

Lawrence Berkeley National Laboratory

Lawrence Berkeley National Laboratory

Title

Deposition Rates of High Power Impulse Magnetron Sputtering: Physics and Economics

Permalink

<https://escholarship.org/uc/item/7cz4g23b>

Author

Anders, Andre

Publication Date

2010-06-02

Peer reviewed

*Presented at Invited Talk at the 52th Meeting of the American Vacuum Society,
San Jose, Nov. 9, 2009*

and

Submitted to

Journal of Vacuum Science and Technology A

Deposition Rates of High Power Impulse Magnetron Sputtering: Physics and Economics

André Anders*

Lawrence Berkeley National Laboratory, University of California, 1 Cyclotron Road, Berkeley, California 94720

November 22, 2009

ACKNOWLEDGMENT

The work was supported by the US Department of Energy under Contract No DE-AC02-05CH11231 with the Lawrence Berkeley National Laboratory.

DISCLAIMER

This document was prepared as an account of work sponsored in part by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor The Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or The Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or The Regents of the University of California.

* **email:** aanders@lbl.gov

Deposition Rates of High Power Impulse Magnetron Sputtering: Physics and Economics

André Anders[†]

Lawrence Berkeley National Laboratory, University of California, 1 Cyclotron Road, Berkeley, California 94720

Abstract

Deposition by high power impulse magnetron sputtering (HIPIMS) is considered by some as the new paradigm of advanced sputtering technology, yet this is met with skepticism by others for the reported lower deposition rates, if compared to rates of more conventional sputtering of equal average power. In this contribution, the underlying physical reasons for the rate changes are discussed, including (i) ion return to the target and self-sputtering, (ii) the less-than-linear increase of the sputtering yield with increasing ion energy, (iii) yield changes due to the shift of species responsible for sputtering, (iv) changes to due to greater film density, limited sticking, and self-sputtering on the substrate, (v) noticeable power losses in the switch module, (vi) changes of the magnetic balance and particle confinement of the magnetron due to self-fields at high current, and (vii) superposition of sputtering and sublimation/evaporation for selected materials. The situation is even more complicated for reactive systems where the target surface chemistry is a function of the reactive gas partial pressure and discharge conditions. While most of these factors imply a reduction of the normalized deposition rate, increased rates have been reported for certain conditions using hot targets and less poisoned targets. Finally, some points of economics and HIPIMS benefits considered.

1. Introduction

High power impulse magnetron sputtering (HIPIMS), also known as high power pulse magnetron sputtering (HPPMS), is an intriguing addition to the family of sputtering technologies because it intrinsically offers ion assistance to film growth^{1,2,3,4,5}. HIPIMS is characterized by a very high power density at the target during each pulse, typically exceeding “conventional” power densities by about two orders of magnitude. Of course, such “abuse” of a magnetron would overheat the device if the duty cycle was high, and therefore HIPIMS is used with low duty cycles. The main motivation for using the extreme power density is to reach conditions that lead to the ionization of the sputtered material. Once at least partially ionized, the flux of sputtered material represent a special means of assistance to film growth on a substrate, which opens significant opportunities for substrate-coating interface engineering⁶ and tailoring film properties^{7,8,9}.

While material scientists are excited about the new process parameters that have the potential of improved film quality and adhesion, the technologists at coating companies are concerned about the deposition rate. In many but not all circumstances it was found that

[†]email: aanders@lbl.gov

HIPIMS processing resulted in lower deposition rates, an issue that is discussed in this contribution.

The manuscript is organized as follows. We will first need to more precisely define what a “deposition rate” is since various normalizations are possible. Then we will have a systematic look at the factors that affect the deposition rate. For simplicity, and making the approach more translucent, the case of pure metal sputtering is discussed in detail, followed by briefly addressing reactive sputtering. The latter subject is more complicated and it should be worked on in the future because of its relevance to industrial use. Finally, some comments are made on the economics, and we will assess HIPIMS having both film quality and economics in mind.

2. Defining “deposition rate”

The term deposition rate is widely used yet different conditions and normalizations are possible, and therefore we need to go through a somewhat academic looking exercise of considering various definitions.

The *static deposition rate* is defined as the growth of film thickness per time. Here, the source of the flux, i.e., the magnetron sputtering gun, and the substrate are essentially in the same relative position during the process (*periodic* substrate motion for improved uniformity does not change the essence of this discussion). The static deposition rate is conventionally expressed in units of nm/s, or equivalent units like nm/min, $\mu\text{m/h}$, or using the non-SI units $\text{\AA}/\text{s}$, or $\text{\AA}/\text{min}$.

The important point is: the static deposition rate is an *absolute* rate, which of course increases as the power to the magnetron is increased. From a practical point of view, there are limits of how much power can be applied without damaging the magnetron or other components. A coatings manufacturer would most likely operate at high power to reduce the time needed for a certain coating to be made. This naturally leads to another quantity, the relative or *normalized static deposition rate*, which is defined as the thickness of deposited film per time and power in units of $\text{nm}/(\text{Ws})$, or equivalent.

We quote here the direct current (DC) rate as a normalization benchmark, although for some materials, typically for deposition from targets of limited conductivity or when reactive gases are present, one should use radio frequency (RF) rates or medium-frequency alternating current (MF-AC) rates. For simplicity we only refer to the DC rates in this contribution and assume that the reader can extrapolate when deposition by MF-AC or RF sputtering is a more appropriate baseline.

So far, the area of the coating did not appear in the consideration but of course it should since industrial manufacturing occurs on large areas. One would generally use, for uniformity reasons, a target that matches or exceeds the area of the substrate. This would lead to a more general normalized rate, the *normalized area static deposition rate*, in units of $\text{nm}/(\text{Ws m}^2)$, or equivalent.

