Chapter 4

Effect of Laser Intensity Profile on the Plasma Formation Mechanism

Abstract

This chapter discusses the influence of energy density profile of ablating laser on the dynamics of laser-blow-off plume by using fast imaging technique. Visualization of the expanding plume in vacuum reveals that geometrical shape and divergence of the plume is highly dependent on the laser intensity profile. Present result demonstrated that the Gaussian profile laser produces a well-collimated, low divergence plasma plume as compared to plume formed by laser having top hat intensity profile. Another interesting feature observed is the formation of laser plasma induced shock wave, when the plasma expands in an ambient medium. Results clearly demonstrate that, highly directional plume produces the strong shock wave in comparison to shock produced by the diverging plume. It has also been found that shock parameters are strongly dependent on the pressure and nature of the ambient gas.
4.1. Introduction

Dependence of the ambient medium and its pressure on the laser blow off (LBO) plume has already been discussed in the previous chapter (Chapter 3). In LBO scheme, the size, shape and divergence of the expanding plume are highly dependent on the thickness of the film, laser spot size and laser fluence. Numerous experimental and theoretical studies have been made to address the effect of film thickness and laser fluence on the LBO generated beam {1-6}. However, little attention has been paid towards investigating the role of intensity profile of laser beam on the geometrical aspect of the LBO plume {7} and its dynamics, which are highly relevant in thin film deposition, neutral beam injection in plasma environment and other LBO induced beam applications.

Several attempts have been made to optimize the expanding laser produced plasma plume (LPP) by varying experimental factors like, ambient gas, focal spot size, laser pulse width, irradiance and wavelength of ablating laser {8-17} General observation is that the lateral velocity of the plume species increases with an increase in the laser fluence. Besides this, the plume expansion becomes cylindrical in shape with increase in the laser spot size.

In view of the above, experiments were conducted to understand the expansion dynamics of the LBO plume formed by two different laser systems having different intensity profiles viz Gaussian and top-hat. In this work, emphasis was given to the comparison of the shape, size, directionality and angular divergence of the LBO plume observed with two different laser profiles by fast time resolved imaging spectroscopy. Study has also been extended to investigate the laser profile dependency of plume formed in an ambient environment and this will be presented in the second part of this chapter.

4.2. Experimental details

A detailed description of experimental setup has been described in chapter 2 {18}. Only the additional parts are briefly summarized here. The experiment was
carried out in a cylindrical stainless steel chamber, evacuated to a base pressure less than $2 \times 10^{-5}$ Torr. The target was composed of uniform layers of $0.05 \, \mu m$ LiF and $0.5 \, \mu m$ thick carbon film, deposited on a 1.2 mm thick quartz substrate.

![Intensity Profiles](image)

**Figure 4.1** Recorded intensity profiles of laser; Gaussian beam profile, “GP” and top hat profile, “THP”.

Nd: YAG ($\lambda = 1064$ nm) lasers having two different intensity profiles, top-hat laser beam profile (referred as ‘THP’) of 8 ns pulse width and Gaussian beam profile (referred as ‘GP’) of 5 ns, were used in this study. Intensity profiles for both lasers are shown in Fig. 4.1. The spot size of the laser beam was $\sim 1$ mm diameter at the target. By adjusting the operating parameters of the laser, fluence $\sim 20$ J/cm$^2$ was set at the target surface for both lasers. An ICCD camera (4 Picos Stanford Computer Optics, Inc.) having variable gain and gating on time, was used to record the time resolved images of the plume luminescence in the spectral range of 350-750 nm. In the present experiment, gate opening time is set at 4 ns. Temporal evolution of the LBO plume has been obtained by varying the time delay from 100 to 4000 ns, between the laser pulse and the opening time of ICCD gate. Five images
were recorded under similar experimental conditions in order to confirm the reproducibility.

