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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,

$$C_m = p/I \quad [\mathrm{N/W}] \tag{1}$$

where p is the ablation pressure in Pa. Since typical C_m values are of order 10–100 μN/W, the portion due to light pressure ($C_{hv} = 2/c = 6.7$ 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 (λ, τ).

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

$$\eta_i = n_i/(n_o + n_i) \approx 1, \tag{2}$$

where n_i is the ion number density. $\{R_{II}\}$ is normally not coincident with $\{R_I\}$. When the conditions for fully forming $\{R_{II}\}$ 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 Φ_o are well-defined, but its results apply to a single wavelength if Φ_o is wavelength-dependent, as the model involves Φ_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 ps < τ < 1 ms and 248 nm < λ <10.6 μm 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

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

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 μm (CO_2) and pulse durations from 1 ms to 100 ps. In Eq. (3),

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

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

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

for the plume "specific impulse," $v_{\mathrm{plume}}/g_{\mathrm{o}}$

and

$$T_{\mathrm{e}} = 2.56 \frac{A^{1/8} Z^{3/4}}{(Z+1)^{5/8}}\left(I\lambda\sqrt{\tau}\right)^{1/2} \quad [\mathrm{eV}]. \tag{6}$$

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],

$$\frac{n_{\mathrm{e}} n_{\mathrm{j}}}{n_{\mathrm{j-1}}} = \frac{2u_{\mathrm{j}}}{u_{\mathrm{j-1}}}\left(\frac{2\pi m_{\mathrm{e}} \mathrm{k}T_{\mathrm{e}}}{\mathrm{h}^2}\right)^{3/2} \exp(-W_{\mathrm{j,\,j-1}}/\mathrm{k}T_{\mathrm{e}}), \tag{7}$$

and writing

$$Z = n_{\mathrm{e}}/n_{\mathrm{i}}, \tag{8}$$

under the obvious normalization constraint

$$\sum_{\mathrm{j}=1}^{j_{\max}} (n_{\mathrm{j}}) = n_{\mathrm{i}} \tag{9}$$

Parameters in the preceding relationships are: $W_{\mathrm{j,\,j-1,}}$ the ionization energy difference in eV between the (j–1)th and jth ionization states of the material; $\mathrm{m_e}$, the electron mass; $\mathrm{k}T_{\mathrm{e}}$, the electron temperature in the plasma plume [eV]; Planck's constant h; 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 jth state; and n_{j}, the number density of each of the ionized states.

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

$$s_{\mathrm{j}} = \frac{n_{\mathrm{i\,j}}}{n_{\mathrm{i,i-j}}} = \frac{8.64\mathrm{E}26}{n_{\mathrm{e}}} \frac{2u_{\mathrm{j}}}{\theta^{1.5} u_{\mathrm{j-1}}} \exp[-W_{\mathrm{j,\,j-1}}/\mathrm{k}T_{\mathrm{e}}] \tag{10}$$

where $\theta = 5040/T_e$, and then computing the array

$$P_j = \prod_{k=1}^{j} S_k = \left[\frac{n_1}{n_o}, \frac{n_2}{n_o}, \frac{n_3}{n_o}, ... \right], \tag{11}$$

and the constants

$$R_1 = \frac{n_i}{n_o} = \sum_{j=1}^{j_{max}} \frac{P_j}{j}. \tag{12}$$

and

$$R_2 = \frac{n_e}{n_o} = \sum_{j=1}^{j_{max}} jP_j \tag{13}$$

from which

$$Z = R_2 / R_1 \tag{14}$$

and

$$\eta_i = (1 + 1/R_1)^{-1}. \tag{15}$$

can be computed, as well as

$$n_e = R_2 \, [(kT_e/p)(1 + R_1 + R_2)]^{-1} \tag{16}$$

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):

$$aI = \rho_v v[C_p(T - T_o) + q + v^2/2] + x_h \rho_s C_v (T - T_o)/\tau + \varepsilon\sigma T^4 + \phi(T, x_h), \tag{17}$$

where a 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 a 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 ε from the half-plane facing the laser, and 6) conduction through the thermal gradient in the heated layer with thickness x_h, ϕ (T, x_h). In Eq. (17), the vapor density is related to pressure p by

$$\rho_v = An_o m_p = \frac{Am_p p}{kT} \tag{18}$$

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

$$v = \left[\frac{\gamma(Z+1)kT}{Am_p} \right]^{1/2}, \tag{19}$$

$$q = q_f + q_v \tag{20}$$

contains the energies of fusion and vaporization,

and

$$x_h = x_{th} + x_v + 1/\alpha \geq \lambda., \tag{21}$$

is the effective thickness of the laser-heated, solid-density layer in the target during ablation. In Eq. (21), $x_{th} = (\kappa\tau)^{1/2}$, κ is thermal diffusivity, x_v is surface recession depth during the laser pulse and α is the optical absorption coefficient [cm^{-1}] at the surface, of the order of $1/\lambda$. For the $\{R_I\}$ 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 γ in Eq. (19) is

