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Dynamics of double-pulse laser produced titanium plasma inferred from thin film morphology and optical emission spectroscopy

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ABSTRACT

In this paper, dynamics of double-pulse laser produced titanium plasma was studied both directly using optical emission spectroscopy (OES) and indirectly from morphological properties of deposited thin films. Both approaches yield consistent results. Ablated material was deposited in a form of thin film on the Si substrate. During deposition, plasma dynamics was monitored using optical emission spectroscopy with spatial and temporal resolutions. The influence of ablation mode (single and double) and delay time τ (delay between first and second pulses in double-pulse mode) on plasma dynamics and consequently on morphology of deposited Ti-films was studied using X-ray reflectivity and atomic force microscopy. Delay time τ was varied from 170 ns to 4 μs. The results show strong dependence of both emission signal and Ti-film properties, such as thickness, density and roughness, on τ. In addition, correlation of average density and thickness of film is observed. These results are discussed in terms of dependency of angular distribution and kinetic energy of plasma plume particles on τ. Advantages of using double-pulse laser deposition for possible application in thin film production are shown.

1. Introduction

Laser produced plasmas (LPPs) have a wide variety of applications since they can be used as a source of photons, electrons, ions, atoms, radicals, molecules, nano-particles, etc. Few of the most common applications are laser induced breakdown spectroscopy (LIBS) which is used for elemental analysis [1–4], extreme ultraviolet (EUV/XUV) source used for development of new generation photolithography [5], ion production for development of table-top accelerators [6] and pulsed laser deposition (PLD) which is used for thin film and nano-structure fabrication [7,8]. Beside numerous advantages of PLD, such as simple and low cost setup, capability of ablation of wide variety of materials, etc., there are also limitations of this technique. These are mainly related to plume content, stoichiometry and propagation. Namely, depending on laser energy density, particulates and debris of nano- and micrometer sizes can be produced and deposited thus making the film dysfunctional. Nano-particle size distribution can be partially controlled by the number of pulses [9] and choice of background gasses at appropriate pressures [10]. The other limitation is related to film uniformity which is affected by angular distribution of ablated plume [11]. In ablation of a multi-component target it is often desired that film stoichiometry remains preserved [12]. Above mentioned issues can be partially or fully overcome by introducing the second laser pulse for ablation, with an appropriate delay time τ after the first one: in a form of double-pulse laser ablation. Generally, plasma produced with double-pulse ablation is angularly more dispersed, more energetic and more atomized which contributes to significant reduction or even absence of deposited particulates [13–15]. Also, with the aim of optimizing deposition process, several possible laser beam configurations have been developed [16–18].

In double-pulse laser ablation, timing of laser pulses is an important parameter [19] since two main competing interactions occur regarding delay time [13]: the interaction of the second laser pulse with primary ablated plasma plume and the interaction on the second laser pulse with target surface heated by the first pulse. The first of these interactions includes absorption and scattering of the second laser pulse inside the primary plasma. This consequently leads to shielding of the target surface and lower ablation rate. On the other hand, overall emission signal is nevertheless enhanced due to enhanced excitation processes (regarding laser-plasma interaction). The second interaction includes higher ablation rate due to both higher absorption and lower ablation threshold. It is well known that surface absorption is proportional to surface temperature [20], while ablation threshold is lowered because the target surface was previously heated by the first laser pulse. Which of these two interactions is dominant depends on delay time τ. The first one dominates for short τ while primary plasma is still dense enough to absorb and scatter the second laser pulse. In fact, it is mostly

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pronounced within the course of the first laser pulse ($\tau \approx$ few ns) and lasts roughly up to few hundreds of ns. In this work such short delay times are not considered. The second interaction dominates for larger τ when plasma plume spreads enough to become transparent to the second pulse so that this pulse can reach target surface. At the same time the surface itself is still at elevated temperature and its ablation threshold is significantly lowered which then leads to enhanced ablation due to decreased ablation threshold. This range of τ (few hundreds of ns $< \tau <$ μs) was used in the present work and further optimization was performed. For sufficiently long delay times, as plasma evolves and targets surface cools down, double-pulse ablation becomes similar to single-pulse ablation in terms of efficiency, ablation rate, emission or absorption enhancement, etc. [13].

This work is an extension of our recent experiments where optical properties of the plume as well as drilling processes on the target were studied with respect to the ablation mode (single- and double-pulse in collinear configuration) and delay time τ [13,21–23]. Here we present the study of influence of those parameters (ablation mode and delay time) on plasma dynamics by means of optical emission spectroscopy (OES) and how this dynamics can be inferred from morphology of deposited thin films, measured complementary by X-ray reflectivity (XRR) and atomic force microscopy (AFM).

