On the plume splitting of pulsed laser ablated Fe and Al plasmas

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A time resolved imaging study of pulsed laser ablated Fe and Al plasma plumes with specific interest in the splitting of plumes into the slow and fast moving components as they expand through the background argon gas at different pressures is reported. The material ablation was achieved using a Q-switched Nd:YAG (yttrium aluminum garnet) laser operating at 532 nm with a pulse duration of ~8 ns full width at half maximum and a fluence of 30 Jcm−2 at the target surface. Typical time resolved images with low magnification show that the splitting occurs at moderate background gas pressures (0.5 and 1.0 mbar for Fe, and 0.2 mbar for Al plasma plumes). The plume splitting did not occur for higher background gas pressures. © 2010 American Institute of Physics. [doi:10.1063/1.3491410]

I. INTRODUCTION

Pulsed laser deposition is a well known thin film deposition technique for a wide variety of materials, especially for the research purposes. It is a simple, versatile, and fast method for depositing thin films. Properties of the deposited film depend on various parameters of the plasma plume such as ion velocity, ion charge, plasma temperature, etc., and also on the background gas pressure. It is important to understand the behavior of the expanding plasma plume so as to optimize thin film growth by controlling various parameters of the laser pulse, background gas pressure, and target-substrate distance. The reported work on thin film deposition shows that the kinetic energy of the ablated species plays a major role in the deposition process. Moreover, it is well established that the kinetic energies of the species in the expanding plasma plume are primarily controlled by the background gas pressure.

Provided the laser energy is in excess of the ablation threshold, the formation of plasma species starts within a few picoseconds of the laser pulse striking the target surface. The removal of material from the target depends on the optical and thermal properties of the material at the selected wavelength, the laser intensity, and the laser pulse duration. The plasma created at the target’s surface expands with high velocity (~104–105 m/s) (Ref. 7) and interacts with the remaining laser pulse (for ns pulse lasers). The laser-plasma interactions further increase the temperature and density of the nascent plasma. For a relatively high ambient gas density, the particle diffusion between the ejected matter (the plume), and the gas is negligible, and a sharp interface can be assumed at least in the early stage of the plume expansion. During that stage, a shock wave is generated in the ambient gas as the plume expands with a velocity larger than the sound speed in the gas at rest. The ambient gas plays a critical role is several applications of laser-produced plasmas, such as thin film deposition, nanoparticle formation, and laser-induced plasma spectroscopy.

After the termination of the laser pulse, the adiabatic expansion begins where the thermal energy gained by the plume from the laser pulse is rapidly converted into the kinetic energy of the plasma plume species. From the perspective of the background gas environment, the expanding plasma species interact with the atoms/molecules of the previously undisturbed background gas. It is well established that the interaction between the ablated species and the background gas strongly influences the properties of deposited thin films in pulsed laser deposition systems.

The plasma plume splitting has been reported by many researchers. showed that for relatively low background gas densities the ablated plasma plume splits during its propagation into fast and slow moving parts. According to Geohegan and Puretzky, this plume splitting is due to the fact that there are high and low energy components in the plasma plume as a weak shock front is formed during the ablation into low background gas pressure. The plume splitting is of great interest because the fast component may damage the growing film or otherwise affect its microstructure. found that in the case of longer duration laser pulses, the adiabatic expansion is disturbed by the continuous electron heating via an inverse bremsstrahlung process. When expanding into a vacuum, the electrons at the plume front escape with high velocity due to the noncollisional flow regime. However, as the remaining plasma has a sustained unbalanced positive charge, the resulting electric potential will accelerate some ions toward the swarm of escaping electrons. Eventually, these accelerating ions can even attain velocities close to those of electrons. It is proposed that this mechanism is responsible for the plume

