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Effects of laser energy fluence on the onset and growth of the Rayleigh–Taylor instabilities and its influence on the topography of the Fe thin film grown in pulsed laser deposition facility

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The effect of laser energy fluence on the onset and growth of Rayleigh–Taylor (RT) instabilities in laser induced Fe plasma is investigated using time-resolved fast gated imaging. The snow plow and shock wave models are fitted to the experimental results and used to estimate the ablation parameters and the density of gas atoms that interact with the ablated species. It is observed that RT instability develops during the interface deceleration stage and grows for a considerable time for higher laser energy fluence. The effects of RT instabilities formation on the surface topography of the Fe thin films grown in pulsed laser deposition system are investigated (i) using different laser energy fluences for the same wavelength of laser radiation and (ii) using different laser wavelengths keeping the energy fluence fixed. It is concluded that the deposition achieved under turbulent condition leads to less smooth deposition surfaces with bigger sized particle agglomerates or network. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4763555]

I. INTRODUCTION

The Rayleigh–Taylor (RT) instability is well known to develop at the interface between superposed fluid layers of different densities, which occurs when the dilute fluid is pushing the denser fluid.1–4 The behaviour of an initially sinusoidal disturbance at the interface was first studied by Rayleigh5 and later by Taylor.6 In that small-amplitude analysis, the sinusoidal perturbations were shown to grow exponentially with time. The Rayleigh–Taylor instability has since been the subject of many investigations, and there are major review articles on this topic by Kull,4 Sharp,5 and Inogamov.8 The phenomenon occurs in a wide variety of situations and length scales. It is a feature of over-turning in the deep ocean, where mixing occurs as a result of the formation of vertical plumes, as described by Lazier et al.7 It is also believed to be the cause of some elaborate structures at galactic length scales, reported in the radio-telescope observations of McClure-Griffiths et al.8

In the present investigation, the plasma plume dynamics of pulsed laser ablation of Fe target is investigated to study the growth and onset conditions of Rayleigh–Taylor instability and its effects on morphology of the deposited thin films. The Fe target is used due to its high carrier concentration, high coupling coefficient (1 − R) where R is the reflectivity, and high absorption coefficient for the incident laser beam which combined with the low overall thermal diffusivity results in low vaporization threshold. If the incident laser energy fluence is above the vaporization threshold, then the laser energy is absorbed in the surface layer and the intense heating leads to the vaporization within the distance of \( \sim (D\tau)^{0.5} \), where \( D \) is thermal diffusivity and \( \tau \) is the duration of the laser pulse. The thickness, \( \Delta x_t \), of the evaporated material per laser pulse is given as

\[
\Delta x_t = (1 - R)(E - E_{th})/\Delta H,
\]

where \( E \) is the incident laser energy, \( E_{th} \) is the threshold energy, and \( \Delta H \) is the volume latent heat of the target material. If \( E_{th} \) is assumed to be independent of \( E \), then this equation shows that there should be a linear increase in the evaporated thickness of the target material, though it has not been observed at high laser energy density.10 However, it is true that more material is ablated at higher laser energy fluence (or energy density) and the laser energy fluence plays a very important role in controlling the thickness as well as the composition11 (for multi component targets) of the thin films grown in pulsed laser deposition (PLD) facility. It is for this reason; we investigated the effects of laser energy fluence on the plasma plume expansion dynamics and RT instability formation using imaging diagnostics to understand their influence on the surface morphologies of the deposited material in a pulsed laser deposition facility. The experimental results of plasma plume dynamics were also used to deduce the ablation parameters using the snow plow12 and shock wave13 models.

II. EXPERIMENTAL SETUP

The details of the experimental setup have been described in Ref. 14. The Fe target disc is ablated using a Q-switched Nd:YAG laser (LOTIS II) at a wavelength of 1064 nm. The laser pulse duration is kept constant at ~8 ns (FWHM) for all the fluences used. The variation in the laser...
fluence was achieved by selecting neutral density (ND) filters of different transmission values, instead of changing the pump energy or laser focal spot size. The laser pulse was focused onto the target surface to a laser spot diameter of 225 µm. The target material disc was mounted on a rotating target holder to achieve a uniform ablation for every shot. Images of the expanding 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 suitable time delays with set intervals between the laser pulse and camera shutter. The image acquisition time, for all delays, was set to 10% of the selected delays.

