharge currents of the Cr target are collected at 500 V and 550 V corresponding to gas dominant discharge stages I and Ⅱ described in our previous works,[3] as shown in Figs. 2(a) and 2(b), respectively. The discharge currents for both discharge voltages increase with increasing the amount of hydrogen and decreasing the amount of argon. The peaks are enhanced significantly suggesting that the gas discharge is strengthened by changing from argon to hydrogen.[16] However, if the proportion of hydrogen is too large, arcing occurs and the discharge becomes unstable.

Self-sputtering of Cu is much easier than Cr and the discharge voltage is smaller. Figures 2(c) and 2(d) show the current curves of Cu at discharge voltages of 560 V and 580 V corresponding to gas dominant discharge stages Ⅱ and metallic dominant discharge stages Ⅲ, respectively.[3] In the weak discharge at a low voltage, the current peaks are unchanged while the current plateau decreases with increasing the hydrogen ratio, indicating a weaker metallic discharge.[16] In the intense discharge at a high voltage, although the amplitudes of the peaks and platforms are the same, the widths of the platforms are smaller implying the metallic discharge decreases. If the hydrogen content is too high, arcing also occurs similar to that observed from the Cr target. The results reveal that sputtering can be reduced by using hydrogen in lieu of argon because hydrogen is lighter and leads to enhanced gas discharge for materials with smaller sputtering yields and the reduced metallic discharge for those with higher sputtering yields.

The HiPIMS discharges of Cr and Cu in a pure hydrogen environment are shown in Figs. 2(e) and 2(f). When a Cr target is used, the current rises sharply in the beginning of the pulse while the discharge stops after 12 μs with the current dropping to zero, indicating that the discharge process is composed of weak arcing. Arcing becomes more severe at a higher discharge voltage resulting in an unstable discharge. With regard to the Cu target, a stable discharge is still observed when the voltage is small, albeit weak, and when the discharge voltage is increased, the current increases rapidly and arcing occurs.

According to Eq. (1), the gas dictates the discharge process in the absence of sputtering. At a low discharge voltage, no or few highly charged gas ions are produced and the discharge current rises to a certain value before becoming steady as manifested by the plateau. When the discharge voltage is high, the initial discharge current is small while increases sharply due to the fact that the plasma is dominated by gas ions and highly ionized ions produced at the high voltage thus improving the secondary electron emission coefficient.[14] Finally, arcing occurs when the discharge current is too high. The process is consistent with the mode I current waveform observed from the discharge of materials with very low sputtering yields such as C.[13] Therefore, the evolution of the current curves in Figs. 2(e) and 2(f) is consistent with the derivation from Eq. (1) revealing the important effect of ion replacement and subsequent decrease in the secondary electron emission coefficient.

In conclusion, a non-sputtering discharge is established with hydrogen instead of argon to verify the effects of ion replacement from gas to metallic ones in addition to the subsequent decrease in the secondary electron emission coefficient in the HiPIMS discharge current curves. When the ion replacement is avoided, the initial peak and following decay in the current curves disappear and arcing occurs frequently. All the discharge current curves are similar to those for HiPIMS discharge of materials with low sputtering yields. Our results demonstrate that the ion replacement during sputtering has a significant effect on the discharge current features in addition to the rarefaction effect.

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