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N.B.: When citing this work, cite the original article.

Original Publication:

J T Gudmundsson, P. Sigurjonsson, Petter Larsson, Daniel Lundin and Ulf Helmersson, On the electron energy in the high power impulse magnetron sputtering discharge, 2009, JOURNAL OF APPLIED PHYSICS, (105), 12, 123302. <u>http://dx.doi.org/10.1063/1.3151953</u> Copyright: American Institute of Physics <u>http://www.aip.org/</u> Postprint available at: Linköping University Electronic Press

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On the electron energy in the high power impulse magnetron sputtering discharge

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(Received 9 May 2009; accepted 12 May 2009; published online 19 June 2009)

The temporal variation of the electron energy distribution function (EEDF) was measured with a Langmuir probe in a high power impulse magnetron sputtering (HiPIMS) discharge at 3 and 20 mTorr pressures. In the HiPIMS discharge a high power pulse is applied to a planar magnetron giving a high electron density and highly ionized sputtered vapor. The measured EEDF is Maxwellian-like during the pulse; it is broader for lower discharge pressure and it becomes narrower as the pulse progresses. This indicates that the plasma cools as the pulse progresses, probably due to high metal content of the discharge. © 2009 American Institute of Physics. [DOI: 10.1063/1.3151953]

I. INTRODUCTION

Plasma based sputtering has found widespread use in various industrial application, in particular, in coating processes. The workhorse of plasma based sputtering applications for over three decades is the magnetron sputtering discharge.¹ In a magnetron sputtering discharge a static magnetic field is applied to confine the secondary electrons in the vicinity of the cathode. In a conventional dc magnetron sputtering discharge, only a few percent of the sputtered atoms are ionized. Initially ionized physical vapor deposition (IPVD) processes were based on a secondary discharge to create a dense plasma, placed between the source (the cathode target) and the substrate, to ionize a large fraction of the sputtered atoms.^{2,3} Recently, IPVD has been achieved by applying a high power unipolar pulse of low frequency and low duty cycle to the cathode to create very high plasma density.^{2,4} This is referred to as high power impulse magnetron sputtering (HiPIMS) or high power pulsed magnetron sputtering. HiPIMS has the advantage of using essentially the conventional magnetron sputtering equipment except for the power supply. The discharge operates with a cathode voltage in the range of 500-2000 V, current densities of $3-4 \text{ A cm}^{-2}$, power densities in the range of 0.5-3 kW cm⁻², frequency in the range of 50–1000 Hz, and duty cycle in the range of 0.5%-5%.^{2,3} Common to all the IPVD techniques is a very high density plasma. There have been several studies of the spatial and temporal variations of the electron density in the HiPIMS discharge using Langmuir probe diagnostics.^{5–11} Measurements of the temporal and spatial behaviors of the plasma parameters in the HiPIMS discharge indicate peak electron density of the order of few times 10^{18} m⁻³ (Ref. 5–7) that expands from the target as an ion acoustic wave.¹² For sputter deposition of thin films, the knowledge of the electron energy distribution function (EEDF) and the plasma parameters in the near-substrate vicinity are of great importance for determining the process parameters and understanding of the ionization mechanism. Here, we apply a Langmuir probe to explore the temporal variation of the EEDF over a wide dynamic range, the effective electron temperature, and the electron density, in a HiP-IMS discharge.

II. EXPERIMENTAL APPARATUS AND METHOD

A standard, slightly unbalanced, planar magnetron source was operated with a copper target 150 mm in diameter. The copper target is directly cooled from the back side while sputtering is in progress. The sputtering target (cathode) was located inside a stainless steel chamber, 450 mm in diameter, and 705 mm long. The base pressure was maintained below 10⁻⁶ Torr with a turbomolecular pump. Argon of purity 99.9997% was used as a discharge gas. The discharge power supply is a pulse generator, SINEX 2, from Chemfilt Ionsputtering. For the measurements reported here the average power was in the range of 215-270 W, corresponding to pulse energy from 4.3 to 5.4 J and pulse length from 80 to 90 μ s, depending on the gas pressure. The repetition frequency was fixed at 50 Hz. A high-voltage probe (Tektronix P 6015A) and a current clamp (Chauvin Arnoux C 160) were used to measure the target voltage and the target current, respectively. Figure 1 shows the voltage and current waveforms obtained for the HiPIMS discharge operated at 3 and 20 mTorr. The exact pulse shape is not only determined by the power supply but also by the load and the discharge formed in the sputtering device.

