Modification of film structure by plasma potential control using triode high power pulsed magnetron sputtering

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We have designed a new triode configuration in a magnetron sputtering apparatus to control the plasma potential of the discharge. An additional chimney electrode was introduced above the conventional sputter gun to apply a positive voltage. The discharge power was provided by a pulse power source to achieve high power pulsed magnetron sputtering operation. We confirmed that the plasma potential increased with increasing positive electrode voltage. Copper films with substantially flatter surfaces could be obtained on a water-cooled and electrically grounded substrate at an Ar gas pressure of 5 Pa.

High-power pulsed (or "impulse") magnetron sputtering (HPPMS/HiPIMS) is an ionized physical vapor deposition (IPVD) technique that has recently attracted significant attention.[1–4] This technique is based on the application of target power as pulses with low repetition frequency and very small duty ratio. By this technique, it is possible to generate the high-density plasma required for IPVD while avoiding heavy thermal load to the target. The HPPMS technique was initially proposed by Kouznetsov et al.[5] in 1999. By using this technique, they obtained a good trench-filling performance without voids, which was ascribed to the high ionization fraction of sputtered particles. Alami and coworkers have also shown that HPPMS is effective in obtaining densified structures of Ta[6] and CrN[7] films.

It is well known that the structure of sputtered films depends on the preparation conditions. A widely accepted zone diagram by Thornton[8] includes substrate temperature and sputtering gas pressure as parameters. More recently, Anders[9] has proposed another structure zone diagram that consists of a "generalized energy", brought by the impinging particles on growing surfaces, instead of the gas pressure. According to these diagrams, a structure with voided defects and a rough surface (Zone I structure) grows at a low substrate temperature and a low kinetic energy (high gas pressure). When a denser film structure with a smoother film surface (Zone T structure) is required, a high substrate temperature and/or a high particle kinetic energy are necessary. When a low substrate temperature is strongly desirable (e.g., deposition on plastic substrates), the particle energy should be high enough.

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This can be achieved if appropriate acceleration potential is introduced in conjunction with the high ionization rate of IPVD/HPPMS.

In our previous study,[10] we have attempted to modify the target voltage during the pulse-off period (V_b) in HPPMS. When films were deposited on water-cooled and electrically grounded substrates, a higher V_b (+50 to +100 V) resulted in more flattened film surfaces and compact cross-section SEM images. With the plasma diagnostic that made use of a Langmuir probe, it was observed that the plasma potential became higher when the negative target voltage was switched to positive at the end of the pulse-on period. It was considered that the space potential of the afterglow plasma increased so that it could shield the plasma from the positive target voltage, and the ionized species within the plasma impinged onto the substrate with higher kinetic energy, reflecting the potential difference between the plasma and the grounded substrate. However, this method has a drawback in that the plasma potential during the pulse-on period is not high. Therefore, the advantage of the high ionization rate of HPPMS is not fully harnessed.

In this study, we have attempted to introduce an additional electrode in the sputtering apparatus in order to configure a triode system. The additional electrode was positively biased to increase the plasma potential for both on and off periods of the HPPMS operation to promote further energetic impingement of ionized species. To understand the function of this additional electrode, a DC discharge experiment was performed as well. By applying different electrode potentials, Cu films were prepared by the DC and HPPMS discharges. We also conducted probe measurements to confirm whether the plasma potential increased following the introduction of the additional electrode, for both pulse-on and pulse-off periods. Usually, a negative bias voltage is applied to the substrate to generate a potential difference between the plasma and the substrate. However, if the positive plasma potential is achieved, the substrate can be grounded. We believe that doing so increases the design flexibility of the sputtering system.

The experiment was conducted with the same sputtering chamber as that used in our previous study;[10] the chamber was a cylinder of 21 cm diameter and 25 cm height. The chamber was evacuated by a turbo molecular pump to a pressure of 4×10^{-5} Pa. The sputter gun was located along the symmetric axis of the chamber. A copper disk target (purity of 4N) with a diameter of 5 cm was used. An additional chimney electrode, 1.5 cm high, was attached on top of the anode cap of the sputter gun, as shown in Fig. 1. For the deposition experiments, Si substrates were set on a water-cooled substrate holder. For plasma diagnostics, the Langmuir probe (Scientific Systems SmartProbe) was introduced, replacing the substrate holder. After the evacuation, 20 sccm of Ar gas was introduced and the pressure was set to 5 Pa by throttling the evacuation valve. A DC source (Advanced Energy MDX-1.5k) and a custom-built pulse power source (Heiwa Dengen)[11] were used as discharge power sources. In this

