Optical Analysis Of Colliding Plasmas For Pulsed Laser Deposition

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Abstract

We report investigation and evolution of interaction zone of two collinearly colliding plumes using fast imaging; Nomarski interferometry and shadowgraphy techniques in laser ablated plasmas used for deposition of thin films. Temporal evolution of shockwave due to lateral shock in the interaction zone of two colliding plumes with phase shift developing in time shows the formation of stagnation layer in the interaction zone. The stagnation layer is simultaneously located and analyzed using optical emission spectroscopy of ZnO. A study on thin films of ZnO deposited using stagnation layer of laser produced colliding plumes is reported. Colliding plumes are reported to give thin films of ZnO having better surface and optical properties. The films are characterized using X-ray diffraction and atomic force microscopy. The optical response of the films is investigated using optical transmission in the UV- visible range of spectrum.

Keywords: *Colliding plasma plumes, Nomarski Interferometry, Shadowgraphy, Stagnation layer, Thin film deposition.*

1. Introduction

"PLD" the acronym for pulsed laser deposition is being used to get the thin films of various materials on different substrates for last couple of years. Thin film have played an important role in revolutionize the current era of modern technology[1]. In PLD the quality of deposited thin films depends on laser parameters, the thermo-physical properties of the target and substrate materials, the coupling between the laser and target materials and their relative configuration. All these affect the quality of thin films by modifying the dynamics and properties of the plasma plume being formed [1-3]. Though, PLD have advantages on other available methods for thin film deposition, but still it has its own disadvantages like particulate formation on the substrate and the uneven deposition on a large area. In recent years, colliding plasma plumes have attracted the attention of the scientific community. The properties of the interaction zone of these colliding plumes depend mainly on the relative distance between the individual plasma (D) and their ion-ion mean free path (L). This interaction zone develops with time and takes the form of stagnation layer with relatively larger temperature, different electron number density gradient and abundance of higher ionic species. Though, these colliding plumes have been studied extensively in various geometrical configurations for different materials [4-10]; understanding of various collisional processes and the plume dynamics of the interaction zone still requires the further investigations.

In the present work, we report the optical characterization of two collinearly colliding plasma plumes of Carbon and ZnO by means of Nomarski interferometry and the optical shadowgraphy to study the electron number density and the shock wave development in various regions of colliding plumes in air. These colliding plumes are further investigated using Fast imaging and optical emission spectroscopy and finally applied to deposit thin films of the ZnO on glass substrate at room temperature.

2. Experimental

Fundamental wavelength of 1064nm from Nd:YAG nano second pulsed laser (Lab 190, Spectra Physics USA) is divided into two parts using a 50/50 beam splitter and focused on the target surface at a distance of 1.3mm from each other to generate the seed plasmas in a vacuum chamber. To study the temporal evolution of the shock wave and the electron number density, we have used the second harmonic of fundamental 1064nm from DCR4 laser system (532nm) as a probe beam in Nomarski interferometer. Schematic of the experimental setup for interferometry and shadowgraphy along with this film deposition is shown in figure 1. The temporal delay between pump and the probe beam is controlled by DG535 delay generator. A camera lens attached to a gated ICCD (Andor) is used for fast imaging of the colliding plume evolution. For optical emission spectroscopy the camera lens is replaced by an optical fiber coupled to a spectrometer i-star 303 from Shamrock Inc. USA (details of the experimental setup for Fast imaging and optical emission spectroscopy are given elsewhere [11]). Thin films of ZnO has been deposited on the glass substrate one at 60mm away from the target surface in the forward direction (sample1) and other at 30mm beneath the stagnation layer at 30mm away from the target surface (sample2) in oxygen ambient at a pressure of 10^{-5}mbar. The deposited samples are characterized using XRD, AFM and analysed for their optical response in UV-visible transmission.