In order to measure the distributions of ablated ions across the expansion axis, an electrical ion probe mounted in front of the plume propagation direction {18}. The ion probe was constructed with a tungsten wire of 0.4 mm diameter. The length of the probe, which was exposed to the plasma and separation between the probe and target plate were set as 3 mm and 45 mm, respectively. In order to get the better spatial resolution in the transverse direction, orientation of the probe was aligned along the axis of plume expansion. The probe assembly was mounted on a linear motion feed-through, which enables the positioning of probe in the plume expansion axis. A negative bias voltage of 22 V was applied to the probe to measure the ion current in the saturation limit. A 2 mF capacitor was used to decouple the measuring circuit from the applied bias voltage. The ion signal across the 50 Ω resistance was recorded on a fast digital oscilloscope.

4.3. Plume formation in vacuum

4.3.1. Fast imaging

Fast imaging of the electronically excited plume species, driven by collisional processes between electrons, ions, and neutrals generated by laser-film interaction provides the two dimensional snap shot of the expanding LBO plume. Expansion dynamics of the species as well as geometrical aspect of the expanding plume such as local structure, directionality and divergence, can be studied by observing these emissions as a function of time. Typical ICCD images of expanding plume formed by THP and GP lasers in vacuum and at various time delays are shown in Fig. 4.2. The visual examination of the plume images reveals following interesting effects.
Figure 4.2 The sequence of images of expanding LBO plume in vacuum formed by THP and GP laser at different time delays. The integration time of ICCD was fixed as 4 ns. Colorbar shows the normalized intensity in arbitrary units.

For both laser beam profiles, plume expands linearly and their intensities gradually diminish with time. The plume expansion in vacuum under the influence of pressure gradient inside ablated plume is treated as adiabatic expansion \cite{19}, where the thermal energy of plume species is rapidly converted into the kinetic energy. This leads to decrease in electron temperature and density with time and hence decrease in electron impact process \cite{6}. Therefore, considerable reduction in the emission intensity of the plume with increase in the time delay can be anticipated. Moreover, linear dependence of the plume front position with time delay confirms free expansion of the plume (Fig.4.3). Average translational velocities of $1.8 \times 10^6$ and $1.1 \times 10^6$ cm/sec for the plume by THP and GP lasers respectively are obtained from the slopes of the curve. However, it is observed that the lifetime of the emissive plume, directionality/divergence and shape are highly dependent on the intensity profile of the laser beam.
Figure 4.3 Observed plume length versus time plot for the THP and GP laser generated plumes. The solid lines represent the linear fit for the experimental data.

In the case of THP, the plume is ellipsoidal in shape i.e. the velocity component along the expansion axis is larger than the lateral direction. The recorded images for THP show a non-uniform intensity pattern parallel to the direction of the plume expansion and are more intense at the leading edge of the plume and at points closer to the target. During the expansion, emission intensity decreases rapidly and finally becomes highly diffused after $t > 1500$ ns and is almost beyond the detection limit of the ICCD.

On the other hand, plume formed by GP laser expands linearly with smaller lateral velocity in comparison to that observed with THP laser. In this case, plume has nearly cylindrical shape. Moreover, the lifetime of the emissive plume is found to be significantly larger and is clearly visible up to a time delay $t > 4000$ ns. Unlike THP produced plume, it has a uniform intensity distribution; of course, it has bright patches at leading and trailing edges of the plasma. Another noteworthy observation is that the overall integrated intensities in the vertical section with a width of
Δz = 0.5 mm of plumes produced by THP and GP laser are nearly the same. The calculated value is within 5% uncertainty at any fixed location of the plume. However, due to confined geometry, images with GP laser look brighter as compared to THP plume.

**Figure 4.4** Variation of plume width as a function of time delay for the both THP and GP laser.

The difference in lateral expansion for these cases is clearly visible in plume width versus time plot as shown in Fig. 4.4. In the case of THP laser, width of the plume gradually increases with time. On the other hand, plume width with GP laser slowly increases up to t = 2500 ns and attains nearly constant value with further increase in the time delay. Figure 4.4 shows that the transverse velocity of THP plume is higher than the transverse velocity of GP plume in the overlapping region; and after t > 2500 ns, GP plume follows one-dimensional motion with negligible transverse velocity component.
4.3.2. Divergence measurement

