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Influence of pulse duration on the plasma characteristics in high-power pulsed magnetron discharges

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High-power pulsed magnetron discharges have drawn an increasing interest as an approach to produce highly ionized metallic vapor. In this paper we propose to study how the plasma composition and the deposition rate are influenced by the pulse duration. The plasma is studied by time-resolved optical emission and absorption spectroscopies and the deposition rate is controlled thanks to a quartz microbalance. The pulse length is varied between 2.5 and 20 μs at 2 and 10 mTorr in pure argon. The sputtered material is titanium. For a constant discharge power, the deposition rate increases as the pulse length decreases. With 5 μs pulse, for an average power of 300 W, the deposition rate is $\sim$70% of the deposition rate obtained in direct current magnetron sputtering at the same power. The increase of deposition rate can be related to the sputtering regime. For long pulses, self-sputtering seems to occur as demonstrated by time-resolved optical emission diagnostic of the discharge. In contrary, the metallic vapor ionization rate, as determined by absorption measurements, diminishes as the pulses are shortened. Nevertheless, the ionization rate is in the range of 50% for 5 μs pulses while it lies below 10% in the case of a classical continuous magnetron discharge. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2159555]

I. INTRODUCTION

High-power pulsed magnetron (HPPM) discharges have drawn much interest these last couple of years. The technique offers the possibility to strongly ionize the flux of sputtered metal atoms and, consequently, to increase the control on the thin-film synthesis.[1–3] The technique is based on the production of high peak power glow discharges (usually in the kilowatt to the megawatt range). Those discharges are created in a short period to avoid target overheating and melting but with a repetition frequency high enough to obtain average powers in the range of classical direct current magnetron (mdc) sputtering devices.

One of the major drawbacks of the technique is the lower deposition rate in comparison to mdc sputtering systems.[4,5] It is believed that the deposition rate decrease is mainly due to two physical phenomena. Firstly, the self-sputtering phenomenon could be invoked.[5] If the pulses are too long, the metallic ions produced in the dense plasma created above the sputtering target can be recaptured on the cathode surface by the high negative voltage. As an obvious consequence, metal ions do not reach the substrate surface and if the self-sputtering yield is inferior to unity, the deposition rate decreases.[6] Secondly, metal ions must be able to diffuse freely towards the substrate. To fulfill that condition, the target-to-substrate gap must be conductive enough. In other words, the space-charge density of the metal ions must be compensated by surrounding electrons. This situation can be achieved by filling the cathode-to-substrate gap by a plasma, for example, using an inductively coupled radio-frequency antenna.[7]

It is therefore necessary to study in which discharge conditions the self-sputtering regime is reached. As the ionization of the metal atoms is submitted to kinetic mechanisms that require sufficient time to induce quasicomplete ionization of the sputtered vapor, we have focused, in this paper, our attention on the study of the pulse duration and on its influence on the plasma characteristics, especially on the deposition rate and the ionization rate.

II. EXPERIMENTAL SETUP

The only difference with the previously presented experimental setup (see Ref. 8) is that the dc power supply is replaced by a high-power pulsed generator.[9,10] This generator is designed to allow working with electric pulses developing high peak power up to tens of kilowatts. This way, high ionization rate of the sputtered metallic species can be achieved.[7,10] The pulse duration can be varied between 2 and

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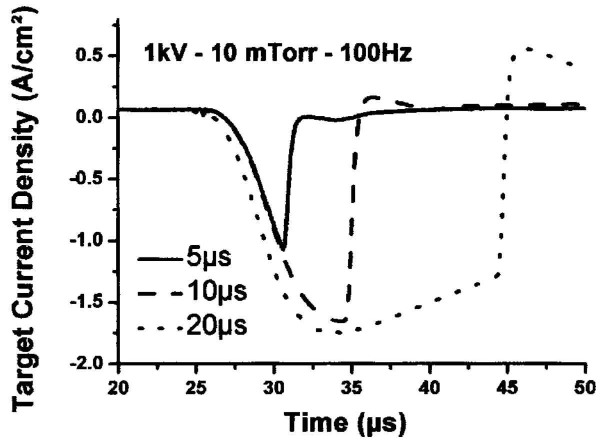
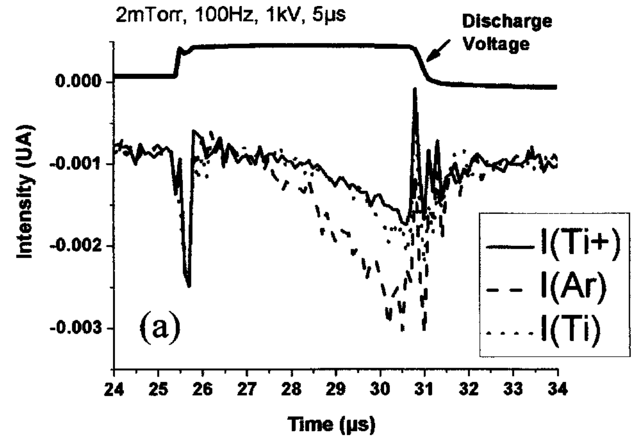