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splitting, i.e., separation of charged particle into the fast and slow populations.\textsuperscript{17,18} Another process of particle acceleration (heating) is related to the transformation of energy mismatch after ion-electron recombination into heat\textsuperscript{19} that can be responsible for the acceleration of front propagation even after the decay of the laser pulse. Bulgakov and Bulgakova\textsuperscript{20} reported the observation of a double layer in a laser created graphite plasma during charged-collector probe measurements and they explained the formation of a double peak structure in ion time-of-flight distribution as due to the formation of an ambipolar electric field in the expanding plume. The plume sharpening behavior suggests that the higher kinetic energy particles are emitted closer to the target surface normal. Harilal \textit{et al.}\textsuperscript{21} reported that the ions of highest ionization state dominate in the direction normal to the target, and that their concentration falls sharply in directions away from the normal. Chen \textit{et al.}\textsuperscript{22} reported that the partial ionization of the vapor due to temperature increase near the shock wave front can also result in an acceleration of the flow. The addition of kinetic energy to the laser-induced flow through absorption of incident laser energy will result in a more moderate deceleration of the shock wave velocity than the predicted blast-wave theory.\textsuperscript{24} Simultaneously, the part of the ablated species that collide with background gas will lose their kinetic energy. Thus, the plume itself might be split as indicated in the experimental observation. When the background gas pressure is sufficiently high, the plume is continuous and no such plume splitting is observed. Wood \textit{et al.}\textsuperscript{25} used a multiple-scattering/hydrodynamical model together with experimental results to explain plume splitting. They also stated that the background gas plays a major role in the process. Another possible cause of plume splitting, particularly for high laser fluences, is attributed to phase explosion.\textsuperscript{26} A laser pulse of comparatively short duration (approximately a few nanoseconds) with high irradiance ($\sim$GW cm$^{-2}$) fluence can raise the target surface temperature to 0.90 $T_c$ ($T_c$ being the thermodynamic critical temperature).\textsuperscript{27,28} A homogeneous bubble nucleation occurs, and the target makes a rapid transition from superheated liquid to a mixture of vapor and equilibrium liquid droplets. The process as a whole is termed either phase explosion or explosive boiling.\textsuperscript{29,30} The aim of the present work is to study the effect of background gas pressure and target material on plasma plume splitting. Laser ablation has been performed for Fe and Al targets in an Ar background gas. A fast gated intensified charge-coupled device (ICCD) camera based imaging system was used to capture the time resolved images of the expanding plasma plume.

\section{II. EXPERIMENTAL SETUP}

Details of the experimental setup have been described in a previous paper.\textsuperscript{3} The Fe and Al targets were ablated using a Q-switched Nd:YAG (yttrium aluminum garnet) laser (LOTIS II) at a wavelength of 532 nm with a pulse duration of $\sim 8$ ns full width at half maximum (FWHM). The laser pulse with energy of 47.5 mJ was focused onto the target to a spot diameter of 225 $\mu$m. The target material was placed on a rotating target holder to achieve uniform ablation for every shot. A schematic of the experimental setup is shown in Fig. 1.

Images of the plasma plumes were captured using a fast gated ICCD camera (PI-MAX, Princeton Instruments, model: 7397-0013). A programmable timing generator (PTG, ST-133, Princeton Instruments) was used to provide time delays with set intervals between the laser pulse and camera shutter. For each delay, the image acquisition time was set to 10% of delay duration. The raw images were processed using the IMAGE$^{\text{TM}}$ software.

\section{III. RESULTS AND DISCUSSION}

Time resolved fast gated images of Fe and Al plasma plumes expanding through the atmosphere of Ar gas for the pressure range of 0.2–3 mbar are shown in Figs. 2 and 3. It is observed (Figs. 2 and 3) that at relatively low pressure (0.2 mbar), the plume expansion is rapid and its visibility vanishes very quickly, while high background pressures cause the plume confinement and its luminosity lasts for a longer period of time. The plasma plume expansion dynamics is found to be controlled mainly by the background gas pressure.

