Vacuum 124 (2016) 1–4

Contents lists available at ScienceDirect

Vacuum

journal homepage: www.elsevier.com/locate/vacuum

Short communication

Peak amplitude of target current determines deposition rate loss during high power pulsed magnetron sputtering

G. Greczynski*, L. Hultman

Thin Film Physics Division, Department of Physics (IFM), Linköping University, SE-581 83, Linköping, Sweden

ARTICLE INFO

Article history:
Received 1 October 2015
Accepted 6 November 2015
Available online 12 November 2015

Keywords:
HIPIMS
Growth rate
Deposition rate
Magnetron sputtering
Ionized PVD

ABSTRACT

Film growth rates during DCMS and HIPIMS sputtering in Ar are measured for ten technologically-relevant elemental target materials: Al, Si, Ti, Cr, Y, Zr, Nb, Hf, Ta, and W, spanning wide range of masses, ionization energies, and sputter yields. Surprisingly, the ratio of power-normalized HIPIMS and DCMS rates α decays exponentially with increasing peak target current density J_T^{max} for all metals. The effect of J_T^{max} on α is dramatic: $\alpha \approx 1$ in the limit of lowest J_T^{max} values tested (0.04 A/cm^2) and decreases to only 0.12 with J_T^{max} ~3 A/cm^2. With the exception of Al and Si, $\alpha(J_T^{max})$ curves overlap indicating that the debated rate loss in HIPIMS is to large extent determined by the peak amplitude of the HIPIMS target current for all tested metals. Back attraction of ionized target species is responsible for such large variation in α.

High power pulsed magnetron sputtering (HIPIMS or HPPMS) [1] is a valuable addition to magnetron-sputtering-based PVD techniques [2–5]. HIPIMS is particularly attractive for the growth of transition metal (TM) nitride layers for two main reasons: (i) the ability to ionize up to 90% of the sputtered metal flux with an option to control the ionization degree by varying the peak target current density [6,7], and (ii) the time separation of metal- and gas-ion fluxes from the target [8]. The former effect is ascribed to the effective electron-impact ionization in the high-density plasma region [9] formed, for a short period of time, 20–100 μs, in front of the sputtering target operated in HIPIMS mode. The latter effect, caused by a strong gas rarefaction taking place during the high-power pulse [10–12] allows the control of metal-ion momentum and energy arriving onto a growing film, through the synchronization of the substrate bias pulse to the metal-ion rich part of the sputter-deposited flux [13,14], with decisive influence on film nanostructure, phase content, and stress state [15,16].

It was postulated early that the benefits of HIPIMS in terms of high ionization of the target material come at the expense of a lowered deposition rate [17–20], implying worse process economics which is currently a perceived obstacle for industrial implementation. Numerous phenomena have been proposed to account for this behavior including (i) the so-called "return effect" - an effective capturing of the ionized portion of the sputter-ejected flux by the cathode field [21,22], (ii) an enhanced azimuthal ion transport (across the magnetic field lines) [23], (iii) a "yield effect" since usually HIPIMS operates at higher target voltage V_T than in conventional DC magnetron sputtering (DCMS) one may expect lower power-normalized rate due to the fact that sputter yields are not directly proportional to V_T [24], and (iv) "species effect" related to the change of effective sputter yield once gas ions are replaced with the back-attracted target ions in the later phase of the pulse [25].

The reported values for the loss of power-normalized HIPIMS deposition rate $\overline{R}_{HIPIMS}$ show large spread even for the same material system. For example, if referred to the power-normalized DCMS rate $\overline{R}_{DCMS}$, the ratio $\overline{R}_{HIPIMS}/\overline{R}_{DCMS}$ varies from 15% to 75% for Ti, and from 37% to 80% in the case of Cu [2,26]. This may be due to different HIPIMS settings and prevents systematic comparisons between material systems, as well as, identification of key parameters for the here contested effect of HIPIMS rate loss.