FIG. 1. The shape of the target current density at 10 mTorr, 1 kV, and 100 Hz changes as the pulse is lengthened. For a 20-μs-long pulse, the curve exhibits a maximum. The current density is calculated using the race track surface (30 cm^2).



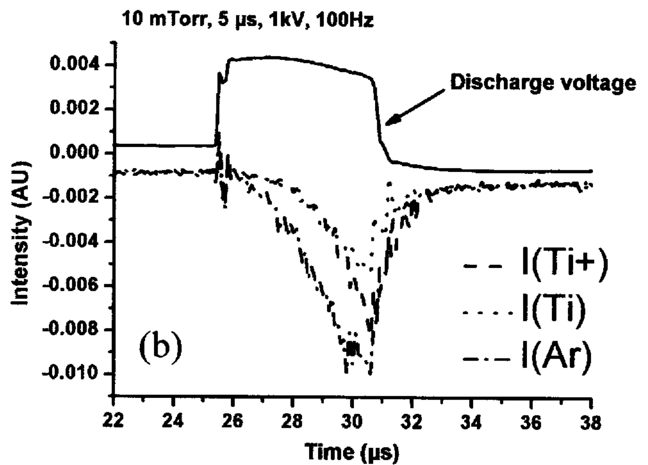


FIG. 2. tr-OES of the 5-μs-long pulsed discharge running at (a) 2 and (b) 10 mTorr.

50 μs. As the pulse duration is diminished, the repetition frequency is increased (up to 12 kHz) in order to keep the same time-averaged effective power. The term effective power is related to the power effectively delivered to the plasma. This power is calculated thanks to the following integral:

$$P_{\mathrm{eff}} = v \int_0^{\tau} I(t) V(t) dt. \quad (1)$$

In this relation, τ is the pulse duration and v is the repetition frequency. $I(t)$ and $V(t)$ are, respectively, the discharge current and voltage. $I(t)$ is measured thanks to a high-current probe (Tektronix TCP220) and $V(t)$ is measured thanks to a high-voltage probe (Tektronix P6015). Both signals are recorded by a digitizing oscilloscope (Tektronix TSD420A). In Fig. 1 is represented the evolution of the discharge current density (taking into account a race track surface of 30 cm^2) for pulse lengths of 5, 10, and 20 μs at 10 mTorr. The discharge voltage is set to 1 kV. One can notice the sharp breakdown delay of the discharge current (inferior to 1 μs). This feature enables a perfect control of the pulse duration. The current decreases to zero as rapidly as the discharge voltage is switched off. The shape of the current curves is changing as the pulse is lengthened. One can assume that the plasma characteristics (electron density and temperature, gas temperature and composition, etc.) will change with the pulse duration. For long pulse (>10 μs), the discharge must be rich in metal ions which can be used to sputter the Ti target with a lower yield than argon ions in a stationary discharge. The current density at the end of the pulse decreases.

In order to study the pulsed discharge, which is run in pure argon, at a pressure of either 2 or 10 mTorr (0.26 and 1.3 Pa, respectively), absorption and emission spectroscopies are used. Both diagnostic tools can be used in either a time-resolved or in a time-averaged mode. Time-resolved absorption measurements are possible thanks to the pulsed hollow cathode used as optical source. The hollow cathode signal can be synchronized with the pulsed magnetron discharge.[7] The second advantage, which has been previously exploited for the diagnostic of a rf amplified magnetron discharge,[8] is that the pulsed hollow cathode allows doing absorption on Ti^+ ions. Both diagnostics are done 5 cm above the magnetron target near the substrate holder. A quartz microbalance used to estimate the deposition rate is placed on the same plane than the substrate holder 8 cm above the target.

