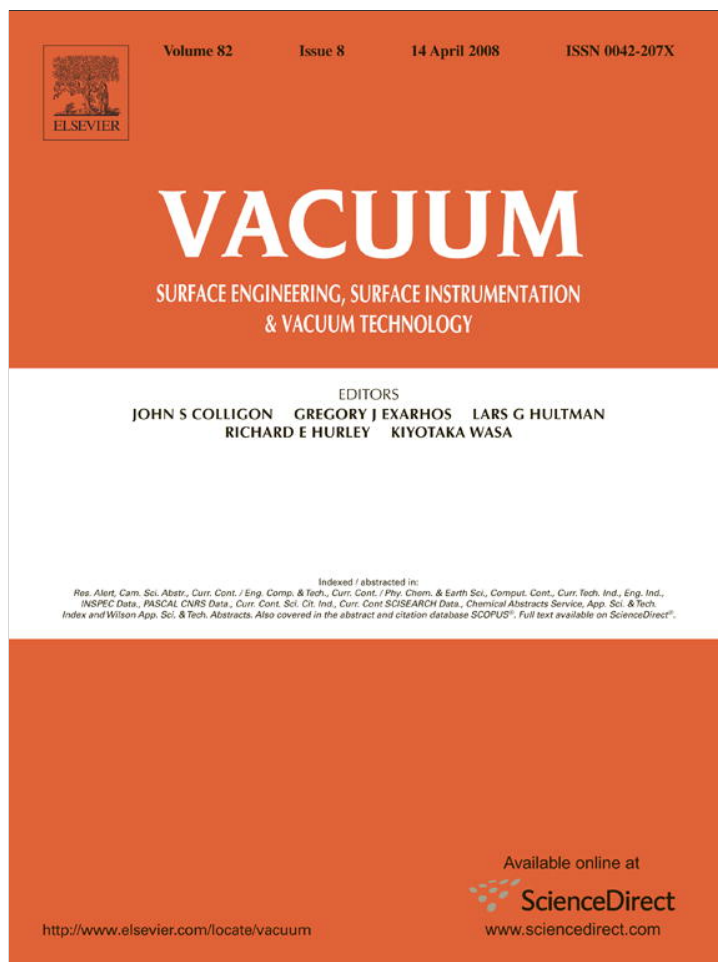


Provided for non-commercial research and education use.
Not for reproduction, distribution or commercial use.



This article appeared in a journal published by Elsevier. The attached copy is furnished to the author for internal non-commercial research and education use, including for instruction at the authors institution and sharing with colleagues.

Other uses, including reproduction and distribution, or selling or licensing copies, or posting to personal, institutional or third party websites are prohibited.

In most cases authors are permitted to post their version of the article (e.g. in Word or Tex form) to their personal website or institutional repository. Authors requiring further information regarding Elsevier's archiving and manuscript policies are encouraged to visit:

<http://www.elsevier.com/copyright>



Short communication

The physical reason for the apparently low deposition rate during high-power pulsed magnetron sputtering

Jens Emmerlich*, Stanislav Mráz, Rony Snyders, Kaiyun Jiang, Jochen M. Schneider

Materials Chemistry, RWTH Aachen University, Kopernikusstr. 16, D-52074 Aachen, Germany

Received 21 August 2007; received in revised form 30 October 2007; accepted 31 October 2007

Abstract

In high-power pulsed magnetron sputtering, a large power density is applied giving rise to a high degree of ionization. From an application point of view, the major drawback of this technology is the considerably lower deposition rate as compared to DC magnetron sputtering. Using transport-of-ions-in-matter simulations, we show that the apparently low deposition rate can be understood based on the non-linear energy dependence of the sputtering yields. Our calculations are consistent with deposition-rate measurements on Cu films as well as with published deposition-rate data for Ti [Konstantinidis S, Dauchot JP, Ganciu M, Ricard A, Hecq M. *J Appl Phys* 2006;99:013307].

© 2007 Elsevier Ltd. All rights reserved.

