TIME RESOLVED TUNABLE DIODE LASER ABSORPTION SPECTROSCOPY ON Al AND Ar\textsuperscript{M} ATOMS IN HIGH POWER PULSED MAGNETRON SPUTTERING*  

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Time resolved tunable diode laser absorption spectroscopy (TD-LAS) on Al and metastable Ar atoms in High Power Pulsed Magnetron Sputtering was performed in order to study the time and space dependency of both species in the afterglow of pulsed plasma. In doing that synchronizing of the high power pulse frequency with the scanning frequency of the diode laser was necessary and performed.  

Key words: optical absorption spectroscopy, laser diode, TD-LAS, time resolved, HPPMS, HiPIMS.  

1. INTRODUCTION  

Magnetron sputtering is a well established technique for the deposition of thin films [1]. For many applications, an increase of the ionization of sputtered atoms is desirable to control the energy and direction of the impinging species on the substrate [2]. One way for answering this requirement is the High Power Pulsed Magnetron Sputtering (HPPMS) technique [3, 4], which is an emerging ionized physical vapor deposition (IPVD) process. Within the short pulse duration (50-200 µs) the high power density sustains very dense plasma near the cathode surface which induces efficient sputtered vapor ionization.  

Recently, a new HPPMS process operating with pre-ionized gas and microsecond range (1-50 µs) was developed [5, 6]. The current pulse duration and shape can be adapted for every magnetron configuration, in order to obtain maximum ion-to-neutral ratios without electric arc development, to control the transition towards the self-sputtering regime, and to optimize the reactive mode  


operation [7, 8]. In order to understand and optimize the deposition process, it is important to control creation and transport of all the species in the reactor. The first characterization with spatial and temporal resolution of the sputtered particles flux was achieved by optical absorption spectroscopy (OAS) using a pulsed hollow cathode lamp [9]. Laser-based techniques have higher performances than OAS in terms of sensitivity and precision. Recently available diode laser systems [10, 11], emitting in the blue wavelength range permit to perform experimental measurements on the ground state of metal species. Absorption measurements have been already used to determine the temperature and density of thermalized particles in DC magnetron discharges [12], and the time averaged density and temperature of aluminum atoms for a HPPMS system [13].

In this paper we present experimental results regarding the time evolution of absorbing species density in a pre-ionized HPPMS system operating in microsecond range, with 200 Hz repetition frequency.

2. EXPERIMENT AND RESULTS

The experimental set-up essentially consisted of a vacuum chamber housing the magnetron plasma source, and the external optical arrangement of the laser system (figure 1). The vacuum chamber was a cylindrical stainless steel reactor closed by optical windows at each side. The chamber was turbo molecular pumped down to a residual pressure of $2.6 \times 10^{-5}$ Pa. Argon was used as working gas at the pressure of 2.6 Pa. During experiments, the target was water-cooled. To prevent aluminum contamination of the windows, internal diaphragms have been used.

![Fig. 1 – Experimental setup.](image)

The planar magnetron cathode is circular one (5.6 cm diameter), connected to a high power adapted pulsed generator [5-9]. Typical pulse shapes of the applied voltage and current intensity are presented in figure 2. The pulse has the following
characteristics: the pulse duration is 5 µs, the repetition frequency is 200 Hz, the pre-ionization current intensity is 4 mA (−170 V), the cathode peak voltage can reach −1000 V and the peak current intensity 50 A.

The laser system consists of two independent tunable diode lasers and control units for diode temperature and diode current (Toptica DL 100), one emitting in the blue and the other one in the red spectral domain. The laser beam was guided through the plasma reactor and optic measurement system by using two beam splitters and two mirrors as shown in figure 1. For argon metastable atom measurement, the laser was tuned linearly around 811.531 nm. For aluminum atom measurement the laser source was a tunable single-mode blue-laser diode system, centered at 394.512 nm.

By synchronizing the HPPMS pulse frequency with the scanning frequency of the diode laser we are able to obtain the absorption profile in virtually any instant on the pulse period. For 200 Hz repetition rate of the pulsed power supply, during a laser period (~ 0.5 s) about 100 HPPMS pulses occur. During a post-discharge period (5 ms) the wavelength variation is 0.08 pm and could be considered as constant. So, we may assume that to each HPPMS pulse period (duration between two successive discharges) corresponds to a given absorption wavelength so that the recorded signal gives the time evolution of the absorbance at the specific wavelength. Due to the relatively low gas pressure and plasma density one may assume that the main broadening process of the spectral lines in the discharge is Doppler one so that information about the thermal energy of the particles can be obtained.

Density and temperature measurements of sputtered Al atoms and metastable argon atoms were achieved using a similar method that described in [13, 14]. The temperature variation of Ar metastables during the afterglow does not exceed 50 K (can be higher only very close to the target immediately after the pulse is turned off),
so the absorbance at resonance wavelength will show properly the relative evolution of metastables density.