III. RESULTS AND DISCUSSION

The investigation of the plasma plume dynamics for different experimental parameters is done using time resolved gated images of the laser ablated plasma at various laser energy fluences of 9.4, 23, 51, 60, and 85 J cm⁻².

A. Time resolved imaging results and Rayleigh–Taylor instability

The time resolved images for different laser energy fluences are shown in Figure 1. The aforementioned laser fluences on target were used because of the available combinations of ND filters. The images captured at 50 and 100 ns (Figure 1), particularly the one taken at 50 ns, clearly show two strongly fluorescent regions, the so called plume splitting, for all the five energy fluences used in this experiment. Since the background gas pressure is relatively high, the slow core plasma staying near the target is thin and not distinguishable at later timings. One can quickly notice that at any given time instant, the width of the plume increases with the increase in the laser energy fluence which is in line with the expectation that the more material is ablated with in the increase in the laser energy fluence.

It is also observed that for all five fluences, the plume front shape up to 150 ns is similar, well defined and smooth. As the plume moves farther from the target (refer images captured at instants of about ∼200 ns), its front boundary shows irregularities or kinks. These instabilities on the front edge of the expanding plume, according to other researchers, are attributed to the Rayleigh–Taylor instability. The RT instability arises due to the difference in the density of two immiscible fluids at the interface. This instability manifests in the form of growth of some kind of perturbation at the interface.

It is an established fact that the RT instability occurs (i) when the heavy fluid accelerates through the light fluid or (ii) when a light fluid pushes through a heavy fluid with the acceleration pointing to lighter fluid, i.e., the lighter fluid is decelerating.

In case of pulsed laser ablation, the initial internal pressure and the density of the ablated plasma are very high (kbar and near solid density, respectively) near the target surface just after the ablation. The high density high pressure ablated plasma then expands through the vacuum or the background gas and hence initially the condition is that a heavy fluid (initially ablated plasma) accelerates through the light fluid (vacuum or ambient gas in PLD chamber) and hence the RT instability can set-in on the expanding plasma plume front and several researchers have worked on this problem particularly for inertial confinement fusion by investigating the growth rate for ablative RT instability [and Refs. 1–14, therein]. However, we are not interested in our work in this initial ablative phase rather we are more interested in later stages when the plasma plume expands through the background gas as it may affect the material that is deposited on the substrate surface in a PLD facility. Moreover, it has been shown by several authors [refer to Refs. 1–14 of Ref. 20] that the ablation process itself leads to a reduction of instability growth rate by the so called ablative stabilization that is probably why we do not observe any RT instability at initial time instants (refer Figure 1). As the ablated plasma plume expands, the plasma density of the plume decreases due to expansion and recombination. Now, if the pulsed laser ablation is done in near vacuum (or low background gas pressure) then even though the density of plasma in the plume decreases but its density remains higher than that of background and then it is always the case of heavier fluid (plasma plume) pushing through a lighter fluid.
(vacuum like or low background pressure) fluid but the heavier plasma plume is still decelerated by the lighter fluid and hence the conditions RT instabilities are not able to set in and hence no RT instabilities were observed in imaging diagnostics of expanding plasma plume done at lower background pressure.\textsuperscript{14} However, as the background gas pressure is increased then at certain higher background gas pressure there arise a situation, due to decrease in plasma density in expanding plume and higher background gas pressure, where a light fluid (expanding plasma plume with sufficiently decreased density) is pushing through the heavy fluid (due to high background gas pressure used) and that too with substantial deceleration (i.e., with acceleration pointing from heavy background gas to lighter plasma plume) due to increased background pressure, resulting in conditions required for the onset of RT instability at the interface of the two fluids, i.e., at the plasma plume front edge; as observed in images of Figure 1. The images in Figure 1 seems to suggest that RT instability starts somewhere about 350 ns. The enlarged images of plasma plumes at three different laser energy fluences are shown in Figure 2 from which it is quite clear that the distance of the plume front from the target at which the instability starts to occur increases with the increase in laser energy fluence (or laser beam energy). This trend is similar to the results obtained by Sharma and Thareja.\textsuperscript{16} The increase in the distance of the onset of the RT instabilities with the laser energy fluence can be understood from the fact that with increasing laser energy fluence more material is ejected and that too with greater energy (refer to the discussion coming later) and then it would require greater distance for the density of the ablated material to decrease significantly with sufficient deceleration to achieve the conditions for the onset of RT instability.

