An Alternate Treatment of the Vapor-Plasma Transition

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Abstract
In this paper, we address the problem of predicting $p(I)$, the variation of surface ablation pressure vs. incident pulsed laser intensity $I$, from the onset of ablation through the transition to its mediation by laser-induced plasma in vacuum. Despite its simplicity, the recently published approach of Sinko and Phipps [1] to this problem describes momentum coupling for many laser-target interactions quite well, for one material at a single wavelength where the ablation fluence threshold is clearly defined. Alternatively, if vapor pressure vs. temperature data $p(T)$ is available for a material, e.g., using the SESAME tables, a different model can be used. In addition to the $p(T)$ data, this model only requires knowledge of basic parameters for the material, such as specific heat, thermal conductivity, optical absorptivity, atomic weight, and its ionization state energies and their partition functions. Since all these parameters, except for optical absorptivity, are independent of laser wavelength, it is possible to calculate a material’s mechanical impulse response to pulsed laser irradiation with broad applicability. We show that our model agrees with published data on momentum coupling in aluminum from KrF to CO$_2$ laser wavelengths to within a factor of two.

1. INTRODUCTION
When a laser pulse is incident on a target in vacuum, mechanical impulse is produced by the pressure of photoablation at the target surface. The figure of merit for this interaction with pulsed laser intensity $I$ is the mechanical coupling coefficient $C_m$,

$$C_m = p/I [N/W]$$

where $p$ is the ablation pressure in Pa. Since typical $C_m$ values are of order 10–100 $\mu$N/W, the portion due to light pressure ($C_{hp} = 2/c = 6.7$ nN/W) is relatively ignorable.

Because of its crucial importance to the design of laser space propulsion engines and applications [2–4], it is crucial to be able to predict the intensity at which maximum $C_m$ will occur for a given material in vacuum at any of the customary combinations of laser wavelength and pulse duration ($\lambda$, $\tau$).

As intensity is increased, ablation begins in the neutral vapor regime (which we term $|R_i|$) and progresses to the fully-formed plasma regime ($|R_{||}$) where the ionization fraction in the ablation plume

$$\eta_i = n_i/(n_o + n_i) = 1,$$

where $n_i$ is the ion number density. $|R_{||}$ is normally not coincident with $|R_i|$. When the conditions for fully forming $|R_{||}$ obtain, the optical spectrum and heat flux transferred to the target surface are entirely mediated by the plasma layer and laser light does not reach the target surface. Since this is a complex transition, treatments prior to [1] treated the two extremes separately, without providing a method of predicting surface pressure through the transition, between them. Model [1] treated the transition well for single organic compounds where ablation thresholds $\Phi_o$ are well-defined, but its results apply to a single wavelength if $\Phi_e$ is wavelength-dependent, as the model involves $\Phi_o$ explicitly.
On the other hand, for elemental surface absorbers such as Al, for which $p(T)$ tables such as the SESAME tables exist, a more general approach can be used to advantage, which we introduce in this paper. We restrict consideration to the range $100 \text{ps} < \tau < 1 \text{ms}$ and $248 \text{nm} < \lambda < 10.6 \mu\text{m}$ and intensities below the inertially confined fusion regime treated by Lindl [5]. We do not treat effects in atmosphere here, nor CW laser irradiation, which is the subject of a subsequent paper.

2. PLASMA REGIME

It was shown by Phipps, et al. [6] that the simple relationship

$$C_{mp} = 184 \frac{\Psi^{9/16}}{A^{1/8} \left( I \lambda \sqrt{\tau} \right)^{1/4}} \text{[\mu N/W]}$$  (3)

derived from inertial confinement fusion compression physics describes $C_m$ to within a factor of two for surface absorbers in the plasma-dominated regime for 23 metals and opaque nonmetals in vacuum for wavelengths from 248 nm (KrF) to 10.6 $\mu\text{m}$ (CO$_2$) and pulse durations from 1 ms to 100 ps. In Eq. (3),

$$\Psi = \frac{A}{2 \left[ Z^2 (Z+1) \right]^{3/4}}$$  (4)

"Vacuum" in the context of this paper can be taken to mean any ambient pressure $p_o < 0.001 \text{ Pa}$, although Beverly and Walters [7] showed that the ambient has small effects on momentum coupling up to $p_o < 1 \text{ Pa}$. There also resulted

$$I_{mp} = 442 \frac{A^{1/8} \Psi^{9/16} \left( I \lambda \sqrt{\tau} \right)^{1/4}}{[\text{s}]}$$  (5)

for the plume "specific impulse," $\nu_{plume}/g_o$ and

$$T_e = 2.56 \frac{A^{1/8} Z^{3/4} \left( I \lambda \sqrt{\tau} \right)^{1/2}}{\left( Z+1 \right)^{3/4}} \text{[eV]}. $$  (6)

where $A$ is the average atomic mass number and $Z \geq 1$ is the average ionization state in the laser-produced plasma plume, which is, in turn, determined by applying Saha’s equation [8],

$$n_e n_i = \frac{2u_j}{u_{j-1}} \left( \frac{2\pi m_e kT_e}{\hbar^2} \right)^{3/2} \exp(-W_{j-1} / kT_e).$$  (7)

and writing

$$Z = n_e/n_i,$$  (8)

under the obvious normalization constraint

$$\sum_{j=1}^{j_{\text{max}}} (n_j) = n_i$$  (9)