We performed film growth rate measurements under identical conditions for ten elemental targets representing widely different masses, melting temperatures, sputtering yields, ionization potentials, and secondary electron emission yields during HIPIMS and DCMS in Ar atmosphere. The HIPIMS peak target current density J_T^{max} is varied by two orders of magnitude, from 0.04 to 3 A/cm^2, by means of changing the average power settings at constant pulsing frequency. We find that materials that are prone to high target current densities during HIPIMS discharge have second ionization

* Corresponding author.
E-mail address: grzgr@ifm.liu.se (G. Greczynski).

http://dx.doi.org/10.1016/j.vacuum.2015.11.004
0042-207X/

potential IP_T^2 lower than the first ionization potential of Ar IP_{Ar}^1. This results in higher concentrations of doubly-ionized sputter-ejected species that, in turn, enable effective potential emission of secondary electrons during the metal (self-sputtering) phase and, hence, enhanced ionization of sputtering gas. For all materials, the ratio of power normalized HIPIMS and DCMS rates α decays exponentially with increasing peak target current density J_T^{max}. Surprisingly, with the exception of Al and Si, $\alpha(J_T^{max})$ curves overlap indicating that the rate loss in HIPIMS is to large extent determined by the peak amplitudes of the HIPIMS target currents.

Aluminum, Si, Ti, Cr, Y, Zr, Nb, Hf, Ta, and W films are grown in a CC800/9 CemeCon AG magnetron sputtering system using rectangular 8.8 × 50 cm^2 targets that are virgin to exclude possible influence of the target history. The same magnetron is used in the experiments to ensure the same magnetic field strength and distribution. Prior to film growth each target undergoes a sputter-in sequence consisting of (1) DCMS operation with the average power $\langle P\rangle_T$ increasing from 1 to 8 kW in steps of 1 kW and 5 min per step, followed by (2) 1 h DCMS with $\langle P\rangle_T = 4$ kW, and (3) HIPIMS with average power set to 2 kW and a frequency f of 1000 Hz resulting in rather low target current density values which increase as f is gradually decreased to 500, 300, 200, and 100 Hz. Finally, frequency is set to 200 Hz (as for the film growth) and $\langle P\rangle_T$ is scanned from 0.4 to 4 kW during less than 15 min in order to assure stable operation in a wide current density range.

Si(001) substrates, 2 × 1 cm^2, are cleaned sequentially in acetone and isopropyl alcohol, and mounted at a distance of only 6 cm from the target to minimize the influence of gas scattering [27]. The growth chamber is degassed prior to deposition during a 2-h-long heating cycle so that the system base pressure is 2.3×10^6 Torr (0.3 mPa). Targets are operated in pure Ar with the gas flow f_{Ar} set at 420 cm^3/min, resulting in the total pressure of 3 mTorr (0.4 Pa). The substrate temperature is 400 °C.

Two multilayer films are grown for each material system, one in HIPIMS and one in DCMS mode, with the average target power $\langle P\rangle_T$ varied from 0.4 to 4 kW in steps of 0.4 kW. During HIPIMS experiments the pulsing frequency f is fixed at 200 Hz, thus variation in $\langle P\rangle_T$ corresponds to changing the pulse energy E_p from 2 to 20 J in steps of 2 J ($\langle P\rangle_T$ and E_p are related by $\langle P\rangle_T = f \times E_p$). While the preset pulse length T_{ON} is 200 μs, the effective pulse length τ, defined here as the time period during which $J_T(t) > 0.1 \times J_T^{max}$, may vary depending on $\langle P\rangle_T$ and target material. Resulting film thickness of individual layers is in the range from 350 to 650 nm. A 50-nm-thick TiN marker layers are deposited after each step in order to facilitate the layer thickness determination by cross-sectional scanning electron microscopy (XSEM) analyses. For the TiN growth in mixed Ar/N_2 atmosphere, the target is protected by cathode shutters to avoid cross-contamination from the neighboring cathode equipped with Ti target. All tested targets are pre-sputtered behind close shutters to remove possibly formed surface nitride layer following each TiN deposition step. A negative dc substrate bias, $V_s = -60$ V is used during DCMS film growth, while pulsed bias voltage with the same amplitude and 4% duty cycle, synchronized with the HIPIMS pulse, is used for HIPMS experiments [13,14].

Target current $I_T(t)$ and target voltage $V_T(t)$ waveforms during film growth are recorded with a Tektronix 500 MHz bandwidth digital oscilloscope. The target current density $J_T(t)$, a more generic quantity, is obtained by dividing $I_T(t)$ with the entire target area $A = 440$ cm^2.

