OPTICAL SPECTROSCOPY AND COMPARATIVE BEHAVIOR STUDIES IN THE LASER-PLASMA INTERACTION OF N_2-LASER ABLATION FROM Zr AND Ti SURFACES

Ricardo Castell and Francisco Lira

Laser Spectroscopy Laboratory, Physics Department, Universidad Simón Bolívar,
Caracas 1080, Venezuela, e-mail: rcastell@usb.ve

1. Introduction

Plasmas induced by the incidence of UV laser radiation on metallic surfaces show interesting features from the neutral and ionized atomic states transitions. Despite very extensive experimental work has been reported [1,6], many effects of laser beam interaction with the metallic surface and the generated plasma are still not satisfactory. Furthermore, none of them has considered so far the spatial and temporal evolution of the transitions emitted during the interaction. The high degree of ionization reached when duration of the laser pulse exceeds the lifetime of the upper level has been explained [7] by relating the strong increase in the rate of energy that accumulates with a sharp decrease of the breakdown threshold when resonance between the radiation and the atomic system was established.

In this work we have used a N_2-laser capable of producing nanosecond pulses of intense UV radiation to study its interaction with the generated plasma. The experiments reported here were done both in vacuum and air with Zr and Ti targets. Time measurements with a resolution of $\pm$ 2 ns for both neutral (Ti I, Zr I) and ionized (Ti II, Zr II) species were taken as a function of the distance from the surface with a spatial resolution of 20 μm.

2. Experimental setup

The N_2-laser used in our experiments was constructed in the Institut für Experimentalphysik V, Ruhr-Universität Bochum, Germany and described previously [8, 9]. It is capable of producing 15-20 ns FWHM pulses of 337,1 nm radiation with energies between 85 and 100 mJ for a gas mixture of 95% N_2 and 5% SF_6. The laser radiation was focused onto the target by a quartz lens with a focal length of 250 mm producing a sharp (0.4 mm x 0.3 mm) spot. Spectroscopically pure titanium and zirconium samples were used as the target material with its surface perpendicular to the laser beam axis. The plasma produced was imaged 1:1 onto the entrance slit (30 μm) of a 1-meter monochromator (McPherson-2051) by two plano-convex UV lenses mounted on a precise translator stage with a three-mirror system that allowed us to rotate 90^0, and scan the image of the plasma across the slit in a direction perpendicular to the target surface. A spatial resolution, in the direction normal to the surface,

of about ±15 μm at the target was obtained. The selected transition was detected by a photomultiplier (RCA 1P28A) coupled to the monochromator. The current pulses from the photomultiplier were monitored with a digitizing oscilloscope (HP 54502 A) with a maximal sampling rate of 400MHz. The oscilloscope was triggered by a current pulse from a Si photodiode (EG&G FND - 100A) which had a rise time less than 1 ns and was illuminated by one edge of the laser beam. Each laser pulse produced a time-resolved spectrum that was stored in a computer using a interface board (AT-GPIB/NI-488). Intensity variations in the laser line were averaged by taking the average of ten consecutive laser shots. The contour curves finally were obtained by scanning the slit of the monochromator in steps of 0.02 mm starting from the metallic surface.
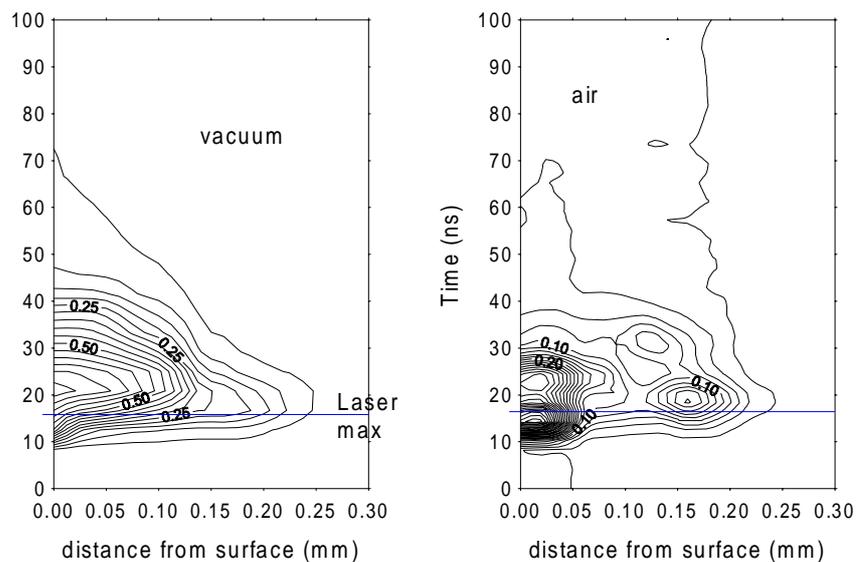

Figure 1. Spatial and temporal behavior of the Ti II (336,61nm) line in vacuum (10^{-5} mbar) and in air.

