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Hysteresis and process stability in reactive high power impulse magnetron sputtering of metal oxides

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Abstract

In the further development of reactive sputter deposition, strategies which allow for stabilization of the transition zone between the metallic and compound modes, elimination of the process hysteresis, and increase of the deposition rate, are of particular interest. In this study, the hysteresis behavior and the characteristics of the transition zone during reactive high power impulse magnetron sputtering (HiPIMS) of Al and Ce targets in an Ar-O_2 atmosphere as a function of the pulsing frequency and the pumping speed are investigated. Comparison with reactive direct current magnetron sputtering (DCMS) reveals that HiPIMS allows for suppression/elimination of the hysteresis and a smoother transition from the metallic to the compound sputtering mode. For the experimental conditions employed in the present study, optimum behavior with respect to the hysteresis width is obtained at frequency values between 2 and 4 kHz, while HiPIMS processes with values below or above this range resemble the DCMS behavior. Al-O films are deposited using both HiPIMS and DCMS. Analysis of the film properties shows that elimination/suppression of the hysteresis in HiPIMS facilitates the growth of stoichiometric and transparent Al_2O_3 at relatively high deposition rates over a wider range of experimental conditions as compared to DCMS.

Keywords: Reactive sputtering, HiPIMS, Al_2O_3, CeO_2, hysteresis, process stability

1. Introduction

Reactive magnetron sputtering is a physical vapor deposition (PVD) technique which is widely employed for the growth of compound films allowing for good film uniformity, reliable control over the film properties, deposition on large area substrates, and cost efficiency [1]. Typical feature in reactive magnetron sputtering processes is that compound formation does not only take place on the substrate but also on the target surface (referred to as target poisoning). Deposition from a fully poisoned target (referred to as the compound sputtering mode) allows for growth of stoichiometric compound films, i.e. films with sufficient incorporation of the reactive gas atoms [2]. At these conditions deposition rates significantly lower than those obtained from an elemental (e.g. metallic) target are commonly observed, since the sputtering efficiency of the compound material is typically lower than the sputtering efficiency of the corresponding metal [2]. Growth of stoichiometric compound films with relatively high rates can be facilitated in the transition regime between the metallic and the compound mode [3]. However, the relationship between reactive gas flow and process parameters is complex leading to an unstable transition zone and a hysteresis in the process parameters, e.g. decrease and increase in the deposition rate does not occur at the same value of the reactive gas flow [2]. Therefore, operation in the transition zone using only the reactive gas mass flow as the control parameter is in most cases impossible [2,3]. The latter is particularly pronounced during reactive sputtering of metal oxides [2,3]. It has been shown that stabilization of the transition zone or elimination of the hysteresis can be achieved by feedback control systems [3], increase of the pumping speed [4,5], reduction of the target area [6], and for some metal oxide systems by the addition of nitrogen into the sputtering atmosphere [7].

High power impulse magnetron sputtering (HiPIMS) is a PVD technique in which the power is applied to the target in short unipolar pulses of low duty cycle (<10 %) and frequency (<10 kHz) [8]. This mode of operation allows for maximum target power density values during the

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pulse on-time (referred to as peak target power density) in the order of several kWcm^{-2}, while the average target power density is maintained in the order of several tens of Wcm^{-2} comparable to conventional magnetron sputtering processes [9]. The high peak target power densities in turn facilitate the generation of dense plasmas (electron densities up to 10^{19} m^{-3}) resulting in high degree of ionization for both gas and sputtered species [8-12]. These plasma conditions have been shown to allow for the deposition of films with superior properties as compared to those obtained by conventional magnetron sputtering techniques. HiPIMS in reactive mode has been extensively employed for the deposition of metal nitrides and oxides [12]. In the particular case of metal oxides, investigations of the process characteristics by Wallin and Helmersson [13] and Sarakinos *et al.* [14] during reactive sputtering of Al-O and Zr-O, respectively, have shown that, in contrast to direct current magnetron sputtering (DCMS), HiPIMS allows for elimination of the hysteresis and stabilization of the transition zone without the use of a feedback control systems. The observed behavior has been attributed to several mechanisms, i.e. depletion of reactive gas in front of the target during the pulse on-time [13,15,16] (referred to as gas rarefaction [17-21]), limited target oxidation during the pulse off-time [13], and higher target erosion rate due to the higher target voltage used in HiPIMS as compared to DCMS [14]. However, the question regarding the origin of the process stabilization in reactive HiPIMS still remains. Moreover, the process stabilization has only been reported for a narrow range of experimental parameters [13,14]. At the same time, Sproul *et al.* [3] have reported the need for a feedback control of the reactive gas partial pressure using HiPIMS, while Audronis *et al*. [22] have claimed that there is no evidence to support the hysteresis suppression/elimination using HiPIMS.

