On the use of shockwave models in laser produced plasma expansion

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Abstract. Interaction of medium to high peak power laser pulses with solid materials produces a plasma that expands supersonically. Expansions of such plasmas have been studied and several models have been proposed to describe it. This work presents a study of the expansion of laser produced plasmas in both vacuum and gas environment by using Langmuir probe and photography. It compares some of the most used models to identify that which better describes the expansion process. In vacuum, such process is properly described by the Anisimov model. However when expanding in a background gas it is found that the Sedov-Taylor model fits properly the position of generated shockwave but overestimates both kinetic energy and pressure of the expanding plasma. Such problem is solved by using a modification of the Freiwald-Axford model. Finally it is demonstrated that after the plasma stopping distance the plasma inter in a diffusive regime.

1. Introduction

Studies on the expansion of laser produced plasma can be traced back to as early as the 60s, where both the in vacuum [1] and in gas background [2] expansion was analyzed. Different models have been proposed to describe these expansions [3-6], most of them using a gas-dynamic approach and describing both temporal and spatial behavior of macroscopic parameters, such as density, temperature and expansion velocity. Knowledge of these parameters is important not only to gain inside on the nature of the plasma it also allows to define how plasma species interact among them and with surrounding gas or solid. Such interactions are of great importance in processes like pulsed laser deposition (PLD) where chemical reactions or sputtering are important factors to produce high quality thin films.

One of the most used [7-12] model for describing laser produced plasma expansion in a gas environment is the Sedov-Taylor (S-T) model for the description of shock waves in nuclear explosions [13]. This can be understood, firstly, by analyzing the basic assumptions in the strong explosion theory: (i) a large amount of energy $E_p$ is released instantaneously from a small (negligible) volume, (ii) the mass of the energy source $m_p$ is negligible compared with the mass of the gas background swept by the shock wave and (iii) the pressure exerted by the explosion over the gas should be greater than the pressure in the unperturbed background gas. Secondly to a straightforward to use set of equation, given by this model, relating shock wave expansion velocity and the plasma parameters mentioned before.
2. Experimental setup

The work was carried out in a stainless steel chamber with a base pressure of $8 \times 10^{-6}$ mbar using a Pfeiffer-Balzers TSH 050 turbo molecular system (50 l/s). When working in a background gas, 5.0 grade oxygen (Air Products) was fed through a Whitey needle-valve into the chamber. A Balzers Penning was used as pressure gauge for the $10^{-5}$ to $10^{-3}$ mbar range. For the region of 0.2-1 mbar an Edwards Pirani gauge was used. The latter was calibrated using a Hastings HPM-2002-OBE wide range gauge, which consists of a dual sensor unit containing a piezoresistive direct force sensor and a thin film Pirani type sensor.

A KrF (248 nm, 26 ns) excimer laser was used for ablation. The laser pulses were focused on the rotating target using a 50 cm focal length lens obtaining a rectangular spot of 0.6 x 0.1 cm dimensions obtaining a fluence of 1 J/cm$^2$. The laser pulse was incident at an angle of 45° to the normal of the target. The average energy per pulse ($E=0.06$ J) was measured using a Scientech thermopile joulemeter.

Images of the plasma expansion were acquired by placing an Andor ICCD camera parallel to the plasma expansion axis and using a combination of two lenses to image the plasma onto the detector of the camera. The axis of the delivery system cut the plasma expansion axis at a distance of 2 cm from the target surface in order to cover a total distance of 4 cm. Both delay and acquisition (3.8 ns) times were established using a Stanford delay generator.

The ion signals for the in vacuum TOF analysis were collected using a planar probe (0.053 cm$^2$), biased at -30 V and capable of both translates over the plasma expansion axis (Z) and rotates on either Z-X or Z-Y plane, see figure 1.

3. Results and discussion

3.1. Expansion in vacuum

Anisimov et al. [3] treated the adiabatic expansion of a one component vapor cloud into vacuum using a particular solution of the gas-dynamic equations, which applies when describing flows with self similar expansion. It is assumed that the formation time of the vapor cloud is much less than its expansion time and that the focal spot of the laser has an elliptical shape with semi-axes $X_0$ and $Y_0$.