Keywords: Pulsed plasma; Magnetron sputtering; Deposition rate

1. Introduction

Ehiasarian et al. [1] reported two operation modes for magnetron sputtering based on the current–voltage discharge characteristic. The dependence of the discharge current, I , versus discharge voltage, U , can be described by a power law where $I = kU^n$ [2]. For conventional DC magnetron sputtering (DCMS), n equals 7–14 [2]. At target current density values of $>600 \text{ W/cm}^2$, the exponent n is approximately 1 and the mode of operation is changing into the so-called high-power pulsed magnetron sputtering (HPPMS) mode [1]. HPPMS was first reported by Kouznetsov et al. [3] in 1999 estimating the degree of ionization for sputtering Cu in Ar to be in the order of 70%. As a consequence of the large dissipated power, plasma density values of $\sim 10^{12} \text{ cm}^{-3}$ can be obtained [4,5]. The advantages of utilizing ionized vapor deposition techniques from an application point of view have recently been reviewed by Helmersson et al. [6], while the structure evolution during ion-assisted growth has been discussed by Petrov et al. [7]. Examples for advantages of the HPPMS

technique include uniform deposition of structures with high aspect ratio [3], interface modification through ion irradiation [8], and increased film density [9,10].

These advantages are generally associated with a large degree of ionization of the sputtered flux. Macák et al. [4] confirms that, besides the sputtering gas, a substantial amount of sputtered Ti is ionized ($\sim 40\%$). The fact that HPPMS is characterized by a large degree of ionization of the sputtered flux is well established [1,11–13].

Helmersson et al. [6] reviewed the available deposition-rate data and summarized that the rates are typically in the order of 25–35% of the rates in DCMS at similar time-averaged power values. From an application point of view, this is probably the largest drawback of the HPPMS technique. Christie [14] suggested that self-sputtering significantly affects the deposition rate during HPPMS, while Konstantinidis et al. [15] stressed the importance of the plasma conductivity. Furthermore, Bugaev et al. [16] and Böhlmark et al. [17] identified that the magnetic confinement of the sputtered flux influences the deposition rate.

As pointed out above, generally, HPPMS and DCMS deposition rates are compared at similar time-averaged power values. HPPMS discharges are driven with cathode

*Corresponding author. Tel.: +49 241 8025974.

E-mail address: emmerlich@mch.rwth-aachen.de (J. Emmerlich).

potentials in the range of -600 to -3000 V [1], exceeding the target potentials commonly employed in DCMS considerably. According to sputtering theory, the sputtering yield, Y , in the here-discussed energy range, depends on the target potential, U , as follows [18–20]:

$$Y \sim U^{1/2}. \quad (1)$$

The fact that the sputter yield is non-linearly energy dependent (see Eq. (1)) during DCMS and HPPMS has been ignored in the previous analysis of the deposition-rate data.

Macák et al. [4] showed, based on time-resolved optical emission spectroscopy data, that the plasma chemistry is a function of time. At the beginning of the pulse, the Ar^+ population is dominant, while later in the pulse Ti^+ is in the majority. The existence of a time separation between the Ar and metal-ion fluxes has been confirmed [17,21].

With this in mind, we have calculated the effect of non-linearly energy-dependent sputtering yields (for the Ar^+ ion scenario as well as for the metal ion part of the pulse) on the deposition rate during HPPMS. The data are then compared to the DCMS deposition rate obtained at identical time-averaged power values. The argumentation is based on TRansport-of-Ions-in-Matter (TRIM) simulations as well as experimental data. The effect of the energy-dependent secondary electron emission yield on the deposition rate is not considered here. A discussion of the effect of electrons on the deposition rate is omitted, due to the lack of data describing metal-ion-induced secondary electron emission during self-sputtering.

2. Experimental details

The TRIM software [22] was employed for the simulation of the sputtering yield of Ar^+ and Cu^+ (self-sputtering) of Cu target atoms as a function of the ion energy range of 100 – 2000 eV. A $1\text{-}\mu\text{m}$ -thick Cu target was bombarded with 9999 Ar^+ and Cu^+ ions, respectively, under perpendicular incidence.

