Langmuir probe diagnosis of laser ablation plasmas

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Abstract. For laser ablation plumes which are significantly ionised, Langmuir probes have proved to be a relatively simple and inexpensive tool for measuring the plume shape, ion energy distribution and electron temperature. In this paper we describe some recent work on the development of Langmuir probes for laser ablation plume diagnosis. Typically in laser ablation plasma the flow velocity is supersonic, which complicates the interpretation of the I-V probe characteristic. We describe some new work on the behaviour of a flat probe lying parallel to the plasma flow. We also compare our measurements with theoretical models of laser ablation plume expansion and draw some conclusions as to which model is more appropriate for the low temperature plasmas which arise in pulsed laser deposition.

1. Introduction
In the investigation and application of laser ablation of solids it is helpful to have a theoretical model of the expansion of the laser ablation plume both for the interpretation of laser ablation experiments and optimizing process applications such as pulsed laser deposition. While there has been a considerable amount of numerical simulation of ablation plume hydrodynamics for specific materials and irradiation conditions, it is more helpful to have analytical (or semi-analytical) methods which can be applied more generally.

In this context, the adiabatic expansion models of Anisimov et al [1] and Singh and Narayan [2] have proved to be very useful for the understanding and interpretation of laser ablation experiments. Both models consider the adiabatic expansion of a small volume of hot gas into vacuum. The dimensions of the volume of gas are chosen to correspond to the dimensions of the vaporised material at the time when evolution and heating of the ablated material is complete. For nanosecond (ns) ablation this time corresponds to the end of the laser pulse, while for femtosecond (fs) ablation the electron-phonon relaxation time seems to be an appropriate choice. The Anisimov model considers the expansion of the plume to be isentropic, which essentially means that there is no heat conduction between parts of the plume. On the other hand the Singh and Narayan model makes the assumption that the plume is isothermal at all times. There is clearly a need to establish which of these extreme constraints is more appropriate. Though the expansion model of Anisimov was developed for the expansion of a neutral gas, it seems to agree with Langmuir probe measurements in ablation plumes [3, 4].

In this paper we will consider these two aspects of modeling the expansion of laser ablation plumes. We show that both models can be applied to ionised ablation plumes and that the isentropic model is the more appropriate choice when the plume temperature is less than about 12 eV.
2. Theoretical considerations

In laser ablation with ns laser pulses heating of the solid leads to intense evaporation for the duration of the laser pulse. It is also now clear that the vaporised material absorbs the laser and is heated to temperatures of several eV resulting in significant ionisation. For a metal where the laser fluence is not far above the ablation threshold the depth of ablated material is about 10 nm. This material expands at the sound velocity during the laser pulse, so that at the end of the pulse the vaporised material exists as a 10 – 100 μm layer of plasma, which has a density of about $10^{19}$ cm$^{-3}$ and a temperature of several eV. It is this material which then expands adiabatically into vacuum [5].

The density gradients in the plasma drive the expansion which leads to supersonic flow. Since the gradient is greatest normal to the target, the acceleration of flow is greatest in the same direction resulting in the usual forward directed ablation plume. Considering the expression for the electron-ion equipartition given by Spitzer [6], it is clear that the electron-ion collision rate is sufficient to maintain the equality of temperature between the electrons and ions, i.e. $T_e=T_i=T$. Thus the plasma pressure will be:

$$ P = n_i(Z+1)kT $$

(1)

where $n_i$ is the ion density, $Z$ is the mean ion charge, $k$ is the Boltzmann constant and $T$ is the temperature. Since the Debye length is much smaller than the plasma dimension we can ignore the influence of charge separation on the expansion dynamics. Following the analysis of Attwood’s, it can be seen that there are two contributions to the gradient of plasma pressure. The first is due to the ion density gradient:

$$ \frac{\partial P}{\partial x} = \gamma kT \frac{\hat{\partial} n_i}{\partial x} $$