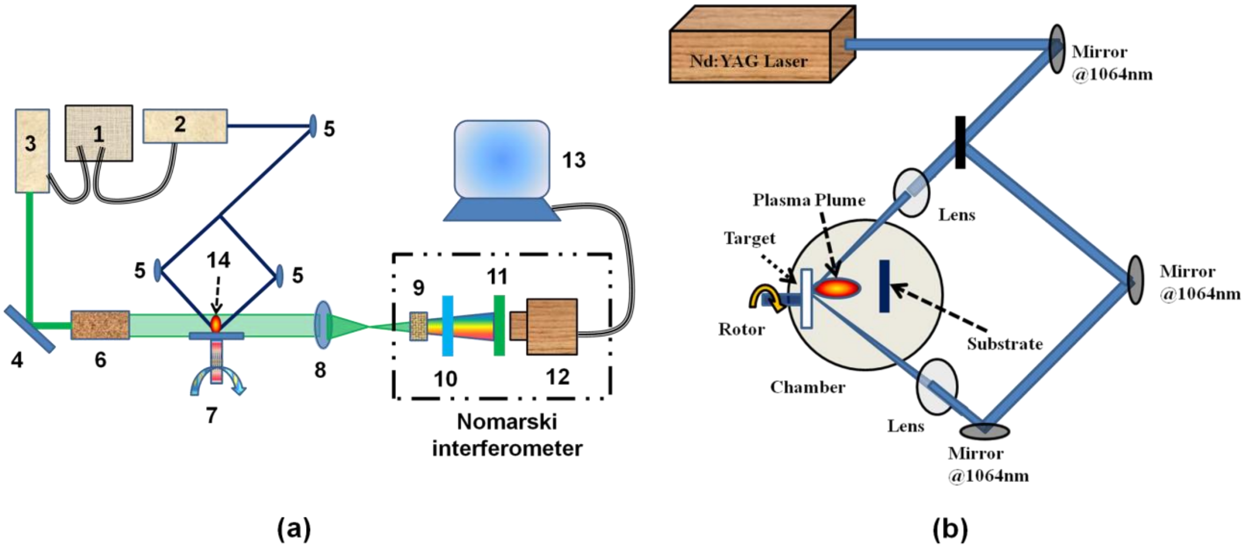

Figure 1. a) Experimental Setup for interferometry and shadowgraphy; 1- DG535 delay generator, 2- Lab 190 Nd:YAG laser@1064nm, 3- DCR4 Nd:YAG laser@532nm, 4-mirror@532nm, 5-mirror@1064nm, 6-beam expander, 7- rotor, 8- focusing lens (f = 10cm), 9- Wollaston prism, 10- Polariser, 11- laser line filter@532nm, 12- CCD, 13- Computer display, 14- Plasma plume. Components 9-12 constitute *Nomarski interferometer*; Blue and red strips indicate beams with mutually perpendicular polarization and the yellow strip indicates the overlapping zone of the two. b) Experimental setup for thin film deposition.

3. Results and Discussion

We have used the two collinearly colliding plumes for present study. When laser beam is focused on to the target, it generates the primary plasma plumes known as seed plasma plumes. These seed plasmas evolve with time and interact with each other along the lateral direction. The expansion velocity of in this direction is slower as compared to the forward direction. This implies that the ion-ion mean free path (L) will be smaller and hence there is an increased possibility of having stagnation in the interaction zone of the colliding plumes.

