Ionization mechanism in the high power impulse magnetron sputtering (HiPIMS) discharge

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Abstract. The ionization mechanism and the temporal behavior of the plasma parameters in a high power impulse magnetron sputtering (HiPIMS) discharge are investigated using a time dependent global (volume averaged) model. The metal ion fraction and the ionized flux fraction are shown to be very high, the sputtered metal is almost fully ionized. During the pulse on period electron impact ionization is the most effective process in creating metal ions while charge exchange becomes the dominant process in creating metal ions after the pulse is off.

1. Introduction

Recently magnetron sputtering discharges have been developed that can generate highly ionized plasma most commonly by adding a secondary discharge between the cathode target and the substrate [1]. An alternative method, referred to as high power impulse magnetron sputtering (HiPIMS) [1], has been shown to give high degree of ionization of the sputtered material [2, 3, 4] and very high plasma densities ($\sim 10^{19}$ m$^{-3}$) [5]. The discharge is created by applying a high power unipolar pulse of low duty cycle to the cathode target. The pulse length is typically 50 - 500 µs and the pulse frequency 1 – 1000 Hz. The high power pulse has a peak cathode voltage in the range 500 – 2000 V which gives peak power densities in the range 1 – 3 kW/cm$^2$ [6]. The exact pulse shape is determined by the load, the discharge formed in the sputtering device, and thus depends on the gas type and gas pressure as seen in figure 1 (after [5]). A high fractional ionization has been demonstrated and values higher than 90 % at pulse energy of 2 J for Ti target have been reported [4]. The measured ionized flux fraction from Cu target was estimated roughly 70 % [2], from a Ti$_{0.5}$Al$_{0.5}$-target around 40 % [3], and from C and Al target 4.5 % and 9.5 %, respectively [7]. Thus, the reported measured values are highly inconsistent. Here, we explore the ionization mechanism and the temporal behavior of the plasma parameters in a HiPIMS discharge and estimate the fractional ionization.

2. Model of the ionization process

To explore the plasma parameters and the ionization mechanism for highly ionized magnetron sputtering a simple time dependent global model was developed, based on the time dependent global model for argon discharge [8] and extended to include metal species as described by Hopwood [9]. The discharge is assumed to consist of electrons, Ar atoms in the ground state, metastable Ar atoms, Ar$^+$ ions, metal atoms, M, and metal ions, M$^+$. 

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The power balance equation, which equates the absorbed power $P_{\text{abs}}$ to power losses due to elastic and inelastic collisions and losses due to charged particle flow to the discharge walls is given as

$$\frac{d}{dt} \left( \frac{3}{2} e n_e T_e \right) = P_{\text{abs}} - e \mathcal{E}_e k_{iz} n_{Ar} n_e - e k_{\text{wall,\,Ar}^+} (\mathcal{E}_e + \mathcal{E}_i) n_{Ar^+}$$

(1)

where $\mathcal{E}_e$ is the mean kinetic energy per electron lost and $\mathcal{E}_i$ is the mean kinetic energy per ion lost and $k_{\text{wall,\,Ar}^+} = 2u_B (h_L R^2 + h_R RL)/R^2 L$ accounts for the flux of Ar$^+$ ions to the chamber walls and $u_B = (e T_e/m_i)^{1/2}$ is the Bohm velocity for argon ions, $m_i$ is the argon ion mass, and $n_{Ar^+}$ is the density of argon ions. Here, $\mathcal{E}_c(T_e)$ is the collisional energy loss per electron-ion pair created and is defined as

$$\mathcal{E}_c = \mathcal{E}_{iz} + \sum_i \mathcal{E}_{x,i} \frac{k_{ex,i}}{k_{iz}} + \frac{k_{el}}{k_{iz}} \frac{3 m_e T_e}{m_i}$$

(2)

where $\mathcal{E}_{iz}$ is the ionization energy, $\mathcal{E}_{x,i}$ is the threshold energy and $k_{ex,i}$ is the rate coefficient for the $i$-th excitation process, respectively, $k_{iz}$ is the ionization rate coefficient for single step ionization $e + \text{Ar} \rightarrow \text{Ar}^+ + 2e$ and $k_{el}$ is the elastic rate coefficient (see [10]). The ratios of the density at the sheath edge to that in the bulk are for the axial and radial directions, $h_L = 0.86[3 + L/2\lambda_i]^{-\frac{1}{2}}$ and $h_R = 0.8[4 + R/\lambda_i]^{-\frac{1}{2}}$, respectively [11], where $\lambda_i = n_{Ar}\sigma_i$ is the ion-neutral mean free path and $\sigma_i$ is the ion-neutral scattering cross section. The combined ionic momentum transfer cross section for these two processes is roughly $1 \times 10^{-18}$ m$^2$ for the thermal energies of interest.