Fig. 1(a–d) shows $J_T(t)$ and $V_T(t)$ waveforms recorded with the pulse energy $E_p = 18$ J during the HIPIMS operation of different metal targets in Ar. Data presented reveal large differences with respect to current–voltage performance between the materials. Based on the appearance of $J_T(t)$ and $V_T(t)$ curves as well as their evolution with E_p (not shown) the target materials can be divided

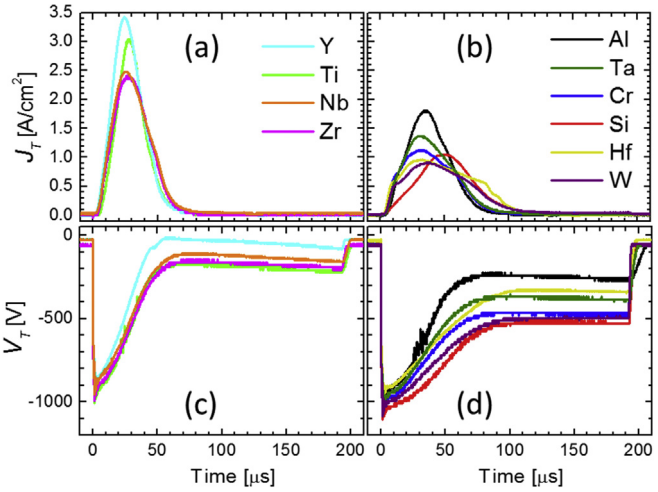

Fig. 1. (a–b) Target current density $J_T(t)$, and (c–d) target voltage $V_T(t)$ waveforms recorded during HIPIMS process with pulse energy of 18 J for Al, Si, Ti, Cr, Y, Zr, Nb, Hf, Ta, and W targets.

into two categories. The first category, including Ti, Zr, Nb, and Y (Fig. 1(a) and (c)), is characterized by narrow, high-amplitude current pulses, and relatively short triangular voltage waveforms that posses low amplitudes towards the end of the HIPIMS pulse [28]. For materials in the second category (Si, Cr, Hf, Ta, and W), current pulses are relatively broad with low-amplitudes, while $V_T(t)$ traces are characterized by higher voltage values towards the end of the pulse. Al target constitutes an intermediate case between category I and II metals. Trends outlined here are independent of the average power setting.

In the case of category I metals the peak target current density J_T^{max} reaches 2.4 A/cm^2 for Zr and Nb, 3 A/cm^2 for Ti, and 3.4 A/cm^2 for Y. The effective pulse lengths τ is in the range from 51 to 59 μs. As a consequence of these extremely high currents PS capacitors get almost completely emptied resulting in low V_T values towards the end of the pulse (cf. Fig. 1(c)). Materials in this category are characterized by rather low second ionization potential IP_T^2 values (cf. Table 1), all satisfying the criterion $IP_T^2 < IP_{Ar}^1$ ($IP_{Ar}^1 = 15.76$ eV) [29] meaning that there is a significant electron population in the discharge with energies in the range $IP_T^2 < E_e < IP_{Ar}^1$, i.e., too low to ionize Ar, yet high enough to produce doubly-ionized target ions [30]. This is of key importance in HIPIMS plasmas, since the primary ionization mechanism is by electron impact [9]. As pointed out by Anders et al. [31] the presence of higher charge state species is essential for the secondary electron emission during the metal-dominated (self-sputtering) phase of the HIPIMS pulse, when concentrations of inert gas ions is lowered due to rarefaction effects [10–12]. For relatively low ion energy used in sputtering, secondary electrons are predominantly ejected by potential emission, which requires that the condition [32].

$$0.39IP > \phi_T \tag{1}$$

in which ϕ_T is the target work function [33], is satisfied. It follows from data presented in Table 1 that all IP_T^1 values are too low to fulfill Eq. (1) and doubly-charged species are necessary for the potential emission to set in, in the absence of gas ions. Eq. (1) is readily satisfied in the case of category I metals, for which higher concentrations of doubly-ionized species are indeed expected, leading to effective secondary electron emission during the metal (self-sputtering) phase, resulting in thorough ionization of the working gas (diluted due to rarefaction) and high target current