For large area coatings, using the typical in-line coating approach, the substrate is moved relatively to the magnetron in a linear fashion, and therefore one needs to introduce the *dynamic absolute deposition rate*, which refers to thickness deposited per time and length, in units $\text{nm}/(\text{s m})$, or equivalent.

Corresponding to the definitions for static deposition, it also makes sense to introduce the *dynamic normalized deposition rate* as thickness per time, length, and power, in units $\text{nm}/(\text{Ws m})$, and the *dynamic normalized area deposition rate*, in units of $\text{nm}/(\text{Ws m m}^2)$, or more compact $\text{nm}/(\text{Ws m}^3)$, although it should be noted that $\text{nm}/(\text{Ws m m}^2)$ is preferred because it much better conveys the origin of the definition.

Looking at all those various definitions it becomes clear that statements in the literature comparing deposition rates are not necessarily trivial, and one has to have a closer look which of the definitions is actually meant.

3. Some HIPIMS deposition rates from the literature

This section is not intended as a review of rates, rather, it should only serve to convey a sense for the significance of the deposition rate issue.

Even before the seminal HIPIMS paper by Kouznetsov and coworkers¹ appeared, Bugaev and coworkers¹⁰ stated in their report on HIPIMS that the deposition rate for copper and titanium was about 50% lower than DC rates.

Konstantinidis and coworkers¹¹ found that the reduction depends on the pulse length: very short pulses of only 5 μs do not allow much of self-sputtering and other processes to evolve, and so the rate for Ti deposition was about 70% of the DC rate, while it went dramatically down to 20% when using longer pulses of 20 μs (though still very short by HIPIMS standards). Even greater reductions down to 15% were observed for TiO_2 deposition¹², although here the average pulse power was determined before the switch module (see section 4.6).

Of course, the deposition rates are closely related to sputtering yields, target surface conditions, and many other factors. For example, using the same setup but different targets, Glocker¹³ found that HIPIMS rates are reduced to about 25% for Al_2O_3 but about the same for ZrO_2 coatings.

Some of the coating applications require films on complex shapes such as around the edges of cutting tool inserts. Here one generally distinguishes rates on the flank and rake. Ionization of the sputtered flux offers much enhanced (improved) conformal coating. Bobzin and coworkers¹⁴, depositing (Ti,Al)N and (Ti,Al,Si)N, showed an almost equal HIPIMS flank rate (parallel to the target surface) and an about 1.6 times higher rake rate (perpendicular to the target surface) compared to the DC case.

More about rate data under various conditions can be found in reviews of the field^{7,8,15}. The examples show that there is a great variety of results indicative for several factors at work.

4. Physics of HIPIMS deposition rates

4.1 Some power considerations for HIPIMS

HIPIMS operates with pulses of very high power:

$$P(t) = V(t)I(t), \quad (1)$$

where $V(t)$ and $I(t)$ are the time-dependent discharge voltage and current, respectively. The extremely high deposition rate during the pulse (and slightly after the pulse, due to time-of-flight effects) are of importance to the actual film growth mechanism, and therefore, even if the time-averaged deposition HIPIMS rate equaled the rate of a comparable DC system, the film properties will be different^{9,16,17}.

Many practitioners focus on the time-averaged power-normalized deposition rate, static or dynamic. To do this, the average power of the process needs to be calculated,

$$\bar{P} = \int_0^{t_D} I(t)V(t) dt / t_D \quad (2)$$

where t_D is the total deposition time. Alternatively, and more practical, one may chose to average over one pulse and use the duty cycle $\delta = t_{on} / (t_{on} + t_{off}) = t_p f_p$, where t_p is the duration of one pulse and f_p is the pulse repetition frequency, to obtain

$$\bar{P} = \delta \int_0^{t_p} I(t) V(t) dt / t_p = f_p \int_0^{t_p} I(t) V(t) dt . \quad (3)$$

4.2 General rate considerations

A discussion of fluxes and rates follows similar ideas laid out for reactive sputtering by Berg¹⁸ and for special HIPIMS cases (excluding self-sputtering runaway) by Christie¹⁹. It is necessary to establish the role of voltage and current not only for power, as done in section 4.1, but also for the sputtering yield and related absolute deposition rates.

A large portion of the voltage between cathode (target) and anode drops in the thin cathode sheath above the target. Ions at the sheath edge are accelerated towards the target by the electric field. If we assume that the sheath is smaller than the mean free path between collisions, the ion energy E_i (in eV) corresponds to the voltage drop V_{sheath} (in V). The charge state number, Q , is a multiplier in the case we deal with multiply charged ions, that is,

$$\Delta E_i = Qe \Delta V_{sheath} , \quad (4)$$

where e is the elementary charge. Besides the voltage drop in the sheath, and due to the presence of a magnetic field, another significant portion of the voltage drops in the “magnetic presheath”. For the propose of discussing the dependencies, one can approximately write for the total kinetic energy of ions impacting the target

$$E_{i,tar} = Qe V . \quad (5)$$

The deposition rate at the substrate, R_{sub} , is generally proportional to the sputter yield caused by ions of type k with energy $E_{i(k),tar}$ impacting the target. We neglect here, for the time being, that some sputtered atoms will be ionized and return later to this point. Furthermore, we can further simplify by averaging over the target and substrate areas and thereby reducing the model to 1 spatial dimension. Since more than one type of ions can be involved in the sputtering process, the deposition rate is proportional to the flux of sputtered atoms, which is caused by impact of various ions of type k :