III. EXPERIMENTAL RESULTS

A. Evolution of optical emission intensities with the pulse duration

Line emission intensities of neutral argon, of titanium neutral, and ions have been recorded versus time using time-resolved optical emission spectroscopy (tr-OES) for pulse lengths of 5, 10, and 20 μs at 2 and 10 mTorr. The 750.4 nm line is used for neutral argon atoms while resonant transitions at 363.5 and 338.4 nm are recorded for Ti and Ti^+, respectively. Figures 2(a) and 2(b) represent the cases of a 5-μs-long pulse for 2 and 10 mTorr, respectively. For short pulses, argon line is the first to rise and dominates up to the end of the pulse. As a general observation, argon line is always the first to rise as argon atoms are the first species to be excited by the highly energetic electrons at the beginning of the pulse. Titanium neutral line appears 1 or 2 μs after. Titanium ionic line appears lastly. This global behavior shows the kinetic mechanisms of metal ionization. First, argon is excited and ionized. Argon ions are used to sputter

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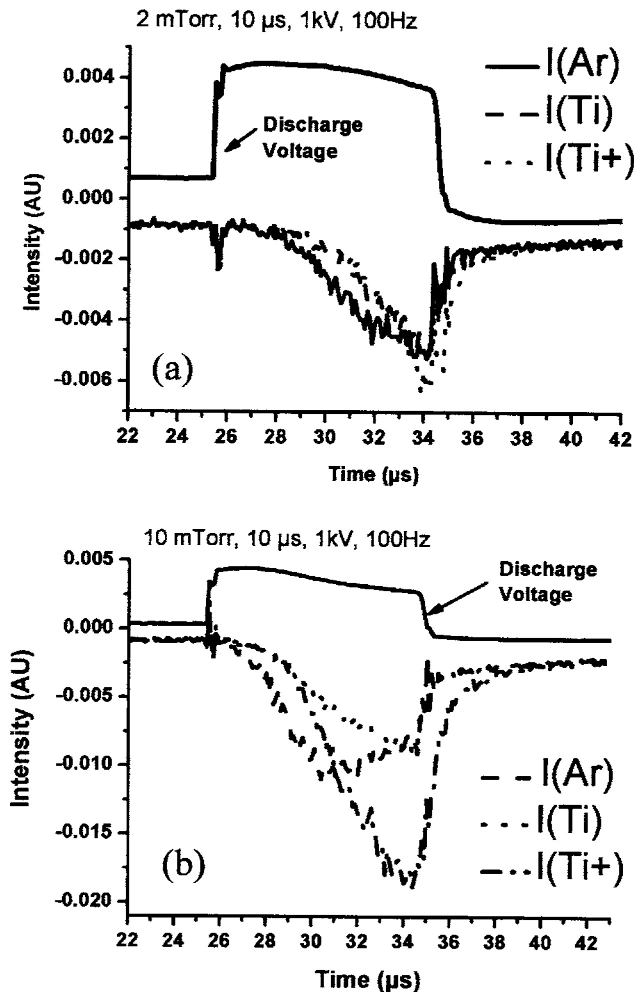

FIG. 3. (a) tr-OES of the discharge running with a pulse of 10 μs long at 2 mTorr. (b) tr-OES of the pulsed discharge at 10 mTorr, 10 μs long pulse. One can remark the decrease of the I(Ar) line after ~ 5 μs, indicating an important decrease of the argon density and/or the electron temperature in front of the sputtering target.