For the images captured at later instants, i.e., from \(-500\) to \(4950\) ns (Figure 1), the plume images show two sets of distinct features: (i) for \(9.4\) J cm\(^{-2}\) laser fluence the plume shape is elongated and comparatively smooth and (ii) for the fluence range from \(23\)–\(85\) J cm\(^{-2}\) the plume shapes are elongated but with very irregular/ unstable boundaries due to Rayleigh–Taylor instability, and also the irregularities are found to increase with increase in laser energy fluence.

\begin{table}[h]
\centering
\begin{tabular}{|c|c|}
\hline
Laser fluence (J cm\(^{-2}\)) & Time delay 350 ns \\
\hline
9.4 & \multicolumn{1}{l|}{\textsuperscript{a}} \\
51 & \multicolumn{1}{l|}{\textsuperscript{a}} \\
85 & \multicolumn{1}{l|}{\textsuperscript{a}} \\
\hline
\end{tabular}
\caption{The plasma plume image at 350 ns for various laser energy fluences, showing that the starting distance of instability is different for different energy fluence.}
\end{table}

\subsection*{B. Estimations of laser ablation parameters using snowplow and shock wave models}

The variation in plasma plume front edge positions with time, using the raw images given in Figure 1, are plotted in Figure 3 for each of the laser energy fluences used in this experiment. The experimentally plotted values are simulated using the snow plow\textsuperscript{,12,14} and the shock wave models.\textsuperscript{13,14} The simulated results of these models are fine tuned to fit the experimental data as close as possible by adjusting the laser and plasma ablation parameters. Hence, the values for the laser energy deposited to the ablated material, mass ablated per laser shot, laser spot radius, and the background gas density can be estimated by fitting the experimental data with the snow plow and shock wave models. To fit the experimentally obtained plume front position (from time resolved imaging), by the snow plow model, the parameters laser spot size, \(R_s\), was measured and the ambient gas density \(\rho_1\) was calculated using the measured ambient gas pressure \(P_1\). As mentioned in one of our previous papers,\textsuperscript{14} the ablated target mass, \(m_a\), at the time \(t = 0\) was estimated by measuring the average mass loss for \(30\) 000 irradiation shots for \(30\) J cm\(^{-2}\) laser energy fluence and was found to be \(6.0 \times 10^{-11}\) kg. In the present experiment, we did not measure the ablated mass per shot for each of the different laser energy fluences, rather as an approximation we used the same value which was obtained for \(30\) J cm\(^{-2}\) and used it in the present simulation and hence the estimated ablated masses mentioned in Table I for different laser energy fluences are relative to this value. As before, the amount of laser energy, \(E_s\), deposited to the ablated material at the time \(t = 0\) cannot be measured experimentally and hence it was deduced by fitting the simulation results to the experimentally measured values of plume front positions.

The estimated values of laser ablation parameters are shown in Table I. These values show the percentage of agreement of experimental values with the simulated values for the ablation parameters. The estimated laser energy deposited to
FIG. 3. Position (R) versus time (t) evolution plots of the visible front edge of the expanding Fe plasma plume, ablated with 532 nm laser of pulse width of ~8 ns, at 1 mbar of Ar gas pressures at different laser energy fluences of (a) 9.4, (b) 23, (c) 51, (d) 60, and (e) 85 J cm$^{-2}$ fitted with the snow plow and the shock wave models.
the ejected material, in both the models, is 70% of the measured value for each of the laser energy fluences. As the measured values were estimated at the target position, the attenuation in laser energy deposited to the ablated material can be attributed to factors like reflection from the Fe target surface, thermal effects, and stress generation at the target, etc. Most interesting point to note is that the estimated value of ablated mass, for snow plow model as obtained from simulation results, is found to vary quite significantly by about 6 times from low value of 18% at 9.4 J cm\(^{-2}\) to relatively high value of 110% at larger laser energy fluence of 85 J cm\(^{-2}\). This is expected because, as mentioned above, these values at different laser energy fluences are relative to fixed ablated mass of \(\frac{6 \times 10^{-11}}{\text{kg}}\) measured for 30 J cm\(^{-2}\) laser energy fluence (as mentioned before) and hence as the incident laser energy fluence is increased/decreased then the corresponding ablated mass is increased/decreased due to the increase in thickness of the evaporated material layer, \(\Delta x\).