Parameters in the preceding relationships are: $W_{j-1}$, the ionization energy difference in eV between the $(j-1)$th and $j$th ionization states of the material; $m_e$, the electron mass; $kT_e$, the electron temperature in the plasma plume [eV]; Planck’s constant $\hbar$; the neutral vapor density $n_o$, $c$, the speed of light; $I$ the incident laser intensity [W cm$^{-2}$]; the plume electron total number density $n_e$[cm$^{-3}$]; $u_j$ the quantum-mechanical partition functions for the $j$th state; and $n_i$, the number density of each of the ionized states.

It is convenient to implement (7–9) numerically (see Allen [9]) by forming

$$s_j = \frac{n_{ij}}{n_{i+1-j}} = \frac{8.64 \times 10^{26} 2u_j}{n_e \theta^3 u_{j-1}} \exp(-W_{j-1} / kT_e)$$  (10)
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where \( \theta = 5040/T_e \), and then computing the array

\[
P_j = \prod_{k=1}^{j} S_k = \begin{bmatrix} n_1 & n_2 & n_3 & n_4 & \ldots \end{bmatrix},
\]

(11)

and the constants

\[
R_j = \frac{n_j}{n_0} = \sum_{j=1}^{\infty} \frac{P_j}{j}
\]

(12)

and

\[
R_2 = \frac{n_2}{n_0} = \sum_{j=1}^{\infty} jP_j
\]

(13)

from which

\[
Z = R_2 / R_1
\]

(14)

and

\[
\eta_i = (1 + 1/R_i)^{-1}.
\]

(15)

can be computed, as well as

\[
n_e = R_2 [(kT_e/p)(1 + R_1 + R_2)]^{-1}
\]

(16)

for a new iteration in Eq. (10).

3. VAPOR REGIME

We consider absorbed laser intensity \( aI \) to be expended in the six processes of Eq. (17):

\[
aI = \rho_v \left[ C_p(T - T_o) + q + v^2/2 \right] + x_h \rho C_v(T - T_o)/\tau + \epsilon \sigma T^4 + \phi(T, x_h).
\]

(17)

where \( \rho \) is target surface absorptivity, so that \( aI \) is absorbed intensity. Taking these six terms in order, these energy sinks are 1) heating the vapor to temperature \( T \), 2) providing the energy \( q \) to create a vapor of atomic species 3) accelerating the vapor to the sound speed \( v = c_s \) at target surface temperature \( T \), 4) heating a surface ablation layer of thickness \( x_h \) to temperature \( T \) from room temperature \( T_o \), 5) black body emission with emissivity \( \epsilon \) from the half-plane facing the laser, and 6) conduction through the thermal gradient in the heated layer with thickness \( x_h \), \( \phi(T, x_h) \). In Eq. (17), the vapor density is related to pressure \( p \) by

\[
\rho_v = \frac{An_m p}{kT}
\]

(18)

\( v \) is the vapor velocity at the target surface (where momentum is transferred to the target),

\[
v = \left[ \frac{\gamma(Z + 1)kT}{Am_p} \right]^{1/2},
\]

(19)

\[
q = q_f + q_v
\]

(20)

contains the energies of fusion and vaporization,

and

\[
x_h = x_{ih} + x_v + \lambda / \alpha \geq \lambda
\]

(21)

is the effective thickness of the laser-heated, solid-density layer in the target during ablation. In Eq. (21), \( x_{ih} = (xT)^{1/2} \), \( \kappa \) is thermal diffusivity, \( x_v \) is surface recession depth during the laser pulse and \( \alpha \) is the optical absorption coefficient [cm\(^{-1}\)] at the surface, of the order of \( 1/\lambda \). For the \( R_1 \) vapor regime, \( Z \approx 0 \) in Eq. (19). Where \( C_p \) and \( C_v \) are, respectively, the specific heat of the target material at constant pressure and volume, the quantity \( \gamma \) in Eq. (19) is

\[
\gamma = C_p/C_v
\]

(22)
Now, we work backwards, requiring an intensity balance between incident laser intensity $I$ and interaction parameters to define what $I$ must have been. To do this, we substitute

$$\frac{k}{C_p} = \left( \frac{\gamma}{\gamma - 1} \right) \rho T,$$  \hspace{1cm} (23)$$

$$\rho_v C_p T = \left( \frac{\gamma}{\gamma - 1} \right) \rho v,$$  \hspace{1cm} (24)$$

and

$$\frac{v^2}{2C_p T} = \frac{\gamma - 1}{2}$$  \hspace{1cm} (25)$$
in Eq. (17) and rearrange it in a more convenient form to give:

$$I = \frac{p v}{a} \left( \frac{\gamma}{\gamma - 1} \right) \left[ 1 - \frac{T_o}{T} + \frac{q}{C_p T} + \frac{\gamma - 1}{2} \right] + \frac{\sigma T^4}{a} + B(\tau)$$  \hspace{1cm} (26)$$

where

$$B(\tau) = \frac{1}{a} \left[ \phi(T, x_h) + \frac{x_h \rho \omega C_v (T - T_o)}{\tau} \right],$$  \hspace{1cm} (27)$$

We can relate the quantity $p$ in Eq. (26) to $T$ by using the Riedel equation [10] in conjunction with the SESAME equation-of-state database for Al maintained at Los Alamos National Laboratory [11], for $T \leq 7890$ K, its triple point.

