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# Interaction of laser pulse with confined plasma during exit surface nanosecond laser damage

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## ABSTRACT

Interpretation of spatial and time resolved images of rear surface ns laser damage in dielectrics requires understanding of the dynamic interaction of the incoming laser beam with the confined expanding plasma in the material. The detailed kinetics of the plasma, involving both expansion and retraction, depends on details of reflection and absorption in the hot material. The growth of the hot region is treated using a model previously developed to understand laser peening. The pressure is found to scale as the square root of laser intensity and drops off slowly after energy deposition is complete. For the conditions of our experimental observations in fused silica, our model predicts a pressure of about 9 GPa and a surface expansion velocity of about 1.5 km/sec, in good agreement with experimental observation.

**Keywords:** Laser-induced damage, laser-induced breakdown, laser ablation

## 1. INTRODUCTION

The optical damage induced by a high-power ICF class laser systems is initiated by small defects localized within a few microns below the surface [1-3]. Recently, the processes accompanying the damage event were observed with high spatial and temporal resolution [4]. In the present paper, we relate a physical picture of the damage process to the experimental observations.

The experimentally observed evolution of the damage process is presented in Figs. (1-4). Initially, the absorption takes place at submicron defects positioned within a few microns below the surface. The runaway absorption and heating process produces a plasma confined by the surrounding material. The high plasma pressure, causes the material to bulge and eventually break, forming an outgoing shockwave in the air and ejecting a high velocity stream of particles. These features are clearly visualized in the experimental observations [4].

Local energy deposition during laser damage by nanosecond pulses produces high temperature and pressure plasmas, which ultimately determine the extent of damage. It is difficult to measure the pressure and temperature directly. In the present paper, we try to correlate the experimental result with the simple theoretical model developed and verified for laser peening to get insight regarding nature of the damage.

## 2. LASER INTERACTION WITH PLASMA IN CONFINED GEOMETRY

For exit surface laser damage, the location of the initial laser energy deposition is typically determined by a defect structure that is embedded in the near surface layer, located at a depth  $d$ , initiating plasma formation and an absorption front that moves toward the laser (into the bulk). Specifically, we will discuss experiments [4], where damage was induced by 8 nsec pulses of 355 nm light with fluence  $\sim 50$  J/cm<sup>2</sup>. The calculations demonstrate almost complete absorption of laser radiation by plasma for 355 nm laser light [5]. We approximate the motion of the material and the associated pressure fronts along the axial direction (normal to the surface) as initially one-dimensional when the laser spot size is much larger than the absorption length, which is the case in our experiments. This situation, physically, is similar to that in laser peening experiments [6,7], a material process in which a laser pulse propagates through a thin layer of water to produce a high pressure confined plasma at the water-metal interface which hardens the metal. A simple physical model [6,7] was developed and experimentally verified to describe the laser peening process and we use this model below. The typical laser intensity and pulse duration in the present work are similar to those of laser-peening experiments thus further justifying the use of the model.

The model [6,7] considers plasma as an ideal gas whose thermal energy  $E_T$  is a constant fraction  $\alpha$  of the plasma internal energy  $E_i$ . Experimentally, the value  $\alpha$  is determined to be about 0.25. For the plasma pressure, assuming the flat in time temporal pulse, the model gives

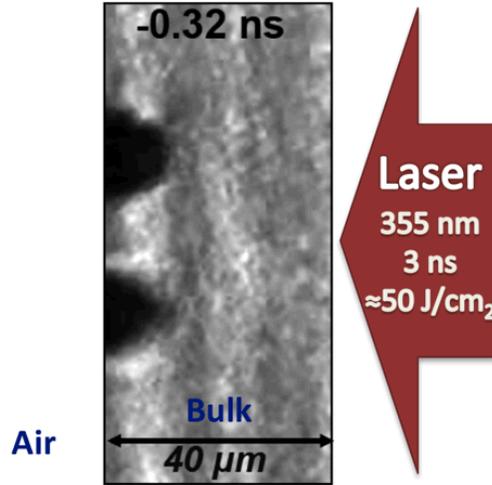


Figure 1. Shadowgraph microscopic image of plasma propagating through bulk back toward the laser beam

$$P(\text{kbar}) = 0.1 I^{1/2} (\text{GW} / \text{cm}^2) \left( \frac{\alpha}{3} \right)^{1/2} Z^{1/2} (\text{g} / \text{cm}^2 \text{s}) \quad (1)$$

Here  $Z = \rho u$ , the material impedance,  $\rho$  is the mass density and  $u$  is the speed of sound. The results are not sensitive to the pulse shape. For fused silica  $Z \approx 1.3 \cdot 10^6 \text{ g/sec cm}^2$ . For our experiments, the laser intensity is  $I \sim 7 \text{ GW/cm}^2$ ; using  $\alpha = 0.25$ , we estimate a pressure  $P \sim 8.5 \text{ GPa}$ . For known pressure and using the relation  $P = knT$  one can estimate the plasma temperature. Specifically, assuming a density  $6 \cdot 10^{22} \text{ cm}^{-3}$  and the above pressure, the plasma temperature is about 0.9 eV.

