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ABSTRACT

High power impulse magnetron sputtering (HIPIMS) of an Al target in Ar/O_2 mixtures has been studied. The use of HIPIMS is shown to drastically influence the process characteristics compared to conventional sputtering. Under suitable conditions, oxide formation on the target as the reactive gas flow is increased is suppressed, and the hysteresis effect commonly observed as the gas flow is varied during conventional sputtering can be reduced, or even completely eliminated, using HIPIMS. Consequently, stoichiometric alumina can be deposited under stable process conditions at high rates. Possible explanations for this behavior as well as a model qualitatively describing the process are presented.

Keywords: Reactive Sputtering; High Power Impulse Magnetron Sputtering; Alumina; Process modeling.

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High power impulse magnetrons sputtering (HIPIMS) is a novel ionized physical vapor deposition technique in which high power pulses with low duty cycle are applied to a conventional sputtering magnetron [1]. In this way, a dense plasma is created, while the average power and thereby the target heating is kept at a moderate level. As a consequence of the dense plasma, a high ionized fraction of the sputtered species can be achieved [1]. Recently, reactive sputter deposition of both nitride [2] and oxide [3,4,5] thin films using this technique has been shown feasible. For example, Sarakinos et $al.$ [4] studied reactive HIPIMS of a $TiO_{1.8}$ target in an Ar/O_2 environment with results indicating that the deposition rate achieved under some conditions is higher than during DC sputtering with the same average current. They also observed that the target voltage change associated with the oxidation of the target occurred at higher O_2 flows in the HIPIMS case. In this letter, the reactive HIPIMS process has been investigated using the challenging example of reactive sputtering of an Al target in Ar/O_2 mixtures as model system. Previous studies have shown that reactive HIPIMS deposition of stoichiometric alumina is possible [5]. In the present work, the impact of the pulsed target power on the reactive process has been closely investigated showing that the use of HIPIMS can lead to many beneficial process properties such as reduced or eliminated hysteresis effects, higher deposition rates, and more stable process conditions.

Experiments were carried out in an ultra high vacuum system with a base pressure < 2×10^{-6} Pa, equipped with a planar circular magnetron (50 mm in diameter). A three mm thick Al disc (99.999 % purity) was used as sputtering target. The Ar (99.9997 % purity) sputtering gas as well as the reactive O_2 gas (99.9995 % purity) were introduced into the chamber through mass flow controllers. The Ar gas flow was

adjusted so that a total pressure of 0.8 Pa was achieved. Measurements were performed for both ordinary DC power delivered by an MDX 1K power supply (Advanced Energy) operating in constant power mode (70 W), as well as for HIPIMS power supplied by a SPIK 1000A pulsing unit (Melec) fed by a constant voltage from an MDX Pinnacle DC supply (Advanced Energy). During the HIPIMS measurements the voltage and current was monitored using a digital oscilloscope. The pulsing unit used produces approximately square voltage pulses with a specified length and repetition frequency. In the measurements presented here, a pulse length of 35 μs and a repetition frequency of 1 kHz were used. The magnitude of the discharge voltage during pulses was adjusted so that the average power delivered to the target was kept constant as the reactive gas flow was varied. In this work, peak voltages in the range of 600 − 800 V were applied, resulting in peak currents of 7 − 25 A. As the O_2 gas flow was varied, the O_2 partial pressure and the deposition rate were monitored. The rate of deposited mass was measured using a quartz crystal microbalance rate monitor (Leybold Inficon), while the O_2 partial pressure was deduced by monitoring the total pressure and the Ar flow. Figure 1 shows the variation in mass deposition rate and O_2 partial pressure as the O_2 gas flow was varied during both HIPIMS and DC sputtering. In order to facilitate comparisons of the behavior of the reactive process, the average power in the HIPIMS measurements was adjusted so that the same deposition rate for pure metal as for DC sputtering was obtained. This means that a higher average power (~ 60 %) had to be used for HIPIMS in order to compensate for the lower deposition rate generally observed with this technique [6]. The DC measurements resemble the behavior expected from previous studies of reactive sputtering [7]. As the reactive gas

