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Technical Note

Emission study of alumina plasma produced by a KrF laser

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ABSTRACT

We report on the plasma emission formed from an α-alumina target when irradiated by laser into vacuum and through oxygen gas. Two diagnostic tools have been used: ICCD camera fast imaging and optical emission spectroscopy. The alumina plasma was induced by a KrF laser beam at a wavelength of 248 nm and pulse duration of 25 ns. The laser fluence was set to 8 J/cm^2 and the oxygen pressure was varied from 0.01 to 5 mbar. By using the ICCD camera, two dimensional images of the plasma expansion were taken at different times. Depending on oxygen pressure and time delay, the expansion behavior of the plasma showed free expansion, plume splitting, shock wave formation, hydrodynamic instability and deceleration of the plume. Using optical emission spectroscopy, the plasma emission revealed the presence of neutral Al I, Al II, Al III into vacuum and under oxygen ambiance. The molecular emission of aluminum oxide (AlO) was detected only in oxygen ambiance. It should be noted that no oxygen lines were observed. Finally, the evolution of the electronic temperature along the normal axis from the target surface, into vacuum, was estimated using the Boltzmann plot method.

1. Introduction

Laser ablation is now a well-established technique for the deposition of a wide range of materials in thin film form [1]. As well alumina (Al_2O_3) is one of the most interesting ceramic materials with great interest owing to their availability, low cost, high melting temperature, high hardness, wear resistance, biocompatibility, chemical stability and other useful characteristics advantageous for structural applications [2–4]. Alumina exhibits chemical and mechanical stabilities even at temperatures up to 1000 °C [5], therefore, it is widely used in various high temperature applications such as catalysis, coatings and microelectronics [6].

In pulsed laser deposition, the properties of thin films depend on the laser fluence, gas pressure, target-to-substrate distance and substrate temperature. This necessitates an optimization study of the plasma before deposition. The structural characteristics of thin films are governed by the kinetic energy of species in the plasma plume.

Several well established diagnostic techniques are used for characterizing laser-induced plasma such as optical emission spectroscopy [7–11], laser induced fluorescence [12], inductively coupled plasma [13] and the Langmuir probe [14].

Several studies investigated the spectral characteristics of aluminum and alumina targets [7,11,15–20]. The effect of ambient air and nitrogen pressures on the expansion dynamics of aluminum plasma has been discussed by Harilal et al. and Sharma et al. respectively [7,11]. Other authors [15] have investigated the spatial evolution of aluminum plasma in atmospheric pressure and showed the difference in the excitation temperature and number density using ultra-violet (UV), visible and infra-red (IR) wavelengths. Others studied the origin of AlO molecules formation from both aluminum and polycrystalline Al_2O_3 targets irradiated by laser [16,17]. Salik et al. have carried out the ablation of alumina target in vacuum generated by Nd:YAG laser at 1064 nm and 532 nm [18]. The variation of plasma parameters (electron temperature and density) as a function of laser energy was studied [18]. In another work, the optical emission spectroscopy of alumina plasma into vacuum induced by the third harmonic of Nd:YAG laser for fluences ranging from8 to 100 J/cm^{2}has been investigated, and splitting of the plasma was observed [19].

In this study, we report an experimental analysis of alumina target irradiated by KrF laser at a fluence of 8 J/cm^2. This fluence value has been chosen for two reasons: first to be far from the onset of phase explosion and second to increase the laser fluence (by increasing the kinetic energy of ablated species) in order to decrease the substrate temperature. In order to obtain crystallized alumina films, Pillonnet et al. [21] and Balakrishnan et al. [22] used high substrate temperature and low laser fluence(3 J/cm^2).

In order to obtain information on the ablated species, their velocities and plasma stopping distance, we propose, as preliminary work, to analyze the alumina plume formed from ablated α-Al_2O_3 target before thin film synthesis. Using fast imaging and optical emission spectroscopy (OES), the plasma generated by a fluence of 8 J/cm^2 into vacuum and under different oxygen pressures was analyzed. The OES analysis

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enables the identification of the species present in the plasma as well as their spatial–temporal evolution.

2. Experimental set-up

The experiment consists of focusing a KrF excimer laser (Lambda Physik Compex 102) at 248 nm, 25 ns pulse width and an incidence angle of 45°, on a rotating polycrystalline ceramic α-alumina target. The focusing optical system is composed of two cylindrical lenses (L1; L2) having a focal length of 100 cm and 50 cm, respectively. The spot area was 0.8 $mm^2$and the laser fluence was set to be 8 J/cm^2. The plasma emission was collected along the perpendicular direction to the ejected matter through a fast intensified charge-coupled device camera (ICCD, Princeton Instruments PI-MAX, 1024 × 256 pixels, pixel size = 24 × 24 μm). A set of spherical (SM), planar (PM) mirrors and an objective was used to image the plasma onto the detector surface. The plasma pictures are captured through a micro channel plate MCP with spectral response of 190 to 850 nm. The time delay *t* between the laser pulse and the observation gate is varied by means of a programmable timing generator (PTG) as shown in Fig. 1 and recorded on the computer using the WinSpec software package.