(2)

Secondly the gradient of electron density gives rise to an ambipolar electric field:

$$ E = -\frac{1}{e} \frac{\partial P_e}{\partial x} = -\frac{1}{e} \frac{\partial \gamma kT \hat{\partial} n_e}{\partial x} $$

(3)

which of course will act on the ions. The ion momentum equation is given by:

$$ m_i n_i \left[ \frac{\partial}{\partial t} + v_i \frac{\partial}{\partial x} \right] v_i = -(Z+1)\gamma kT \frac{\hat{\partial} n_i}{\partial x} $$

(4)

Where $m_i$ is the ion mass, $v_i$ is the ion flow velocity and $\gamma$ is the ratio of specific heats. This is the same starting point as used by Anisimov in his analysis of the expansion of a small volume of hot vapour. Thus it is valid to use the models developed by Anisimov and Singh and Narayan to describe the expansion of ionised laser ablation plumes. However the following question remains; is the expansion isentropic or isothermal? Essentially we want to know if electron heat diffusion is sufficiently rapid to keep pace with the plasma expansion. Taking the Spitzer formula for the thermal conductivity of plasma it can be shown that heat will diffuse a distance, $s$, in time, $t$, given by:

$$ s^2 = \frac{9.3 \times 10^{19} T^{5/2}}{Z(Z+1)n_i} t \quad [\text{cm}^2] $$

(5)

where $T$ is in eV and $n_i$ in cm$^{-3}$. The rate of plasma expansion is of the same order of the ion sound speed, $c_s$, and in one dimension, the size, $l$, of a freely expanding plasma after time, $t$, is approximately:

$$ l = c_s t = 9.79 \times 10^4 \left( \frac{n_i}{M} \right)^{1/2} t \quad [\text{cm}] $$

(6)

where $M$ is the atomic weight. For as long as the expansion is approximately one-dimensional; $n_i l = n_s d$, where $n_s$ is the atom density in the target and $d$ is the ablation depth. Thus:

$$ n_i = \frac{n_s d}{l} $$

(7)
Substituting for \( n_s \) in (5) leads to:

\[
\frac{s^2}{Z(Z+1)n sd^3} = 9.3 \times 10^{-9} T^{5/2} 
\]

For an isentropic plasma expansion we require that \( s < 1 \), which from (6) and (8) yields:

\[
T^2 < 1.05 \times 10^{-14} \gamma^{1/2} Z^{3/2} (Z+1)n_s d M^{-1/2} 
\]

Taking \( \gamma = 5/3 \), \( Z = 1 \), \( d = 10^{-6} \text{ cm} \) and for Ag, \( n_s = 5.86 \times 10^{22} \text{ cm}^3 \) and \( M = 108 \) we get:

\[
T < 12.4 \text{ eV} 
\]

Thus it would seem that for typical \( n_s \) ablation conditions the plume expansion may be treated as isentropic if the plasma temperature is less than about 12 eV, which is normally the case.

The Anisimov model of the expansion of a laser ablation plume is a solution of the gas dynamic equations under adiabatic and isentropic conditions. The starting point for the model is the small semi-ellipsoidal volume of hot material that has been produced by the ablation process at the end of the laser pulse and has dimensions \( X_0, Y_0 \) \( \text{ and } Z_0 \), where \( Z_0 \) is normal to the target. At all times the expansion is self-similar and the various plasma parameters are constant on semi-ellipsoidal surfaces. For example the temperature at any given point is given by:

\[
T(x,y,z) = (5/3)(\gamma - 1)/(2\gamma) \frac{E_p m}{k M_p} \left( \frac{X_0 Y_0 Z_0}{XYZ} \right)^{\gamma - 1} \left[ 1 - \frac{x^2}{X^2} - \frac{y^2}{Y^2} - \frac{z^2}{Z^2} \right]^{-1/2} 
\]

where \( E_p \) is the actual energy in the plume, \( M_p \) is the plume mass, \( m \) is the atom or ion mass and \( X, Y \) and \( Z \) are the semi-axis of the plume front. The model can be used to predict various aspects of the expansion, which can be compared with experiment. In particular the aspect ratios of the plume at large distances and the particle flux on to a detector have been measured using Langmuir probes and compared with theory [3,4]. In this paper we concentrate on how the measured time variation of electron temperature for a laser ablation plume compares with the prediction of the Anisimov model.