We assume that metal ions are created by electron impact ionization $e + M \rightarrow M^+ + 2e$ with rate coefficient $k_{\text{mix}} = 1.23 \times 10^{-13} \exp(-7.23/T_e)$ m$^3$/s [9], by Penning ionization, collisions of metal atoms with electronically excited argon atoms, $\text{Ar}^* + M \rightarrow M^+ + \text{Ar} + e$ with a rate coefficient $k_P = 5.9 \times 10^{-16}$ m$^3$/s [13] and through charge exchange $\text{Ar}^* + M \rightarrow M^+ + \text{Ar}$ with a rate coefficient $k_{\text{chexc}} = 1 \times 10^{-15}$ m$^3$/s [13]. The particle balance for the metal ions gives

$$\frac{dn_{m^+}}{dt} = k_{\text{mix}} n_e n_m + k_P n_{Ar^+} n_m + k_{\text{chexc}} n_{Ar^+} n_m - k_{\text{wall,m}} n_{m^+}$$

(3)

where $n_m$ is the neutral metal density, $n_{m^+}$ is the metal ion density, $n_{Ar^+}$ is the density of metastable argon atoms, $k_{\text{wall,m}} = 2u_{B,m} (h_L R^2 + h_R RL)/R^2 L$ accounts for the flux of metal ions to the chamber walls, and $u_{B,m} = (e T_e/m_{i,m})^{1/2}$ is the Bohm velocity for metal ions, and $m_{i,m}$ is the metal ion mass. The particle balance for metal atoms is

$$\frac{dn_m}{dt} = \frac{h_L r_T^2}{R^2 L} \left( \gamma_{\text{sput}} u_B n_{Ar^+} + \gamma_{\text{selfsput}} u_{B,m} n_{m^+} \right) - k_{\text{mix}} n_e n_m - k_P n_{Ar^+} n_m - k_{\text{chexc}} n_{Ar^+} n_m - k_{\text{diff,m}} n_m +$$

(4)

where $\gamma_{\text{sput}}$ is the yield of sputtered atoms per incident argon ion, $\gamma_{\text{selfsput}}$ is the yield of sputtered atoms per incident metal ion, $r_T$ is the target radius. The loss of metal atoms includes electron impact ionization, Penning ionization and charge exchange with argon ions and the diffusion loss of metal atoms given by $D_m/\Lambda^2$ where $\Lambda = [(\pi/L)^2 + (2.405/R)^2]^{-1/2}$ is the effective diffusion length of neutral species and $D_m = e T_e \lambda_i/m_{i,m} v_{th}$ is the metal atom diffusion coefficient, where $v_{th} = (8 e T_e/\pi m_{i,m})^{1/2}$ is the mean neutral speed and $m_{i,m}$ is the metal atom mass. The Penning ionization depends on the density of the metastable argon atoms which are produced by electron impact excitation $e + \text{Ar} \rightarrow \text{Ar}^* + e$ with rate coefficient $k_{\text{exc}}$ (see [10]) and its loss is due to deexcitation by Penning ionization, electron impact deexcitation with a rate coefficient $k_{\text{deexc}} = 4.3 \times 10^{-16} T_e^{-0.74}$ m$^3$/s [8], diffusion losses and electron impact ionization $e + \text{Ar}^* \rightarrow \text{Ar}^+ + e$ with rate coefficient $k_{\text{exc,iz}}$ [12]. The particle balance equation for generation and loss of metastable argon atoms is

$$\frac{dn_{Ar^+}}{dt} = k_{\text{exc}} n_e n_{Ar} - k_{\text{exc,iz}} n_e n_{Ar^+} - k_{\text{deexc}} n_e n_{Ar^+} - k_{\text{loss}} n_{Ar^+} n_{Ar^+} - k_P n_{Ar^+} n_m$$

(5)
The argon ions are created by electron impact ionization from argon atoms and are lost through charge exchange with metal atoms and as flux to the chamber walls. The particle balance for argon ions is

\[
\frac{dn_{Ar^+}}{dt} = k_{i\alpha} n_e n_{Ar} + k_{exc,i\alpha} n_e n_{Ar^*} - k_{chexc} n_m n_{Ar^+} - k_{wall,Ar^+} n_{Ar^+}
\]  

(6)

The temporal variation of the particle density and the electron temperature was obtained by solving the differential equations (1), (3), (4), (5) and (6) simultaneously and self-consistently. Once the density of Ar\(^+\) and M\(^+\) ions is found the quasi-neutrality condition gives the electron density \(n_e = n_{Ar^+} + n_{m^+}\).

**Figure 1.** The power applied to the discharge versus time from the pulse initiation. The target was made of tantalum 7.5 cm in radius.

**Figure 2.** The sputter yield of Ar\(^+\) on Al and Al\(^+\) on Al versus time. The target voltage pulse was experimentally determined at 10 mTorr.