The shadowgram mimics the shock wave produced by the plasma species due to their interaction with the ambient particles and gives the information about refractive index gradient [12] in the plasma. On the other hand, observed fringes in the recorded interferogram shows shift owing to the density of the plasma [12]. As the refractive index of the plasma is proportional to the density of the plasma, both these techniques complement each other to give information of the density and its gradient inside the plasma plume. In shadowgraphy, the refractive index gradient in the plasma plume causes the deflection of the probe beam. The shockwave induces the density gradient in the plasma plume which in turn results in refractive index gradient and is detected the CCD as the shadowgram [13]. Usually, the plasma acts as the rarer medium then the atmospheric ambient and different portions of probe beam gets refracted from different parts of the shock front which then interfere with each other giving rise to a refraction fringe pattern. This pattern follows the symmetry of the medium and diffraction maximum will give information about the shock wave front. In interferometry, a phase shift is induced due to the refractive index of the plasma plume which is manifested as the shift in the interference fringes in recorded interferograms. The direction of the shift determine the abundance of the contributing species in the plasma as forward shift i.e. the shift away from the target corresponds to the electron density and that towards the target corresponds to density of the neutrals/bound electrons; and the amount of shift gives the measure of the dimension of the medium[14]. At initial times, when the density of the plasma is above the critical density $\left(\frac{1.114\times10^{27}}{\lambda_{nm}^{2}} = 3.94\times10^{21}\,cm^{-3}\right)$ [15] for the probe beam $\left(\lambda_{nm} = 532\right)$, we see a dark region near to the target surface and at later times we only see a change in the contrast. The seed plasmas evolve with time and start interpenetrating to each other. This interpenetration increases and takes the form of the stagnation layer with increased density as evidenced in the shadowgrams and the interferogram for Carbon and ZnO in figure 2 and 3 respectively. Formation of the stagnation layer is clearly indicated by increased shift in interferogram and shockwave signature in corresponding shadowgram for both Carbon and ZnO targets. A larger negative shift observed in interferograms, in interaction zone, indicates the abundance of neutral species in the ZnO colliding plasma as compared to colliding Carbon plasma plumes at later times. These studies established the formation of the stagnation layer in both Carbon and ZnO colliding plasma plumes.

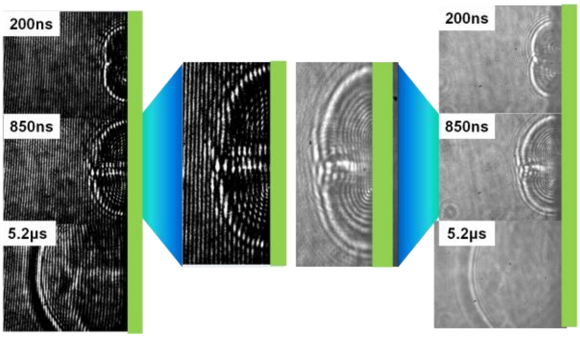

Figure 2. Time resolved interferogram and shadowgrarm for Carbon colliding plasma.

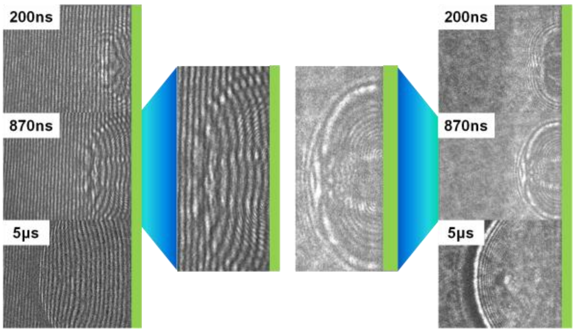

Figure 3. Time resolved interferogram and shadowgrarm for ZnO colliding plasma.

Now to apply these colliding plumes to thin film deposition, we have studied the interaction of the colliding plumes of ZnO in oxygen ambient at 10^{-5}mbar pressure using fast imaging and the optical emission spectroscopy. It is observed that the colliding plumes start interaction with each other at ~20ns and finally takes the form of the stagnation layer ~150ns [11]. These colliding plumes are also reported to be rich source of ZnO nanoparticles [16].

After establishing the formation of the stagnation layer at lower pressure, we have deposited thin films on glass substrates at two different positions with respect to the stagnation layer one in front of the stagnation layer (sample1) and other at below of the stagnation layer (sample2). These samples have been characterized by XRD and the AFM for their structural and morphological characterization respectively. Figure 4 shows the room temperature XRD of deposited thin films. The obtained XRD pattern is then indexed using Pearson Crystallographic Database. SampleI is having only one diffraction peak corresponding to (110) direction of ZnO and sample2 indicates the formation of polycrystalline ZnO. The observation of the (110) oriented film is attributed to the modifications imparted due to the highly mono energetic plasma species reaching the target surface.