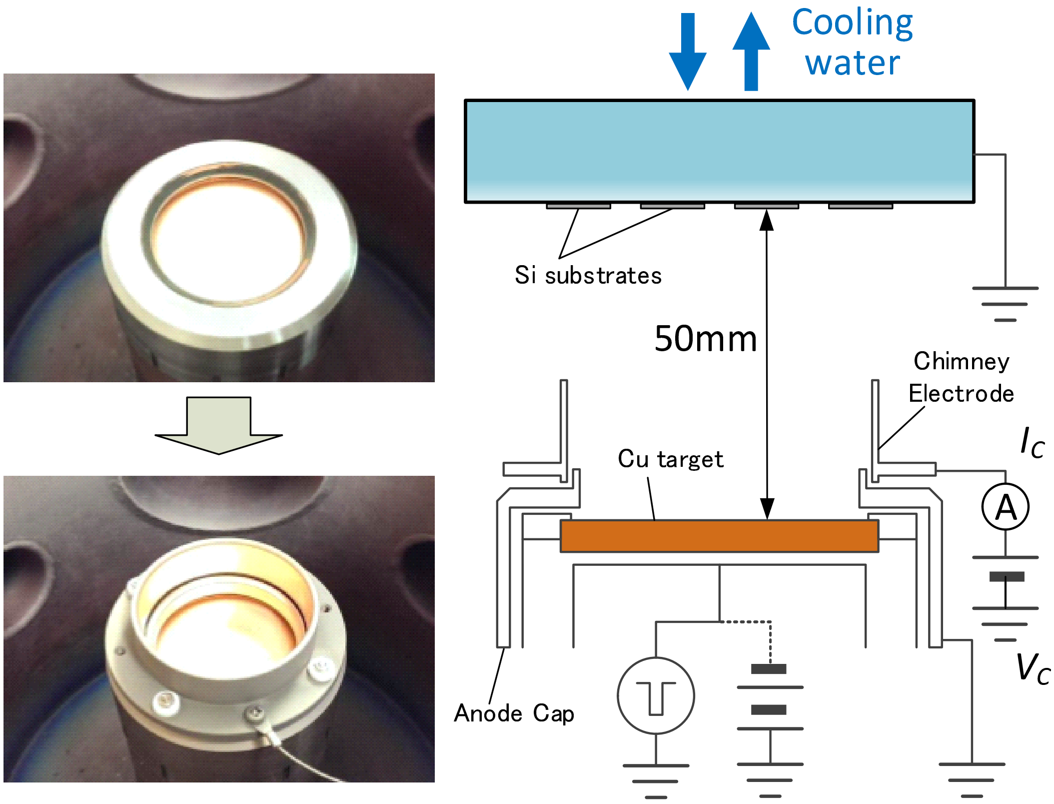

Fig. 1. Experimental setup that includes the chimney electrode above the sputter gun. Left: before and after the introduction of the chimney electrode. Right: cross section of the electrode configuration.

experiment, the pulse power source was driven with a repetition frequency and a duty ratio of 200 Hz and 5%, respectively. The pulse-off target voltage was 0 V. During the pulse power discharge, the waveforms of target current and voltage were monitored and recorded using an oscilloscope (Agilent DSO 3062A). The discharge sources were driven in a constant current mode. Initially, the target power source was set to apply the (time-averaged) power of 100 W with $V_C = 0$, and later the voltage V_C was increased up to +40 V. By doing so, the power that was applied to the discharge volume (from the target source and the V_C source) was nearly the same under all conditions. With the DC and HPPMS discharges, copper films were deposited on HF-treated low-resistivity Si(111) substrates. The substrate holder was electrically grounded and water-cooled to maintain it at room temperature. After the deposition, the films were observed using a secondary electron microscope (SEM; JEOL JSM-6510). In another experiment, in which the Langmuir probe was used, current-voltage characteristics were obtained and analyzed using the method described by Chen,[12] and plasma parameters were obtained. When applied to HPPMS, the Langmuir probe was operated in a boxcar mode for time-resolved measurements triggered by a timing signal from the pulse power source.

Plan-view SEM images of the Cu films deposited on Si substrates are shown in Fig. 2. The scale