Visible inspection of Fig. 4.2 reveals that there is a significant difference in the divergence of the expanding plume formed by THP and GP lasers. The divergence of plume can be estimated by measuring the diameter of the plume at two separate points 'd_i' and 'd_f' separated by a distance 'x' and using the relation,

\[
\text{Divergence} = A \tan^{-1}\left(\frac{d_f - d_i}{2x}\right)
\]  

(4.1)

Divergence measurement has been carried out over one set of plume images formed at \( t = 800 \) ns. The portion of the plume has been considered up to the maximum acquired diameter, which is nearly equal to half of the total length of the plume. Diameter of the plume at different locations of the plume and separation between them are used to estimate the divergence of plume for both laser systems. The estimated values of divergence of plumes formed by THP and GP lasers at \( t = 800 \) ns are 1.5 rad and 40 mrad respectively. This indicates that transverse expansion in the case of THP produced plume is larger in comparison with GP-plume by a factor of ~ 40. The measured divergence, 40 mrad with GP laser further reduces at later stages (\( t > 2500 \) ns) where the plume width is almost invariant with time. This is an important finding in the sense that GP produces low divergence and long lived highly collimated plume. The persistence of low divergence for longer time is highly suitable for producing collimated atomic/ionic beams.

Further, while comparing the directionality of THP and GP laser induced plumes; it is worthwhile to see the angular dependence of ejected species in the respective plumes extracted from the recorded images \{20\}. The recorded image is divided into radial slices of specific width and the emission intensity is integrated along each axis. A typical intensity distribution obtained by normalizing to the maximum intensity of LBO plumes formed by THP and GP laser respectively at a time delay, \( t = 800 \) ns is shown in Fig. 4.5.
Figure 4.5 Angular dependence of emission intensity of the LBO plumes produced by THP and GP laser at 800 ns time delay.

It should be noted that, the difference in angular distributions for THP and GP is not as prominent as observed in the visible inspection of the plume images, shown in Fig.4.2. This discrepancy is due to the limitation of the adopted method, where the major portion of the angular slice is lying in the intense portion of the plume close to the target, especially at higher angles, which gives wrong information at these angles. In spite of this, some features are still comparable for these two cases and we have observed some differences as well. For both the cases, one can see that distribution of species is highly peaked in the forward direction and shows isotropic behavior. Almost similar distributions are observed for different time delays. The intensity distribution profile of THP plume appears to have two types of distributions (i) a narrow one lying near the central position and (ii) a broader one appearing as a shoulder. On the other hand, species in the GP-plume lie in a narrow angular region and vary smoothly with the angle of incidence.
4.3.3. Ion probe studies

Since the ICCD images provide the information only about the excited plume species, it is worthwhile to verify the above results with other diagnostic techniques. In this regard, the ion distributions across the expansion axis have been measured using an electrical ion probe. Experimental details for this studies has already been discussed in chapter 2.

![Figure 4.6](image)

**Figure 4.6** Ion distributions of the LBO plumes as function of radial distance for the both THP and GP laser. The distance between the ion probe and target plate was set as 45 mm.

The total intensity of ions at any probe position is obtained by the area under the temporal profile of ion current. This has been normalized with the maximum intensity and the intensity variation of ejected ions as a function of radial distance for both THP and GP lasers are shown in Fig. 4.6. It clearly shows that the ion distribution is peaked in the forward direction for both the laser systems and also the ions formed by GP laser have narrow distribution as compared to that of THP laser. Thus, the ion probe results further support the fact that the plume formed by GP laser is of low divergence.
4.3.4. Plume Formation Mechanism

It has already been mentioned earlier that the THP plume has larger divergence in comparison to plume formed by GP laser. For similar power density range, Singh & Narayan \cite{19} and others \cite{21} have modelled the plume formation and its expansion for the conventional solid ablation. According to their proposed models, the pressure driven acceleration is larger along the direction where the plume has smaller size. Since at the initial stage, plume size along the expansion direction is small. Therefore, the velocity component along the expansion axis is much larger than the lateral one and hence the plume acquires an ellipsoidal shape. It is observed that the plume shape during expansion is elliptical in the case of THP, which is in close agreement with the proposed model. However, the plume shape is close to cylindrical for GP plume. The plume size in the transverse direction varies negligibly with time, especially at higher time delays, which could not be fully predicted by the above model. This difference in the divergence of both plumes during their expansion should be related to the difference in plume formation.