The time resolved images in Figs. 2 and 3 show that the increase in background gas pressure leads to significant changes in the plume shape and the propagation velocity. The visible shapes and velocities of Fe and Al plasma plumes obtained are different due to different physical and chemical properties of their bulk. For the similar laser energy fluence, the ablated plasma plumes of Fe (at 0.5 and 1.0 mbar) and Al (at 0.2 mbar) have shown peculiar features in their shapes with the plume separating into two luminous parts (refer to Figs. 2 and 3); commonly referred as plume splitting. Figure 2(a) shows that for Fe plasma in the background gas pressure of 0.2 mbar there was no splitting, while
The Al plasma plume clearly shows splitting that starts at ~150 ns and can be observed up at ~350 ns [refer to Fig. 2(a)]. It can be observed in Figs. 2(b) and 3(a) that the splitting of the Fe plasma plume occurs for the background argon gas pressures of 0.5 and 1.0 mbar. The plume splitting of Fe plasma in 0.5 mbar of Ar gas is seen to begin at ~200 ns and ends at 450 ns [refer to Fig. 2(b)], and as the pressure is increased to 1 mbar the separation starts earlier at ~150 ns and ends up at around ~300 ns [refer to Fig. 3(a)]. The shift in Fe plasma plume splitting duration toward an earlier stage with the increase in background pressure may be explained by considering the increased pressure results in slowing down a faster moving component due to a high degree of interaction of the plume species with the background gas that results in diffusion of the slower component into the background gas. It can be noticed that in the case of Al for the background argon gas pressure of 0.5 mbar and above, the rear portion of the plasma plume is not visible to indicate that there is no observable plume splitting for these cases as the plasma plume does not seem to have components with different velocities. The absence of a rear portion of the plasma plume seems to indicate that there is no low velocity component formation for the operating gas pressure of 0.5 mbar and above. The low velocity component for Al is seen to form only for low operating pressure of 0.2 mbar or may be also be present at pressures lower than this for which we did not do the imaging. While for Fe, the low velocity component is very prominently observed for the operating gas pressures of 0.5 to 1.0 mbar, whereas it is observed for a short duration at early time instants for higher operating gas pressure of 3 mbar.

Hence, for Fe, we clearly notice that at a low background gas pressure, there is no splitting and with the increase in argon background pressure to some moderate values of 0.5 and 1.0 mbar a very clear and distinctive plume splitting is observed and it starts to disappear with further increase in pressure. The trend for the dependence of plume splitting with background argon pressure is probably the same for Al except for the fact that the pressure regime shifts to lower pressure region, i.e., for Al the distinctive plume splitting probably occurs at lower argon background gas pressure range of about 0.1 to 0.2 mbar while for Fe it occur at slightly higher background gas pressure of 0.5 to 1.0 mbar.

Based on the above observations, we recognize three regimes that occur for the propagation of plume in the background gas with the increasing pressure. For lower pressures, the plume expansion is vacuumlike, then with the increase in pressure there is a transition regime where the plume splitting into fast and slow component can be observed. This transition regime is not unique and it does depend on the...
target material and we found it to be different for Fe and Al targets used in the present experiments. According to Harilal et al., the pressure range where the plume splitting is observed falls within the transition from collisionless to collisional interaction of the plume species with the background gas that in their view is supported by enhanced radiation emission. With the further increase of background gas pressure, the plume splitting disappears and the plume is significantly slowed down [compare plume front position in Figs. 3(a) and 3(b)] and according to Harilal et al. at higher pressures, the plume eventually stops as a result of shock wave formation in the diffusionlike regime. We have also noticed that at a higher background pressure, the shock wave models fits/explains the plume dynamics. It may be highlighted here that the plume splitting into fast and slow components is background-induced, although it may not be easily detectable under vacuum condition if plasma luminosity based imaging technique is used. Bulgakova et al. observed plume splitting in vacuumlike conditions using a time-of-flight technique.