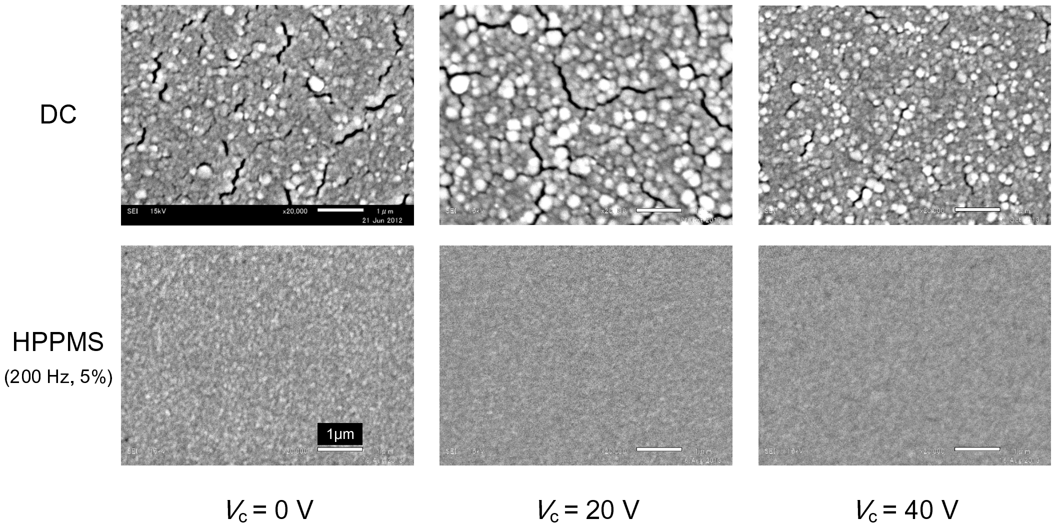

Fig. 2. Plan-view SEM images of deposited Cu films with different discharge sources (DC/HPPMS) and different chimney electrode voltages (V_C) at an Ar gas pressure of 5 Pa. In HPPMS deposition, the repetition frequency and duty ratio were 200 Hz and 5%, respectively. The target power was applied in a constant current mode to achieve 100 W (see text for the detailed procedure). The scale bar at the right bottom corner of each figure represents 1 μm.

bar at the right bottom corner of each figure represents 1 μm. These samples were deposited for 18 min. The deposition rate was measured prior to the actual deposition using a quartz crystal microbalance monitor and found to be 2.0 nm/s, independent of V_C and of whether the power was DC or pulse. Therefore, the thickness of these films was expected to be approximately 2 μm. The target voltage in DC sputtering was −390 V, and the pulse-on voltage in HPPMS was −550 V (both are at $V_C = 0$).

Cu films that were deposited by a DC discharge at $V_C = 0$ and 20 V appeared to have very rough surfaces with large grains and voids, which reflected the columnar structure of Zone I. This structure is specific to high gas pressures (low-energy particles) and low substrate temperatures. At $V_C = 40$ V for a DC discharge, the voids were rather subtle and the film roughness was slightly reduced. However, the films still appeared to contain grains that were larger than 100 nm. On the other hand, in the case of HPPMS, the film surface was already much smoother at $V_C = 0$ than in the DC cases. The size of a typical grain was reduced to 30–50 nanometers. We consider that this occurred because the HPPMS yielded a higher ionization fraction, retaining a kinetic energy as high as 5–7 electron volts provided by a plasma sheath. By increasing V_C to 20 and 40 V, grains became finer and the film surface was significantly flattened.

Figure 3 shows the results of the Langmuir probe measurement. In this measurement, the substrate holder was removed and the probe was introduced along the symmetric axis. The end of the probe tip

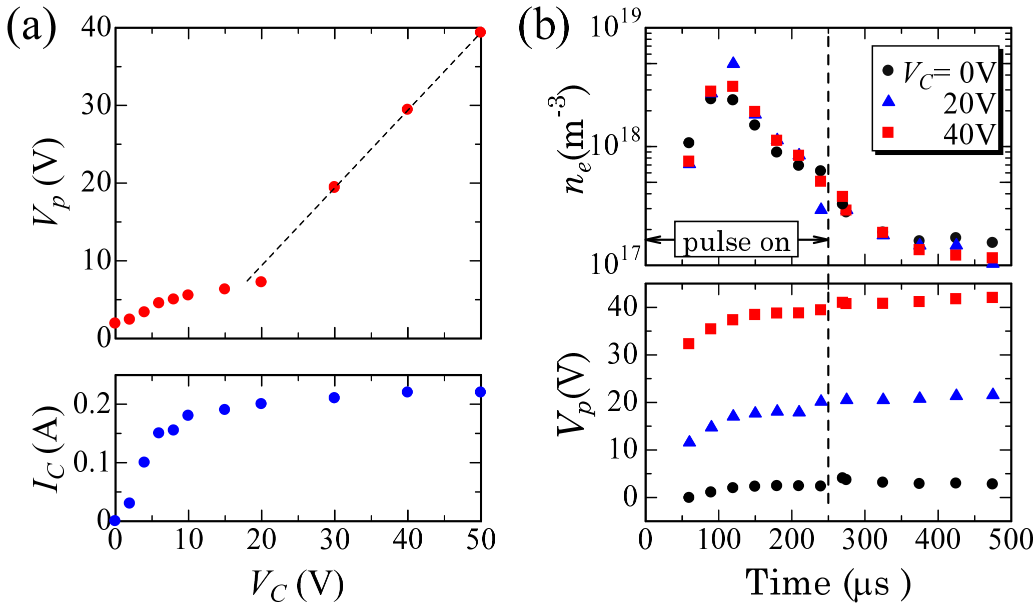

Fig. 3. (a) Dependence of plasma potential V_p and chimney electrode current I_C on the chimney electrode voltage V_C in DC discharge. (b) Time resolved plasma parameters (electron density n_e and plasma potential V_p) in HPPMS discharge with V_C as a parameter.