The expansion is modeled as a triaxial gaseous semi-ellipsoid (figure 2.1) whose semi-axes are initially equal to $X_0$, $Y_0$ and $Z_0 \approx v_{\text{sound}} t_p$, where $t_p$ is the duration of the laser pulse and $v_{\text{sound}}$ is the sound speed in the vaporized material.

As mentioned before plasma initial parameters were obtained making use of the Anisimov model following the procedure explained by Hansen et al. [14]. The expansion front velocity was calculated from the measured arrival time of the plasma front to different positions using the ion signals from Langmuir probe measurements. In all cases it was used the time at which 10% of the maximum signal is reached. The expansion velocity was calculated to be ($v_f=2.3\times10^6$ cm/s). Numerical calculations were performed for different $\gamma$ values. The calculated current flows were compared in each case to the
experimentally obtained ion currents in order to obtain the best $\gamma$ value. The best fit for both time of arrival and signal shape was obtained for $\gamma=1.1$.

Using this $\gamma$ value, the total initial energy of the plasma was estimated to be $E_p=0.053 \text{ J}$, which is a reasonable result if we take into account the energy losses during the evaporation of the material. Here, it was assumed that the energy required for evaporation of the target material is the sum of the energy expended in heating up the material $E_{\text{heat}}$ and the vaporization energy $E_{\text{vap}}$. Thousands of degrees Kelvin are temperatures typically reached on the target material during PLD process. For ZnO at the pressures used in this work, vaporization temperatures are in the range of 1000 to 1500 K [15].

The energy needed to bring a certain mass of material at room temperature to the above mentioned temperatures can be estimated as $E_{\text{heat}} = mc\Delta T$; with $c = 0.5 \text{ JK}^{-1}\text{g}^{-1}$ and a mass of 1.6 $\mu$g, which was the average evaporated mass per pulse, a value of $E_{\text{heat}} = 0.001 \text{ J}$ is obtained. Using 464 kJ mol$^{-1}$ as the vaporization energy per mol [15] and knowing the number of moles in the evaporated mass ($1.23 \times 10^{-8} \text{ mol}$ of ZnO), the energy expended for the vaporization will be $E_{\text{vap}} \sim 0.006 \text{ J}$. The total energy required for the evaporation is then ~0.007 J, adding this value to the energy obtained from the model (0.053 J) gives a total energy of 0.06 J which was the average energy used in the experiments.

On the other hand the angular dependence of the number of particles arriving per unit area normal to the flow, $J(\theta)$, can be obtained as well from the Anisimov model. It is defined as [3]:

$$J(\theta) = (1 + \tan^2 \theta)^{\frac{3}{2}} \left[ 1 + \left( \frac{Z_{\text{inf}}}{Y_{\text{inf}}} \right)^2 \tan^2 \theta \right]^{\frac{1}{2}}$$

The plasma angular distributions in both the X and Y directions were measured by rotating the probe in the x-z and y-z planes respectively. Figure 2 shows the normalised ion angular distributions. For the y-z plane the radius of the described circumference was 4 cm from the target, while for the x-z plane it was 7.8 cm. Both data were fitted, using equation (1).

![Figure 2. Ion angular distribution, measured by a Langmuir probe at different angles with respect to the plasma expansion axis. Top: at a distance from target of 7.8 cm in the x-z plane. Bottom: at a distance of 4 cm and in the y-z plane. Squares are experimental values and solid lines are the fits obtained using equation. (1).](image)

### 3.2. Expansion in a gas environment

Under this condition the analysis of the plasma expansion is definitively more complex than the vacuum case. Now the expanding plasma particles will interact with the molecules of the gas, new
processes of energy exchange appear speeding up the reduction of the total energy of the expanding plasma.

In supersonic expanding plasmas, as developed during PLD, translational energies are greater than the internal energies of the ions; from this the importance of being able to characterize the plasma expansion is evident. Depending on the pressure of the gas, plume splitting and total braking of the expansion occurs. Plume splitting is characterized by the detection of two peaks in TOF studies of the plasma flow.