The GP-plume moves with smaller velocity as compared to the THP-plume, which can be understood in terms of a qualitative mechanism for the sequence of the processes in the formation of the LBO plume \cite{1} e. g. melting of the film after the laser strikes the front surface, propagation of the melt front towards the back surface and propelling of the material due to increased vapour pressure. In the case of thin film target where skin depth attaining time is less than laser pulse duration, all these events are completed before the termination of laser pulse. Therefore, the propelled material further interacts with the laser and forms the plasma plume. The time required for the melt front to reach the back surface is called melt-through. This parameter plays an important role in the film removal process, which should depend on the power density of the laser, thickness and thermal properties of the target film.
In the case of THP laser, uniform heating of target film within the irradiated region is expected. For the considered power density, the super-heated front surface having high vapor pressure causes explosive rupture of the vacuum-film interface. Therefore, the propelled material is propagated in the forward direction with significant transverse velocity component. On the other hand, in the case of GP produced plume, a non-uniform heating/melting of the film favors bubble formation in between the substrate and film interface \cite{22}. With increase in vapor pressure, ultimately an opening may be formed to release the material rather than the occurrence of explosive rupture. Since peak intensity of the GP laser is higher than the intensity of the THP, a smaller region of the film near the Gaussian peak will reach melt-through rapidly and propulsion may start through a small area. This will reduce the build-up of vapor pressure. Therefore, the material removal will take place through a small orifice similar to gas expansion through a smaller size nozzle but with less stagnation pressure, which may result in a relatively low velocity collimated plume with GP.

4.4. Conclusions

The above discussed results clearly showed the dependence of characteristic expansion of the LBO plume on the laser beam intensity profile. The geometrical shape, velocity and directionality of the plumes formed by the GP and THP laser were significantly different. One of the important observations reported in the present work was the very low divergence plume produced by GP in comparison to the plume generated by THP. The present observations are explained on the basis of the formation mechanism of plasma using two different intensity profiles. However, more theoretical work is required to understand it quantitatively. The present observations will be of significant importance in shaping laser generated plasma plumes and understanding and controlling the geometrical aspect of the LBO generated atomic/ionic beam.
4.5. Plume expansion in an ambient medium: Effect of laser intensity profile

In the previous section, we have seen that laser intensity profile significantly controls the LBO plume geometry. The fast imaging spectroscopy of the LBO plume in vacuum has revealed that the ablating laser having the Gaussian energy density profile produced the LBO plume of cylindrical geometry and expands with little lateral divergence as compared to plume formed by top-hat intensity profile laser (Sec. 4.3). In this context, we were motivated by the fact that the geometrical shape and divergence of plume play the crucial role in plume-ambient atom interaction, especially in shock front formation (23). We therefore attempted to look more closely the above phenomena by considering the LBO plume expansion in different ambient conditions. For this, the study was extended to investigate the expansion dynamics of LBO plume for three different ambient gases: helium(He), oxygen(O₂) and argon(Ar).

Compared to the adiabatic expansion into vacuum, the interaction of the plasma plume with an ambient gas is a complex gas dynamic process, which we have already discussed in chapter 3 (6, 24-27). During the initial period after plasma formation, plume has a very high driven pressure and the effect of ambient gas is not significant. In this stage plume expands just as in vacuum. However, expansion dynamics is fully controlled by the interaction between the ablated species and the ambient atoms at the time delay of hundreds of nanoseconds depending on the pressure and atomic weight of the ambient gas. After the termination of laser pulse plume, particles move with high velocity into the surrounding gas with continuous interaction of ambient atoms. When the mass of the ablated plume is equivalent to the mass of the ambient gas, plume material compresses the ambient gas and as a result, strong shock wave is formed. The strength, shape and velocity of the shock front are highly dependent on the velocity and angular distribution of the plume and nature of the ambient gas.
In view of the above, experiments were conducted using the fast imaging to understand the dynamics of LBO plume and shock formation in He, O₂ and Ar environment. In this study, emphasis was given to the shape, strength and velocity of the shock wave and their dependence on the pressure and nature of the ambient gases. Helium and argon as ambient environment has been chosen for comparison due to the large difference in their atomic masses and ionisation potentials whereas oxygen provides the information regarding the chemically reactive aspect.