The base pressure in the chamber was $\sim 5 \times 10^{-4}$ Pa. A Cu target ($\varnothing 90$ mm) was at a distance of 120 mm from the substrate. The films were deposited on grounded Si wafers using HPPMS (Chemfilit Ion sputtering, SINEX 2.0-AS14) and DCMS (MKS Instruments, ENI RPG-100E) in an Ar atmosphere of 1 Pa. In the HPPMS plasma, the voltage was set at -750 V with voltage-pulse duration of $50 \mu\text{s}$ and a frequency of 100 Hz. The corresponding target current was measured using a current probe (Tektronix, A6303) in combination with an amplifier (Tektronix, TM502). Time-dependent evolutions of voltage and current were measured by an oscilloscope (Tektronix, TDS 3014B) and the time-averaged power was calculated to be 78 W. A power of 77 W was applied during DCMS (-308 V and 250 mA). The deposition rate was determined from thickness-profile measurements using a surface profilometer (Dektak-3030).

3. Results and discussion

Fig. 1 shows the simulated sputtering yield extrapolated from the target potential during DCMS, -308 V, assuming that the yield is linearly proportional to the target potential (denoted “+”). This extrapolation suggests a 100% efficient sputtering process with respect to DCMS at any target potential. This assumption is the erroneous basis of the comparison between DCMS and HPPMS deposition rates at constant time-averaged power. Additionally, the simulated sputtering yield for Ar^+ (denoted “□”) and Cu^+ (denoted “■”) bombardment on a Cu target is plotted as a function of target potential in the range of 100 – 2000 eV. The investigated discharge conditions for DCMS and HPPMS are marked at 308 and 750 eV, respectively. For Ar^+ sputtering, the Cu sputtering yield increased from 1.74 (DCMS, $Y_{\text{Ar}^+}^{\text{DC}}$) to 3.24 (HPPMS, $Y_{\text{Ar}^+}^{\text{HPPMS}}$), which agrees well (within 20% deviation) with experimental data [23], while the linearly extrapolated sputtering yield, $Y^{\text{extrapolated}}$, increases to 4.24 . The maximum relative deposition rate, defined as HPPMS rate/DCMS rate, of an Ar^+ plasma determined by the $Y_{\text{Ar}^+}^{\text{HPPMS}}/Y_{\text{Ar}^+}^{\text{extrapolated}}$ ratio is 76%. Based on the Christie model [14], the maximum relative deposition rate of a metal (M^+) plasma (no Ar^+ contribution) is given by the ratio:

$$\frac{Y_{\text{M}^+}^{\text{HPPMS}} - 1}{Y^{\text{extrapolated}}}. \quad (2)$$

The term “ -1 ” represents the metal ion employed in the self-sputtering event and its incorporation in the target. The relative deposition rate for the pure metal plasma (Eq. (2)) is only applicable for materials exhibiting a self-sputtering yield higher than 1. For materials not fulfilling this requirement, contribution to sputtering by both Ar

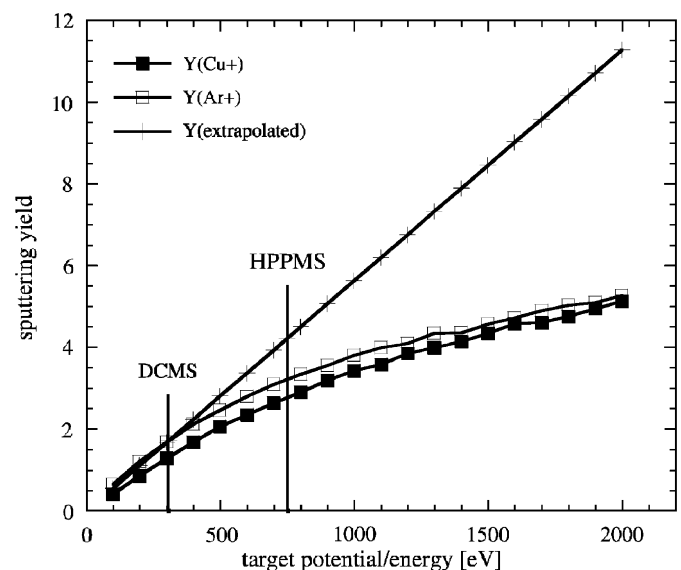


Fig. 1. Simulated sputtering yield for Cu target bombarded with Ar^+ (□), Cu^+ (■) ions, and an extrapolated yield (+) (linearly extrapolated from the Ar^+ yield at 308 eV) as a function of target potential.

and metal ions is required to avoid plasma extinction. Eq. (2) for a Cu^+ plasma with $Y_{\text{Cu}^+}^{\text{HPPMS}} = 2.84$ yields 43%.