The Langmuir probe wire radius *a* has to be greater than the electron Debye length, $a > \lambda_{De} = (\epsilon_0 T_e / e n_e)^{1/2}$, where n_e is the electron density and T_e is the electron temperature. Here the Debye length is in the range of 5–25 μ m. Thus, the Langmuir probe is made of a stainless steel wire, 200 μ m in diameter that was placed inside a ceramic tube for insulation extending out 5 mm. For the measurements presented here the Langmuir probe is located 80 mm away

0021-8979/2009/105(12)/123302/3/\$25.00

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FIG. 1. (Color online) The applied target voltage V_T and the applied target current I_T for an argon discharge at 3 and 20 mTorr. The target is made of copper 150 mm in diameter.

from the target surface and 40 mm off the central axis (under the race track). The Langmuir probe was biased in the range -20-+25 V in 0.01–0.05 V steps. For each voltage step the current drawn by the probe was measured as a function of time from initiating the pulse to the discharge target. The time step was 320 ns. The current was measured as a voltage drop over a 1 Ω shunt resistance. These current traces, at a fixed voltage, are then used to construct *I-V* curves for each time step. The probe voltage was measured over a voltage divider to adjust the voltage for an A/D converter, Pico ADC 212 oscilloscope module that has a 12-bit resolution. The fine voltage steps and the 12-bit resolution are essential for resolving the EEDF over the wide dynamic range presented here for the electron energy probability function (EEPF) and is significantly improved from our previous work.^{5,8}

The second derivative of the Langmuir probe I-V characteristics is obtained by numerically differentiating and filtering¹³ the measured I-V curve. The EEDF is then determined from the Druyvesteyn formula (Ref. 14, p. 191) and found by

$$g_e(V) = \frac{2m}{e^2 A} \left(\frac{2eV}{m}\right)^{1/2} \frac{d^2 I_e}{dV^2},$$
 (1)

and the EEPF is given by

$$g_P(\mathcal{E}) = \mathcal{E}^{-1/2} g_e(\mathcal{E}), \tag{2}$$

where \mathcal{E} is the electron energy, and the change of variables $\mathcal{E}=\frac{1}{2}mv^2/e$ has been introduced. Once the EEDF is known, the electron density is found by

$$n_e = \int_0^\infty g_e(\mathcal{E}) d\mathcal{E}.$$
 (3)

The effective electron temperature is then calculated from the average electron energy or

$$T_{\rm eff} = \frac{2}{3} \langle \mathcal{E} \rangle = \frac{2}{3} \frac{1}{n_e} \int_0^\infty \mathcal{E}g_e(\mathcal{E}) d\mathcal{E}.$$
 (4)

III. RESULTS AND DISCUSSION

The temporal variation of the effective electron temperature $T_{\rm eff}$ is shown in Fig. 2(a) and indicates a significant



FIG. 2. (Color online) (a) The effective electron temperature $T_{\rm eff}$ and (b) the electron density n_e vs time for an argon discharge at 3 and 20 mTorr. The Langmuir probe is located under the race track 80 mm away from the target surface. The target is made of copper 150 mm in diameter.