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is introduced, the rate of deposited mass first increases as both the metal and oxygen
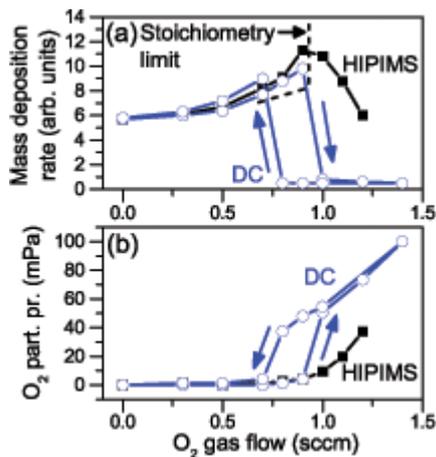

Figure 1. Rate of deposited mass (a) and O₂ partial pressure (b) as a function of O₂ gas flow for HIPIMS and DC sputtering. The approximate limit for deposition of stoichiometric alumina is shown in (a). Measurements for higher O₂ flows than indicated for HIPIMS were unreliable due to target arcing. Lines are guides for the eye only.

deposits onto the substrate [Fig. 1(a)]. However, as oxide compound starts to form on the target surface, the sputtering and deposition rates drop due to a drastic decrease in sputter yield as well as a drop in target voltage due to an increased ion induced secondary electron emission coefficient [8]. As the target surface is covered with oxide (commonly referred to as poisoned mode), the deposition rate levels out and varies only slightly as the reactive gas flow is increased further. Since a fairly low pumping speed was used, the DC process expectedly experiences a hysteresis behavior [9]; as the gas flow is reduced after the target has reached the poisoned mode, the target stays in the poisoned mode until the flow is reduced to a point below where the deposition rate first dropped. The corresponding effect can be observed for the O₂ partial pressure [Fig. 1(b)]; as the reactive gas is first introduced, the O₂ partial pressure stays low due to consumption of gas at chamber walls and other collecting areas. However, as a certain critical gas flow is reached, the partial pressure experiences a sudden increase and then varies linearly with the flow. Also here, a

hysteresis effect can be observed in the DC case. Measurements done using the HIPIMS technique exhibit a distinctly different behavior. The transition to the poisoned mode is considerably less abrupt, as seen both in the deposition rate [Fig. 1(a)] and the O_2 partial pressure [Fig. 1(b)], and the maximum deposition rate is higher. For the O_2 flows studied, no hysteresis was observed in the HIPIMS case, but it should be noted that reliable measurements for a completely oxidized target were impossible due to target arcing at higher target oxide coverage. Deposition of stoichiometric alumina coatings usually occurs in the region of the transition to compound mode as indicated in Fig. 1 [7] (The approximate stoichiometry limit was in this work investigated by studying the optical transparency of depositions made onto Si substrates. A tendency of obtaining stoichiometric films for lower O_2 partial pressures with HIPIMS than DC was observed. The effect was rather small and should be investigated further, but could be an indication of an increased reactivity of the reactive gas in the HIPIMS case.) Hence, to be able to achieve stoichiometric films at reasonable deposition rates, operation in, or close to, the unstable transition region is required. As a consequence, industrial-scale deposition processes of alumina are subject to process instabilities and most often special measures has to be taken in order to control the process. Possible solutions include using higher total pressures [10] or controlling the reactive gas flow with respect to O_2 partial pressure (as determined by, e.g., mass spectrometry or optical emission spectroscopy) or the target voltage [7]. Our results show that the use of HIPIMS can provide a remedy to this problem, thereby reducing the need for feedback control systems. In addition, the wider process window obtained might, e.g., make the process less unstable with respect to different process fluctuations and improve the stoichiometry homogeneity at different positions in the deposition chamber. The highest deposition rates reported