$$R_{sub} = C_1 \Phi_{a,sputtered} = C_1 \sum_k \gamma_k (E_{i(k),tar}) \Phi_{i(k),tar} , \quad (6)$$

where C_1 is a geometry factor that could also include scattering of sputtered atoms by the process gas if the pressure is relative high. The symbol $\Phi_{i(k),tar}$ is the flux of ions of type k arriving at the target; $\Phi_{i(k),tar}$ is related to the ion current density by

$$j_{i(k),tar} = eQ_k \Phi_{i(k),tar} . \quad (7)$$

Taking the yield of secondary electrons, $\gamma_{SE(i)}$, into account, the total current density at the target is

$$j_{tar} = \sum_k (j_{i(k),tar} + j_{SE(k)}) = \sum_k [1 + \gamma_{SE(k)}] j_{i(k),tar} , \quad (8)$$

resulting in the discharge current

$$I_{tar}(t) = \int_{A_{tar}} j_{tar} dA . \quad (9)$$

Any increase in voltage will lead to an increase in sputtering yield, as shown in Fig. 1. It is well known, and evident from Fig. 1, that the yield scales with the ion energy in a less than proportional manner. The yields can be calculated with the TRIM²⁰ or similar Monte Carlo

Code. Table 1 shows that the result for each combination of ion type and target material can be reasonably fitted by an allometric function

$$\gamma_k = a_k E_k^{b_k}. \quad (10)$$

An acceptable approximation is $b_k=b=1/2$ for all curves, which simplifies further presentation. The fit is quite good for medium mass elements but shows noticeable deviations for carbon. For self-sputtering, the exponent b is best approximated by

$$b = 0.106 M^{0.387}, \quad (11)$$

where M is the atomic mass number of the self-sputtering element, however, we will stick to the simpler $b \approx 1/2$.

Using Eqs. (10), (9), (7) and (5), the rate can be expressed via the ion current to the target and the discharge voltage in the relatively simple form

$$R_{sub} = C_1 \sum_k a_k V^{1/2} I_{i(k),tar}. \quad (12)$$

This expression will be further evaluated in the next sections.

4.3 The return effect

The perhaps most important and widely recognized effect for reducing the power-normalized rate in HIPIMS is due to the fact that a large fraction of sputtered atoms is ionized, and some of the newly formed ions return to the target (Fig. 2). There they contribute to current (i.e., power included in the normalization of rates) and cause self-sputtering.

Self-sputtering has been known long before HIPIMS and considered for sputtering of liquids²¹, high-yield solids^{22,23}, and for pseudosparks²⁴. Following the nomenclature of Hosokawa and co-workers²⁵, let us designate the probability of ionization of a sputtered atom with α , and the probability that the newly formed ion returns to the target with β , where of course $0 \leq \alpha \leq 1$ and $0 \leq \beta \leq 1$. Let us further consider that the deposition rate is the sum of the deposition rates by condensable atoms and ions, which in turn are proportional to the corresponding fluxes of atoms and ions to the substrate, hence:

$$R_{sub} = R_{sub,atoms} + R_{sub,ions} = C_2 (\xi_a \Phi_{a,sub} + \xi_i \Phi_{i,sub}), \quad (13)$$

where ξ_a and ξ_i are the sticking coefficients. In order to not make this discussion overly complicated, we simplify by setting $\xi_a = 1$ and $\xi_i = 1$:

$$R_{sub} = C_2 (\Phi_{a,sub} + \Phi_{i,sub}), \quad (14)$$

The goal of a model is to combine the processes on the target with the deposition rate on the substrate, and here the probabilities α and β can be used. In conventional sputtering, $\alpha \approx 0$ and thus $\Phi_{i,sub} = 0$ and $\Phi_{a,sub} = \Phi_{a,sputtered}$. Under HIPIMS conditions, $\alpha > 0$ and

$$\Phi_{a,sub} = (1 - \alpha) \Phi_{a,sputtered}, \quad (15)$$

$$\Phi_{i,sub} = \alpha(1 - \beta) \Phi_{a,sputtered}. \quad (16)$$

From Eqs. (14) to (16) we can readily assess the return effect dictated by the probabilities α and β . The effect becomes larger with increasing α and β . Let's illustrate this by an extreme case: if the ionization of sputtered atoms become almost complete, $\alpha \rightarrow 1$, no neutral atoms arrive at the substrate, $\Phi_{a,sub} \rightarrow 0$. If conditions are such that almost all ions return to the target, $\beta \rightarrow 1$, also no ions arrive at the substrate, $\Phi_{i,sub} \rightarrow 0$. In essence, the deposition rate can

almost vanish, $R_{sub} \rightarrow 0$! In other words, at least theoretically in the extreme case of $\alpha \rightarrow 1$ and $\beta \rightarrow 1$, the return effect can reduce the deposition rate by almost 100%.

The values of α and β will strongly depend on the HIPIMS system, target material, and the actual discharge parameters. Therefore, it is not surprising at all that data on normalized deposition rates vary greatly. For example, the ionization probability will greatly depend on the sputtering yield because it determines the fluxes of neutrals and thereby, indirectly, the electron temperature. For low yield materials, like Ti, where some gas is needed to sustain the HIPIMS discharge, the degree of ionization of Ti can be pushed rather high, and even multiple ionization is significant²⁶. The deposition rates should be expected to be very low. For high yield materials, like Cu, sustained self-sputtering can be achieved, even without gas, and here the return probability β is close to unity at the self-sputtering threshold and much reduced if the system operates far beyond the threshold²⁷. This leads to the prediction that operation at higher HIPIMS voltage and current leads a higher deposition rates than operation near the self-sputtering threshold, which is a different trend than the yield effect predicts in the next section.