3. Experiment
A 248 nm, 26 ns, KrF excimer laser at normal incidence was used for ablation. The laser fluence was 1.5 J cm\(^{-2}\) on a spot of 1 x 0.5 mm\(^2\). A plane Langmuir probe with dimensions 13 x 3 mm\(^2\) was positioned close to the target normal at a distance of 9.5 cm. To record the ion flux the probe was oriented normal to the flow and biased at \(-10 \text{ V}\) to reject electrons. Since the flow is supersonic, the ion current is dominated by the ion flow velocity and is given by \( I = A \eta \nu_i \), where \( A \) is the probe area. Langmuir probes have also been used to measure \( T_e \) in ablation plumes [8-10]. However since typical ion flow velocities correspond to energies of 10-200 eV the ions will still reach the probe in the electron retarding region, which complicates the extraction of a value of \( T_e \). To measure \( T_e \), we have adopted the procedure of orienting the probe surface parallel to the plasma flow to eliminate the ion current due to flow [11]. By recording the probe signals for a range of bias voltages, choosing a particular time, and then plotting the current against the bias voltage, the I-V characteristic is obtained.

4. Results and discussion
Figure 1(a) shows an ion signal obtained with the probe surface oriented perpendicular to the plasma flow at a bias of \(-10 \text{ V}\). The TOF of the maximum ion flux corresponds to an ion energy of about 117 eV. This signal also yields a value of \( 3.8 \times 10^3 \text{ m s}^{-1} \) for the velocity of the plume front normal to the target. Figure 1(b) shows how the electron signal depends on bias for the probe lying parallel to the flow. The I-V characteristic obtained with the probe in the parallel position, for the peak ion time-of-flight (TOF) of 5.5 \(\mu\)s is shown in Figure 2. The slope of the linear part of a semi-logarithmic plot (inset) yields a value of about 0.3 eV for \( kT_e \). Figure 3 shows the variation of the measured \( kT_e \) as the plasma flows past the probe. The temperature is seen to increase to a maximum of approximately 0.35 eV. This initial increase in temperature as the plasma arrives is consistent with the assumption that the plume expansion is both adiabatic and isentropic.
Figure 1. (a) Ion signal for the probe oriented perpendicular to the flow. (b) Electron signal for various biases for the probe in the parallel position.

From the measured plume front expansion velocity we can derive a value for $E_p/M_p$ in the initial plasma, which can be used in (11) to predict the temperature variation at the probe position. The initial value of $E_p/M_p$ corresponds to 60 eV per ion. This predicted temperature at 9.5 cm is shown in figure 3, and has the same behaviour as the measurement, though the magnitude is lower.

Figure 2. The I-V characteristic corresponding to the peak ion TOF (5.5 μs). The semi-logarithmic plot is shown in the inset where $kT_e$ is obtained from the slope of the linear part.

Figure 3. The variation of the measured electron temperature as the plasma flows past a probe at 9.5 cm compared to the calculated plasma temperature using (11).

5. Conclusion
In conclusion we have shown that the adiabatic gas dynamic model of laser ablation plume expansion developed by Anisimov and Singh and Narayan can be used to describe an ionised plume. Further we have shown that the isentropic model of Anisimov is appropriate when $T_e \leq 12$ eV. We have found good agreement between the prediction of the model and experiment for the time varying electron temperature in plasma produced by laser ablation of Ag.
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