The time resolved images, some of which are shown in Figs. 2(a) and 2(b), of Fe and Al plasma plume at 0.2 and 0.5 mbar, respectively, are used to estimate the plume front positions for the fast and slow moving components and are shown in Fig. 4. It may be noted from images shown in Figs. 3(a) and 3(b) that after certain instants it is not possible to identify the slow moving component as it diffuses significantly.

Figure 5 shows that the plots of the velocities of the faster and slower components of the visible plasma plume, which are measured from the data shown in Fig. 4. It may be seen that, in the observable window, the velocity of the faster component of the plume is almost twice that of its slow component.

To understand the phenomenon of plume splitting in a certain background pressure regime and to explain the shift of this pressure regime to lower pressure side for Al plume as compared to that of Fe plasma plume, we look into some of the fundamental processes involved in pulsed laser ablation. For the laser fluence used by us in present the experiment, a small amount of material is ablated from the metal target surface at the earlier part of the laser pulse. This ablated material is heated by the remaining part of the incoming laser pulse to thermally ionize the material and making it opaque to the incident radiation beyond the critical density region where the plasma frequency is higher than the incoming electromagnetic radiation; \( \omega_p > \omega_c \). The initially ablated material in front of the target surface absorbs most of the energy of the laser pulse and the target surface is effectively cut off from the incoming radiation for a large fraction of the incoming laser pulse. The properties of this initially ablated plasma must be dependent on the target material, energy and profile of the laser pulse, etc. The initially ablated plasma then expands rapidly away from the target surface. The observed plume splitting can be explained in two different ways.

The initially ablated plasma, which also absorbs more energy from the remaining part of the laser pulse, becomes hotter and hence not only starts to expand rapidly but also
begins to emit intense radiation. Due to the direct collisional interaction between this initially ablated plasma and the surface of the target, and also because of the intense radiation, a second vaporization of the target surface takes place. The initially ablated plasma produced directly by the incident laser, expanding out in the background gas or vacuum away from the target surface forms the fast component while the vaporization at a later stage by energetic plasma and radiation results in the slower component of the ablated plasma plume.

The generation of the fast and slow components from the target surface can also be explained using the concept of double layer (DL) formation in the initially ablated plasma which starts to expand. The DL is formed due to the generation of hot electrons that obtain additional energy in the initially expanding ablated plasma plume. The mechanisms responsible for generation of hot electrons are three body recombination and absorption of remaining part of the laser pulse due to inverse bremsstrahlung. These hot electrons move with higher velocities as compared to that of heavier ions, creating a spatial charge separation (breaking the quasineutrality due to this DL formation) in expanding plasma plume resulting in electric potential and a self-consistent ambipolar electric field. The ions that enter the region of potential drop (the DL region) will accelerate, resulting in formation of the fast component of expanding plasma plume whereas the rest of the nonaccelerating core plasma forms the slow component.

The generation of fast and slow moving components in the expanding plasma leads to the observable plasma plume splitting in particular regime of the background gas pressure. It may be pointed out that the formation of fast and slow moving components should not be a function of the background gas pressure (although it will require an appropriate laser pulse fluence as observed by Harilal et al. and should occur over the entire range of background pressure. However, it can be anticipated that the increasing background gas pressure will have a different effect on the two components. The fast moving shell of the plume will come in contact with the background gas while the slow moving core does not feel the effect of background gas directly. The so called plume splitting, which we and several other researchers have observed using imaging techniques, refers to the observation of two distinct luminous fronts in certain background pressure regime (i) is mainly due to the formation of fast and slow components and (ii) is limited to certain pressure regime (mostly in moderate range) because in the moderate background gas pressure region the strong fluorescence is observed due to particle collision both in the plume body (the slow plume core) and in the plume expansion front (the fast plume shell) with the plume front edge defined quite sharply due to the presence of a shocked gas front. In other words, while the slow core plasma is radiating initially due to gas collisions between the plume ejecta in the high pressure region of the initial expansion; in the moderate gas pressure range there exist a shocked gas region at the front edge of fast plume shell with the shocked gas temperature rising to several thousands of Kelvin with significantly enhanced optical emission from excited species in the shocked gas plasma; leading to the formation of two distinct luminous fronts, i.e., the so called plume splitting.