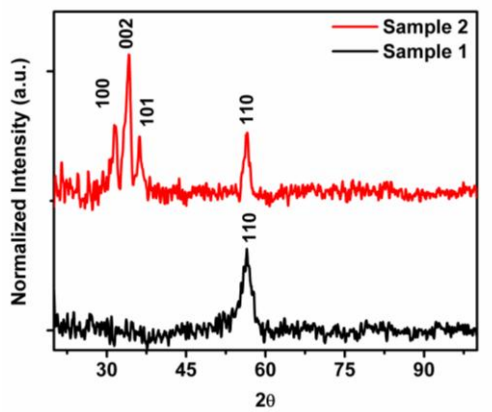

Figure 4. Room temperature XRD of in front (sample1) of and beneath (sample2) the interaction zone of colliding plumes.

The AFM results (figure 5) show the sample1 to have better surface morphology and smaller particle size than sample2. These films are further analyzed for their optical response using the UV-Visible transmission spectroscopy. Figure 6 showing the transmission of the samples1 and 2 infer that the sample1 is ~60% transparent throughout the visible range whereas the sample2 shows an average transparency of ~70% above 400nm with clear band edge signature ~380nm. These observations confirm the better quality of the thin films using 1064nm as ablation wavelength in pulsed laser deposition.

4. Conclusions

We have successfully analysed the colliding plumes of Carbon and ZnO plasma and established the formation of stagnation layer through shadowgraphy and interferometry. The shockwave signatures and the shift in the interference fringes observed in between the seed plasma plumes are attributed to increased density due to formation of the stagnation layer. We have also observed the larger abundance of the neutrals in colliding ZnO plumes as compared to colliding Carbon plumes at later time delays. The thin films having better structural, morphological and optical properties have been deposited using colliding plumes.

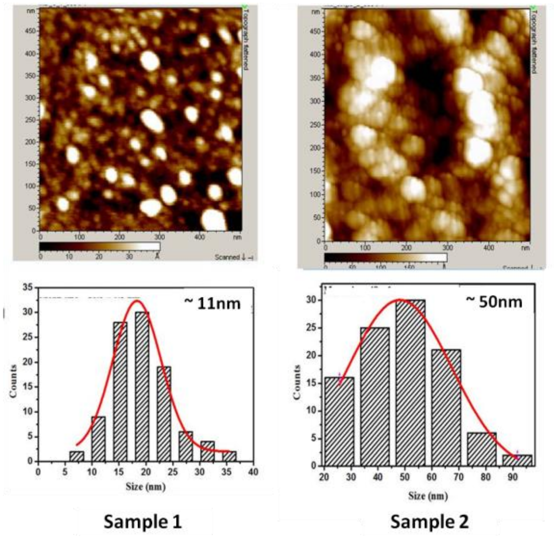

Figure 5. AFM images and corresponding particle size histograms for sample1 and sample2.

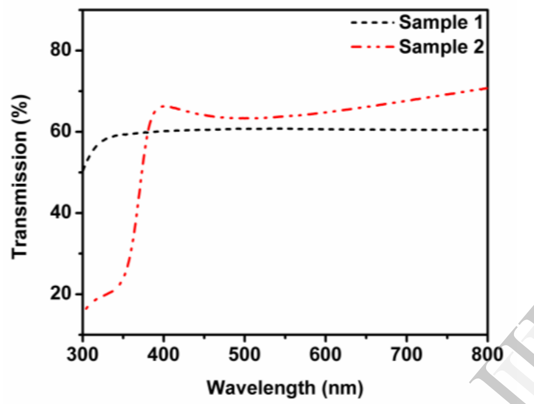

Figure 6. Optical transmission between 300nm to 800nm for deposited thin films.

5. References

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