The previously discussed optical emission spectroscopy data revealed a transition from an Ar^+ -dominated to a metal-ion-dominated plasma [4]. Based on this and the here-discussed non-linear energy-dependent sputtering yields (Eq. (1)), it is reasonable to expect a maximum relative deposition rate for Cu of 76%, assuming that the entire Cu flux is generated by sputtering of a Cu target by Ar^+ ions and transported without any loss to the substrate. The minimum deposition rate that can be expected is 43%, assuming that the entire Cu flux is sputtered by Cu^+ ions and transported without loss to the substrate. These data strongly suggest that a comparison of HPPMS and DCMS deposition rates is physically meaningful only if the non-linear energy dependence of the sputter yield (Eq. (1)) is taken into account. To expect similar values for the HPPMS and DCMS deposition rates at similar time-averaged power values is not compatible with sputtering theory.

To test our calculations, Cu thin films were deposited by HPPMS and DCMS in an Ar atmosphere. Due to the high pressure (1 Pa) and the long pulses (50 μs) applied in our experiments, it is reasonable to assume that the main part of the pulse is Cu^+ dominated [21] and thus the relative deposition rate should be close to the theoretical limit determined for a pure Cu^+ plasma. The relative deposition rate based on thickness measurements was determined to be 32%. This is somewhat lower than the predicted maximum rate based on sputtering of Cu by Cu^+ ions, but this calculation is based on 100% efficient transport. Due to the high degree of ionization of the sputtered material, a certain amount of the metal-ion flux may be guided away from the path to the substrate and hence does not contribute to film growth. This is consistent with the work of Konstantinidis et al. [21]. Böhlmark et al. [17] showed that the deposition rate can be increased by 80% with the variation of the magnetic field.

Our theoretical considerations are also consistent with the HPPMS deposition-rate data reported by Konstantinidis et al. for Ti films in an Ar^+ -dominated plasma with 5 μs pulses at 0.27 Pa [21]. In Fig. 2, the simulated sputtering yields for Ar^+ (denoted “□”) and Ti^+ (denoted “■”) sputtering, and the yield extrapolated from DCMS at 300 eV (denoted “—”) are presented. Good agreement between the reported relative deposition rate of approximately 70% and our simulated value of 67% based on the sputtering yields of Ti [$Y_{\text{Ar}^+}^{\text{DCMS}}(300\text{ V}) = 0.45$; $Y_{\text{Ar}^+}^{\text{HPPMS}}(820\text{ V}) = 0.82$; $Y^{\text{extrapolated}}(820\text{ V}) = 1.23$] is achieved. However, for the HPPMS Ti plasma with 5 μs pulses operated at an increased pressure of 1.3 Pa, the relative deposition rate dropped to 43%. This is consistent with the optical emission data indicating the presence of a Ti^+ -dominated plasma later in the pulse and hence a significant deposition-rate reduction has to be expected.

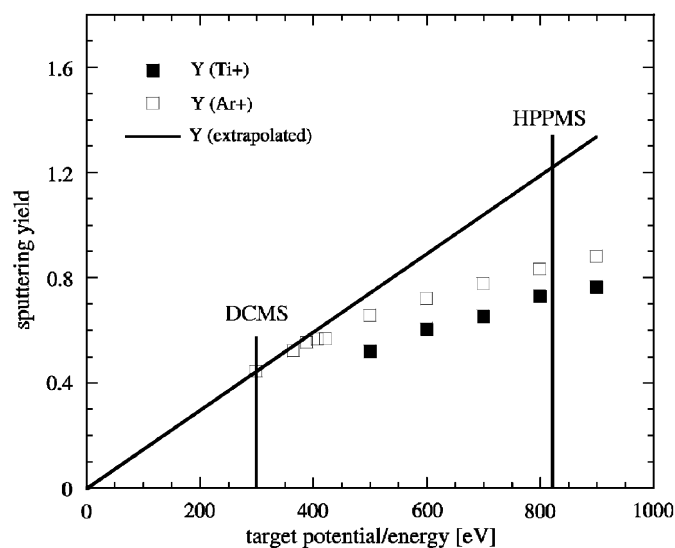


Fig. 2. Simulated sputtering yield for Ti target bombarded with Ar^+ (□), Ti^+ (■) ions, and an extrapolated yield (—) (linearly extrapolated from the Ar^+ yield at 300 eV) as a function of target potential.