4.4 The yield effect

Voltage and current are *proportionally* used in the calculation of power, Eq.(3), and therefore any *increase* in operational voltage necessarily implies a *decrease* of the averaged current in order to keep the same averaged power, even as the actual current during pulses is much higher than in the DC case. As pointed out by Emmerlich and co-workers²⁸, the deposition rates of DC and HIPIMS operation, at equal average power, could only be the same if the sputtering yields were exactly proportional to the voltage, which they are not, see (10). The theory of sputtering necessitates a reduction of the normalized deposition rate when going to HIPIMS operation unless additional new effects come into play, which are discussed later in this section.

Equation (12) can be simplified by executing the summation and using a different constant C_3

$$R_{sub} = C_3 V^{1/2} I_{i,tar} . \quad (17)$$

Strictly speaking, this expression should be interpreted as the instantaneous rate *during* a HIPIMS pulse,

$$R_{sub}^{HIPIMS}(t) = C_3 V^{1/2}(t) I_{i,tar}(t) , \quad (18)$$

or as a relation for the DC rate

$$R_{sub}^{DC} = C_3 V_{DC}^{1/2} I_{i,tar,DC} . \quad (19)$$

In order to compare deposition rates at equal power, the HIPIMS rate needs to be averaged over time, similar to the averaging done for the power, *cf.* (Eq. (3)),

$$\overline{R_{sub}^{HIPIMS}} = f_p \int_0^{t_p} R_{sub}^{HIPIMS}(t) dt . \quad (20)$$

We can write the ratio of the power-normalized average HIPIMS and DC rates as

$$\rho = \frac{\overline{R_{sub}^{HIPIMS}} / \overline{P_{HIPIMS}}}{R_{sub}^{DC} / P_{DC}} . \quad (21)$$

To evaluate this expression, we make use of the fact that HIPIMS pulses use a higher voltage than the voltage of typical DC operation in order to drive the high power discharge pulse. A

particularly easy case for evaluation is when the HIPIMS system operates with a constant voltage supply because we can write that the HIPIMS voltage is a factor K higher than the DC voltage:

$$V_{HIPIMS} = K V_{DC} \quad (22)$$

with $K > 1$. The average rate can then be written as

$$\overline{R_{sub}^{HIPIMS}} = C_3 f_p (K V_{DC})^{1/2} \int_0^{t_p} I_{i,HIPIMS}(t) dt. \quad (23)$$

For the evaluation we utilize that the average HIPIMS power is equal to the DC power (that was the premise of comparing rates):

$$P_{DC} = V_{DC} I_{i,tar,DC} (1 + \gamma_{SE}) = \overline{P_{HIPIMS}} = K V_{DC} f_p (1 + \gamma_{SE}) \int_0^{t_p} I_{i,HIPIMS}(t) dt. \quad (24)$$

Combining (19) to (24) we finally arrive at a surprisingly simple expression for the ratio of power-normalized HIPIMS and DC rates:

$$\rho = K^{-1/2}. \quad (25)$$

We see that the more the HIPIMS voltage is enhanced compared to the DC voltage, the greater is the reduction of the normalized HIPIMS deposition rate. This result is simply due to the fact that the sputtering yield does not scale linearly with the energy of ions, and it has nothing to do with the return effect.

To see how big the yield effect is, let us look at an example. DC sputtering of copper could be done with 350 V, and the HIPIMS pulse could be run with 700 V, a factor $K = 2$ higher. The yield effect gives a reduction to $2^{-1/2} = 0.707$, i.e. a reduction by about 30%.

4.5 The ion species effect

The sputter yield is largely determined by the target material (more precisely, by the surface binding energy). The curves for Ar ion and self-ion sputtering (Fig. 1) are close but not identical. For many metals, we can see a slight change, about 10-15% for the most relevant range of ion energy, when the process transitions from argon sputtering to self-sputtering. As self-sputtering becomes more prominent or even dominant as the pulse progresses, the rate for self-sputtering should be considered.

A change of the type of sputtering ion will also cause a change in the secondary electron yield. For the relatively low energy mostly used in sputtering, $E_i < 1$ keV, secondary electrons are primarily ejected by potential emission. Here, the reduction of secondary electron emission can be quite dramatic because the singly charged metal ions have less than twice the target's work function²⁹, while noble gas ions, like Ar^+ , fulfill this criterion³⁰. Yet, the effect of this reduction on normalized rates is limited because the secondary electrons only appear in the discharge current, Equ. (24), representing a very small percentage of the total discharge current.

Overall, for most elements, the change caused by the change of the type of sputtering ions is not very important. The perhaps biggest difference is for carbon.

4.5 Film effects

In this subsection, we turn to the changes on the substrate. The whole point of considering HIPIMS was to provide ion assistance to film growth, affecting film density, texture, stress, etc. It is clear that denser films imply a somewhat thinner film thickness at equal number of deposited atoms (or ions) per unit area. Therefore, going from conventional sputtering (especially at high pressure) to HIPIMS implies an apparently reduced deposition rate, which is

generally a small effect. Additionally, one should also consider that the sticking coefficients may be reduced, energetic neutral atom bombardment is enhanced, and that self-sputtering can occur on the substrate. The latter can be very significant when the substrate is negatively biased³¹. One should recall that HIPIMS plasmas can be used for ion etching⁷.