The experimental data points, shown in five different plots of Figure 3, are merged together in a single combined plot in Figure 4(a) along with their snow plow model fitted results. The simulated snow plow fit are very good and using the simulated data points of snow plow mode (not using the actual data point), the positions of plume front against incident laser energy fluences at different time instants is estimated and plotted in Figure 4(b). It can be seen from Figure 4(a) and more clearly from Figure 4(b) that at initial instants (refer plume front displacements at 45 ns in Figure 4(b)) the plume front positions are very much the same for different laser energy fluences, i.e., even though the energy fluence is increased by almost an order of magnitude (by more than 9 times from 9.4 to 85 J cm\(^{-2}\)) the plume expansion are nearly the same. This leads to the inference that at initial instants, as long as the laser energy fluence is well above the threshold energy fluence, which is reported\(^{10,21}\) to be about 0.11 to 0.40 J cm\(^{-2}\), required for vaporization of the target material, the initial plume expansion is almost independent of the incident laser energy fluence. It may be attributed to the fact that the initial expansion of ablated plasma near the target surface is mainly driven by the very high driving pressure (~1 kbar)\(^{22}\) and hence there is hardly any effect of background gas pressure or laser input energy and the plume expansion is normally referred as “vacuum like.” Only as the plume moves further away from the target, its expansion dynamics is controlled by other parameters such as laser energy fluence, amount of material ablated, the interaction processes between the ablated material and the ambient gas, etc.

Another important point to note in Figure 4(b) is that plots of plasma plume front positions for different laser energy fluences at any given time instant can be fitted very well by linear trend line and the slopes of these trend lines increase with increasing time instants pointing to the fact

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|c|c|c|c|c|}
\hline
Laser fluence (J cm\(^{-2}\)) & \multicolumn{2}{c|}{9.4} & \multicolumn{2}{c|}{23} & \multicolumn{2}{c|}{51} & \multicolumn{2}{c|}{85} \\
\hline
\textbf{Parameters} & \textbf{Experimental values} & \textbf{SP} & \textbf{SW} & \textbf{SP} & \textbf{SW} & \textbf{SP} & \textbf{SW} & \textbf{SP} & \textbf{SW} \\
\hline
\(E_s\) (J) & \(\frac{4.75 \times 10^{-2}}{\text{J}}\) & 70 & 70 & 70 & 70 & 70 & 70 & 70 & 70 \\
\(m_0\) (kg) & \(\frac{6 \times 10^{-11}}{\text{kg}}\) & 18 & \ldots & 40 & \ldots & 60 & \ldots & 75 & \ldots \\
\(R_s\) (m) & \(\frac{2.25 \times 10^{-4}}{\text{m}}\) & 100 & \ldots & 100 & \ldots & 100 & \ldots & 100 & \ldots \\
\(p_1\) (kg m\(^{-3}\)) & \(\frac{1.615 \times 10^{-3}}{\text{kg m}^{-3}}\) & 7 & 18 & 15 & 40 & 7 & 20 & 6.5 & 22 \\
\hline
\end{tabular}
\caption{Comparison of experimental and the calculated ablation and expansion parameter values for different laser energy fluences. For the experimental measurements, Fe used as target and the background Ar gas pressure was set at 1 mbar. SP: snow plow; SW: shock wave; \(E_s\): laser energy deposited to ablated plasma in model; \(E_s\): experimental value of laser energy at the target; \(m_0\): experimentally measured by collecting ablated material for large number of laser pulses; \(R_s\): experimentally measured laser spot size; \(p_1\): density of argon estimated from filling gas pressures which were measured experimentally.}
\end{table}
that plume expands at increasingly higher speeds with increasing laser energy fluence. Assuming that the similar fraction of incident laser energy (for different input laser energy fluences) goes into the ablated plasma plume, which seems to be the case as seen from results tabulated in Table I as for all cases it is tagged at 70% of laser energy at the target, then the thermal energy of the ablated plasma (and the corresponding velocity and the plasma plume front position) must increase with the increasing laser energy fluence; as observed in Figure 4(b). However, one can easily note that the relative increase in the plume front positions is very marginal compared to the relative increase in laser energy fluence. This is because with the increasing laser energy fluence more mass is being ablated, as inferred through the modelling results; and there is about 6 fold increase in ablated mass from the lowest to the highest energy fluence, and hence even though there is more energy available with ablated plasma but the mass of ablated material itself is higher. Since with the increase in laser energy fluence the ablated plasma plume has faster expansion; one can deduce that the relative increase in energy deposited to ablated plasma is higher than the relative increase in ablated mass (i.e., there is increase in energy available per unit mass or per ablated particle) with increasing laser energy fluence. This explains the trends observed for plasma plume expansion with the increasing laser energy fluence.