3. Experimental results

The spectra from titanium and zirconium plasmas were taken separately both in vacuum (p = 2 x 10^{-6} mbar) and in air (normal pressure) between 300 nm and 500 nm. Both spectra displayed strong lines from the ionized and neutral species. In the case of plasmas produced in air a rather strong continuum showed up at shorter wavelengths and Stark broadened lines [10] were observed.

The Ti II transition (a^2P-z^2P^0) at 336,6 nm was chosen here to show the typical behavior of the single ionized species. This line is close to the laser line wavelength and has a lower level that does not decay further by photon emission. The contour curves (Fig. 1) show a comparative pattern in high vacuum and in air. The line intensity follows rather close the laser reaching its maximum value at the same time or even before the laser pulse does. In the case of ablation in air the confined plasma shows a second peak away from the surface as a result of the expanding shock wave front. The same behavior was observed in several Zr II transitions.

The typical evolution of Zr I and Ti I transitions is shown in Fig. 2 for the Zr I (389,03 nm) line. In vacuum, and near the metallic surface, it can be observed that the intensity starts to build up some 30 ns after the laser pulse peak and reaches its maximum value some 60 ns later. As the plasma plume expands away from the surface the maximum intensity appears later in time. Time history measurements can be detected up to 350ns after the laser pulse impacts the surface. Plasma velocities of a few kilometers per second are deduced. For the Ti I transitions the whole process starts about 35 ns earlier than for the Zr I lines.
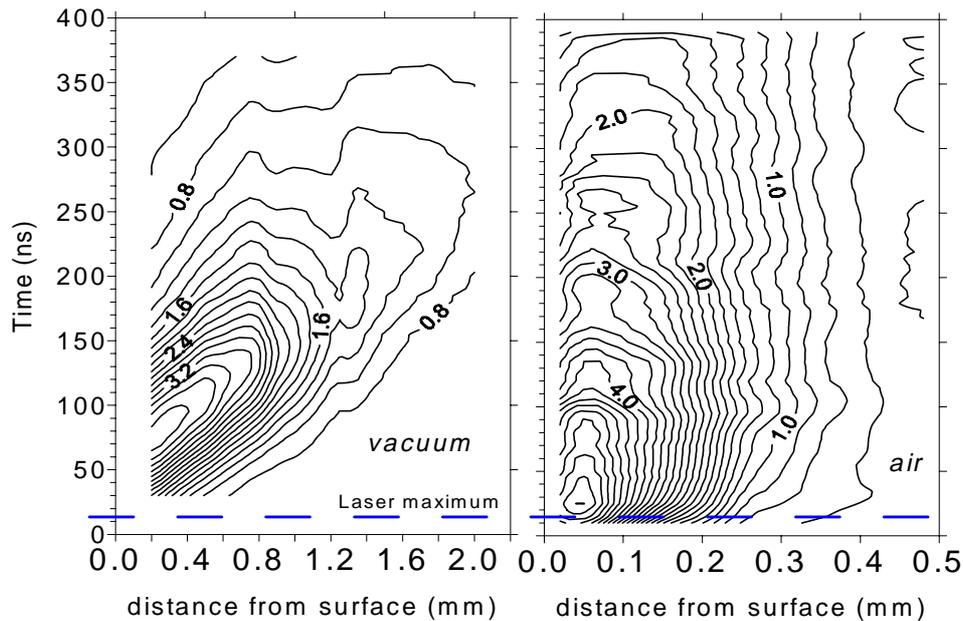

Figure 2. Contour curves for the Zr I (3890,32 A) line emission from the laser generated plasma in high vacuum (2 x 10^{-6} mbar) and in air. Dotted line show the laser pulse peak.