The goal of the present study is to explore the feasibility of HiPIMS to stabilize transition zone and eliminate the hysteresis during reactive deposition over a wide range of experimental parameters. In addition, we seek to contribute to the understanding of the fundamental mechanisms that determine the process characteristics in reactive HiPIMS

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processes. To this purpose we study the process characteristics during reactive HiPIMS and DCMS of Al and Ce in an Ar-O_2 ambient using a variety of experimental parameters with respect the pulsing frequency and the pumping speed. The results obtained are discussed in light of the mechanisms suggested in the literature. Finally Al-O films where grown and their optical properties was compared for the two deposition techniques.

2. Experimental procedure

Experiments were performed both in DCMS and in HiPIMS modes in an ultra high vacuum stainless-steel chamber with a base pressure below 10^{-6} Pa. Discs 50 mm in diameter and 3 mm in thickness made of Al (99.9995% purity) and Ce (99.9% purity) were used as sputtering targets. Ar gas with a purity of 99.9997% was introduced into the chamber through a mass flow controller and the Ar flow was adjusted to maintain a constant partial pressure ranging from 0.65 to 0.9 Pa depending on the experimental conditions. O_2 gas (99.9995% in purity) at various flows was introduced into the chamber through a fast solenoid valve connected to a partial pressure feedback system. An O_2 lambda probe (Zirox XS22.3H) was used as an O_2 partial pressure sensor. This experimental arrangement enabled to maintain control over the O_2 partial pressure at all values of O_2 flow employed and therefore operate within the transition sputtering zone. In order to monitor the O_2 partial pressure, a Spectra vision 1000-P differentially pumped mass spectrometer was used. In this experiment, the O_2 partial pressure values were calculated by measuring mass 32 and mass 40 of O_2 and Ar, respectively and using a relative sensitivity factor of Ar/O_2 = 1.2 [23]. For the experiments carried out in DCMS mode power was applied to the target by an MDX 1 K Pinnacle dc generator operated at constant power mode. In the HiPIMS case, unipolar pulses with a length (t_{ON}) of 35 μs and a frequency between 1 kHz to 10 kHz were used supplied by a SPIK 1000 A pulsing unit fed by the MDX generator. These relatively short pulse lengths were employed to minimize the probability for the occurrence of arcing during the process [18]. The discharge voltage was adjusted as the reactive gas flow was varied in order to keep the average power delivered to

the target constant. For experiments performed in DCMS mode, the target voltage, current and power were directly obtained from the readout of the dc generator. In the case of HiPIMS process both target current and voltage are time dependent quantities which were measured using a Chauvin Arnoux C 160 current clamp and 1:100 voltage divider, respectively, and monitored in a Tektronix TDS 520 C digital oscilloscope. The power was subsequently obtained by the product of the voltage and the current signals. The measurements of the time dependent target voltage revealed nearly rectangular waveforms. To determine the width of the hysteresis and thus evaluate the process stability at the various deposition conditions the target (discharge) voltage, the O_2 partial pressure, and the mass deposition rate (monitored by a quartz crystal microbalance) were recorded as functions of the O_2 flow. It is known that the hysteresis effect and the process stability are also influenced by the pumping speed of the system and the hysteresis may be eliminated if the pumping speed is higher than a critical value [4]. Therefore, the process characteristics during reactive DCMS and HiPIMS of the Al target were studied by varying the pumping speed from 20 to 50 l/s employing a throttle valve at the entrance of the turbomolecular pump. In this particular case the average power in the HiPIMS mode was adjusted so that the same mass deposition rate from pure metallic target (i.e. without the presence of a reactive gas) as for DCMS was obtained.