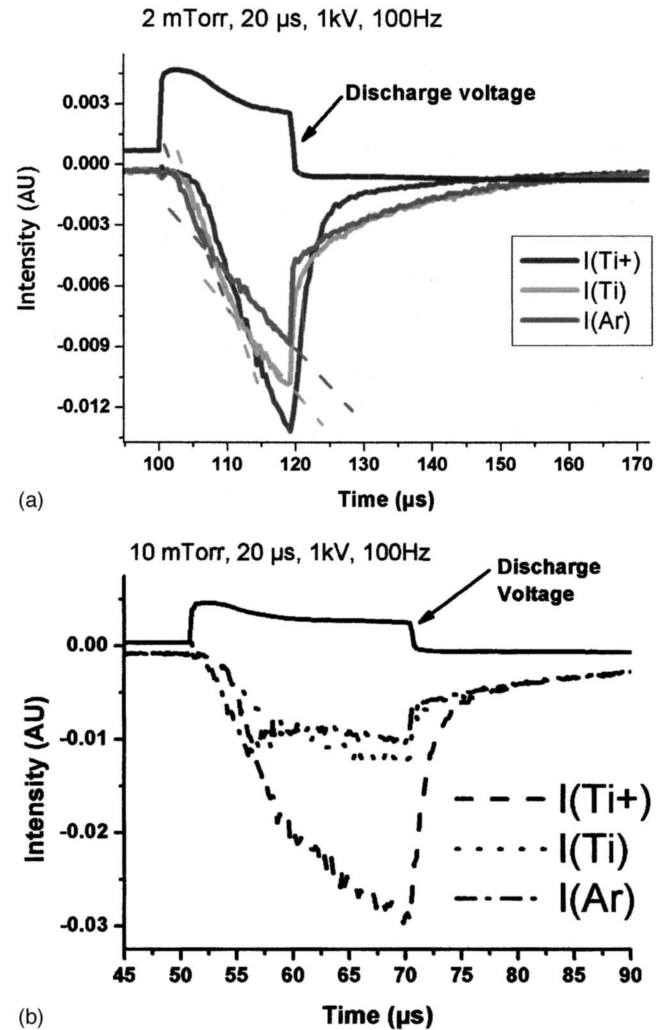

FIG. 4. (a) Evolution of line intensities for a pulse of 20 μs long at 2 mTorr. As the discharge is running on, the slope of the intensity emitted by the argon and titanium changes. This modification of the slope is underlined by the addition of two tangential lines to the I(Ti) and I(Ar) curves. (b) Evolution of line intensities for 20-μs-long pulses at 10 mTorr. As the argon atoms are swept away due to the intense sputtering wind, the magnetized plasma is rich in Ti^+. The metal ions can be used for sputtering and the discharge enters in a regime of self-sputtering. The intensity of Ti^+ tends to saturate as this sputtering regime sets up.

metal atoms which have to stay in the dense magnetized plasma that is building up in front of the cathode to be finally ionized. Of course, to increase the ionization/excitation probabilities, sputtered atoms have to be slowed down by an increase of the working pressure. As shown by measurements in Figs. 2(a) and 2(b), as the pressure goes from 2 to 10 mTorr, the mean free path is reduced and as a result line intensities are amplified. At 10 mTorr, ionization seems more efficient. $I(Ti^+)$ to I(Ti) ratio is higher than at 2 mTorr.

When the pulses are longer, titanium atoms are spending more time in the plasma and, then, their probability to be ionized is higher (see Fig. 3). The $I(Ti^+)$ to I(Ti) ratio is increased compared to shorter pulses. Moreover, one can observe that in Fig. 3(b), argon line intensity dramatically decreases approximately 5 μs after the beginning of the pulse. As the optical line intensity is proportional to the concentration of ground-state level and to the electron density and temperature (in the case of an excitation induced by electron collisions),[11] two phenomena have to be taken into account. First, one can invoke the sputtering wind that sweeps away background gas atoms.[12] This phenomenon induces a local decrease of the argon density due to the intense target sputtering provoked by the large ionic current density. The second phenomenon is the cooling of the electron temperature that occurs as more and more titanium atoms are injected into the magnetized plasma as the pulse is running on.[13] Titanium atoms have a low ionization potential (6.8 eV) compared to argon (15.6 eV).

When the pulses are even longer [20 μs, Figs. 4(a) and 4(b)], one can expect that the argon atom density will be reduced in front of the cathode. Then titanium ions, which are more and more numerous, will be recaptured because of the target high negative voltage (~ -1 kV). In these conditions, the discharge enters a regime of self-sputtering. This effect is less pronounced at 2 mTorr [Fig. 4(a)] than at 10 mTorr [Fig. 4(b)] where a variation of the slope of the line intensity as a function of time can clearly be observed. The population of titanium ions tends to saturate as they are

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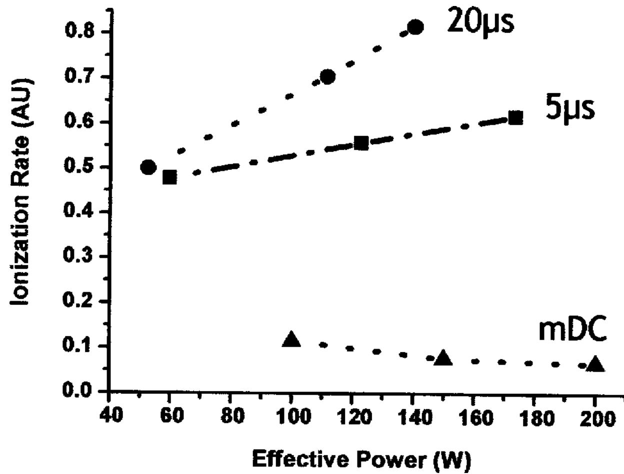

FIG. 5. Ionization rate of the sputtered metallic vapor as determined using the Ti and Ti^+ absorption coefficients (for HPPM, 20 and 5 μs pulse lengths and mdc). The highest ionization rates are obtained with HPPM device. The pressure is kept at 10 mTorr and the initial cathode voltage is 1 kV for HPPM.

used to sputter the target. Those observations can be correlated to the shapes of the target current-density curves reported in Fig. 1.