$$\gamma = C_p/C_v \tag{22}$$

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

$$\frac{\mathrm{k}}{C_\mathrm{p}} = \left(\frac{\gamma}{\gamma - 1}\right) A m_\mathrm{p}, \tag{23}$$

$$\rho_\mathrm{v} v\, C_\mathrm{p} T = \left(\frac{\gamma}{\gamma - 1}\right) p\, v, \tag{24}$$

and

$$\frac{v^2}{2 C_\mathrm{p} T} = \frac{\gamma - 1}{2} \tag{25}$$

in Eq. (17) and rearrange it in a more convenient form to give:

$$I = \frac{pv}{a}\left(\frac{\gamma}{\gamma - 1}\right)\left[1 - \frac{T_\mathrm{o}}{T} + \frac{q}{C_\mathrm{p} T} + \frac{\gamma - 1}{2}\right] + \frac{\sigma \varepsilon}{a} T^4 + B(\tau) \tag{26}$$

where

$$B(\tau) = \frac{1}{a}\left[\phi(T, x_\mathrm{h}) + \frac{x_\mathrm{h} \rho_\mathrm{s} C_\mathrm{v} (T - T_\mathrm{o})}{\tau}\right]. \tag{27}$$

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 $T \le 7890$ K, its triple point.

Eqs. (26) and (27) are wavelength-dependent only as λ affects the surface absorptivity a. For the infrared to ultraviolet range studied here, we used $0.05 \le a \le 0.24$ 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

$$C_\mathrm{mv} = p_\mathrm{v} / I. \tag{28}$$

Vapor specific impulse is

$$I_\mathrm{sp\ v} = v / g_\mathrm{o}. \tag{29}$$

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

$$\ln(p / p_1) = \left[\mathrm{C} \frac{\Delta H}{\mathrm{k}}\left(\frac{1}{T_1} - \frac{1}{T}\right)\right] \tag{30}$$

where the subscript "1" refers to a 1-bar reference condition, T is the vapor temperature, ΔH is the enthalpy of melting, vaporization and dissociation and C is a fitting constant. ΔH 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

$$C_\mathrm{m} = p/I = \left[\left(1 - \eta_\mathrm{i}\right) p_\mathrm{V} + \eta_\mathrm{i} p_\mathrm{P}\right] / I = (1 - \eta_\mathrm{i}) C_\mathrm{mv} + \eta_\mathrm{i} C_\mathrm{mp}. \tag{31}$$

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.

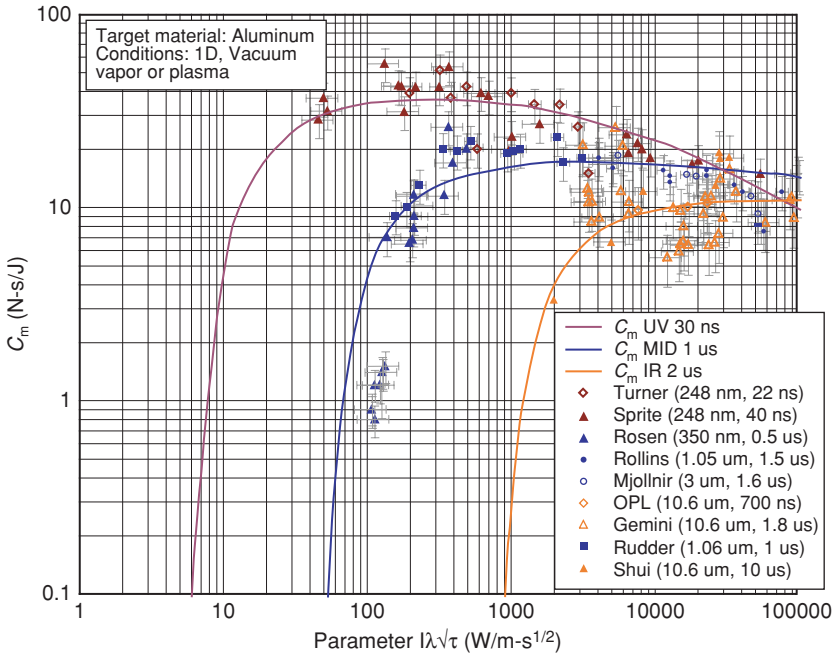

Figure 1. Fitting nine sets of mechanical coupling data from KrF to CO_2 laser wavelengths with our combined model. References are: Turner [6], Sprite [6], Rosen [14], Rollins [15], Mjøllnir [6], OPL [6], Gemini [6], Rudder [16] and Shui [15].

