Collisional formation of atomic oxygen in pulsed laser ablation

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In the last years, a great deal of work has been devoted to understand the basic physical mechanisms involved in pulsed laser ablation (PLA), one of the most promising techniques for depositing thin films and multilayers of innovative materials. Methods typical of atomic physics can be succesfully employed to investigate phemnomena occurring during expansion of the ablated material (the plume) from target to substrate. Phenomena of great interest in applications of PLA to ceramics (including, e.g., high-T_c superconductors, ferroelectrics, ferrimagnets) are those leading to collisional dissociation of O_2 gas injected into the deposition chamber during the process. As a metter of fact, collisionally-induced production of oxygen-containing species, involving O_2 dissociationas an intermediate step, is known to be essential in achieving the correct stoichiometry in ceramic thin films.

Recently [1, 2], we have developed a diagnostic method based on transient absorption spectroscopy of atomic and molecular oxygen, which enables a detailed investigation of O_2 dissociation during plume expansion in a molecular oxygen environment. In our diagnostics set-up, a probe laser beam, produced by diode lasers tuned on atomic and molecular oxygen transitions, is sent through the plume, and the transmitted intensity is recorded as a function of time after the arrival of the ablation pulse onto the target (Fig. 1). Probe beam delivery into the deposition chamber is accomplished by using a single-mode optical fiber ending with a collimation optics mounted on a micrometer translator, whose position can be adjusted in order to investigate different regions of the plume. The atomic oxygen line investigated is the 3s $^5S_2 \rightarrow$ 3p $^5P_3$, a metastable transition lying around 777 nm, widely investigated in the past [3], whereas for molecular absorption measurements different rotational transitions of the atmoshperic band X $^3\Sigma_g^+$ (ν''=0) $\rightarrow$ b $^1\Sigma_g^+$ (ν'=0), at $\approx$ 760 nm, are employed. Due to the electrical quadrupole character of the involved transitions, detection of molecular absorption requires a special care to improve the sensitivity of the measurements.

Measurements carried out with our diagnostics allow us to derive a relative density mapping for O and O_2 as a function of the process parameters (fluence of ablation laser, partial pressure of the O_2 environment), and allow us to infer different infromation relavnt for both PLA understanding and process optimization. For instance, in the deposition of YBCO superconductor, O absorption gets maximum values when O_2 environment pressure is kept around 0.2–0.3 mbar, i.e., in the range that empirical (trial and test) analysis indicates as the most suitable for obtaining good quality superconductive films. In addition, molecular and atomic

absorption turn out to be strictly time and space correlated each other, demonstrating that O_2 is efficiently dissociated during plume expansion.

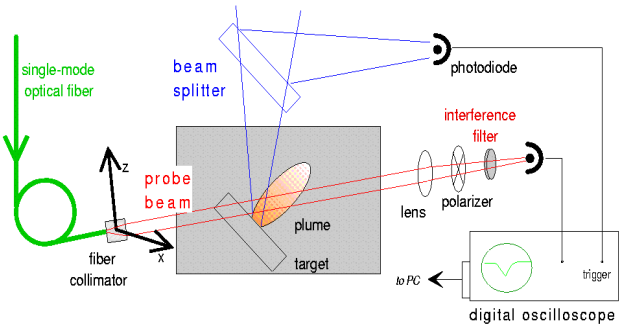

Fig. 1. Scheme of the configuration adopted for absorption spectroscopy measurements during PLA.

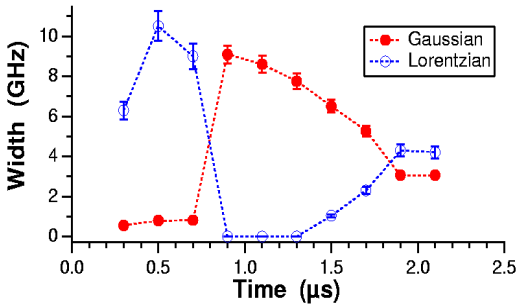

Fig. 2. Results of line profile analysis for atomic absorption during PLA of a metal target at $z = 9$ mm.

As concerns the dissocation mechanisms, two main channels can be envisioned. One of them. expected in the typical conditions of PLA experiments, involves the partial formation of a shock–wave layer, where ablated material and oxygen molecules are compressed and heated up to temperatures $\sim$ 10-20 $\times$ 10^3 K. The other one, unexpected and not reported so far, is probably related to prompt ejection of fast electrons from the target and subsequent collisional dissociation through electron bombardment [4]. The occurrence of these two channels is confirmed by our findings. For instance, the dynamical behavior of the corresponding absorption signals is completely different for the two cases, as well as the dependence on the O_2 pressure and on the ablation laser fluence. Moreover, analysis of the atomic absorption line profile, performed thanks to the tunability ($\pm$ 10 Ghz) of our probe beam source, reveals that absorption line profile is mostly Lorentzian at early times and Gausian at later times, correspoding to collisional processes involving fast electrons and shock-wave driven dissociation, respectively. An example of the results obatined in line profile analysis is shown in Fig. 2, where the Lorentzian and Gaussian widths obtained by a best-fit procedure to a Voigt function are reported for atomic absorption acquired as a function of time at a distance z of 9 mm from target.

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[3] G.M. Tino et al., *Phys. Rev. Lett.* **64** 2999 (1990).
[4] A. Camposeo et al., *Appl. Phys. Lett.*, submitted.