4.6 Power loss in the switch module

The normalization of rates using power should use the actual pulse shapes as indicated in Eq. (3); this requires monitoring the time dependent current and *voltage at the target*. In absence of a suitable time-resolving current monitor and voltage divider, the average power may be read at the charging power supply used for the HIPIMS process (e.g.^{12,32}), however, this would also include the power losses in the switch module. Switching losses are especially significant when very short (< 30 μ s) pulses are used. The power normalized rates may appear worse than they actually are given that the power loss in switching elements (usually insulated gate bipolar transistors, IGBTs) was included in the normalization, and not just the power dissipated at the magnetron. In extreme cases, this underestimate can exceed 30%³³.

4.7 Magnetic unbalancing and guiding effect

One generally distinguishes balanced and unbalanced magnetrons, which refers to the closing of the magnetic field lines in front of the target. In an unbalanced situation, fields extend far beyond the target and help plasma to escape and more efficiently assist in film growth than in the balanced case. As we compare HIPIMS and DC, one needs to recognize that the high discharge current, in conjunction with a high azimuthal drift or Hall current³⁴, will affect the magnetic field distribution. This will affect the local plasma density, ionization probability and return probability, and the plasma transport of material from the target to the substrate. Extensive modeling is required to grasp those complicated interactions, and no simple statement can be made other than that in most cases, one would expect increased transport of ions to the substrate. Application of external magnetic fields, like from a coil, can redirect the plasma flow and lead locally reduced and elsewhere enhanced deposition^{35,36}.

4.8 Hot target effects

It should be mentioned that the HIPIMS rates are not always reduced but can be enhanced. Most notably, the surface of the target, and especially the race track zone, may be heated to such degree that sublimation (from a solid) or evaporation (from a liquid) sets in. Of course, melting of the target is usually not desired, and we also have to worry about overheating the magnets which are located on the backside of the target.

There is some discussion in the literature whether or not the sputtering yield is temperature dependent, i.e., whether rate enhancement effects are due to enhanced sputtering or onset of sublimation/evaporation. Vaulin and co-workers³⁷ found an increase in the yield for copper when the temperature of the target exceeded about 800°C; this work was cited by Fortov³⁸ (p. 119). Bohdanský and coworkers³⁹ showed that this increase is related to evaporation rather than sputtering. Behrisch and Eckstein^{40,41} generally agree but acknowledged that the surface binding energy has a nonlinear effect on the yield. The surface binding energy can be derived from the heat of sublimation *at a given temperature*. Even as the heat of sublimation decreases with temperature, evaporation is overtaking the loss rate at high temperature⁴⁰ by a wide margin. The vapor pressure of the different materials⁴² is of course the right gauge for estimating the relevance of sublimation/evaporation. As pointed out by Doerner and

coworkers⁴³, ion bombardment can induce stimulated sublimation/evaporation that is temperature dependent in a highly non-linear manner. Experiments with lithium and beryllium showed that higher rates can be ion-stimulated at temperatures below the usual onset for evaporation (Li) and sublimation (Be). This hot target approach has not yet been fully explored but efforts are underway⁴⁴.

4.9 Rates for reactive systems

Reactive sputtering deserved an extra paper and cannot be fully addressed here. The greatest concern is of course the well-known “poisoning” of the target surface followed by a rapid decrease of the deposition rate^{18,45,46}. HIPIMS has been done with reactive gases, and much lower^{12,32}, slightly lower^{13,36}, and higher^{47,48,49,50} rates than DC operation have been reported. Clearly, all of the so-far discussed effects apply plus the effects related to target poisoning. The most interesting fact, however, is that higher rates are possible, which could be associated with a reduced hysteresis allowing for tighter control of the operational point (gas flow versus partial pressure). Sarakinos et al.⁴⁷ pointed out that the high pulse power leads to stronger gas rarefaction, which was confirmed by unrelated HIPIMS experiments⁵¹. Stronger rarefaction reduces the flux of reactive gas to the target, hence less poisoning is found.

Bombardment of the film by the reactive gas in-between pulses allows us to more readily obtain stoichiometric films than in continuous operation. Gas bombardment is a troubling feature for the deposition of ultrapure metal films, which are exposed to residual gas between deposition pulses, but it is a highly desired feature for compound deposition because it allows us to reduce the partial pressure of the reactive gas. As a consequence, at least in some cases, HIPIMS processing can be done such that the target remains in metal mode while stoichiometric compound films are deposited. In these cases, the HIPIMS rate can be large compare to rates otherwise obtained with a poisoned target.

For example, Chistyakov^{48,49} demonstrated a HIPIMS version with modulated power that perhaps utilizes the combined effect of high target temperature and hysteresis control. By stepwise increasing the power within 1-2 ms, high normalized rates were obtained for Al₂O₃, TiN, and other materials, exceeding the normalized DC rates. Another example is the deposition of ZrO₂ where tight control of the partial pressure allowed the rate to be two times higher than the comparable DC rate⁵⁰.

Reactive deposition is often accompanied with the formation of energetic negative ions, especially when using oxygen in the deposition of oxide films^{52,53}. The effect on rates by sputtering and densification of the growing films is usually small, though it should be mentioned as yet another effect that is noticeable, especially when the substrate is not moved relative to the target.

5. Economical considerations

HIPIMS is an emerging technology still in the early stage of adoption, and its wider use will depend on the issue of the often-reported reduction of power-normalized rate, which gave the motivation for this paper. From a broader economical point of view one has to look at the return of investment, considering the cost of acquisition, the cost of ownership, and the cost of operation normalized to the output. A balanced assessment needs to also take into account what advantages one would have, especially if such advantages are enabling.