To unravel the effect of the process characteristics on the films properties, Al-O films were deposited onto Si (100) substrates located at a distance of 11 cm from the target employing both reactive DCMS and HiPIMS. Prior to the deposition the Si substrates were cleaned ultrasonically in acetone and isopropanol. Film uniformity was assured by substrate rotation. The Ar partial pressure was maintained at a fixed value of 0.9 Pa while the O_2 partial pressure was varied to grow films in the metallic, the transition, and the compound sputtering zones. Based on the deposition rate data the deposition time was adjusted to grow films with thickness of 200 nm. The effect of the deposition conditions on the atomic composition of the films was determined by means time-of-flight elastic recoil detection analysis (ToF-ERDA)

using a 40 MeV $^{127}I^{9+}$ ion beam. A detailed description of the ToF-ERDA experimental set-up has been presented elsewhere [24]. Film thickness and as a consequence the deposition rate was determined by scanning electron microscopy (SEM) using a LEO 1550 Gemini. The effect of the deposition conditions on the crystal structure was investigated by means of grazing incidence X-ray diffractometry (GIXRD). The GIXRD measurement was performed with Philips PW1830 diffractrometer operated at 40 kV and 40 mA with a Cu anode (Cu Kα, $\lambda = 1.540597$ Å). The incident beam angle, ω, was 1° while the scanning range in 2θ was 15 - 85°. In order to investigate the optical properties of the films, spectroscopic ellipsometry measurements were performed [25] using a dual rotating compensator ellipsometer (RC2) from J. A. Woollam Co., Inc. The ellipsometric data were recorded at angle of incidence of 45, 55, and 65° in the spectral range of 300 to 1700 nm with an increment of 1 nm. The optical properties and thickness of thin film may be derived by fitting of the relative amplitude change (Ψ) and the relative phase change (Δ) using the CompleteEASE software from J. A. Woollam Co., Inc. The optical response of the Al-O layer (refractive index, n, and extinction coefficient, k) was described using the Cauchy dispersion formula [26]. The thickness of the Al-O layer was obtained by the iteration process and compared to the value measured by SEM.

3. Results and discussion

3.1 Process characteristics

The discharge voltage, mass deposition rate, and O_2 partial pressure curves as functions of the O_2 flow during reactive DCMS of the Al target are shown in Fig. 1. An Ar partial pressure of 0.8 Pa, a pumping speed of 20 ls^{-1} and a constant power of 45 W were used. A typical hysteresis behavior can be observed. When the O_2 flow is increased, the discharge voltage exhibits an abrupt decrease at an O_2 flow value of about 0.61 sccm indicating the transition from the metallic to the compound sputtering mode (Fig. 1(a)). The transition from the compound back to the metallic mode (when the O_2 flow is decreased) takes place at an O_2

flow of about 0.58 sccm. Hysteresis is also observed for the mass deposition rate (Fig.1 (b)) and the O_2 partial pressure (Fig. 1 (c)). Feedback control of the reactive gas partial pressure was used, making all operating points inside the transition zone accessible. This results in the typical S-shaped curves shown in Fig. 1. In the case of a mass flow controlled process, no stable process parameters values can be obtained in the O_2 flow range designated by the vertical dotted lines in Fig. 1. The abrupt change at the critical gas flows occurs due to changes of the target surface composition, when a substantial fraction of the oxide is formed at the target. The change in discharge voltage is related to the change in secondary electron yield from oxide covered surface [27].