In general both features, plume splitting and plasma deceleration, are related with the appearance of both a shock wave and a contact front. The shock wave is the propagation of a perturbation created by the sudden impact of the expanding plasma with the gas background. It defines a boundary layer separating the unperturbed gas in front of the shock wave from the gas that went through this layer and is perturbed. The contact front, then, establishes a boundary between this perturbed gas and the plasma.

The formation and propagation of shock waves has been widely studied [13, 16]. It is well known that the formation of such a feature is related to the ratio of the pressure (density) of the perturber and the gas and that the speed at which it propagates through the gas is defined by the thermodynamic properties of the gas. On the other hand, a full formulation has been given in which parameters such as pressure, density and temperature of the perturbed gas behind the shock wave are expressed as a function of the speed at which this wave propagates through the gas. A more specific work on shock waves developed for the modeling of strong explosions [13] has been widely used for the description of the laser produced plasmas [7-12].

3.2.1. Sedov-Taylor model. The Sedov-Taylor (S-T) theory defines the shock position $R$ by:

$$R = \varepsilon_0 \left( \frac{E_s}{\rho_0} \right)^{\frac{1}{2(\gamma+1)}} t^{\frac{2}{2(\gamma+1)}}$$

where $\varepsilon_0 = 1.08 \left( \frac{\gamma+1}{2} \right)^{\frac{1}{2(\gamma+1)}}$

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where $\rho_0$ is the density of the gas background and $\gamma$ its specific heats ratio. The parameter $\nu$ will take values of 1, 2 or 3 for expansions with planar, cylindrical or spherical symmetry. The flow speed $U_{bs}$, density $\rho_{bs}$, peak pressure $P_{bs}$ and temperature $T_{bs}$ behind the shock (bs) are given by:

$$U_{bs} = \frac{2}{\gamma + 1} \frac{dR}{dt}$$

$$\rho_{bs} = \frac{\gamma + 1}{\gamma - 1} \rho_0$$

$$P_{bs} = \frac{2}{\gamma + 1} \rho_0 \left( \frac{dR}{dt} \right)^2$$

$$T_{bs} = \frac{2\gamma}{\gamma + 1} \left[ \frac{\gamma - 1}{\gamma + 1} M^2 + 1 \right] T_0$$

where $M = \frac{1}{v_{sound}} \frac{dR}{dt}$ is the Mach number with $v_{sound} = \sqrt{\frac{P_0}{\rho_0}}$.

In practice this model is commonly used to extract the plasma expansion velocity by fitting curves of shock position versus time measured during the experiment. Langmuir probe and ICCD images are the main characterization techniques used to measure the experimental values.
Figure 3 shows time resolved images, which were recorded using the ICCD camera as explained before. The images were recorded for two different pressures of oxygen, 0.06 mbar and 0.4 mbar. The low pressure case is characterized by a broader and homogeneous distribution of the hotter regions in the plasma plume. As the plasma expands, the hotter region starts concentrating toward the leading edge of the plume. The higher pressure case shows the presence of the confined region occurring earlier in time during the expansion. For longer periods of time this region begins to broaden. For both cases the appearance of such a region defines the formation of a contact front, which in turn is related to the formation of a shock wave.

**Figure 3.** Images acquired for different time delay as the plasma expands in gas background at 0.06 mbar and 0.4 mbar. The direction of the expansion is denoted on the top-left image with length scale of 4 cm. The target position is the left border in all cases. Images are normalized to their maximum intensity.
Figure 4 shows the measured TOF using the Langmuir probe for the 0.06 and 0.1 mbar oxygen pressures. The plume splitting is clearly present at all distances and at the two pressures, though its resolution increases with the increase of both distance and pressure. For clarity the peaks are labelled in the bottom-right graph.

**Figure 4.** Ion currents measured in the TOF study of the plasma expansion at 0.06 mbar and 0.1 mbar of oxygen. Two peaks are detected. The first peak expands through the gas background with a vacuum-like constant velocity, while the second peak decelerates. First and second peaks are labeled in the right-bottom graph for clarity. Examples of the fits (red lines) are presented for TOF measured at 4 cm for both pressures.

Figure 5 shows the TOF of the ion signals as measured using the Langmuir probe at a distance of 2, 3 and 4 cm from the target surface and at a pressure of 0.4 mbar of oxygen. At this pressure plume
splitting is no longer detected. There is a stronger peak value reduction with increasing distance and a broadening of the distribution.