Scattered laser light from the ablated plume was also measured in air and in vacuum at several distances from the surface. Measurements taken with lower energy pulses (20 mJ/pulse) are shown in Fig. 3 for the Ti plasma. Careful alignment of the optical detecting system was done to make sure that only light reflected or emitted by the plasma reached the detecting system.

4. Discussion and conclusions

A model based only upon the interaction between the laser beam and the metallic surface, as the source of the plasma emitting species, could not be fitted to the experimental results. An additional process that populates the upper states of the observed transitions has to be considered and has been already reported [11].
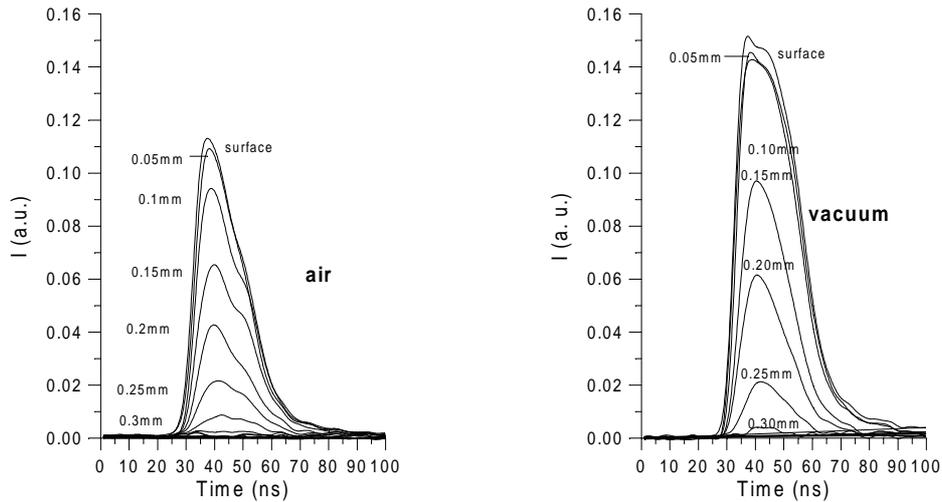
Figure 3. Scattered light at the laser wavelength: 337,1 nm measured at different distances from the surface in a plasma plume generated by laser ablation in titanium. Maxima coincide with the laser pulse peak..

From our results we conclude that the incidence of the laser radiation creates in vacuum a plasma that expands with an average velocity ~8 km/s, and thereafter the duration of the laser pulse radiation allows its interaction with the atoms in the upper energy levels by introducing an additional source of plasma radiation for the ionized (Ti II and Zr II) transitions. On the other hand, the source of the Ti I and Zr I radiation, is mainly due to the recombination process and not to direct excitation from the laser.   The shielding of the laser light by the laser generated plasma in air and in vacuum implies peak electron densities of the order of 10^{21} cm^{-3}.

Pulsed UV radiation energy ablates Ti faster ($\approx$ 30 ns) than Zr from pure solid surfaces in vacuum. In air no time difference was observed.

References

[1]   Künnemeyer, R. and Kock, M.: J. Phys. B: At. Mol. Phys. **16**, L607 (1983)

[2]   Rothenberg, E. and Koren, G.: Appl. Phys. Lett. **44**, 664 (1984)

[3]   Dreyfus, R.M., Kelly, R. and Walkup, R.E.: Appl. Phys. Lett. **49**, 1478 (1986)

[4]   Herman, J., Boulmer-Leborgne, C., and Dubreuil, B.: Appl. Surf. Sci. **46**, 2291 (1990)

[5]   Gingelbach, R.M. and Wentzek, P.L.G.: Appl. Phys. Lett. **58**, 1597 (1991)

[6]   Walkup, R.E., Jasinski, J.M. and Dreyfus, R.M.: Appl. Phys. Lett. 48, 1690 (1986)

[7]   Measures, R.M. and Cardinal, P.G.: Phys. Rev. A23, 804 (1981)

[8]   Cubbedo, R. and Curry, S.M.: IEEE J. of Quant. Elect. QE-9, 499 (1973)

[9]   Rebhan, U., Hildebrandt, J. and Skopp, G.: Appl. Phys. **23**, 341 (1980)

[10] Andreic, Z., Henc-Bartolic, V. and Kunze H.-J.: Physica Scripta **47**,405 (1993)

[11] Castell, R., and Lira, F.: Astrophysics and Space Science (*accepted: may/98*)