The investment, or cost of acquisition, refers primarily to the power supply because the rest of the deposition system is very similar to conventional sputtering. The cost of the power

supply is today about a factor 3 higher than the cost of a DC supply of equal power. Existing deposition system can be retrofitted and modified with relatively little effort and money, mostly addressing feedthroughs, cabling, in-situ diagnostics, etc. Additionally, in order to fully utilize the new opportunities of HIPIMS associated with the ionization of the flux, implementation of a capable bias power supply should be considered. In contrast to bias supplies used in conventional sputter deposition, HIPIMS bias supplies need to have much higher current capabilities, and synchronized pulsed supplies might be considered, similar to what has been used in cathodic arc plasma deposition⁵⁴.

The cost of ownership is not very different from the cost of ownership of any other magnetron system, with the slight difference that the HIPIMS power supply may be a larger and may require a somewhat greater footprint on the floor. In the overall picture, looking also at the vacuum chamber, parts handling and storage, the difference is negligible.

The cost of operation per output, i.e. coatings produced as measured by the absolute rate, is the main concern. The option of increasing the power is limited. Many of the listed factors affecting the power-normalized rate lead to a reduction - implying lost productivity and profitability. This can be acceptable if the coating is superior to what is done with DC sputtering because a premium can be charged for the higher value. More interesting, however, is to follow up on cases where rates have been found that are comparable or even higher than the DC rates. It still remains to be shown that this can be implemented on a large scale. If successful, HIPIMS is very likely to grow beyond niche applications.

The processes described in sections 4.1 to 4.6 occur with all HIPIMS systems; they generally lead to reduced rates. Processes of magnetic guiding, hot targets, and certain reactive deposition conditions may lead to higher rates. This is schematically summarized in Fig. 3. The combination and interplay of those individual effects give a resulting rate, which determines the economics.

6. Conclusions

The physics of sputter processes suggests that a rate reduction should be expected when the mode of operation is switched from DC to HIPIMS. In this comparison, the time-averaged power was used for normalization, which is arbitrary from a physics point of view but reasonable from an application / business point view. Several different effects have been considered to explain the differences seen between DC and HIPIMS rates. Higher rates are possible under special circumstances such as very high target temperature.

It is argued that the value added should be taken into account, especially that coatings have improved quality, conformity, adhesion, surface smoothness, etc., when making a decision to utilize HIPIMS. Although HIPIMS is not likely to generally replace conventional sputtering due to higher equipment costs and often reduced normalized rates, it will find application where it provides enabling advantages. HIPIMS will gain great importance for those cases where high film quality can be obtained at rates matching or exceeding the usual DC or RF rates.

Acknowledgment

This work was supported by the Assistant Secretary for Energy Efficiency and Renewable Energy, Office of Building Technology, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

References

- ¹ V. Kouznetsov, K. Macak, J. M. Schneider, U. Helmersson, and I. Petrov, *Surf. Coat. Technol.* **122**, 290 (1999).
- ² K. Macak, V. Kouznetsov, J. Schneider, U. Helmersson, and I. Petrov, *J. Vac. Sci. Technol. A* **18**, 1533 (2000).
- ³ J. T. Gudmundsson, J. Alami, and U. Helmersson, *Surf. Coat. Technol.* **161**, 249 (2002).
- ⁴ J. Vlček, P. Kudláček, K. Burcalová, and J. Musil, *J. Vac. Sci. Technol. A* **25**, 42 (2007).
- ⁵ A. P. Ehiasarian, W. D. Munz, L. Hultman, U. Helmersson, and I. Petrov, *Surf. Coat. Technol.* **163-164**, 267 (2003).
- ⁶ A. P. Ehiasarian, J. G. Wen, and I. Petrov, *J. Appl. Phys.* **101**, 054301 (2007).
- ⁷ A. Ehiasarian, in *Plasma Surface Engineering Research and its Practical Applications*, edited by R. Wei (Research Signpost, Kerala, India, 2008), p. 35-86.
- ⁸ U. Helmersson, M. Lattemann, J. Bohlmark, A. P. Ehiasarian, and J. T. Gudmundsson, *Thin Solid Films* **513**, 1 (2006).
- ⁹ S. Mahieu, P. Ghekiere, D. Depla, and R. De Gryse, *Thin Solid Films* **515**, 1229 (2006).
- ¹⁰ S. P. Bugaev, N. N. Koval, N. S. Sochugov, and A. N. Zakharov, *Investigation of a high-current pulsed magnetron discharge initiated in the low-pressure diffuse arc plasma*, XVIIth Int. Symp. Discharges and Electrical Insulation in Vacuum, Berkeley, CA, edited by S. Anders and A. Anders, LBNL, 1074-1076 (1996).
- ¹¹ S. Konstantinidis, J. P. Dauchot, M. Ganciu, A. Ricard, and M. Hecq, *J. Appl. Phys.* **99**, 013307 (2006).
- ¹² J. A. Davis, W. D. Sproul, D. J. Christie, and M. Geisler, *High Power Pulse Reactive Sputtering of TiO₂*, 47th Annual Technical Conference, Society of Vacuum Coaters, Dallas, TX, April 24–29, 2004, SVC, 215-218 (2004).
- ¹³ D. A. Glocker, M. M. Romach, D. J. Christie, and W. D. Sproul, *High power pulsed reactive sputtering of zirconium oxide and tantalum oxide*, 47th Annual Technical Conference, Society of Vacuum Coaters, Dallas, TX, April 24–29, 2004, SVC, 183-186 (2004).
- ¹⁴ K. Bobzin, N. Bagcivan, P. Immich, S. Bolz, J. Alami, and R. Cremer, *J. Mat. Proc. Technol.* **209**, 165 (2009).
- ¹⁵ K. Sarakinos, J. Alami, and S. Konstantinidis, *Surf. Coat. Technol.*, In Press (2009).
- ¹⁶ I. Petrov, P. B. Barna, L. Hultman, and J. E. Greene, *J. Vac. Sci. Technol. A* **21**, S117 (2003).
- ¹⁷ A. Anders, *Thin Solid Films*, In Press (2009).
- ¹⁸ S. Berg, H.-O. Blom, M. Moradi, and C. Nender, *J. Vac. Sci. Technol. A* **7**, 1225 (1989).
- ¹⁹ D. J. Christie, *J. Vac. Sci. Technol. A* **23**, 330 (2005).
- ²⁰ J. F. Ziegler and J. Biersack, “Monte Carlo code SRIM2006.02, downloadable from <http://srim.org/>.” (2006).
- ²¹ R. C. Krutenat and W. R. Gesick, *J. Vacuum Sci. Technol.* **7**, S40 (1970).
- ²² N. Hosokawa, T. Tsukada, and T. Misumi, *J. Vac. Sci. Technol.* **14**, 143 (1977).
- ²³ W. M. Posadowski and Z. Radzimski, *J. Vac. Sci. Technol. A* **11**, 2980 (1993).
- ²⁴ A. Anders, S. Anders, M. A. Gundersen, and A. M. Martsinovskii, *IEEE Trans. Plasma Sci.* **23**, 275 (1995).
- ²⁵ N. Hosokawa, T. Tsukada, and H. Kitahara, *Effect of discharge current and sustained self-sputtering*, Proc. 8th Int. Vacuum Congress, Le Vide, Cannes, France, 11-14 (1980).
- ²⁶ J. Andersson, A. P. Ehiasarian, and A. Anders, *Appl. Phys. Lett.* **93**, 071504 (2008).
- ²⁷ J. Andersson and A. Anders, *Phys. Rev. Lett.* **102**, 045003 (2009).