cooling of the electrons in the HiPIMS discharge. Early in the pulse the effective electron temperature is in the range of 1.5-2 V and falls as the pulse progresses. The effective electron temperature at about 90 μ s into the pulse reaches roughly a constant value of about 0.7 V at 3 mTorr and 0.3-0.4 V at 20 mTorr, that remains for the following 150 μ s. This is consistent with the findings of Vetushka and Ehiasarian⁹ which record a peak electron temperature early in the pulse and then relatively constant values of 0.4 and 0.8 eV after the pulse is off for at least 300 μ s at 2 mTorr for Cr and Ti targets, respectively. The effective electron temperature in a conventional dc magnetron sputtering discharge is in the range of 2–4 V,^{15–17} significantly higher than observed for the HiPIMS discharge. The electron density is shown versus time for an argon discharge at 3 and 20 mTorr in Fig. 2(b). The electron density increases sharply with time and peaks at roughly 100 μ s into the pulse. The electron density decays faster at the lower pressure. This is consistent with earlier measurements that have shown very high plasma densities in the HiPIMS discharge^{5,6} or about two to three orders of magnitude higher density than what is commonly observed in a conventional dc magnetron sputtering discharge.^{15–17} Generally a monotonic rise in plasma density with discharge gas pressure⁸ and applied power¹⁸ and linear increase in electron density with increased discharge current¹⁰ is observed. In contrast to our earlier reports, the oscillations in the electron density observed at low pressure

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FIG. 3. (Color online) The EEPF for various times from initiating the pulse for an argon discharge at 3 (dotted line) and 20 (solid line) mTorr. The Langmuir probe is located under the race track 80 mm from the target surface. The target is made of copper 150 mm in diameter.

are absent. That remains to be explored if the discharge enters instability regimes at certain pressures and powers or if these are artifacts of a power supply.

Figure 3 shows the temporal variation of the EEPF for an argon discharge at 3 and 20 mTorr. The EEPFs are graphed on a semilog plot to display them over a wide dynamic range. In this representation $\ln(g_P)$ is linear with \mathcal{E} for Maxwellian-like EEPF. The measured EEDF is a Maxwellian-like with a depleted high energy tail in the range $60-150 \ \mu s$ from initiating the pulse. At high electron densities electron-electron Coulomb collisions are an important energy transfer mechanism that leads to equalization of the distribution temperature. For the electron-electron collisions the collision frequency scales linearly with the electron density or $\nu_{ee} \propto n_e \mathcal{E}^{3/2}$. Thus, high electron density leads to a Maxwellian-like low energy part of the EEPF. The depletion in the high energy part is due to the escape of high energy electrons to the chamber walls and inelastic collisions of high energy electrons. The EEPF is broader for the lower discharge pressure of 3 mTorr. This is consistent with the fact that at higher neutral gas pressure, we would expect increased inelastic collisions with the neutral gas and thus increased depletion of the high energy electrons. Furthermore, the EEPF becomes narrower as the pulse progresses at both 3 and 20 mTorr. This indicates that the plasma cools off as the pulse progresses. There is a significantly higher density of metal atoms in a HiPIMS discharge compared to a conventional dc magnetron sputtering discharge. This has been observed both by optical emission spectroscopy^{19,20} and mass spectroscopy,^{21,22} which show that the discharge develops from an argon dominated to a metal dominated discharge during the pulse. For example, Vlček et al.²² claimed that the Cu⁺ ions dominate the ion flux (92% of the total ion flux) in the substrate vicinity when operating at maximum power density of 950 W/cm² and pressure of 3 mTorr. This is expected to cool the EEPF due to electron impact excitation and ionization of the metal atoms that have much lower excitation thresholds and ionization potential than the argon

sputtering gas. The bi-Maxwellian distribution we thought we saw^{5,8} we no longer believe to be correct. However, recent report by Pajdarová *et al.*⁷ indicate that bi-Maxwellian electron energy distribution may be observed in the initial stages of the pulse, in particular, at lower power densities. That is consistent with a conventional dc magnetron sputtering discharge where the EEDF is commonly seen to be bi-Maxwellian.^{15–17}

IV. CONCLUSION

It can be concluded that the high electron density in HiPIMS discharge leads to a Maxwellian-like EEDF. Furthermore, the high plasma density leads to a higher fraction of metal produced in the HiPIMS discharge compared to a conventional dc magnetron discharge. It also leads to a high ionization fraction of the sputtered species due to electron impact ionization of metal atoms which significantly cools the discharge.

ACKNOWLEDGMENTS

This work was partially supported by the Icelandic Research Fund, the University of Iceland Research Fund, and the Swedish Research Council.

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