As discussed above, one of the obvious conclusion that one can have from the greater expansion rate of plasma plume with increasing laser energy fluence (as noticed from increasing gradients of trend lines with increasing time instants or with relatively wider displacement of data points at higher laser energy fluence in Figure 4(b)) is that average energy that goes to ablated plasma species particle is higher at higher laser energy fluence. To prove this, the instantaneous energy content of the plume (\(E_{\text{sp}}\)), as it expands in space at different times, is estimated using equation \(E_{\text{sp}} = \frac{\Omega}{2} \left( \frac{R}{c} \right)^5\) given in Ref. 19 where the symbols have their usual meaning as described in the corresponding reference. The corresponding ratio of the instantaneous energy content of the plume to the laser energy initially deposited to the ablated mass, i.e., \((E_{\text{sp}}/E_a)\) is estimated. The ratio \(E_{\text{sp}}/E_a\) represents the fraction of the initial deposited laser energy that is contained in the expanding plasma plume at different times. The variation of \(E_{\text{sp}}\) and \(E_{\text{sp}}/E_a\) with time is shown in Figures 5(a) and 5(b), respectively. It is interesting to note that the fraction of initially deposited laser energy that remains with the expanding plume is more for high laser energy fluence as compared to the low energy fluence, as shown in Figure 5(b). It is because of this enhanced fraction of initially deposited laser energy that is available with expanding plasma plume, one observes the faster plume expansion at higher laser energy fluence even though the amount of ablated mass is also high.

The raw images (Figure 1) of the expanding plasma plume were used to plot the maximum plume intensity versus time graph for various laser energy fluences as shown in Figure 6. It may please be noted that the optical arrangement to capture the images (i.e., the lens set, filter combination, distances, and magnification used and the gain of the ICCD camera, etc.) were all identical. The plot gives an instant idea of the effect of laser energy fluence on the intensity of the plasma plume at later timing of the plume expansion. The increase in intensity of the plasma plume with increasing laser energy

![FIG. 5. (a) The fraction of laser energy contained in the plume versus time at different fluences. (b) The ratios of laser energy contained in the plume to the laser energy deposited to the ablated mass versus time.](image)

![FIG. 6. The time resolved maximum intensity profiles of plasma plume for various laser energy fluences.](image)
fluorescence can be attributed to two facts derived and discussed before through the fitting of snow plow and shock wave model to imaging diagnostics results which are (i) the increase in the ablated mass (Table I) leading to more plasma (or radiating) particles in the plume and (ii) the increase in fraction of energy that is being coupled to the expanding plasma plume (higher $E_{wp}/E_{w}$ ratio seen in Figure 5(b)) should lead to higher internal energy and hence enhances the radiation from the expanding plasma plume. Another interesting observation in Figure 6 is that of two distinct regions: (i) the first region before about 1 μs where the intensity of the plume decreases due to fast initial expansion of the plasma plume leading to lowering of the plume temperature in bulk of the plasma and dilution of plasma density due to volumetric expansion and (ii) the second region is after about 1 μs, where plume intensity generally increases first before starting to drop again after about another microsecond. The increase in maximum intensity of the plume in second region of the plume is due to the substantial accumulation and heating of the plasma in shocked gas region at the front edge of the plume along with the start of recombination radiation which again goes down later due to the plasma cooling caused by emission of radiation from the recombination processes.

C. Influence of RT instabilities on surface morphology of Fe thin films

As discussed before, the increase in ablated mass and plasma plume front speed for higher input energy fluences also leads to the occurrence of RT instability at the plasma plume front edge at bigger distance from the target (as observed in Figure 2) or from a smaller distance from the substrate. It was also noticed that more turbulent plasma plume (greater RT instability) was observed with the increasing laser energy fluence. The growth of RT instabilities in the expanding plasma plume will lead to more turbulent plasma and hence the deposited thin films using such a turbulent plasma plume may be different from those of smooth non-turbulent plasma plume. The thin films are expected to have either damages or less uniformity in their morphologies if the plasma plume suffers turbulences due to RT instabilities. The instabilities in expanding plasma plume can be mitigated if the instability growth time is enhanced, i.e., time to grow the R-T instability is increased so much so that it does not occur violently. According to the expression for instability growth $\xi(t) = \xi(0) \exp\left((A_T g k)^{1/2} t\right)$, where $A_T = \rho_1 \rho_2 / \rho_1 + \rho_2$ is the Atwood number for the system to be unstable, i.e., for instability to grow exponential with time $A_T g k > 0$, where $\rho_1 > \rho_2$ with $g$ being positive, i.e., in a gravitational field if the fluid sitting above is heavier than the fluid sitting below then the perturbation at the interface become unstable. This was the set up studied by Rayleigh.