### 4.5.1. Imaging results

LBO plasma plume has been recorded using an ICCD fast imaging system and the results are presented in the coming sections.

![Figure 4.7 Images of LBO plume at a background pressure of 3 x 10⁻⁵ Torr of vacuum and 5 x 10⁻¹ Torr of three ambient gases He, O₂ and Ar at five different time delays after the onset of plasma formation.](image)

In order to understand the effect of pressure and atomic/molecular weight of ambient gas on the geometrical structure and hydrodynamical movement of the expanding LBO plume, the temporal evolution of emitting plume are recorded at different pressure levels of He, O₂ and Ar under the similar experimental conditions.
Figure 4.7 shows the sequence of images recorded at different time delays, varying from 600 ns to 3000 ns at vacuum and ambient pressure of $3 \times 10^{-1}$ Torr of He, O$_2$ and Ar gases. All these images are recorded at a laser irradiance of $\sim 3$ GWcm$^{-2}$ and normalized to their maximum intensity. Each image represents the spectrally integrated emission intensity in the range 350–750 nm emitted from different species within the plume.

As can be seen from Fig.4.7, in vacuum plume propagates linearly and their intensities decrease gradually with time. Intensity distribution is almost uniform throughout the plume volume, though it has bright patches at leading and trailing edge of the plasma. In vacuum, plume expands under the influence of pressure gradient and is treated as adiabatic expansion {28}, where electron temperature and density decrease with time and hence electron impact process also decreases. Therefore, considerable reduction in the emission intensity of the plume with increase in the time delay is observed. Similar studies have already been performed using top-hat profile laser {Chapter 3}. In the present case Guassian profile laser was used and compared to the earlier studies. One of the peculiar observations is that, the plume expands with negligible lateral velocity and has nearly cylindrical shape in comparison with conventionally observed elliptical shape {Chapter 3}. Due to the highly directional nature, the lifetime of the emissive plume is found to be significantly larger and is clearly visible up to $t > 4000$ ns. Very low divergence and long lived highly collimated plume is attributed by film ablation mechanism with laser having Gaussian intensity profile {Sec. 4.3}.

Plume was completely modified upon introducing the ambient gas. These images show distinct features of the plasma plume in $10^{-1}$ Torr of He, O$_2$ and Ar pressures in terms of emission intensity, duration and geometrical shape of expanding plasma plume. Visible examinations of the images clearly demonstrate the effect of nature of the ambient gas, e.g. atomic mass, ionisation energy and thermal conductivity on the expanding plasma plume. In the case of He, major
portion of the geometry is nearly identical to that observed in vacuum and travel almost with the same distance as in the case of vacuum expansion. An additional luminous cone-shaped structure starts to appear in front of the plume in the presence of ambient gas. This added component is attributed to shock front and propagates with a higher velocity as compared to the plume velocity and therefore well separated from plasma plume in He environment.

In the case of heavier ambient gas, e.g. O\(_2\) and Ar, the images are significantly different as compared to the observed images in He environment. Apart from the more intense and sharper contact boundary i.e., the shock front, a drastic enhancement in emission intensity especially in the centre portion of the plume are observed in presence of Ar and O\(_2\). In this case, (P < 10\(^{-1}\) Torr) conical structure remains intact with plume front and not well separated as in the case of He environment.

In order to get more insight into the plume dynamics under various environments, plume images as a function of gas pressure of He, O\(_2\) and Ar are shown in Fig. 4.8. The background gas pressure varies from high vacuum to 3 Torr pressure and time delay is set as 1000 ns. Careful examination of Fig. 4.8 reveals the following general features. Under the same ambient pressure, the shock front moves with higher velocity in lighter gases (He) compared to the velocity in heavier gases like O\(_2\) and Ar.