**Figure 5.** Ion currents measured in the TOF study of the plasma expansion at 0.4 mbar of oxygen. The expansion is characterised by a strong deceleration and a broadening of the signals. Note that the signal corresponding to 3 cm has been increased for clarity in the plots.

In order to extract the profiles of the curves which convolve to form the observed TOF, the curves were fitted using shifted half Maxwellians (see red lines plots in figure 4) and then both front and peak arrival time were extracted. The plots of the arrival time at each position for both peaks at the three different pressures are shown in figure 6. As mentioned before, the first peak shows a TOF which indicates a linear vacuum-like flow. The velocities, in each case extracted from the slope of the plots, were similar for both pressures and half the value obtained for the in-vacuum expansion in the previous section. The second peak shows a decelerated expansion with a greater deceleration at the higher pressure. In this case the fit was performed using the Sedov-Taylor model, equation (2).

As mentioned above the figure 6 corresponds to the data extracted using the Langmuir probe. Dashed and dotted lines were obtained by setting fixed values of both $E_p$ and $\rho_0$ to 0.053 J and $9\times10^{-5}$ kg/m$^3$ respectively allowing the fitting procedure to find the best $\varepsilon_0$ and $\nu$. The values of $E_p$ and $\rho_0$ correspond to the energy predicted by the Anisimov model in the previous section and the background gas density for a pressure of 0.06 mbar respectively. The solid lines were obtained by setting fixed values of parameter $\varepsilon_0$, choosing $\gamma = 1.28$, and $\nu = 3$ corresponding to a spherical symmetry as explained before.
Figure 6. Plots of the arrival time of ion TOF at each position for both peaks at 0.06, 0.1 and 0.4 mbar of oxygen. For 0.06 and 0.1 mbar, where plume splitting is observed, the first peak shows a vacuum-like expansion. Both front and peak arrival times at each position show a decelerated expansion. Dashed, dotted and solid curves represent the fits performed using equation (2).

The analysis performed to the data extracted using the ICCD camera is presented in the figure 7. The data represents the position of the front edge of the images presented in figure 3. The fitted curves were obtained following the same idea as for the fit of the data presented in figure 6.
Figure 7. Plots of the arrival time of the images front edge at each position for both pressures 0.06 and 0.4 mbar of oxygen. For the two background pressures the expansion is decelerated. Both dashed and dotted fit lines were obtained following the same procedure as explained for figure 6.

In an overview of the results presented in both figure 6 and 7 it can be corroborated that in general the expansion of laser produced plasmas is satisfactorily reproduced by a general shock wave law of the type of equation 2. However a close comparison reveals that different values of the parameters of the equation are obtained for the same experimental data. At 0.06 mbar of oxygen background, figure 6, the fit of the front position (dashed line) suggests that the plasma expands with a cylindrical symmetry while the fit of the peak position (dotted line) is closer to a spherical one. Similar problems arise if comparing the previous mentioned results with the 0.06 mbar of oxygen case but for the data measured using ICCD images (figure 7).

3.2.2. Modified Freiwald-Axford model. This model is a modification of Freiwald-Axford model [17]. Details of modification can be seen in [19]. Final equation is:

\[
\left( \frac{dR}{dr} \right)^2 = \frac{E_p}{\rho_0 R_p^3 + \frac{8.37}{\gamma+1} \rho_0 \gamma^\frac{\gamma-1}{2} R_p^3 + \frac{8.37}{\gamma+1} \rho_0 R_p^3}
\]

Knowing the values of the energy \(E_p\), density \(\rho_0\) and radius of the plasma \(R_p\) before starting the expansion the position of the plasma front as a function of time can be obtained by numerical
evaluation. Once the plasma front velocity is known the rest of the parameters can be calculated using equation (3).

