## Towards deeper understanding of an HiPIMS discharge by time-resolved optical plasma diagnostics

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As a promising pulsed sputter deposition technique, High-Power Impulse Magnetron Sputtering (HiPIMS) required an extensive time-resolved plasma characterization. Being essentially non-intrusive, optical plasma diagnostics methods posses in particular the time- and space- resolution(s) which normally exceed the typical needs of a typical HiPIMS geometry [1] and, therefore, can be readily applied for characterization of the high-power pulsed discharges.

Our recent progress on time-resolved study of a HiPIMS discharge is presented in this work. The selected Optical Emission Spectroscopy (OES), Resonance Optical Absorption Spectroscopy (ROAS), and Laser-Induced Fluorescence (LIF) results are reported. Several parameters, such as the absolute densities of the sputtered species, gas temperature, velocity distribution function of the sputtered species are described.

The schematic representation of the studied magnetron discharge system for the implementation of LIF diagnostics [2] is given in Fig. 1. The analyzed plasma regions in case of LIF are nearly cylindrical with the width of about 5 mm and length of about 10 cm. For OES and ROAS diagnostics the probed plasma volume had the width of about 1 cm and was located at about 5 cm above the magnetron target.



Fig. 1. Schematic representation of the HiPIMS plasma together with the probed volume by LIF.



Fig. 2. Time-resolved evolution of the absolute densities during the HiPIMS on- (20  $\mu$ s) and off-(980  $\mu$ s) time measured by ROAS. Pressure = 20 mTorr.



Fig. 3. Time-evolution of the Ti sputtered atoms density measured by LIF in a HiPIMS plasma at three different distances from the magnetron target. Dashed lines indicate end of the pulse.

Results obtained by ROAS [3] during sputtering of Ti in Ar demonstrate the large differences in the absolute densities for Ti, Ti<sup>+</sup>, Ti<sup>met</sup> and Ar<sup>met</sup>, as shown in Fig. 2. Such an integrated time-resolved representation indicates a highly non-uniform time-evolution of the sputtered Ti, its ions as well as the metastables of Ti and Ar, which are supposed to play an important role in HiPIMS plasmas. The arrival time (to the probed volume) for all the measured species during the HiPIMS pulse is found to be different, as it can be seen from Fig. 2. In addition, this is accompanied by the population inversion of the ground and metastable states energy sublevels during the pulse [3] (not shown). These results are in agreement with the Ti ground state density recently measured by LIF [4] at different distances from the magnetron target (Fig. 3). Here we can clearly observe a "wave" of the sputtered species propagating away from the magnetron target at high pressure, as well as the rarefaction of the Ti density during the 20 µs plasma pulse (Fig. 3(b)). These effects are absent, however, at low pressure (Fig. 3(a)).

Along with the above described measurements, the gas temperature ( $T_{gas}$ ) associated with the thermalized species during the pulse is determined based on the rotational band analysis of N<sub>2</sub><sup>+</sup> (391.4 nm) assuming partial bulk gas thermalization [5].  $T_{gas}$  is found to grow linearly during the HiPIMS pulse being mainly a function of the energy  $E_P$  delivered per pulse. The minimum and maximum gas temperatures attained during the HiPIMS plasma pulse measured in this way are presented in Fig. 4. As it can be seen, gas temperature depends strongly on the target material, and increases linearly with increasing of  $E_P$  for heavier target atoms (W), whereas it saturates quickly for the lighter ones (Ti).

In addition, LIF spectroscopic diagnostics applied for time-resolved characterization of the velocity distribution of the sputtered Ti atoms in HiPIMS reveals a rapid increase of the full width at half maximum (FWHM) of the velocity distribution function (vdf) during the plasma pulse following by a fast decrease of the vdf's FWHM during the plasma off-time [2, 6], as it is summarized in Fig. 5 for 15 mm of the distance from the target. After taking into account possible valuable broadening mechanisms (such as Stark, Zeeman, instrumental, etc.), the described vdf behaviour was finally attributed to the Doppler-shift phenomenon in plasma, i.e. to the rapid broadening (on-time), or relaxation (off-time) of the vdf of sputtered species likely accompanying by either rarefaction [7] (on-time) or refilling [8] (off-time) effects in the bulk gas.



Fig. 4.  $T_{gas}$  attained at the beginning  $(T_{min})$  and at the end  $(T_{max})$  of the HiPIMS plasma pulse calculated based on the linear fits of the  $T_{gas}$  time-evolution during the pulse. The results are presented for Ti (a) and W (b) sputtering at various  $E_p$  values and different  $N_2$  contents. Pulse duration is 20 µs. Ar pressure is 20 mTorr.



Fig. 5. The time-resolved evolution of FWHM of the Ti absorption line measured by LIF after the deconvolution with the side broadening effects determined at two gas pressures. The maximum attained broadening corresponds to about 5 km/s of the velocity component parallel to the magnetron target.

As we can observe in Fig. 5, the FWHM saturates in the second part of the plasma pulse forming a plateau (where FWHM is about 10 pm). This effect is also visible at somewhat larger distances from the target. Based on the FWHM data shown in Fig. 5, it should be noted that, the high broadening values obtained during the plasma pulse at 15 mm are, in our opinion, mainly due to the intensive bulk gas rarefaction in the second half of the plasma pulse which allows observing the "initial" velocity of the sputtered species without their considerable deceleration by collisions with the bulk Ar. At the same time, at higher distances from the target (or at higher pressure) the maximum attainable FWHM values decreases considerably.

Summarizing the diagnostics results, the following time- and space- resolved picture of a HiPIMS discharge can be deduced:

During the plasma pulse, Ti and Ti<sup>+</sup> species are produced first and they are keeping their ground state and metastable sublevels population inverted due to rather high level of the gas excitation. This is following by an increase of the density of Ti and finally Ar metabstables, which also experience the population inversion when reach their maxima.

At the same time, during the HiPIMS pulse, the velocity distribution gets essentially broadened presumably due to the bulk gas rarefaction, and rapidly shrinks far away from the target as well as right after the plasma pulse with a faster-than-exponential decay, which is due to the collisional relaxation of the vdf enhanced additionally by the refilling effect. These effects, as expected, are especially prominent at low pressure and in the vicinity of the magnetron target (z = 15 mm).

The gas temperature associated with the thermalized bulk Ar species in the discharge (measured by the rotational structure of the added nitrogen) increases linearly during the plasma pulse proportionally to the total energy  $E_P$  delivered during the pulse. After the HiPIMS pulse ends, both the ballistically moving and thermalized heated species rapidly dissipate their energy, and the gas cools down to nearly the room temperature during the off-time. The bulk gas temperature is found to be also different for heavy and light sputtered particles due to the different ways of the energy transfer to the bulk gas. Observed "bulk temperature" trends reflect the overall gas heating and relaxation channels in HiPIMS as a function of the applied power, distance from target, sort of the sputtered species, etc.

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