For a case, according to Taylor, where the two fluids, in gravitational field, are accelerating themselves with acceleration $f$ and hence with the effective acceleration $g'$, then the condition for instability to occur modifies to

$$\frac{\rho_1}{\rho_1 + \rho_2} g' k > 0.$$

In such a case, if lighter fluid sits on the top of heavier fluid, i.e., $\rho_1 < \rho_2$ even then the condition for instability can be satisfied if the whole system is accelerating with the effective acceleration $g' < 0$, where $g' = g + f$. This case is equivalent to the situation where the lighter fluid is pushing into the heavier fluid with net acceleration pointing into lighter fluid then the interface of the two fluids becomes unstable.

The growth time for RT instability formation can be increased sufficiently if the factor $(A_T g k)^{1/2}$ is kept low by keeping the magnitude of deceleration, $g'$, of the expanding plasma plume low. The deceleration of the expanding plasma plume depends on the combination of many interrelated parameters and in general is affected by: (i) laser energy fluence, (ii) type of target material used, (iii) back-ground pressure, (iv) photon energy (i.e., the laser beam of wavelength) etc. The point is, if one can mitigate the RT instabilities then probably the surface features (and other properties) of the deposited films can be improved/changed.

Figure 7 shows the thin film samples deposited at three different laser energy fluences with all other parameters being the same, i.e., with argon background pressure of 3 mbar, at substrate to target distance of 3 cm and using 3000 laser pulses. It may be noted that with the increase in the laser energy fluence, the surface of the deposited film is increasingly less smooth with more chaotic patches found on the surface. From the previous discussion, also shown in Figure 2, the distance of onset of RT instability shifts away from the target, i.e., towards the substrate surface with increasing laser energy fluence and hence one may draw an inference that the increasingly patchy chaotic features are the results of shifting of turbulent plasma zone nearer to the substrate surface with increasing laser energy fluence. Another possible explanation for the observation of patchier chaotic surface feature is the increase in the amount of ablated mass with increasing laser energy fluence. The greater the ablated mass, which was observed in the modeling results discussed before, the greater will be the amount of material reaching to the substrate surface with the increasing laser energy fluence which can easily be judged from the images shown in Figure 7.

In other words, the changes in surface features may simply be due to the difference in amount of material being deposited on the substrate surface as the amount of mass ablated by significantly different laser energy fluences are very much different (refer Figure 1) and hence it is not easy to conclude about the direct evidence of the effect of RT instabilities on the surface features of deposited thin films. To clearly establish the effect of RT instabilities on surface morphology of deposited thin films, another experiment was planned where the amount of material ablated is nearly the same while the plasma plume dynamics is significantly different, i.e., for one of the depositions the plasma plume front is turbulent due to the RT instability while for the other deposition the plasma plume is stable and free from RT instability.

D. Influence of laser wavelength on the RT instabilities and the surface morphology of the Fe thin films

As mentioned before, the laser beam photon energy can also lead to different characteristics of plasma plume
(turbulent or smooth) and may affect the morphologies of the deposited material. To investigate this aspect, another experiment was performed using laser beam of different photon energies by using two different wavelengths, 1064 and 532 nm (~8 ns pulse width), while keeping laser energy fluence fixed at ~110 J cm\(^{-2}\) at the same background pressure of 1 mbar of Ar. It is believed that since the input laser energy fluences for the two wavelengths are kept same, the amount of ablated material may not differ too much, as the reflectivity of Fe for 532 nm and 1064 nm radiation is relatively close at 0.56 and 0.65, respectively.\(^2\,3\) Though some difference in amount of ablated material is still expected, not only because of different reflectivities of Fe for different laser wavelengths but also because of dependence of ablated mass on laser wavelength (discussed later in detail), but this difference is going to be much less compared to the case where the amount of laser energy fluence was changed significantly from 9.4 to 85 J cm\(^{-2}\).