Space resolution of \(~1\) mm in the direction perpendicular to the target (axial variation) is also obtained by manually translating the laser path with an external mirror, with \(3\) mm step. By combining the space and time resolution achievable by absorption measurements we focused mainly on the time period between the two pulses (afterglow). In this way the transport phenomena of sputtered species from target surface towards substrate as well as the metastable kinetics processes can be studied.

Figures 3 to 6 show the main results, concerning the time and space variation of the relative density of both investigated species. Figure 3 presents the absorbance of the metastable argon (grey scale) in time (abscissa) and along the normal \(z\)-axis (ordinate). Let us remember that the pulse width is only \(5\) µs and it acting close to the time origin. Hence \(\text{Ar}^M\) density is maximum (black) in front the target \((z \sim 10\) mm\) just after the pulse cut off and it decays towards the minimum reached \(~1.5\) ms later. Then the \(\text{Ar}^M\) density starts to increase in all the plasma volume due to the action of the pre-ionization. Its secondary maximum is located \(~10\) mm in front of the target \((t > 4\) ms\) corresponding to the maximum of the plasma density trapped by magnetron field configuration.

Figure 4 shows, for the sake of clarity, the time evolution of absorbance at the resonance wavelength at several axial positions obtained as line sections of the Fig. 3 at constant \(z\). Four representative positions were selected in the plasma: close to the target \((z = 1\) mm\), in the dense magnetron plasma \((z = 12\) mm\), in the diffusion plasma \((z = 24\) mm\), and far from the target \((z = 45\) mm\). One can easily see the different decay rates of the \(\text{Ar}^M\) density at the beginning of the afterglow, depending on the axial position. \(\text{Ar}^M\) loss is sharp (more than 1 decade) and fast very close to the target \((z \leq 12\) mm\) due to at least two kind of processes: (i) the electron impact quenching and (ii) diffusion and surface losses (cathode, anode ring, walls, …). Far away \((z > 40\) mm\) the decay is much slower and the amplitude of the density variation is restricted to about 30 %. For intermediary positions the \(\text{Ar}^M\) behavior is a mixture of the two situations discussed above (typical shape – Fig. 4, \(z = 24\) mm).

At \(1.5\) ms, the minimum of the \(\text{Ar}^M\) density is reached being consistent with the typical decay time of the electron density decay in the afterglow \(~2\) ms with one decade [15]. The \(\text{Ar}^M\) quenching is active even at low electron energy (far in the afterglow) because the energy variation between the 4s (metastable level) and the 3d (radiative level) is about few tens of meV. On the other hand, at this moment the pre-ionization is well established (full black line – Fig. 4) and consequently the electron as well as \(\text{Ar}^M\) densities start to increase.
Figure 3 – Time and space evolution of maximum absorption signal, at 811.531 nm corresponding to ArM absorption line.

Figure 4 – Time evolution of maximum absorbance at λ=811.531 nm for different distances from target surface. Al Target, P=2.6 Pa.

Figure 5 shows the equivalent of the time-space representation in figure 3 for Al atoms. The evolution shows an initial maximum placed close to the plasma pulse, followed by a decrease mainly due to diffusive loss of sputtered Al atoms, analogous to the ArM behavior at the beginning of the afterglow. On the contrary, after the pre-ionization voltage is established, the aluminum density is below the detection limit since the magnetron current is only 4 mA. The travel time of Al is compatible with the mean thermal velocity of ~50 m/s, corresponding to a flight distance of ~10 cm from the target to the substrate.
The Ar\textsuperscript{M} kinetics, namely production and destruction mechanisms, are mainly related with the temporal and spatial variation of the electron density, collisions with electrons being the main creation and loss processes. But, surface processes have to be also considered in the Ar\textsuperscript{M} balance the fact emphasized by the time evolution of absorbance at the resonance wavelength close to the target ($z = 1$ mm), which shows decreasing of argon metastable atoms density below the limit of detection. Comparison between time and space evolution of metal and argon species shows that the metal atoms originating from the target are governed by transport phenomena through the plasma volume, while the Ar\textsuperscript{M} are strongly influenced by electron density variation in time and space. The pre-ionization acts as a continuous source of electrons between two successive high power pulses.

3. CONCLUSIONS

A new method, based on laser diode absorption spectroscopy, was used for time evolution measurement of the neutral species density for HPPMS systems. It is then possible to obtain the absorption profile in virtually any instant on the pulse period. This method has proved its effectiveness for both metal sputtered species, Al atoms, and also for the metastable states of argon buffer gas. This method could be extended in the future to laser induced fluorescence measurements using tunable laser diodes.

The space and time behavior of Ar metastables, which is strongly conducted by electron impact processes, suggests that in the short pulse HPPMS discharge with pre-ionization, weakly ionized bulk plasma in the afterglow induced by pre-ionization acts as an important Ar metastable source.
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