B. Evolution of the global ionization rate of the metallic vapor with the pulse length

To qualitatively estimate the global (time-averaged) ionization rate of the metallic vapor near the substrate, atomic absorption spectroscopy has been employed. The term qualitatively is used as, actually, we do not have any idea of how the discharge temperature changes with the pulse length and the repetition frequency. To do absorption measurements onto titanium neutrals and ions, we have used the pulsed titanium hollow cathode lamp.

In order to obtain the time-averaged value of the ionization rate, as the magnetron discharge is operated at 100 Hz and the hollow cathode lamp at 55 Hz, the recordings of line intensities necessary to estimate the absorption coefficient for Ti and Ti^+[8] are made for approximately 2 s. The ionization rate of the metallic vapor ρ is estimated using the following equation:

$$\rho = \frac{A(Ti^+)}{A(Ti^+) + A(Ti)}. \tag{2}$$

In this relation, Ti and Ti^+ absorption coefficients are, respectively, labeled A(Ti) and $A(Ti^+)$. The results of the measurements are reported in Fig. 5 as a function of the effective power delivered to the plasma, for an argon pressure of 10 mTorr, in the cases of the mdc and the HPPM with 5- and 20-μs-long pulses. For the HPPM device, the effective power is increased by only increasing the repetition frequency. For 5 μs pulses, the repetition frequency has to be set between 3 and 12 kHz while for 20 μs pulses, the frequency has to range from 0.1 up to 0.5 kHz. It appears that the highest ionization rate is obtained with long pulses, which confirms the trends given by the line intensity ratios

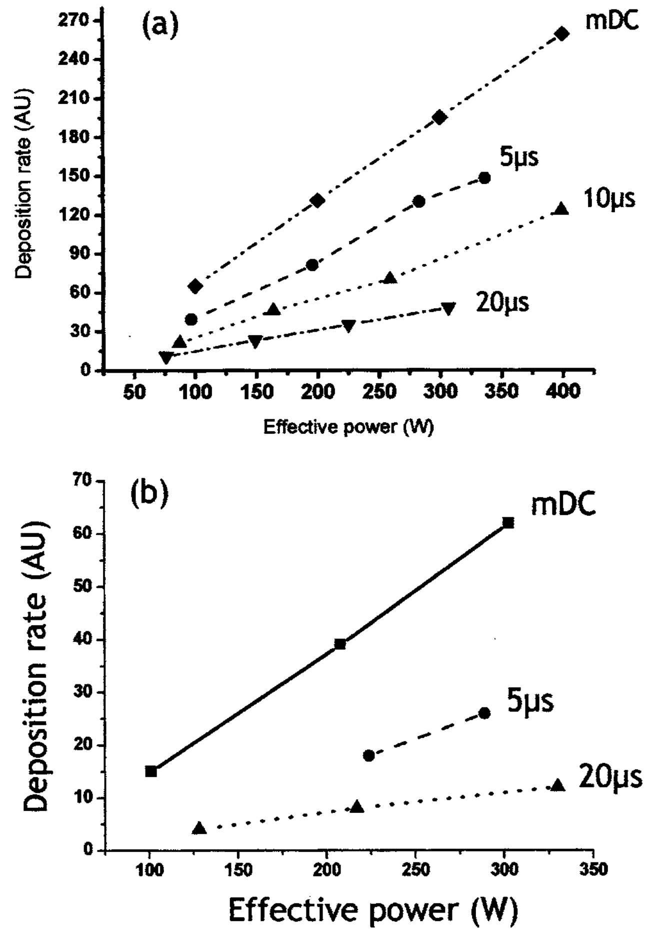

FIG. 6. Influence of the pulse length on the deposition rates measured in HPPM sputtering (a) at 2 and (b) 10 mTorr and comparison with the mdc mode.

recorded by tr-OES (see previous section). The lowest value is obtained with the mdc ($\rho < 10\%$).