At lower background gas pressures, the luminous slow plasma core is there but the luminous fast plasma shell is absent as the shocked gas region is not formed due to insufficient background gas [refer to the images for Fe plasma in Fig. 2(a)]. At higher operating gas pressures, the luminous region of fast plasma shell is always observable as the shocked gas region is formed whereas the luminous slow core does not seem to form [refer to the first two images of Al plasma plume in Fig. 3(b)]; but the reality is that at higher background gas pressure the slow plasma core recedes and disappears much faster. So it appears as if the plume splitting is not taking place at the higher background pressures, as seems to be the case in the images of Al plasma plume in Figs. 3(a) and 3(b) and also for Fe plasma in Fig. 3(b). The expectation is that the luminous slow core should be observable if the magnified images are captured closer to the target surface at earlier time instants and if the fast plasma core is also able to form at earlier instants, which it should, then we should be able to observe the so called plume splitting (two distinct luminous fronts) at earlier time instants in the magnified images closer to the target surface.

Let us now try to understand the reasons for the shift in the pressure regime in which plume splitting is observed to the lower pressure side for Al plume as compared to that of Fe plasma plume. The shift can be attributed to differences in (i) atomic weights and (ii) vaporization energies of Al and Fe. It may be important to point over here that as the laser pulse is shot on different target materials, then different target materials will start to vaporize at different time instants as they have different heats of vaporization. For example, the vaporization energies of Al and Fe are 293.4 and 349.6 kJ mol$^{-1}$, respectively. The Al, with the lower vaporization energy, starts to evaporate at earlier time instants of laser pulse as compared to that of Fe; the heavier Fe species that desorbs later from the target surface is more energetic as it can vaporize only at higher target surface temperature (or with higher energy imparted from incident laser pulse). In other words, the particles of Fe plasma are not only heavier but also more energetic as compared to that of Al plasma. Hence, the background gas at lower pressure will be able to cause the thermalization of lighter Al species at fast plume front resulting in the observation of distinct luminous fronts (plume splitting) at the lower background gas pressure.

IV. CONCLUSIONS

The pressure dependent plasma plume splitting refers to the observation of two distinct luminous regions in pulsed laser ablation of Fe and Al targets using time resolved imaging has been investigated. The separation of the plume into the faster and slower components for the ablation of Fe was observed for 0.5 and 1 mbar of Ar background gas pressures, while for the Al plume the splitting was found to be observed at 0.2 mbar. It is observed that the splitting of the plume occurs in a relatively earlier time range ($\sim$150 to $\sim$400 ns). The maximum duration between the leading edge of the faster and the slower components of Fe plasma, for a back-
ground gas pressure of 0.5 mbar, was found to be roughly 300 ns.

The plasma plume’s splitting is found to be strongly influenced by the background gas pressure and the target material. We recognized three regimes of background gas pressure. (i) For lower pressures, the plume expansion is vacuumlike without any formation of visible frontal edge and hence no observable plume splitting, and (ii) then with the increase in pressure there is a transition regime where the plume splitting into fast and slow component was observed, however the background gas pressure window for this transition regime found to depend on the target material. The shift in the pressure regime in which plume splitting is observed to the lower pressure side for Al plume as compared to that of Fe plasma plume was attributed to differences in atomic weights and vaporization energies of Al and Fe.

The plume splitting, in the appropriate background gas pressure range, is explicable in terms of the strong fluorescence from (i) the gas collisions between the plume ejecta in the high pressure region of the initial expansion in the slow core plasma and (ii) from excited species in shocked gas plasma formed at the front edge of the fast moving plasma shell. The slow plasma core and the fast shell plasma are formed due to either the double vaporization of target material from a relatively long duration laser pulse or to the formation of a double layer at the initially expanding plasma from the hot electrons being energized by inverse bremsstrahlung and three body recombination.

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