using DC sputtering with feedback control of the reactive gas is around 70 % of the metal rate (in terms of thickness) [7]. Assuming densities of 2.7 and 3.6 g/cm^3 for Al and (amorphous) Al$_2$O$_3$, respectively, the corresponding value for HIPIMS is ~ 140 % for the situation in Fig. 1(a). Thus, when comparing sputtering at similar average target powers, the deposition rates in the two cases are comparable. Furthermore, the ionized deposition flux in the HIPIMS discharge opens new possibilities of optimizing the coating properties as compared to DC sputtering. It should be noted that no attempts of optimizing the pulse parameters were done in this study, and it is plausible that the process properties are fairly sensitive to the choice of pulsing configuration. Thus, further improvements of the process conditions are presumably possible.

We suggest the observed effects on the process characteristics to be due to the pulsed nature of the target erosion; the high erosion rate during the pulses effectively cleans the target surface and displaces the oxidation to higher flows, while target poisoning between pulses, i.e. with limited plasma activation, is expected to be slight, since fairly high levels of gas exposure have been shown necessary in order to cause compound formation in such cases [11]. Hence, the effect can be considered to be similar to that previously reported to occur when applying a high current density to a small, movable erosion zone on the target (e.g. by utilizing a movable magnet) [12], but in the present case being caused by the temporal distribution of the erosion, rather than a spatially distributed one. In order to gain a better understanding of the process, a simple model was created. The model is a modified version of the classical model for reactive sputtering commonly referred to as the Berg model [9]. In this model, a set of balance equation are set up as detailed in ref. 9, describing the flux of metal and reactive gas between target, collecting areas (including substrate), and the pump. In

particular, the equation describing the steady-state compound coverage at the target, θ_t, takes the form

$$\frac{JY_c\theta_t}{q} - \alpha 2F(1-\theta_t) = 0, \tag{1}$$

where J is the target current density, Y_c the compound sputtering yield, q the charge of the incoming ions, α the sticking coefficient for the reactive gas at the target, and F the incoming flux of reactive gas molecules. In order to account for the higher target erosion rate during pulses and the reduced oxidation between pulses as described above, an effective target current density, $J_{eff} > J$, was introduced, replacing J in Eq. (1). Note that an increased effective target current density also accounts for a possible rarefaction of reactive gas in front of the target, previously suggested to occur due to the large flux of material during pulses [4], as manifested by a reduced value of F. The average current is, however, still used to calculate the flux of material from the target to the collecting areas. Assuming a continues averaged flux of material to the collecting areas is, at least for the higher range of frequencies used in HIPIMS processes, justified by the energy spread of the sputtered species and the fact that the time required for the species to reach the substrate is of the order of the pulsing period time [13]. Figure 2 shows results from simulations with parameters resembling the experimental situation shown in Fig. 1 for both DC ($J_{eff} = J_{ave} = 0.2$ A) and HIPIMS conditions ($J_{eff} = 0.7$ A, $J_{ave} = 0.2$ A). As can be seen, the model qualitatively reproduces the experimental observations. The hysteresis behavior occurring in the DC case is eliminated under HIPIMS conditions. [Note that not all points shown in the S-shaped curves are possible to reach unless feedback control of the reactive gas flow is used. If constant gas flow is used, sudden jumps will occur as indicated by the dashed lines in Fig. 2(a).] The target coverage of compound is displaced towards
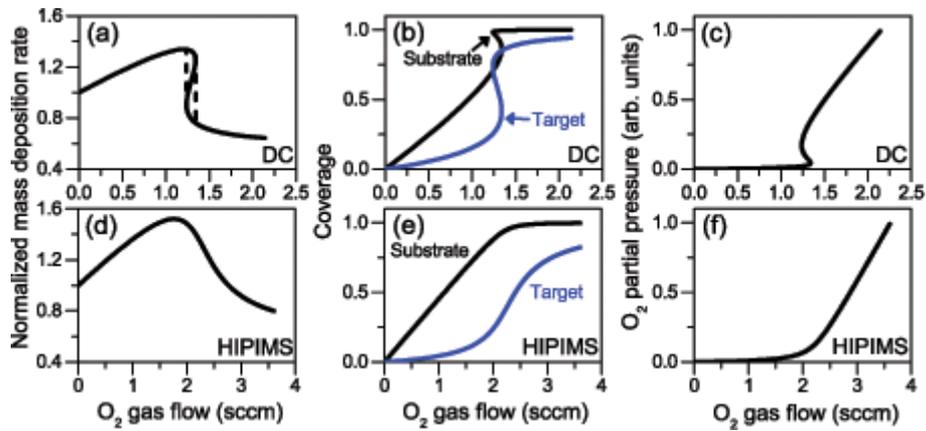
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Figure 2. Modeling results for the rate of deposited mass [(a) and (d)], compound coverage at target and substrate [(b) and (e)], and O₂ partial pressure [(c) and (f)] for both DC sputtering [(a), (b), and (c)] and HIPIMS [(d), (e), and (f)]. Parameters were chosen to mimic the experimental situation in Fig. 1.