Figure 8 shows the results of fitting the plasma front position using equation (4). For both pressures the values of $E_p$ and $\gamma$ calculated from Anisimov model were used while the initial plasma density ($\rho_{p0}$) and radii ($R_{p0}$) were left free. As can be observed for both pressures there is a good agreement, having greatest deviations at early times of expansion. Such feature is reasonable if we take into account that it will be needed a period of time before reaching the necessities conditions to form a shock wave and contact front, i.e. the dimension of the shell where the swept background species are accumulating should be greater than the diffusion length of species in contact or within it [18]. A visual inspection of figure 3 shows that a well defined front (brilliant region) is present around 1.5 µs and 1 µs for the 0.06 mbar and 0.4 mbar of oxygen, respectively, which are the times when respective fits (figure 8) start the best agreement. Finally, it should be noted that best values obtained for $\rho_{p0}$ and $R_{p0}$ are reasonable.

A further step in comparing the previous models was to take into account the following phenomenological analysis. It is well known that when the plasma expands in the presence of a gas background there exists a maximum distance which it will be able to reach [19, 20]. This is due to the reduction of its internal energy as a consequence of the work developed in displacing the surrounding gas. At this distance the pressure exerted by the plasma equals that of the gas background. So a correct model should not only fit the plasma front position but lead to a correct plasma pressure calculation at the stopping distance.

Two different models, the drag model [19] and the Predtechensky model [20], were used to determine the plasma stopping distance for the pressures used in the experiments. Table 1 shows the calculated stopping distances. Both models predicted quite similar distances with differences of 5 mm and 10 mm for 0.06 mbar and 0.4 mbar, respectively. However such differences can’t be depreciated in
processes like PLD where position of substrate with respect to the ablated material plays an important role.

Table 1. Plasma stopping distances calculated using both Drag and Predtechensky models.

| Oxygen Pressure (mbar) | Drag Model | Predtechensky Model |
|------------------------|------------|----------------------|
| 0.06                   | 4.9        | 5.5                  |
| 0.40                   | 3.9        | 3.1                  |

So in selecting the best values the following method was used. If the value for the maximum expansion distance is correct, it is to be expected that from this point on the plasma expands similarly to the spreading of a cloud of ions by diffusion through a gas.

The ionic number density $n$ at a radius $r$ from the release point and time $t$ can be defined as follows [21]:

$$n = \frac{N_0}{(4\pi D(t-t_0))^\frac{3}{2}} e^{-\frac{r^2}{4D(t-t_0)}}$$  (5)

where $N_0$ is the initial number of ions at the point where the diffusion starts and $D$ is the diffusion coefficient. The parameter $t_0$ was introduced in our work for fitting purposes to allow for an offset in the initial time. Using a Langmuir probe placed at 4 cm from the target the ionic current was measured at a background pressure of 0.4 mbar. Figure 9 shows the collected current together with a fit performed using equation (5). The best value obtained for $r$ set the release point at 3 cm which is the stopping distance predicted by the Predtechensky model.

![Image](image-url)

**Figure 9.** Ion current (black squares) measured at 0.4 mbar using a Langmuir probe at 4 cm from the target. Solid line is the fit obtained using equation (5).

Once the plasma stopping distances were defined equations (4) was used to calculate $dR/dt$ and then the pressures were calculated using set of equation (3). Figure 10 shows the obtained results. The pressure plot shows that for both gas backgrounds the F-A model predicts with good agreement the
position at which the plasma exerted pressure equals that of the gas surrounding it. On the other hand pressures from S-T model are too high and do not match the background pressure at the predicted stopping distance.

It is interesting to note that the agreement obtained with equation (5) is indicating that once the plasma reach the stopping distance it enters in a diffusive regime, yet Freiwald-Axford model fits plasma front positions. This can be explained if we take into account that at this point the shock wave becomes a normal sound wave which still expands under the same law. It of course to be expected that with the increase of the distance deviations will appear due to the predominance of the diffusion.

4. Conclusions
This work presented a study of the expansion of laser produced plasmas in both vacuum and gas environment by using Langmuir probe and photography.

In vacuum, the expansion is properly described by the Anisimov model. Plasma gamma value and initial energy as calculated from this model can be used as initial parameters for calculating plasma front position when expanding in a gas background.

It was found that the Sedov-Taylor model fits properly the position of generated shockwave but overestimates both kinetic energy and pressure of the expanding plasma. Such problem is solved by using a modification of the Freiwald-Axford model.

Finally it is demonstrated that after the plasma stopping distance the plasma inters in a diffusive regime.

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