At a fixed time delay, shock front is more luminous in higher atomic/molecular gas and becomes more intense and sharper with increasing the background gas pressure. A second intensity discontinuity just behind the shock front is observed in Ar and O\(_2\) at pressure > 1x10\(^{-1}\) Torr. This effect is not visible at lower pressure of heavier gases and all the considered pressure range of He.
Figure 4.8 Images of LBO plume at different background pressures of three ambient gases He, O\textsubscript{2} and Ar at a time delay of 1000 ns after the onset of plasma formation. Colorbar shows the normalized intensity in arbitrary unit.

Further, the enhancement in emission intensity, especially inside the plume is not much significant with increasing the He pressure. However, drastic enhancement is observed in shock front and inside the plume in the case of both Ar and O\textsubscript{2}. For a fixed time delay (1000 ns) and at pressure 10\textsuperscript{-1} Torr, it has been found that there is an overall enhancement in intensity of 1.52 times in Ar and 1.45 times in O\textsubscript{2} as compared to that in He. As usual, plume experiences more resistive force i.e, drag force, in heavier gas environment in comparison to that of He gas, and
therefore axial as well as lateral confinement is dominant with increasing the pressure of Ar and O₂. This can be clearly seen in the measured plume length for the different pressures of He, Ar and O₂ at fixed time delay as shown in Fig.4.9.

![Observed plume length at 1500 ns for the different pressures of He, Ar and O₂.](image)

**Figure 4.9** Observed plume length at 1500 ns for the different pressures of He, Ar and O₂.

The difference in enhancement in emission intensity inside the plume in different gasses can be explained in terms of electron impact excitation which is the dominant process to excite the plume species {Chapter 3}. Since the ionization energy of He is greater than O₂ and Ar, more increase in electron density during the plume-ambient gas interaction is expected in O₂ and Ar environment. Another mechanism which contributes to the increase in electron density in different ambient gases is the cascade growth of the electron number density and absorption coefficient of the plasma in ambient environment {Chapter 3}.

### 4.5.2. Shock structure

The shape of the plume and its divergence control plume-gas interaction and hence influences the shock front formation and expansion. The highly
directional plume with insignificant lateral movement and cylindrical geometry as in the present case, efficiently compresses the ambient gas and therefore provides better chance to form strong shock front. This can be clearly seen in the Fig. 4.7 where, the appearance of shock wave structure and their visibility in ICCD images are more clear as compared to reported results in the past {6, 24}.

![Figure 4.10](image)

Figure 4.10  (a) plume formed in vacuum $4 \times 10^{-5}$ Torr (b) plume formed in Helium pressure of 1 Torr (c) subtraction of image 'a' from image 'b'.

In order to confirm the presence of shock front, we have subtracted the plume image formed in vacuum from the images observed at 600 ns and 1000 ns in He medium as shown in Fig. 4.10. Interestingly the major portion of the plume is completely eliminated after the image subtraction process and only the cone shaped front portion remains visible. This method confirmed that, the shape and intensity of plume observed in vacuum remains unchanged in He environment even at a
relatively high pressure of 1 Torr and the intense shock front is formed only due to the compression of background gas by the plume front.

The explosive hot plume expands adiabatically, with high velocity creating a high pressure material front and pushing the surrounding gas. Its expansion velocity rapidly decreases and it transfers its energy and momentum to the ambient gas, thus sending out a shock wave which gradually separates from the plume. Taylor-Sedov \cite{29} model the spherical blast wave expansion by considering the blast wave originating from a point explosion where the shock swept the ambient gas mass such that $M_a \gg M_p$, where $M_a$ and $M_p$ are the masses of ambient gas and mass of the ablating species respectively. This model is valid for the strong shock condition where the ambient gas pressure is negligible in comparison with the to pressure behind the shock front. The shock wave model has limiting characteristic distance $R_1$, for a particular gas and pressure, below which this model is not valid \cite{30}. $R_1$ is set by the requirement that the mass of the gas encompassed by the shock wave (in volume $= \frac{2}{3}\pi R^3$) is much greater than the initial ablated mass $m$. This requires

$$R_1 \gg R = \left(\frac{3m}{2\pi \rho_b}\right)^\frac{1}{3} \tag{4.2}$$

Due to the cylindrical geometry in present case, one cannot directly estimate the distance where the shock initiated, but it is clear that in the case of heavier background gas and high-pressure region, shock is initiated at early stage of plasma compared to that of lighter gas and relatively low pressure region. The above inference is clearly visible in the present observation as shown in Fig. 4.8.