### 3. Results and discussion

To explore the ionization processes in a high power impulse magnetron sputtering discharge we assume a discharge chamber of radius \(R = 15\) cm and length \(L = 15\) cm with a target of radius 7.5 cm made of aluminum. We assume the power pulse to be the same as shown in figure 1 and the discharge pressure to be 10 mTorr (after [5]). The electron energy distribution is assumed to be Maxwellian. We assume the sputter yield for argon ion bombardment of aluminum and the self-sputter yield of aluminum to depend on the target voltage according to the data collected by Ruzic [14] and Hayward and Wolter [15], respectively. The sputter yield of Ar\(^+\) on Al and Al\(^+\) on Al versus time for a given voltage pulse is shown in figure 2.

Figure 3 shows the calculated electron density, argon ion density, the aluminum ion density and the aluminum atom density versus time from the pulse initiation. The argon ion density increases sharply when the pulse is initiated and peaks at \(3.4 \times 10^{19}\) m\(^{-3}\) 53 \(\mu\)s after initiating the pulse and then decays. The applied power peaks at 137 kW 22 \(\mu\)s after initiating the pulse. The aluminum ion density peaks at \(9.2 \times 10^{17}\) m\(^{-3}\) 70 \(\mu\)s after initiating the pulse. The aluminum atom density peaks at \(1 \times 10^{17}\) m\(^{-3}\) 41 \(\mu\)s after initiating the pulse. We note that the metal atom density is only apparent during the pulse while the metal is sputtered off the target. The metal ion flux to the surface is \(\Gamma_{m^+} = 0.61 n_{m^+} u_{B,m} m^+\) and the flux of the neutral metal is \(\Gamma_m = \frac{1}{2} u_{nm} n_m\). In discharges that are not in thermal equilibrium the electron temperature \(T_e\) is typically significantly larger than the neutral gas temperature \(T_g\). Thus, the fraction of ionized metal flux at the substrate \(\Gamma_{m^+}/(\Gamma_{m^+} + \Gamma_m)\) is larger than the fraction of ionized metal...
in the plasma $n_{m+}/(n_{m+} + n_{m})$. The integrated metal ion fraction is 0.98 and the integrated ionized flux fraction is 0.99 during the pulse. In figure 4 we see that during the first 100 µs from initiating the pulse electron impact ionization is the most effective process in creating metal ions. At about 100 µs into the pulse charge exchange becomes the dominant process in creating the metal ions. Penning ionization is a negligible process.

4. Conclusion

The metal ion fraction and the ionized flux fraction are very high, the sputtered metal is almost fully ionized. During the first 100 µs from initiating the pulse (roughly while the pulse is on) electron impact ionization is the most effective process in creating metal ions.

Acknowledgments

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References

[1] Helmersson U, Lattemann M, Bohlmark J, Ehiasarian A P, Gudmundsson J T 2006 Thin Solid Films 513 1
[2] Kouznetsov V, Macák K, Schneider J M, Helmersson U, Petrov I 1999 Surf. Coat. Technol. 122 290
[3] Macák K, Kouznetzov V, Schneider J M, Helmersson U, Petrov I 2000 J. Vac. Sci. Technol. A 18 1533
[4] Bohlmark J, Alami J, Christou C, Ehiasarian A P, Helmersson U 2005 J. Vac. Sci. Technol. A 23 18
[5] Gudmundsson J T, Alami J, Helmersson U 2002 Surf. Coat. Technol. 161 249
[6] Ehiasarian A P, New R, Münn W D, Hultman L, Helmersson U, Kouznetzov V 2002 Vacuum 65 147
[7] DeKoven B M, Ward P R, Weiss R E, Christie D J, Scholl R A, Sproul W D, Tomasel F, Anders A 2003 46th Techn. Conf. Proc. (San Francisco, CA) (Albuquerque NM: Society of Vacuum Coaters) p 158
[8] Ashida S, Lee C, Lieberman M A 1995 J. Vac. Sci. Technol. A 13 2498
[9] Hopwood J A 2000 Thin Films: Ionized Physical Vapor Deposition, ed. J A Hopwood (San Diego CA: Academic Press) pp 181–207
[10] Gudmundsson J T 2002 Technical Report RH-21-2002, Science Institute, University of Iceland.
[11] Godyak V A 1986 Soviet Radio Frequency Discharge Research (Falls Church VA: Delphic Associates)
[12] Lee C, Lieberman M A 1995 J. Vac. Sci. Technol. A 13 368
[13] Lu J, Kushner M J 2000 J. Appl. Phys. 87 7198
[14] Ruzic D N 1990 Handbook of Plasma Processing Technology: Fundamentals, Etching, Deposition, and Surface Engineering, eds. S M Rossnagel, J J Cuomo and W D Westwood (Park Ridge NJ: Noyes Publications) pp 70–90
[15] Hayward W H, Wolter A R 1969 J. Appl. Phys. 40 2911