The plasma plume images were captured at 200 and 500 ns with a gate width set at 10\% of the delay time. The imaging results for the two different photon energies (i.e., different wavelengths) are shown in Figures 8 and 9. It may please be noted that the images shown in Figures 8 and 9 are captured at greater magnification as compared to that of images shown in Figure 1. Two images each at the imaging instants of 200 and 500 ns, for each of the wavelengths used, were recorded while keeping other experimental condition same to demonstrate the reproducibility of the plume features. There are clear differences in the plume shapes and features; the plasma plume for target ablation using 532 nm is having strong RT instability at the plume front and while for the target ablation using 1064 nm laser beam, the front of the plume is uniform and the shape of the plume is nearly spherical. The high magnification images captured for 532 nm laser pulse, refer Figure 8, shows the formation of RT instability on the plume front edge so distinctively that one can even measure the wavelength, \(\lambda\), of the perturbation in images captured at 200 ns which is found to be about 0.9 mm. The RT instability is growing as one can observe the increase in the amplitude of the perturbation in the images capture at 500 ns. It may please be noted that for 532 nm laser wavelength the experiments were conducted at single laser energy fluence and hence the detailed analysis (like that of one done for 1064 in Sec. III B) is not performed in present investigation, however, a systematic investigation for ablation of Fe target by 532 nm laser has been earlier performed and reported by us in our earlier reported work.\(^1\,4\)

The Rayleigh–Taylor instability is clearly seen to occur in the plasma plume for 532 nm ablation and its possible effect on the physical features of the deposited films was investigated by doing the depositions with 3000 and 6000 number of laser pulses, at argon background gas pressure of 1 mbar with the target to substrate distance of 3 cm for the two wavelengths of laser beam. A set of field emission scanning electron microscope (FESEM) images of Fe thin films samples deposited in our pulsed lased deposition system is shown in Figure 10. The images are shown with two different magnifications. From the SEM images, in Figure 10, it is clear that the thin films deposited using 1064 nm laser are relatively more uniform in the sense that they have relatively...
smoother background and the patchy structures on them are also comparatively much smaller as compared to that of the one deposited with 532 nm laser beam (compare images (a) with (b) and (e) with (f) in Figure 10) for which RT instabilities were observed.

The major issue to be discussed or settled next is to determine the dominating factor that controls the surface features of thin film depositions shown in Figure 10. As mentioned before, the laser wavelengths of 1064 and 532 nm with similar energy fluences are expected to ablate the target material by similar amounts as the reflectivity of Fe is with 9% of each other for these laser wavelengths. It may also be noted that for 8 ns laser pulse width, the laser intensity in present investigation is about $1.4 \times 10^{10}$ W/cm$^2$. It has been routinely reported that for laser intensity less than and more than $10^{13}$ W/cm$^2$ the mass ablation rate scales as $\lambda^{-4/9}$ (Refs. 24–26) and $\lambda^{-4/3}$, respectively, with $\lambda$ being the wavelength of the laser. This implies that for laser intensity ($\leq 10^{13}$ W/cm$^2$) used in present investigation, about 1.36 times more material would be ablated at 532 nm compared to that at 1064 nm. The results reported in above mentioned Refs. 24–26 are for target materials of C, Al, Si, and Sn; whereas Fe target is used in the present investigation. Moreover, in present investigation the major interest is in estimating the relative amount of material being deposited on substrate surface for these two different laser wavelengths. So, another set of thin film deposition was done.
by ablating Fe target using laser energy fluence of about 110 J/cm² for both laser wavelengths at similar target to substrate distance of about 3 cm for same deposition duration of 20 min. The cross-sectional SEM images, shown in Figure 11, shows that average film thicknesses are about 169 and 136 nm for the depositions conducted using 532 and 1064 nm laser wavelengths, respectively. This indicates that the amount of material deposited at 532 nm is about 1.24 times the material deposited at 1064 nm which is quite close the expected value of 1.36 times based on the references mentioned above.24–26

This confirms that amount of material deposited at laser wavelengths of 1064 and 532 nm is only marginally different with the films deposited with 532 nm being only 24% thicker but not so much that it will dominate the surface morphology of the deposited material.