2. Experimental setup

Experimental setup for studying double-pulse laser produced plasma including optical emission spectroscopy (OES) and pulsed laser deposition of thin films is shown in Fig. 1. It consists of two Q-switched Nd:YAG pulsed lasers for ablation and a vacuum chamber evacuated by roots pump down to a base pressure of 5×10^{-3} mbar. Both lasers operate at 1064 nm with a repetition rate of 5 Hz. Laser beams were focused onto the target surface at the same spot (lens focal length 33.5 cm, radius 4 cm). Laser which was first fired is hereafter referred as laser-1 (Quantel Brilliant, pulse width 5 ns, output energy 300 mJ), and the second one as laser-2 (Quantel Brio, pulse width 4 ns, output energy 100 mJ). After passing through optical guiding elements, laser pulses decrease in energy, namely they fall to the value of 68 mJ and 23 mJ, respectively. Their corresponding spot sizes and fluences at the target surface were as follows: 26.2×10^{-4} cm^2 yielding fluence of 26 J/cm^2 and 15.3×10^{-4} cm^2 yielding fluence of 15 J/cm^2. Target was a 3 mm thick and 10 mm in diameter titanium disc, aligned 45° with respect to the impinging laser pulses, rotated to avoid crater formation and kept on floating potential. The substrate was placed parallel to the target surface at a distance of 65 mm. Each sample of Ti-film was deposited by 2000 pulses in double-pulse mode (1000 pulses per laser) with preselected delay time, or by 1000 pulses in case of single-pulse ablation. The substrate was prime grade Si (100) wafer (p-type B doped, 500 μm thick). Both the substrate and the target were at room temperature. Details of lasers operating process and angular distribution of ablated plumes, described with $\cos^{11}(\theta)$ and cubic Bézier functions sketched in Fig. 1, can be found in [21]. The first function is more forward directed and represents single-pulse laser ablation plume density distribution; the second one is more dispersed (flat top) representing the plume density distribution produced by double-pulse ablation. Each density distribution defines average ejection angle θ of ablated material (ablation angle); this angle was found to be 22° for single-pulse laser ablation, in contrast to 68° for double-pulse laser ablation [21].

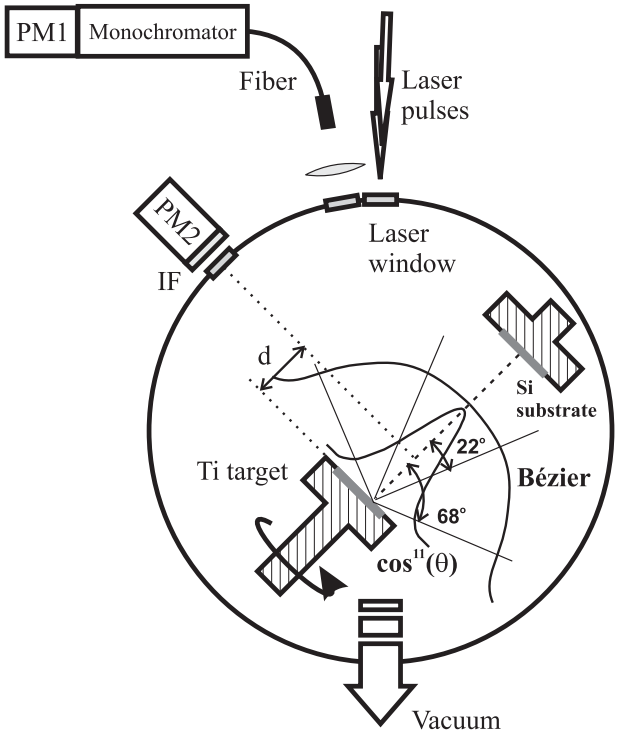

Fig. 1. Experimental setup.

From Fig. 1 it can be seen that setup for optical emission measurements consists of two parts. The first part of emission measurements is related to spectrally and temporally resolved optical emission which was measured using a photomultiplier PM1 (Hamamatsu R2949) attached to a monochromator (MDR23 LOMO) with a slit of 100 μm and a spectral resolution of 0.1 nm. As shown in Fig. 1, light from plasma was collected using borosilicate lens with a focal length of 10 cm and coupled to an optical fiber with a diameter of 6 mm inclined by 35° with respect to the target surface. As the lens was positioned 25 cm away from plasma plume, the light was collected from whole plume (without spatial resolution). Signal from photomultiplier was sent to a box-car where it was integrated within predetermined time-windows, parameters of which are given below.

The second part of emission measurements was aimed on measuring optical emission with spatial and temporal resolutions. It included detecting plasma light transmitted through interference filters (UVIF350 and VISIF625) and detected by means of PM2 (RCA C31034) at different distances d above the target surface. Each of these two filters has a transmitting range such that it only passes through the light corresponding to ionic (Ti^+) or neutral (Ti and impurities) emission, respectively, thus separating their contributions. Maximal available distance d between target surface and optical axis was 20 mm. Spatial resolution was defined by angular width (1.5°) of a solid cone from which the light was collected. As the photomultiplier was located at a distance of 43 cm from the plasma plume, the diameter of detection cone at position of the plume was 15 mm.

In this work, emission from plasma plume was measured in both single- and double-pulse mode. In Fig. 2 firing procedure of both lasers is sketched. Namely, Fig. 2a shows emission obtained during ablation using only laser-1, while Fig. 2b shows emission obtained during ablation using only laser-2. When ablating with only laser-2, laser-1 was used as a trigger (with a certain time delay) for laser-2 but its pulses were blocked. This is the reason why emission in Fig. 2b starts, in this case, at around 1 μs. By summing up emission signals from Fig. 2a and b (two independent ablations) one obtains a signal, Fig. 2c, which corresponds to the one which would be measured in double-pulse mode if there were no interaction between the second pulse and plasma produced by the first pulse. On the other hand, Fig. 2d shows measured emission signal obtained in double-pulse mode, and by comparing it with Fig. 2c one can see that aforementioned interaction cannot be neglected. Time delay τ between two lasers was set by using delay