higher gas flows in the HIPIMS case, whereas the compound coverage at the substrate is more similar for DC and HIPIMS, as shown in Figs. 2(b) and (e). The increase in O_2 partial pressure as the gas flow is increased beyond the point where all gas is consumed by the collecting areas follows a similar trend [Figs. 2(c) and (f)]. For simulations of systems exhibiting very strong hysteresis in the DC case, the hysteresis was reduced, but not completely eliminated, using HIPIMS (not shown).

In conclusion, the process behavior during reactive sputtering using the HIPIMS technique has been experimentally studied and modeled. Under suitable conditions, the use of HIPIMS gives rise to numerous beneficial process properties such as a reduced or eliminated hysteresis effect, less abrupt transition to the poisoned target mode, and a higher maximum deposition rate compared to DC sputtering with the same initial metal rate. The results are of large practical importance for industrial-scale deposition of, e.g., alumina, where the need to operate at an unstable point

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where stoichiometric films are deposited at reasonable rates poses a substantial problem.

[1] U. Helmersson, M. Lattemann, J. Bohlmark, A.P. Ehiasarian, and J.T. Gudmundsson, Thin Solid Films 513 (2006) 1.

[2] A.P. Ehiasarian, W.-D. Münz, L. Hultman, U. Helmersson, and I. Petrov, Surf. Coat. Technol. 163 (2003) 267.

[3] S. Konstantinidis, J.P. Dauchot, and M. Hecq, Thin Solid Films 515 (2006) 1182.

[4] K. Sarakinos, J. Alami, and M. Wuttig, J. Phys. D: Appl. Phys. 40 (2007) 2108.

[5] W.D. Sproul, D.J. Christie, and D.C. Carter in: Proceedings of the 47th Annual Technical Conference of the Society of Vacuum Coaters, April 24–29, 2004, Dallas, TX, USA (2004) 96.

[6] J. Alami, K. Sarakinos, G. Mark, and M. Wuttig, Appl. Phys. Lett. 89 (2006) 154104.

[7] W.D. Sproul, D.J. Christie, and D.C. Carter, Thin Solid Films 491 (2005) 1.

[8] S. Maniv and W. D. Westwood , J. Vac. Sci. Technol. 17 (1980) 743.

[9] S. Berg and T. Nyberg, Thin Solid Films 476 (2005) 215.

[10] M. Kharazzi Olsson, K. Macák, U. Helmersson, and B. Hjörvarsson, J. Vac. Sci. Technol. A 16 (1998) 639.

[11] D. Depla and R. De Gryse, J. Vac. Sci. Technol. A 20 (2002) 521.

[12] T. Nyberg, S. Berg, U. Helmersson, and K. Hartig, Appl. Phys. Lett. 86 (2005) 164106.

[13] L.B. Jonsson, T. Nyberg, I. Katardjiev, and S. Berg, Thin Solid Films 365 (2000) 43.