The most elaborate investigation of the effect of laser wavelength on the kinetic energy of ablated species, for graphite ablation, was done by Murry and Peeler.28 They reported that lower laser wavelength, with greater photon energy, causes the ablated species to be in smaller sized ionic cluster of greater kinetic energy. The most energetic ablated carbon ionic species for laser wavelengths of 1064, 532, 248, and 193 nm were reported to have energies of 5, 38, 55, and 600 eV, respectively.28 It may however be pointed that their experiments were conducted at ultrahigh vacuum of $2 \times 10^{-7}$ mbar while present experiments are conducted at 1 mbar; and no experimental data for Fe target ablation is present at ambient gas pressure of 1 mbar. The comparison of plasma plume front positions, at 500 ns instant in Figures 8 and 9, shows that the plume front position for 532 nm is only marginally ahead of 1064 nm ablation. This indicates that the kinetic energies of ablated species, in present investigation, are almost similar and would not result in any significant differences in surface topography of deposited materials.
The comparison of SEM images of Figure 10(c) against Fig. 10(d) shows that the surfaces of thin film samples deposited for two laser wavelengths are equally dense; rather the sample deposited 1064 nm laser (refer Figure 10(d)) has more densely packed individual particles/grains of about 100–150 nm in size. The surface morphology of sample deposited with turbulent plasma plume of 532 nm laser, as shown in Figure 10(c), exhibits the clustering/agglomeration of smaller sized particles/grains to form bigger sized (0.3 to 1 mm) particle agglomerates which will make the surface to have higher surface roughness; while the particles remain isolated in Figure 10(d) forming a much smoother film surface. The formation of even much bigger particle agglomerates/clusters, several microns in dimensions, for 532 nm laser deposition sample can also be noticed in low magnification image shown in Figure 10(a) in comparison to the one seen in Figure 10(b) for 1064 nm laser deposition. Similar conclusions can also be drawn from the comparison of Figure 10(g) with Fig. 10(h), as one can observe that the network of particles is once again much bigger in size in Figure 10(g) for 532 nm laser deposition. The formation of bigger sized particle clusters/agglomerates/network for 532 nm laser deposition may be related to more turbulent RT instability supported plasma plume for this wavelength. The SEM images of Figure 10 thus help us to conclude the strong role played by RT instability plasma in controlling the surface features of the thin films in PLD depositions.

IV. CONCLUSIONS

The experiment with different laser energy fluences of 9.4, 23, 51, 60, and 85 J cm\(^{-2}\) on Fe target at 3 mbars Ar background pressure show some distinct features in expanding plasma plumes: (i) the slow moving plasma core formation up to 150 ns, (ii) more light emission intensity from expanding plasma plume for increasing laser energy fluence which was found to be related to the energy content in the expanding plume, and (iii) appearance of RT instability.

The expanding plume’s front was found to be well defined and smooth up to 150 ns and later (~200 ns onward) its front boundary shows perturbations which were attributed to Rayleigh–Taylor instability. The distance of the onset of the RT instabilities was found to increase with the increase in laser energy fluence which was attributed to ejection of more material at higher laser energy fluence. It was concluded that the energetic plume with larger mass need greater distance for the density of the ablated material to decrease significantly with sufficient deceleration to achieve the conditions required for the onset of RT instability.

The intensity of the plasma plume was found to increase with increasing laser energy. This was attributed to the increase in fraction of energy that is being coupled to the expanding plasma plume (higher \(E_{\text{m}}/E_{\text{o}}\) ratio). This should lead to higher internal energy and hence enhances the radiation from the expanding plasma plume.

The relative increase in the plume front positions was found to be marginal compared to the relative increase in laser energy fluence because with the increasing laser energy fluence more mass is ablated. It was observed that with the increase in laser energy fluence the ablated plasma plume has faster expansion; indicating that the relative increase in laser energy deposited to ablate plasma is higher than the relative increase in ablated mass with increasing laser energy fluence which was supported by modeling results as well. The comparison of the simulation and experimental results reveals that the fraction of input laser energy deposited to the ablated species is about 70% of the input laser energy at the target for all the laser energy fluences.

The possible affect of RT instability on the topography of the deposited material was investigated for two different experimental conditions: (i) using different laser energy fluences for the same wavelength of laser radiation and (ii) for different laser wavelengths keeping the energy fluence same. It was concluded that (i) for Fe thin film depositions using different energy fluences of 1064 nm laser it is not possible to conclusive infer the effect of RT instabilities on the surface features of thin films as the changes in surface features may simply be due to the significant difference in the amount of material being ablated by significantly different laser energy fluences, and (ii) for Fe thin film depositions using similar energy fluences of 532 and 1064 nm laser wavelengths, however, it was conclusively demonstrated that the deposition achieved under turbulent condition, due to RT instabilities, lead to less smooth deposition surfaces with bigger sized particles agglomerates or networks.

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