- 28 J. Emmerlich, S. Mraz, R. Snyders, K. Jiang, and J. M. Schneider, *Vacuum* **82**, 867 (2008).
- 29 A. Anders, *Appl. Phys. Lett.* **92**, 201501 (2008).
- 30 R. A. Baragiola, E. V. Alonso, J. Ferron, and A. Oliva-Florio, *Surf. Sci.* **90**, 240 (1979).
- 31 A. Anders, *Appl. Phys. Lett.* **85**, 6137 (2004).
- 32 W. D. Sproul, D. J. Christie, and D. C. Carter, *The reactive sputter deposition of aluminum oxide coatings using high power pulsed magnetron sputtering (HPPMS)*, 47th Annual Technical Conference, Society of Vacuum Coaters, Dallas, TX, April 24–29, 2004, SVC, 96-100 (2004).
- 33 J. Andersson, *Measurements of frequency-dependent power losses (personal communication, unpublished)*, (2009).
- 34 S. M. Rossnagel and H. R. Kaufman, *J. Vac. Sci. Technol. A* **5**, 88 (1987).
- 35 J. Bohlmark, M. Ostbye, M. Lattemann, H. Ljungcrantz, T. Rosell, and U. Helmersson, *Thin Solid Films* **515**, 1928 (2006).
- 36 S. Sevvana, S. Chennadi, R. K. Lakkaraju, J. Li, D. M. Mihut, and S. L. Rohde, *Rate enhancement in high power pulsed magnetron sputtering growth using secondary discharge confinement*, 48th Annual Technical Conference Proceedings, Society of Vacuum Coaters, Denver, CO, April 23-28, 2005, 720-725 (2005).
- 37 E. P. Vaulin, N. E. Georgieva, and T. P. Martinenko, *Sov. Phys. Solid State* **19**, 827 (1977).
- 38 *Encyclopedia of Low Temperature Plasma (in Russian)*; edited by V. E. Fortov (Nauka, Moscow, 2000).
- 39 J. Bohdanský, H. Lindner, E. Hechtel, A. P. Martinelli, and J. Roth, *Nucl. Instrum. Meth. Phys. Res. B* **18**, 509 (1986).
- 40 R. Behrisch and W. Eckstein, *Nucl. Instrum. Meth. Phys. Res. B* **82**, 255 (1993).
- 41 *Sputtering by Particle Bombardment*; edited by R. Behrisch and W. Eckstein (Springer, Berlin, 2007).
- 42 R. E. Honig and D. A. Kramer, *RCA Rev.* **30**, 285 (1969).
- 43 R. P. Doerner, S. I. Krashenninnikov, and K. Schmid, *J. Appl. Phys.* **95**, 4471 (2004).
- 44 J. Vlcek, B. Zustin, J. Rezek, K. Burcalova, and J. Tesar, *Pulsed magnetron sputtering of metallic films using a hot target*, 52nd Annual Technical Conf. Proc., Society of Vacuum Coaters, Santa Clara, CA, SVC (2009).
- 45 W. D. Sproul, D. J. Christie, and D. C. Carter, *Thin Solid Films* **491**, 1 (2005).
- 46 *Reactive Sputter Deposition*; edited by D. Depla and S. Mahieu (Springer, Heidelberg, 2008).
- 47 K. Sarakinos, J. Alami, and M. Wuttig, *J. Phys. D: Appl. Phys.* **40**, 2108 (2007).
- 48 R. Chistyakov, “High deposition rate sputtering,” US 6,896,773, (2005).
- 49 R. Chistyakov, B. Abraham, and W. D. Sproul, *Advances in high power pulse reactive magnetron sputtering*, 49th Ann. Tech. Conf. Proc., Society of Vacuum Coaters, Washington, D.C., 88-91 (2006).
- 50 K. Sarakinos, J. Alami, C. Klever, and M. Wuttig, *Surf. Coat. Technol.* **202**, 5033 (2008).
- 51 A. Anders and G. Y. Yushkov, *J. Appl. Phys.* **105**, 073301 (2009).
- 52 S. Mahieu and D. Depla, *Appl. Phys. Lett.* **90**, 121117 (2007).
- 53 S. Mráz and J. M. Schneider, *J. Appl. Phys.* **100**, 023503 (2006).
- 54 A. Anders, *Surf. Coat. Technol.* **93**, 157 (1997).