The discharge voltage, the mass deposition rate, and the O_2 partial pressure as function of the O_2 flow during reactive sputtering of the Al target are plotted in Fig. 2. The process characteristics of the reactive DCMS process (originally presented in Fig. (1)) are also plotted for reference. In all these experiments, the Ar partial pressure was 0.8 Pa at a pumping speed of 20 ls^{-1}. The pulsing frequency in the HiPIMS mode was 1, 2, and 4 kHz and the average target power was adjusted accordingly to match the deposition rate achieved by DCMS from a metallic target at a power of 45 W. In Fig. 2 only measurements for increasing O_2 flow are shown since the HiPIMS discharge was unstable at relatively high O_2 flows. Therefore, experiments were terminated when the discharge extinguished due to excessive arcing. Arcing also limited the energy per pulse that could be used. The process curves in HiPIMS exhibit a behavior completely different than those in DCMS. The transition from the metallic to the compound mode is without the S-shaped curves observed in processes with a hysteresis. This is indicative of suppression/elimination of the hysteresis. Comparison of the slope of the curves at the onset of the transition zone reveals that a frequency of 4 kHz results in a smoother change of the process characteristics allowing for a more gradual transition and hence better performance regarding the hysteresis behavior as compared to processes operated at 1 and 2 kHz. Operation at pulsing frequencies larger than 4 kHz was not possible when the

average target power of 45 W was used as a starting value for the DCMS experiments. The reason was that at frequencies larger than 4 kHz the peak target power measured at the oscilloscope was too low to guarantee an accurate control of the average power. Therefore, and in order to evaluate the effect of frequencies larger than 4 kHz on the process characteristics, discharge voltage-O_2 flow curves were recorded for reactive DCMS and HiPIMS employing a starting DCMS average power of 100 W. For these experiments the partial Ar pressure was fixed at 0.9 Pa, while the pumping speed was 20 ls^{-1}. The measurements are presented in Fig. 3 where it is again seen that HiPIMS processes with frequencies of 2 and 4 kHz exhibit a smooth transition zone as opposed to the DCMS process. However, further increase of the frequency to 10 kHz results again in a steep transition from the metallic to the compound mode similar to that in the DCMS process. It has to be pointed out here that at 10 kHz, the duty cycle is 35 % which is substantially higher than the typical values used in HiPIMS (<10 %). The peak power is low and similar to standard mid-frequency reactive magnetron sputtering. However, we use throughout the manuscript the term HiPIMS for all pulsing frequencies to avoid confusion when comparing the various experimental series. To rule out the possibility that the observed behavior is specific for Al and O system, another set of experiments was carried out using a Ce target. The deposition rate was not monitored in this particular experiment and DCMS and HiPIMS (frequencies of 1, 2, and, 4 kHz) were operated at the same constant average power of 70 W. The Ar partial pressure was 0.65 Pa at a pumping speed of 25 ls^{-1}. The discharge voltage-O_2 flow curves are plotted in Fig. 4. In the DCMS case, a relatively wide hysteresis is observed with critical flows of 0.80 and 0.63 sccm. The HiPIMS process exhibits a hysteresis too. However, the width of the hysteresis is substantially smaller than that DCMS and decreases with increasing frequency from 1 to 4 kHz. Higher frequencies were not evaluated. The shift in the critical flow towards lower O_2 mass flow for 1 and 2 kHz indicates lower deposition rate than in DCMS.

The effect of the pumping speed on the characteristics of the reactive HiPIMS and DCMS process were investigated by recording the target voltage-O_2 flow curves during reactive deposition of Al using a pumping speed of 50 ls^{-1}, a starting DCMS average target power of 100 W and a constant Ar partial pressure of 0.8 Pa. The results are presented in Fig. 5 where it can be seen that both DCMS and HiPIMS (pulsing frequency of 4 kHz) processes are hysteresis free. With HiPIMS however, the onset of the transition is delayed to higher values of O_2 flow and the initial slope of the process curve is more gradual. This fact, in combination with the results presented in Fig. 3, indicate that HiPIMS shift the process towards a reduction of the hysteresis and stabilization of the transition zone irrespective of the pumping speed.