**4.5.3. Shock wave analysis**

In order to compare the shock wave condition in different gases the density ($\rho_s$), temperature ($T_s$) and velocity ($V_s$) in the shocked region in He, Ar and O$_2$ is
estimated by using the mass, momentum and energy conversation, and is given by 
\{31, 32\}

\[
V_s \approx \left(\frac{\gamma + 1}{2}\right) V_a \\
T_s \approx \frac{2\gamma}{\gamma + 1} \left[ \frac{\gamma}{\gamma + 1} \frac{M^2 + 1}{M^2} T_b \right] \\
\rho_s \approx \rho_b \left(\frac{\gamma + 1}{\gamma - 1}\right)
\]

where \(V_a\) is the velocity of expanding plume, \(M\) is the Mach number (\(M = V_a /\text{sound velocity}\)), \(\gamma\) is the specific heat ratios of background gas and \(T_b\) and \(\rho_b\) is the temperature and density of the undisturbed background gas. Using the time-of-flight of plume, velocities of expanding plume at \(10^{-1}\) Torr of He, O\(_2\) and Ar are estimated as \(1.63 \times 10^4\), \(1.36 \times 10^4\) and \(1.26 \times 10^4\) m/sec respectively. Using the relation (4.3, 4.4), the estimated velocity in the shock region in He, O\(_2\) and Ar are \(2.16 \times 10^4\), \(1.62 \times 10^4\) and \(1.67 \times 10^4\) m/sec respectively. The above estimate indicates that at fixed background pressure, shock front moves with highest velocity in He compared to the velocity in O\(_2\) and Ar and therefore clear separation between the plume front and shock front is observed in He environment (Fig. 4.7). Further, the estimated temperature at the shock region in He, O\(_2\) and Ar pressure of \(10^{-1}\) Torr are \(2.07\), \(11.55\) and \(12.23\) eV respectively. The high temperature in O\(_2\) and Ar is correlated with the observed luminosity of the shock front.
Figure 4.11  Intensity profiles for various time delays for the plume formed in (a) Helium and (b) Argon gas pressure of $5 \times 10^{-1}$ Torr.
In order to get a better presentation of the shock propagation, intensity variation of the ICCD images along the expansion axis are binned for the different delay at 10^{-1} Torr of He and Ar as shown in Fig. 4.11.

Figure 4.11 clearly demonstrate most of the evolution features, e.g., intensity variation with time, shock formation and its expansion, plume confinement and their differences in He and Ar environment. Two well separated components corresponding to the expanding plume and shock front are clearly observed in He environment. The separation between the plume and shock front increases with increase in time delay and the intensity of the shock front reduced with time. On the other hand, in the case of Ar, the plume portion and shock front are not resolved as observed in the case of He. However, at lower time delays, one can easily identify the ionized front (discussed later) and the shock front merging in intense plume front with increasing time delay due to the increase in the resistive force with time. The rate of decrease of shock intensity with time is higher in He as compared to decrease in Ar environment. This is because of the large temperature difference in He and Ar environment which is ~ 2 eV and 12 eV in He and Ar respectively. Because of the higher thermal conductivity of He, shock front is rapidly cooling in He environment \{24\}.