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 (λ = 248 nm) to CO_2 (λ = 10.6 μm) 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 $\{R_I\} - \{R_{II}\}$ transition in our parameter range is given by a fixed value of $I\sqrt{\tau}$

$$I\sqrt{\tau} = 4.8\text{E}8 \text{ Wm}^{-2}\text{s}^{1/2} \tag{32}$$

so that C_m data plotted vs. $I\lambda\sqrt{\tau}$ should show peaks distributed according to λ. This result agreed with our numerical modeling, which showed that Z and ηi, which in turn control the $\{R_I\} - \{R_{II}\}$ 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 $\{R_{II}\}$ theory and, if we are treating a model for the $\{R_I\} - \{R_{II}\}$ 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 τ 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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REFERENCES

[1] J. Sinko and C. Phipps, "Modeling CO_2 laser ablation impulse of polymers in vapor and plasma regimes," *Appl. Phys. Lett.,* 2009, 95, 131105-1 to 131105-3.

[2] C. Phipps, M. Birkan, W. Bohn, H.-A. Eckel, H. Horisawa, T. Lippert, M. Michaelis, Y. Rezunkov, A. Sasoh, W. Schall, S. Scharring and J. Sinko, "Review: Laser Ablation Propulsion," *J. Propulsion and Power,* in press (2010).

[3] C. Phipps and J. Luke, "Laser space propulsion – applications at two extremes of laser power," in *Laser Ablation and its Applications,* C. Phipps, ed., Springer Series in Optical Sciences, **129**, Springer, New York, 2007, 407–434.

[4] C. R. Phipps, H. Friedman, D. Gavel, J. Murray, G. Albrecht, E. V. George, C. Ho, W. Priedhorsky, M. M. Michaelis and J. P. Reilly, "ORION: Clearing near-Earth space debris using a 20-kW, 530-nm, Earth-based, repetitively pulsed laser", *Laser and Particle Beams*, 1996, 14 (1), 1–44.

[5] J. Lindl, "Development of the indirect-drive approach to inertial confinement fusion and the target physics basis for ignition and gain," *Phys. Plasmas,* 1995, 2, 3933.

[6] C. R. Phipps, Jr., T. P. Turner, R. F. Harrison, G. W. York, W. Z. Osborne, G. K. Anderson, X. F. Corlis, L. C. Haynes, H. S. Steele, K. C. Spicochi and T. R. King, "Impulse Coupling to Targets in Vacuum by KrF, HF and CO_2 Lasers", *J. Appl. Phys.,*1988, 64, 1083.

[7] R. E. Beverly III and C. T. Walters, "Measurement of CO_2-laser-induced shock pressures above and below LSD-wave thresholds," *J. Appl. Phys.,* 1976, 47(8), 3485–3495.

[8] M. Saha, "Ionization in the solar chromosphere," *Phil. Mag.* 1920, 40, 472.

[9] C. W. Allen, *Astrophysical Quantities*, 3rd edition, Athlone Press, London, 1973, 34.

[10] B. E. Poling, J. M. Prausnitz and J. P. O'Connell, *The Properties of Gases and Liquids*, 5th edition, McGraw-Hill, New York, 2001, 7.9-7.11.

[11] The SESAME equation-of-state database is maintained by group T-1 at Los Alamos National Laboratory (sesame@lanl.gov); see S.P. Lyon and J.D. Johnson, "SESAME: The Los Alamos National Laboratory Equation of State Database," LANL Report No. LA-UR-92-3407, 1992 for additional information.

[12] *Handbook of Optical Constants of Solids II*, Palik E.D. ed., Academic, New York, 1998, 388–91.

[13] O. Knacke, O. Kubaschewski and K. Hesselmann, eds., *Thermochemical Properties of Inorganic Substances* 2nd ed. Springer Verlag New York, 1991.

[14] D. I. Rosen, J. Mitteldorf, G. Kothandaraman, A. N. Pirri and E. R. Pugh, "Coupling of pulsed 0.35 μm laser radiation to aluminum alloys," *J. Appl. Phys.*, 1982, 63(4), 3190–3200.

[15] J. A. McKay and P. M. Laufer, "Survey of Laser-produced pressure and impulse data," Final Report, Contract N00014-86-C-2241, available as ADA197313 from Defense Technical Information Center, Fort Belvoir, VA 22060-6218, 1987.

[16] R. R. Rudder, U.S. Air Force Weapons Laboratory Report No. AFWL-TR-74-100 (1974), available as ADD703343 from Defense Technical Information Center, Fort Belvoir, VA 22060–6218, 1974.

[17] C. R. Phipps, H. Friedman, D. Gavel, J. Murray, G. Albrecht, E. V. George, C. Ho, W. Priedhorsky, M. M. Michaelis and J. P. Reilly, "ORION: Clearing near-Earth space debris using a 20-kW, 530-nm, Earth-based, repetitively pulsed laser", *Laser and Particle Beams*, 1996, 14 (1), 1–44.

[18] C. Phipps, J. Luke, D. Funk, D. Moore, J. Glownia and T. Lippert, "Laser Impulse Coupling at 130fs," *Appl. Surf. Sci.,* 2006, 252, 4838–4844.