A typical feature of the HiPIMS process is the relatively high peak target current which results in depletion (rarefaction) of neutral species in the vicinity of the target [18-21]. The rarefaction affects not only the Ar but also the reactive gas species [12,15]. It has been suggested in the literature [13,16] that the latter has as a consequence a lower effective flux of reactive species towards the target which in turn results in the elimination/suppressions of the hysteresis and the stabilization of the transition zone in reactive HiPIMS processes. The decrease of the pulsing frequency for a constant average power (or current) leads to larger peak target currents and thus more pronounced gas rarefaction. At the same time smaller frequencies imply longer time between the pulses which in turn means more time available for the gas to be replenished in the target's vicinity. These mechanisms may explain the fact that an optimum behavior with respect to the hysteresis width is obtained at intermediate pulsing frequencies (Figs. 3 and 4). Another mechanism that can be used to explain the behavior observed in Figs. 3 and 4 is based on the limited target oxidation between the pulses [13] caused by the reduced reactivity of O_2 under the absence of plasma [27]. Decrease of the frequency enhances the tendency for limited target oxidation during the pulse off-time since the time between pulses becomes longer. This is counteracted by the increase of the peak

target current which in turn leads to a larger plasma density and eventually a larger density of activated (i.e. excited) reactive gas species.

3.2 Film properties

The effect of the process characteristics on the film properties was evaluated by depositing Al-O films at various O_2 gas flows employing DCMS and HiPIMS (pulsing frequency of 4 kHz). Both processes were operated at a constant average target power of 100 W. Characterization of the reactive process by recording the target voltage-O_2 flow curves (not presented here) showed that the reactive HiPIMS process transition zone is stable and hysteresis free as opposed to the DCMS case in agreement with the results presented in Figs. 2 and 3. The chemical composition of the films as a function of the O_2 flow for DCMS and HiPIMS flow is plotted in Fig. 6. There it is seen that stoichiometric Al_2O_3 films are obtained for O_2 flow of 1.28 sccm for both DCMS and HiPIMS. The XRD measurements revealed that all films were X-ray amorphous (not presented here). Figures 7 (a) and (b) show typical ellipsometric spectra of films grown by DCMS and HiPIMS 4 kHz, respectively. The results show excellent agreement between the experimental data (symbols) and the fit based on the Cauchy model (solid lines). In Fig. 7 (c), the calculated optical constants of deposited films are plotted. It can be seen that both films are transparent in the visible spectral range. The refractive index, *n* and extinction coefficient, *k* of the Al-O films at a wavelength of 630 nm are presented in Fig. 8. It is observed that transparent films (i.e. film with a zero extinction coefficient *k*) are obtained for O_2 flows equal to or larger than 1.28 and 1.34 sccm for HiPIMS and DCMS, respectively. This in contrast to ToF-ERDA analysis (Fig. 6) which shows the formation of Al_2O_3 films at O_2 flows of 1.28 sccm for both cases. This indicates that the optical constants are very sensitive to the marginal variations of the composition which cannot be resolved by ToF-ERDA in accordance to previous reports [28]. Furthermore, it can be seen that the refractive index for transparent Al-O films lies in the range 1.55-1.68. These values are smaller than those for crystalline bulk Al_2O_3 ($n \sim 1.76$ [29]) and can be explained by the

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amorphous structure of the films which results in a lower mass density than that of crystalline bulk Al_2O_3. Finally, the effect of the O_2 flow on the deposition (growth) rate of the Al-O films during DCMS and HiPIMS is demonstrated in Fig. 9. While similar deposition rates for both techniques are obtained up to O_2 flow of ~1.4 sccm, above this value the DCMS rate drops abruptly. This implies that HiPIMS allows for the growth of stoichiometric and transparent Al_2O_3 films at relatively high deposition rates over a wider range of experimental conditions as compared to DCMS. The drop in the HiPIMS deposition rate around 1.2 sccm was reproducible and corresponds to a region with unstable discharge voltage where the voltage needed to be continuously increased to maintain a constant power. The duty cycle for 4 kHz, 14 %, is substantially higher than typical values used in HiPIMS. However, there is still a substantial improvement in the film quality, indicated by the refractive index, as compared to DCMS.

