SPECTROSCOPIC AND ION PROBE DIAGNOSTIC OF A LASER CREATED TITANIUM PLASMA IN NITROGEN ENVIRONMENT

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Optimisation and control of pulsed laser reactive deposition of TiN thin films in nitrogen environment require the understanding of the plasma formation, gas pressure effect and the laser flux influence[1]. In this contribution, time and space resolved emission spectroscopy and ion probe measurements were used to study the plasma dynamics in nitrogen gas environment.

1. Experimental arrangement

The plasma is created by a Q switched Nd-Yag laser delivering 200mj energy in 30 ns focused on a rotating pure titanium target through a 40cm focal lens, at 45° angle giving a laser flux of 10^9 W.cm^{-2}. The luminance plume of the plasma is optically imaged at 90° along the target normal on the 100 µm entrance slit of a spectrometer giving a resolution of 0.8 Å. The light emission is collected by a fast photo-multiplier connected to a fast storage oscilloscope. An ion probe consisting on a shielded single circular plate 2mm in diameter and biased at –50 V, is placed along the plume propagation axis. When the gas pressure is increased, a rapid change in plasma expansion and the species kinetic energy across the plume can be expected. The use of a small area probe ensures spatial selectivity of on-axis ion signals exclusively.

2. Results and discussion

Spectroscopic analysis

Time integrated spectra

In recording time integrated spectra in the wavelength range 2000Å -7500Å , we find spectral line emission from Ti^{2+}, Ti^{+} and Ti, molecular nitrogen as well as from neutral and ionised atomic nitrogen. In space resolved time integrated spectrum, we can distinguish two zones of emission. The first close the target surface, is dominated by a strong continuum

emission, making spectroscopic measurement difficult due to the high density, high temperature and its high optical opacity. The second zone characterise the recombination phase of the expanding plasma, where the most quantitative observation will occurs.

Figure 1 shows the maximum emission intensity of the lines Ti $^{2+}$ * at 241.4nm, Ti $^{+}$ * at 350,4 nm and Ti* at 374.1nm. All the lines observed exhibits a relative enhancement in intensity and the spatial extension of these intensity is strongly dependent on the gas pressure. This behaviour has been observed before [2] and has been attributed to different process such as electron impact ionisation of metastable species, collisionnal quenching or confinement of the plasma to a limited region at high pressure gas[3,4]. The former two processes lead to the formation of ionised and excited nitrogen species and induce an increases of their emission as shown in figure 1.

Plasma propagation

The plasma propagation is measured by recording the spectral line kinetics of metal vapour ions and nitrogen for various distances. Figure 3 shows the z-t plot of the time of the maximum emission intensity of Ti III at 241.4nm, Ti II at 350.4nm and Ti I at 374.1 nm against the travelled distance. The curves shows also the time of the maximum emission intensity of NII* an NI* at respectively 399.5nm and 745.2nm. We note that Ti III spectral line emission is observed simultaneously with the N II line in the early plasma phase, while emission from Ti II, Ti I and N I appear later when the plasma temperature decrease.

The N_2^+ 391.4nm line rises rapidly from t= 0 and then above 1 mbar has an apparent second peak. The molecular nitrogen was excited and ionised by xuv emitted during the laser target period, however collisions between the target debris and the background plasma can easily excite the NII line.

The temporal shape of the Ti II line at 350.4 nm and NI line at 745.2nm are characterised by a double peak structure above 2 mm from the target surface at 0.1 mbar. Their first peaks follows exactly the space-time dependence of Ti III line at 241.4 nm and NII line at 399.5 nm respectively. This effect has been observed earlier and is attributed to fast recombination occurring in the plasma expansion[5].

Figure 2 shows also that the drag force model fits very well the time of flight vapour propagation for 0.1 mbar. The initial velocity is 4.5 10^6 cm.s^{-1}, the stopping distance deduced from the fit is 1.3 cm and from this we can deduce the cross section for Ti scattering by nitrogen gas as $\sigma = 7\ 10^{-16}$ cm2.

Figure 3 shows the electron number density determined using Stark broadening of isolated lines of titanium at different gas pressures. The electron density seems to be constant

and equal to 6 10^{17} cm^{-3} at 2 mm from the target surface and increases at intermediate distances as the gas pressure is increased. Elastic collisions between vapour species and nitrogen species prevent the expansion of the plasma, making the electron density to increase, ionisation of the ambient nitrogen gas can contribute to the electron density increase.

The electron temperature is determined assuming local thermodynamic equilibrium. The high number electron density found is favourable for local thermodynamic equilibrium to be valid. The temporal evolution of electron temperature give a value of 2.4 ev at 75 ns from the start of laser pulse and 2 mm from the target surface and 1.6 ev at 400 ns and 7 mm. Temperature appearing higher at earlier time for both distances.

Ion probe analysis

The ion probe signal consist on a photo-electric peak arising at the same time the laser reach the target surface and a broad signal corresponding to the ions arrival. In vacuum, the laser generated plume expand freely at peak speed of 5.10^6 $cm.s^{-1}$. As the pressure is increased, the plasma arrival becomes delayed significantly and the pulse width is broadened also. The ion probe waveforms displays a splitting of the plume into a fast component transmitted with little delay and a component with a pronounced slowing. As the pressure is increased, a single pulse shape is acquired.

Figure 4 shows the drop-off of the ion integrated current with distance at different nitrogen pressures. In vacuum, the signal drop at x^{-2}, expecting solid angle behaviour. As the pressure of the foreign gas is increased, the signal drops at x^{-2} exp(-b x), where b is an attenuation coefficient depending on the gas pressure, the ion following a simple scattering model, where the initial beam intensity is reduced according to $I = I_o \exp(-N \sigma x)$, where N and σ are the number of plasma particles and the scattering cross section respectively.

3. Conclusions

The propagation characteristic of laser ablated Ti plasma in nitrogen gas have been investigated by spectroscopic and ion probe diagnostic technique. Spectroscopic diagnostic reveals an intense N_2^+ emission at 391.4 nm rising promptly at the laser start leading to an ionised volume in front of the target, a second peak due to collisions with target debris appearing above 1 mbar. Another important result is the detection of N II and N I ion propagating with the vapour plasma, N II spectral line emission observed simultaneously with Ti III emission. The slowing down of the ejected species is well described using a drag force model. Temperature and density of the expanding plasma are determined assuming local thermodynamic equilibrium.

References

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