Those first results on the influence of the pulse length on the evolution of the ionization show that the highest ionization rate is obtained in HPPM with long pulses. But tr-OES measurements have demonstrated that as the pulse becomes longer, the discharge regime changes from argon sputtering to self-sputtering mode. This modification of the sputtering mode can obviously modify the deposition rate of the thin films. The evolution of the deposition rate as a function of pulse length is the focus of the following section.

C. Influence of the pulse length on the deposition rate

Deposition rate has been recorded for different effective powers in mdc and in HPPM modes. The results are presented in Figs. 6(a) and 6(b) for 2 and 10 mTorr, respectively. Whatever the pressure is, the mdc exhibits the highest deposition rates. In HPPM regime, for the same effective power (but for different repetition frequencies), the deposition rate increases as the pulse length decreases. At 2 mTorr [Fig. 6(a)], 300 W of effective power, the deposition rate achieved with pulses 5 μs long is ~70% of the one obtained

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with the mdc discharge. At the same pressure, same power, but with longer pulses (20 μs), the deposition rate in HPPM is $\sim$20% of the one measured in mdc.

To explain the difference in deposition rate between the short and long pulse cases, one could at first invoke that for 5 μs pulses, as shown in the previous section, the self-sputtering regime is not clearly established. Then, an important proportion of the ions produced in the magnetized plasma can diffuse towards the substrate and contribute to film deposition. This is not the case for long pulses where self-sputtering is more present. This effect could be an advantage in the case of metals which exhibits self-sputtering yields superior to unity as e.g., copper and tantalum.[6] The second point is that for short pulses at the same average effective power, the frequency is approximately one order of magnitude higher. Then, the target-to-substrate gap is more frequently filled by the electrons and ions produced during the electrical pulses. At high frequencies, the plasma filling the gap between both electrodes has less time for relaxing. This could lead to an increase in charged particle density. This more uniform provision of electric charges would lead to a better time-averaged conductivity of the plasma. It is known that it is important to compensate the space-charge density of the ions to improve their transport.[7,14] An equivalent reasoning can be used to discuss the effect of the gas heating provoked by the intense sputtering wind. For short pulses and higher repetition frequencies, the local decrease of background gas atoms could be more efficient as the relaxation time is smaller than for long pulses and low frequencies. The transport of the sputtered particles would be therefore more efficient. Both these assumptions could explain the increased deposition rate in the case of short pulses.

So using short pulses, it is possible to partially overcome the decrease of the deposition rate encountered while working in HPPM regime.

IV. CONCLUSIONS

The design of a high-power pulsed generator featuring a rapid discharge breakdown delay has allowed us to produce a highly ionized titanium vapor ($\rho > 50\%$ at 10 mTorr) and to especially focus our study on the influence of the pulse duration onto the plasma characteristics.

As a key result, it appears that when the pulse duration is increased from 5 to 20 μs, the discharge evolves from argon sputtering to self-sputtering mode. The result is explained by the enrichment of the magnetized plasma by metal ions and the decrease of argon neutral density.

Time-resolved optical emission spectroscopy measurements done 6 cm above the magnetron target have shown an increase of the line intensity ratio of ionic to neutral titanium with the pulse duration: the ionization rate of the vapor increases.

Time-averaged qualitative atomic absorption spectroscopy measurements have confirmed this trend. This put into evidence the necessity for the sputtered atoms to spend enough time in the plasma to be highly ionized. In other words, atoms have to reside sufficient time for the vapor to be ionized. In consequence, it is possible to tailor the ionization by varying the pulse length.

The second experimental finding is that it is also possible to modify the deposition rate by changing the pulse duration (and simultaneously the repetition frequency in order to keep the same average power). The shorter the pulses are, the higher the deposition rate is. A deposition rate being 70% of the value reached in dc magnetron is obtained with 5 μs pulses at 2 mTorr. It is therefore necessary to balance ionization and deposition rates following the characteristics of the thin films needed.

Some perspectives to increase further the deposition rate would be to apply a reverse (positive) voltage to the cathode, after the primary sputtering pulse, in order to accelerate the metal ions towards the substrate. In the same way, a special combination of electrostatic lenses to extract the metallic ions out of the magnetized plasma could be implemented. Further works should be focused on the comprehension of the variation of the deposition rate with the frequency and pulse length. It should be especially necessary to implement time-averaged electron-density measurements in the target-to-substrate gap and gas temperature measurements to confirm some assumptions.

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