4. Conclusions

In the present study the hysteresis behavior and the characteristics of the transition zone during reactive HiPIMS of Al and Ce targets in an Ar-O_2 atmosphere have been investigated over a large range of experimental parameters with respect to the pulsing frequency and the pumping speed. Our results revealed that HiPIMS allows for suppression/elimination of the hysteresis observed in DCMS and a smoother transition from the metallic to the compound sputtering mode. In the used deposition system, optimum behavior with respect to the hysteresis width has been obtained at frequency values between 2 and 4 kHz, while HiPIMS processes with values below or above this range resemble the DCMS behavior. The relationship between the pulsing frequency and the hysteresis behavior has been tentatively explained on the basis of the gas rarefaction and the limited target oxidation between the power pulses. The elimination/suppression of the hysteresis in HiPIMS has facilitated the growth of stoichiometric and transparent Al_2O_3 at relatively high deposition rates over a wider range of experimental conditions as compared to DCMS.

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Fig. 1. (a) Discharge voltage, (b) mass deposition rate and (c) O_2 partial pressure as a function of the O_2 flow during reactive DCMS of the Al target. Experimental conditions: Ar pressure of 0.8 Pa, constant power of 45 W and the pumping speed of 20 ls^{-1}.

Fig. 2. (a) Discharge voltage, (b) mass deposition rate (c) and O_2 partial as a function of the O_2 flow during reactive HiPIMS of the Al target at pulsing frequencies of 1, 2 and 4 kHz. The process characteristics of the reactive DCMS process (originally presented in Fig. (1)) are also plotted for reference. Experimental conditions: constant Ar pressure of 0.8 Pa at a pumping speed of 20 ls^{-1}. The average target powers in HIPIMS were adjusted to achieve the same mass deposition rate from a metallic target as in DCMS.

Fig. 3. Discharge voltage as a function of the O_2 flow during reactive DCMS and HiPIMS of the Al target. For the HiPIMS process pulsing frequencies of 2, 4 and 10 kHz were employed. The average target power in DCMS was 100 W. The average target powers in HIPIMS were adjusted to achieve the same mass deposition rate from a metallic target as in DCMS. Experimental conditions: constant Ar pressure of 0.9 Pa at a pumping speed of 20 ls^{-1}.

Fig. 4. Discharge voltage as a function of the O_2 flow during reactive DCMS and HiPIMS of the Ce target. For the HiPIMS process pulsing frequencies of 1, 2 and 4 kHz were employed. All processes were operated at a constant average target power of 70 W. The Ar partial pressure was 0.65 Pa at a pumping speed 25 ls^{-1}.

Fig. 5. Discharge voltage as a function of the O_2 flow during reactive DCMS and HiPIMS of the Al target. For the HiPIMS process a pulsing frequency of 4 kHz was employed. The

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average target power in DCMS was 100 W. The average target powers in HiPIMS were adjusted to achieve the same mass deposition rate from a metallic target as in DCMS. Experimental conditions: constant Ar pressure of 0.8 Pa at a pumping speed of 50 ls^{-1}.

Fig. 6. Composition of Al-O films deposited at different O_2 flows by DCMS and HiPIMS 4 kHz as measured by ERDA.

Fig. 7. Typical measured and fitted spectra on the Al_2O_3 films at incident angle 55° deposited by (a) DCMS and (b) HiPIMS 4 kHz. The symbols correspond to the experimental data and the lines to the best fit employing the Cauchy model. (c) Refractive indices, n, and extinction coefficients, k of the Al_2O_3 films deposited by DCMS and HiPIMS 4 kHz, derived from Fig. 7 (a) and (b).

Fig. 8. Refractive index n (solid line) and extinction coefficient k (dot line, open symbols) at 630 nm comparing DCMS and HiPIMS 4 kHz.

Fig. 9. Deposition rate calculated from the thickness of deposited films as a function of the O_2 flow during reactive DCMS and HiPIMS 4 kHz of the Al target.

(a) DCMS

Experiment data (ψ)
Fit
Experiment data (Δ)
Fit

(b) HiPIMS 4 kHz

Experiment data (ψ)
Fit
Experiment data (Δ)
Fit

(c)

n HiPIMS 4 kHz
n DCMS
k HiPIMS 4 kHz
k DCMS

Wavelength (nm)