was located 50 mm away from the target surface. The dependence of the plasma potential V_p on V_C in DC discharge is shown in Fig. 3(a) along with the dependence of the chimney electrode current I_C on V_C. It can be seen that I_C increases gradually and saturates at $V_C = 20$ V. Before the saturation of I_C, the increase in V_p was rather modest, while past the saturation point it increased with a slope of unity. This suggests that the chimney electrode becomes the anode of the discharge at $V_C > 20$ V. The electron density was $(3 - 4) \times 10^{16}$m^{-3} and was almost independent of V_C.

The results of the time-resolved probe measurement in HPPMS discharge are shown in Fig. 3(b). The origin of the horizontal axis denotes the ignition of the pulse voltage, which continues for 250 μs, reflecting the pulse frequency of 200 Hz and the duty ratio of 5%. The electron density n_e was mostly independent of V_C; it increased initially and peaked at a density as high as $3{\times}10^{18}$m^{-3} at approximately 100 μs, and then decreased. The occurence of a peak in n_e corresponded to that in target current, which is commonly observed in HPPMS with modest pulse power.[3,10] On the other hand, the plasma potential V_p showed a different dependence on V_C. It increased as high as V_C in both pulse-on and pulse-off periods as expected. It increased during the initial stage of the pulse-on period and became almost stable after 100 μs, at which point the electron density became maximal. The smooth surface of the Cu films that was obtained using the HPPMS with a positive V_C is ascribed to the positive plasma potential and effective bombardment of positive ions on the grounded substrate surface.

The mechanism by which the plasma potential increases up to the most positive electrode potential is understood to be the dragging of electrons by the positive electrode that causes the plasma to "charge up", resulting in a positive space potential.[13] In this viewpoint, the slight increase in V_p at the end of the pulse [Fig. 3(b)] can be ascribed to the termination of the supply of secondary electrons from the target. The phenomenon can also be explained by the plasma-wall interaction.[14] When the wall voltage V_w is much lower than the floating potential V_f, the current is mainly delivered by ions and is not sensitive to V_w. On the other hand, when $V_w > V_f$, the electron current dominates and it exponentially depends on the difference $V_p - V_w$ of potentials. Therefore, in order for the electrodes to satisfy the current continuity conditions, the floating potential and hence the plasma potential must approach the voltage of the most positive electrode unless a very large difference between electrode areas exists.

The triode sputtering concept was proposed during the very early years of the sputtering method (see Chopra's textbook[15] for examples). However, in those systems, the third electrode was a thermionic electron source that helped the low-gas-pressure discharge. More recently, a triode sputtering system for discharge plasma control has been proposed by Fontana and Muzart,[16] which was fabricated by inserting a grid between the target and the substrate; however, this system does not directly aim to control the plasma potential. An increase in the plasma potential achieved by introducing an additional grid electrode was reported by Jung et al., but the effect was rather limited because it utilized a DC sputtering apparatus.[17] The plasma potential control using the triode configuration and the high ionization rate of HPPMS were crucial for the effective bombardment and film structure modification.

Note that the V_p in this experiment was considerably lower than that used in our previous study.[10] In our previous experiment, the plasma potential was raised by icontrolling the target voltage only after the high power discharge was cut off. Hence, a pulse-off period target voltage of +100 V was necessary to obtain a smooth surface such as that obtained by HPPMS and a V_C of +20 V in Fig. 2. In the present experiment, therefore, it is considered that a large part of ion bombardment occurred during the pulse-on period and that the energy deposition on the growing surface by accelerated ions became more effective following the addition of the chimney electrode. Another approach to preparing a Zone T structure at low substrate temperatures is to reduce the discharge gas pressure so as to prevent the deceleration of the initial energy of sputtered particles by collisions with ambient gases.[8] However, the reduction in gas pressure promotes negative side effects, e.g. super high-energy reflected neutrals.[18] At least in some applications, it is desirable to stop those high-energy particles while bombarding the growing film surface at controlled energy.

In summary, we have developed a triode HPPMS system and studied the effect of an additional electrode on the plasma and growing film structure. In accordance with the increase in the additional electrode voltage, the plasma potential increased and a flatter surface structure was obtained. This suggests a more effective impingement of ionized species onto the growing film surface.

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