The above intensity profile can also be used to estimate the shock strength in different ambient conditions. The shock strength which is the reciprocal of the shock thickness, is obtained by using the density gradient in pre-shock to post shock region \{33\},

\[
\frac{1}{S} = \delta = \frac{I_{\text{max}} - I_0}{Z_a - Z_b}
\]  

(4.6)

where, \(I_{\text{max}}\) is the peak intensity, \(Z_a\) and \(Z_b\) are the distance from the target where the intensity is 75% and 25% of the peak intensity. \(I_0\) is the pre-shock intensity, \(\approx 0\). Figure 4.12 shows the typical intensity profile at 600 ns time delay in
He adopted for calculating the shock strength. From the intensity profile of the plume images, the second peak away from the target is considered as shock region and inverse of the width of 25% and 75% of the falling edge of the lobe represent the shock strength. The shock strength at different pressure levels of He and Ar plotted as a function of time are shown in Fig 4.13.

![Shock Strength Calculation](image)

**Figure 4.12 Method adopted to calculate shock strength from intensity profile plot.**

This gives information regarding the development of different stages of shock front propagation in time domain. The calculated shock parameter values clearly shows the dependency of shock wave formation and its dynamics over the ambient gas medium and are highly influenced by the mass of ambient gas. From the first sight it is clearly visible that shock strength increases with rise in pressure of surrounding gas medium. Upon introducing the gas, shock wave forms in the medium and continuously interact with the gas and decreases with time. Maximum shock strength is observed at 3 Torr pressure in both He and Ar environment. Increase in pressure cause more resistance from the external medium which causes increase in blast pressure, and hence maximizes the shock strength. During the
initial period of plume formation $t < 1500$ ns, the higher plume pressure causes larger shock strength and then gradually decreases with further increase in time delay. Obviously, the shock strength is maximum in Ar because of heavier atoms.

As already mentioned, a sharp and more intense boundary, just behind the shock front is observed in Ar and O$_2$ at pressure $> 1 \times 10^{-1}$ Torr. This can be

![Figure 4.13 Shock-strength calculated from the ICCD images recorded for plume formed in (a) Helium and (b) Argon ambient gases.](image-url)
explained in terms of elastic scattering of plume front species with the ambient atoms/molecules. The highly directional plume species are scattered in collision with the ambient atoms and deviate from the original direction. Scattering angle depends on the mass and radii of the plume and background species. The scattering angle of the carbon atoms which is the major constituent in the plume has been estimated and it is observed that there is a change in direction by ~15°, ~75° and ~78° in single collisions with He, O₂ and Ar atoms respectively {34}. Since the temperature in the shock region is very high ~ 12 eV in O₂ and Ar, there is a finite chance of ionization of scattered plume species. Therefore, due to the higher scattering angle and higher shock temperature, the second intense boundary behind the shock front is attributed to ionized plume front. Due to low scattering angle and shock temperature ~2 eV, the ionized front is not visible in He environment. Here we ignore the inelastic scattering and chemically reactive scattering, especially in the case of O₂ gas as reported in past {35}. In the case of inelastic scattering or oxidation of plume species i.e., chemically reacted with dissociated oxygen, width of the plume in O₂ is narrower than plume width in Ar because of low scattering angle of heavier molecules. The reduction of plume width and length with increasing pressure is mainly due to the increase of resistive force of ambient gas and hence this effect is maximum in heaviest gas like O₂ and Ar.

4.6. Conclusions

In summary, the characteristic expansion of LBO plasma plume and the formation of shock wave in different gas pressure, ranging from high vacuum to 3 Torr of He, O₂ and Ar have been studied experimentally with fast gated ICCD camera. The enhancement in emission intensity, structure formation, drag force and confinement effect is maximum in Ar environment in comparison to He and O₂ environment. The associated physical processes responsible for the production of these differences were briefly discussed. The present observations suggest that highly collimated, low divergence plume produces substantially strong shock wave
in comparison to the previously reported shock produced by the diverging plume. It has been found that shock velocity is highest in He and the shock front is completely detached from the plasma plume. However, the shock temperature and strength is maximum in high pressure and heaviest ambient gas. We could not observe any significant difference in the plume shape in Ar and O$_2$ environment and therefore ignore the possibility of chemically reactive processes in the present case. Further, an ionizing front just behind the shock front is observed in Ar and O$_2$ at pressure $P > 1 \times 10^{-1}$ Torr and this effect is not visible for the entire pressure range in the case of He. We feel that this study will give additional information to explore the above subject.
4.7. References