Eqs. (26) and (27) are wavelength-dependent only as $\lambda$ affects the surface absorptivity $a$. For the infrared to ultraviolet range studied here, we used $0.05 \leq a \leq 0.24$ for aluminum [12].

We now have a numerical solution which relates $p$ and $v$ to $I$ over the range corresponding to our $p(T)$ data, and we can compute the vapor regime coupling coefficient as

$$C_{mv} = p_v/I.$$  \hspace{1cm} (28)$$

Vapor specific impulse is

$$I_{spv} = v/g_{vo}$$  \hspace{1cm} (29)$$

Where limited extrapolation from $p(T)$ data is required, we can write a Clausius-Clapeyron equation for the surface pressure.

$$\ln(p/p_i) = \left[ C \frac{\Delta H}{k} \left( \frac{1}{T} - \frac{1}{T_i} \right) \right]$$  \hspace{1cm} (30)$$

where the subscript “1” refers to a 1-bar reference condition, $T$ is the vapor temperature, $\Delta H$ is the enthalpy of melting, vaporization and dissociation and $C$ is a fitting constant. $\Delta H$ can be found in statistical physics references [13].

### 4. COMBINED MODEL

Having results for the two physical extremes of vapor and plasma, the question arises of how to make a smooth transition between the models. To do this, we use the approach in [1], writing

$$C_m = p/I = \left[ (1 - \eta) p_v + \eta_p p_p \right]/I = (1 - \eta) C_{mv} + \eta C_{mp}.$$  \hspace{1cm} (31)$$

Specific impulse during the transition can be obtained in the same way.

Figure 1 shows the results we obtained for aluminum in vacuum, together with nine data sets which fit the model to within an rms standard deviation of a factor of two in $C_m$. 

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5. DISCUSSION

We showed the successful fitting of the momentum response of a single material vs. incident intensity at three different combinations of wavelength and pulse duration, across the transition from the vapor to the plasma regime. This is important because it permits us, for the first time, to determine the intensity for peak momentum coupling for any material for which we know \( p(T) \), a critical determination for laser space propulsion applications, such as the ORION concept [17].

Unlike [1], which treated material response at one wavelength, we have modeled mechanical coupling data spanning the whole range from KrF (\( \lambda = 248 \text{ nm} \)) to CO\(_2\) (\( \lambda = 10.6 \mu\text{m} \)) lasers with a single model in which only surface absorptivity \( a(\lambda) \) varies. Our results are shown in Figure 1.

Although the parameter \( I\lambda\sqrt{\tau} \); is not involved explicitly in the vapor regime analytical model, we plotted the data vs. \( I\lambda\sqrt{\tau} \) for two reasons. In the first place, we have shown [18] that the threshold for the \{RI\} – \{RII\} transition in our parameter range is given by a fixed value of \[ I\sqrt{\tau} = 4.8E8 \text{ Wm}^{-2}\text{s}^{1/2} \] so that \( C_m \) data plotted vs. \( I\lambda\sqrt{\tau} \) should show peaks distributed according to \( \lambda \). This result agreed with our numerical modeling, which showed that \( Z \) and \( \eta_i \), which in turn control the \{RI\} – \{RII\} transition, are numerically dependent on \( I\lambda\sqrt{\tau} \) because of their dependence on \( I\sqrt{\tau} \).

Second, \( I\lambda\sqrt{\tau} \) is the controlling variable in the plasma regime theory, and we have shown before [6] that plasma regime \( C_m \) data from many wavelengths, pulse durations and materials coalesce in that representation, as predicted by the \{RII\} theory and, if we are treating a model for the \{RI\} – \{RII\} transition, it makes sense to use that plotting variable. In Figure 1, data enters the plasma regime for \( I\lambda\sqrt{\tau} > 120, 560, 5,000 \) (left to right) for the three model fits.

Because of the different wavelengths, it is impossible for the Figure 1 data to follow a single trend in the vapor regime, and this is what we see: trends separated horizontally by the magnitude of \( \lambda\sqrt{\tau}/a \) for the various data sets. To make this meaningful, we have taken care to use data with similar \( \tau \) in the vapor regime.

Figure 1 and the associated analysis shows an advantage for shorter wavelengths and pulse durations for achieving larger peak coupling coefficient \( C_m \), in aluminum.

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