4. Conclusions

We have shown that the apparent decrease of the relative deposition rate discussed in the literature may be understood by considering the non-linear energy dependence of the sputtering yields. Our results strongly suggest that a comparison of high-power pulsed magnetron sputtering (HPPMS) and DC magnetron sputtering (DCMS) deposition rate is physically meaningful only if the non-linear energy dependence of the sputtering yield is taken into account. To expect similar values for the HPPMS and DCMS deposition rate at similar time-averaged power values is not compatible with sputtering theory. These results are of general relevance for ionized physical vapor deposition, utilizing larger target potentials as employed during DCMS.

Acknowledgment

We acknowledge financial support from DFG (Sch 735/15-1, “Adaptive Oberflächen für Hochtemperatur-Anwendungen”).

References

- [1] Ehiasarian AP, New R, Münz W-D, Hultman L, Helmersson U, Kouznetsov V. *Vacuum* 2002;65:147.
- [2] Westwood WD. *Sputtered deposition*. New York: AVS; 2003. p. 58.
- [3] Kouznetsov V, Macák K, Schneider JM, Helmersson U, Petrov I. *Surf Coat Technol* 1999;122:290.
- [4] Macák K, Kouznetsov V, Schneider JM, Helmersson U, Petrov I. *J Vac Sci Technol A* 2000;18:1533.
- [5] Gudmundsson JT, Alami J, Helmersson U. *Appl Phys Lett* 2001; 78:3427.
- [6] Helmersson U, Lättemann M, Böhlmark J, Ehiasarian AP, Gudmundsson JT. *Thin Solid Films* 2006;513:1.

- [7] Petrov I, Barna PB, Hultman L, Greene GE. *J Vac Sci Technol A* 2003;21:S117.
- [8] Ehiasarian AP, Münz W-D, Hultman L, Helmersson U, Petrov I. *Surf Coat Technol* 2003;163–164:267.
- [9] Alami J, Persson POÅ, Music D, Gudmundsson JT, Böhlmark J, Helmersson U. *J Vac Sci Technol A* 2005;23:278.
- [10] DeKoven BM, Ward PR, Weiss RE, Christie DJ, Scholl RA, Sproul WD, et al. In: Proceedings of the 46th annual technical conference of the Society of Vacuum Coaters. Albuquerque: SVC; 2003. p. 158.
- [11] Böhlmark J, Alami J, Christou Ch, Ehiasarian AP. *J Vac Sci Technol A* 2005;23:18.
- [12] Vlček J, Kudláček P, Burcalová K, Musil J. *J Vac Sci Technol A* 2007;25:42.
- [13] Vlček J, Kudláček P, Burcalová K, Musil J. *Europhys Lett* 2007; 77:45002.
- [14] Christie DJ. *J Vac Sci Technol* 2005;23:330.
- [15] Konstantinidis S, Dauchot JP, Ganciu M, Hecq M. *Appl Phys Lett* 2006;88:021501.
- [16] Bugaev SP, Koval NN, Sochugov NS, Zakharov AN. In: Proceedings of the XVIIth international symposium on discharges and electrical insulation in vacuum, 21–26 July, Berkeley, CA, USA, 1996, p. 1074.
- [17] Böhlmark J, Östbye M, Lattemann M, Ljungcrantz H, Rosell T, Helmersson U. *Thin Solid Films* 2006;515:1928.
- [18] Wilson WD, Haggmark LG, Biersack JP. *Phys Rev B* 1977;15:2458.
- [19] Zalm PC. *J Vac Sci Technol B* 1984;2:151.
- [20] Steinbrüchel Ch. *Appl Phys A* 1985;36:37.
- [21] Konstantinidis S, Dauchot JP, Ganciu M, Ricard A, Hecq M. *J Appl Phys* 2006;99:013307.
- [22] Ziegler JF, editor. *The stopping and range of ions in solids*, vol. 1. New York: Pergamon; 1985. <<http://www.srim.org/>> [accessed 01.03.07].
- [23] Laegreid N, Wehner GK. *J Appl Phys* 1961;32:365.