The spectral measurements of the plasma emission are carried out by imaging the plasma onto a 100 μm entrance slit of a spectrometer (Acton 750, 1800 lines/mm holographic grating) with a spectral resolution of 0.6 Å. The transmitted light is collected by a photomultiplier (Hamamatsu R294, 2 ns) connected to a plotting table that records the spectra of alumina plasma. Then, the emitting species are identified. Again, the photomultiplier is connected to an oscilloscope (Tektronix TDS3032, 5GS/s, 300 MHz) so as to record the TOF signal of the emitting species by tuning the spectrometer to the selected wavelength line. The TOF signal was finally recorded on a computer using the Wavestar software (see Fig. 1). The system has been calibrated in wavelength and intensity using standard calibration procedures.

3. Results and discussion

3.1. Ablated mass

The first experiment performed was the measurements of the mass removal per pulse. This is achieved by measuring the target weight before and after irradiation with 18,000 laser shots while varying the irradiance into vacuum.

The plot of the ablated mass per pulse versus the irradiance is expected to give information on the ablation regime. In PLD experiment one has to avoid the formation of droplets as indicated in a previous work [23].

Fig. 2 reports the ablated mass per pulse as a function of the laser irradiance. The experimental data follow an evolution described by a function of the type I^n (I is the laser irradiance and n a constant). The best fit was obtained for n = 0.48. According to Russo et al. [24] and Liu et al. [25] this range of laser irradiance can be attributed to the case in which phase explosion is not the mechanism of ablation. Furthermore, extrapolating the experimental data gives a threshold laser irradiance of 8×10^7 W/cm^2 (fluence of 2 J/cm^2). Using the equation presented by Peruzzi et al. [26] for low fluence $\Delta h = B\,(I - I_{th})$, where Δh is the ablated layer thickness, B is a constant, I_{th} is the laser threshold irradiance, which was estimated as 7.5×10^7 W/cm^2 (fluence threshold of 1.88 J/cm^2). This value is slightly higher than the fluence threshold given by Orlianges (1 J/cm^2) [27]. In the present work, it was considered that the thickness of the ablated layer per pulse is proportional to the ablated mass per pulse.

In the following study, the irradiance is fixed to 3.5×10^8 W/cm^2 (fluence of 8 J/cm^2).

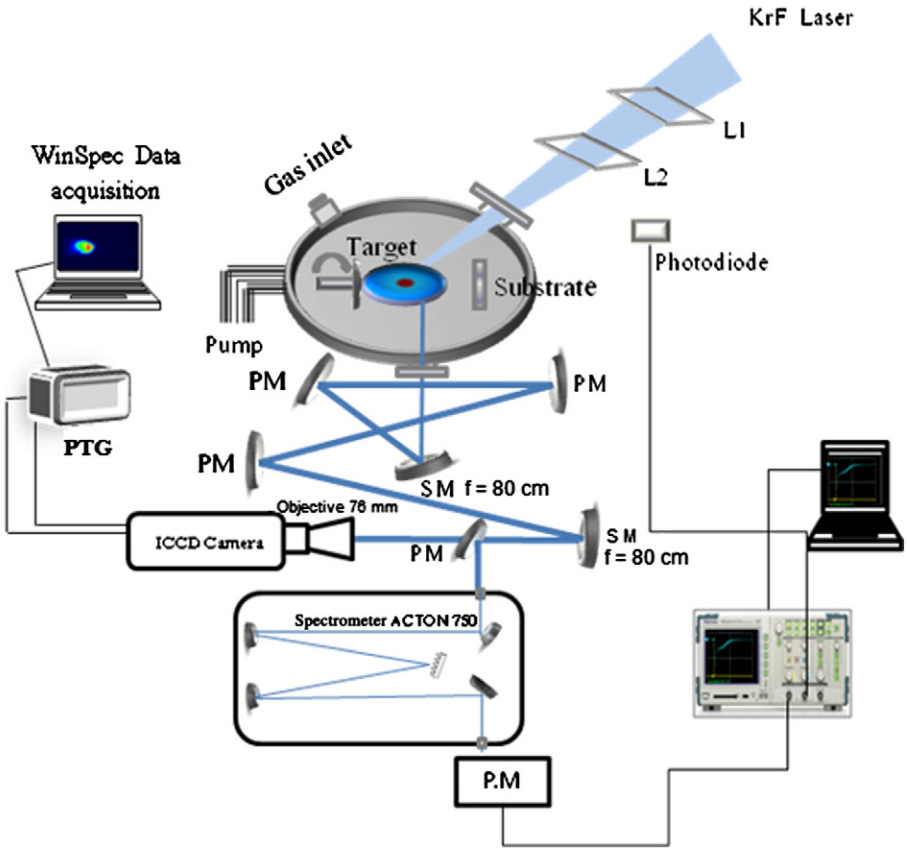

Fig. 1. Schematic of the experimental set-up.