Table and Table Captions

Table 1: Fit parameters a and b of sputter yields, calculated by the TRIM code, assuming a $\gamma_k = a_k E_k^{b_k}$ dependence; R is the correlation coefficient.

	<i>Ion mass</i> (amu)	a_k	b_k	R^2
Ar→C	39.9	0.0021	0.687	0.981
C→C	12.0	0.0562	0.224	0.820
Ar→Al	39.9	0.0296	0.512	0.981
Al→Al	27.0	0.1042	0.370	0.957
Ar→Ti	39.9	0.0425	0.443	0.977
Ti→Ti	47.9	0.0285	0.484	0.978
Ar→Cr	39.9	0.0861	0.457	0.980
Cr→Cr	52.0	0.0458	0.531	0.982
Ar→Cu	39.9	0.1421	0.468	0.980
Cu→Cu	63.5	0.0691	0.556	0.985
Ar→W	39.9	0.0429	0.521	0.984
W→W	183.8	0.0066	0.770	0.994

Figure Captions

Fig. 1 Sputtering yields for copper and titanium as a function of ion energy, as calculated by the TRIM2006 code, exemplifying the approximate $E_i^{1/2}$ dependence of the sputtering yield.

Fig. 2 Schematic presentation of the fluxes involved in the deposition by HIPIMS under conditions when the plasma is dominated by metal sputtered from the target; α , β , γ are the ionization probability, the return probability, and the sputtering yield, respectively; for further explanations see text.

Fig. 3 Illustration of the changes to HIPIMS rates in relation to DC rates caused by some of the effects discussed in the text. The numbers are for orientation only, and actual values may significantly depart from the displayed depending on the specifics of the deposition system.

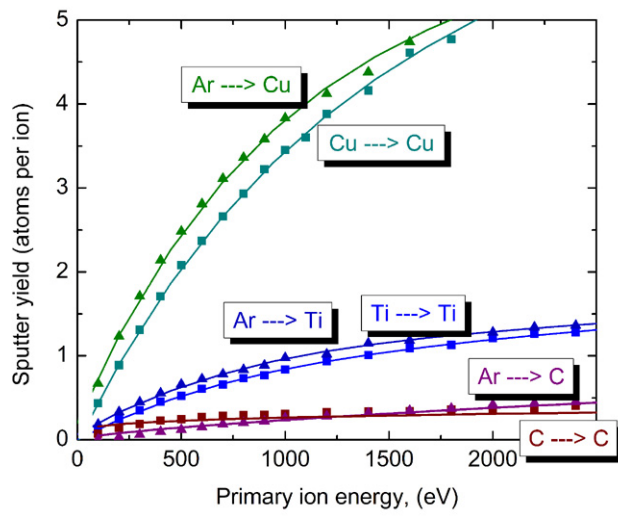


Fig. 1

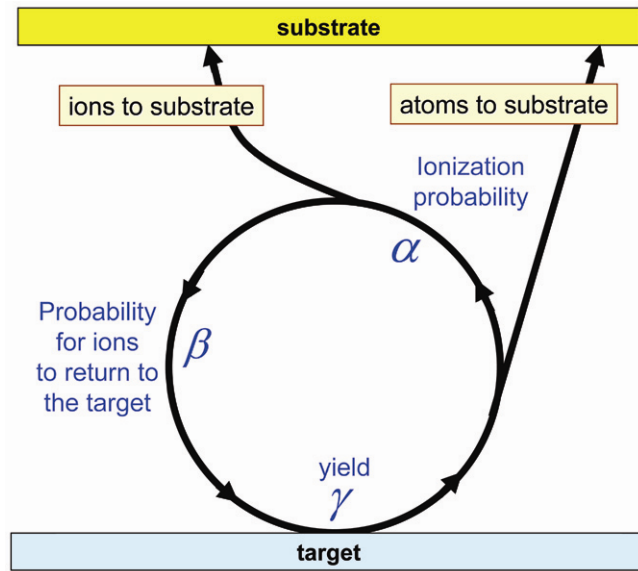


Fig. 2

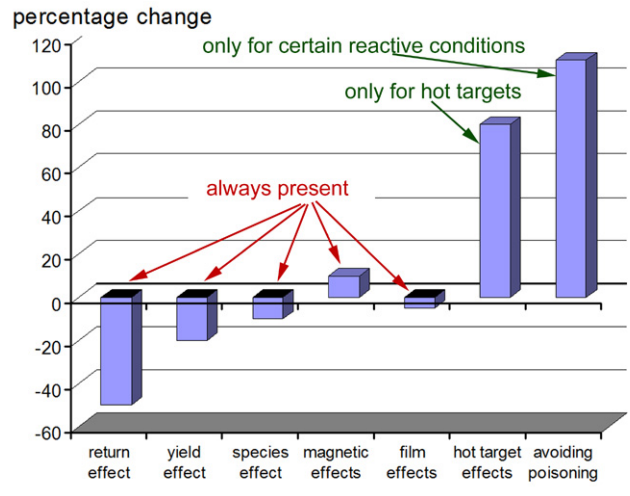


Fig. 3