For the plasma expansion toward the beam, the model predicts a velocity  $v$

$$v \left( \frac{\mu\text{m}}{\text{nsec}} \right) = \frac{2 \cdot 10^4}{Z \left( \frac{\text{g}}{\text{cm}^2 \text{sec}} \right)} P(\text{kbars})$$

The speed of the plasma zone expansion (dark area in our experiments shown in Figure 1 for the above parameters is estimated to be about  $1.3 \mu\text{m/ns}$ . This value corresponds to the axial speed of expansion of the modified region in our experiments and is only slightly lower than that observed in the experiments [4].

The pressure on the thin layer between the plasma and free surface causes the bulging up to about 25 ns delay and break up and particle ejection at delays longer than about 35 ns as shown in Fig 2. The velocity  $V$  of this layer with thickness  $d$  can be expressed as

$$V(t) = \frac{\int P(t) dt}{\rho d} \quad (2)$$

When the layer starts to move, the plasma expands, and the pressure drops from the value given by Eq(1) and the acceleration stops. This moment is determined by the time needed by the rarefaction wave reflected by the free surface to return to the plasma region. This suggests that we can use  $2d/u$  (where  $u$  is the speed of sound) instead of the pressure pulse duration in Eq. (2) to estimate the maximal velocity, which can be written as

$$V \sim \frac{2P}{\rho u} \quad (3)$$

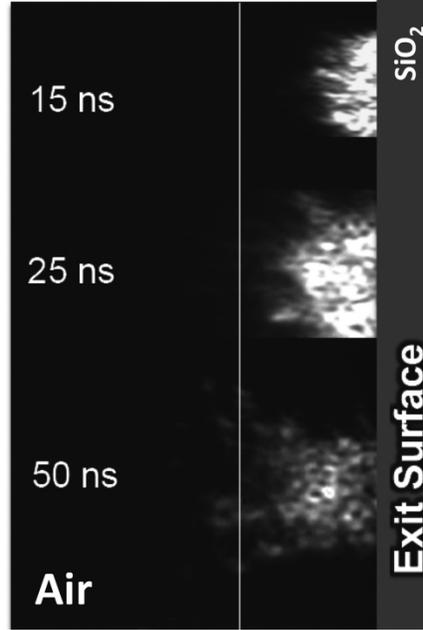


Figure 2.:Backscattering microscopic image of surface bulging arising from increased pressure of the laser induced plasma in SiO<sub>2</sub> surface. The delayed ejection of the first particles is observed in the 50 ns delay image.

Note that the velocity given by Eq. (3) is independent of the thickness  $d$  of the initially absorbing layer. Substituting  $P$  from (1) we find

$$V = 0.2 I^{1/2} (GW / cm^2) \left( \frac{\alpha}{3} \right)^{1/2} Z^{-1/2} (g / cm^2 s) \quad (4)$$

As the exact profile of the pressure pulse is not known, Eq. (4) provides an estimate of the swelling velocity at early times. This velocity will continue to increase with time after the break up, but this additional increase will be a fraction of this estimated velocity at early times. Using  $P \sim 8.5$  GPa, the swelling velocity is estimated to be  $V \sim 1.45$  km/sec for fused silica samples. This is only slightly lower than  $\sim 1.6$  km/s that was observed in the experiments [4].

Another important result of the experiments and simulations of laser peening discussed in ref. [6,7] is that the pressure pulse duration is 2-3 times longer than the laser pulse duration due to confinement. For the 8 ns pulse used in this specific set of experiments [8], the above suggests that the temporal duration of the pressure wave is on the order of 25 ns. After that the plasma pressure drops. The internal pressure produces hoop stresses inducing crack growth. The termination of pressure arrests crack growth. This appears to be in agreement with the termination of the generation and/or expansion of the circumferential cracks as well as the termination of the surface swelling in the experiments [4]. It is therefore possible that the underlying mechanism for the processes associated with these experimental observations is the termination of the residual pressure wave following laser energy deposition.

### 3. SHOCK AND EJECTA EXPANSION

Bulging and then high velocity ejection of material creates a shock in the surrounding air. The position of the shock front in air is clearly visible in shadowgraphic microscopy due to the density jump at the interface which causes a localized change of the index of refraction. The ejection of particles having diameter on the order of 1  $\mu$ m or larger is also visualized with shadowgraphic microscopy. However, the observation of the ensuing gaseous material arising from the evaporation of the affected material exposed to temperatures well above the boiling is much more difficult and not visible for most dielectric optical materials.

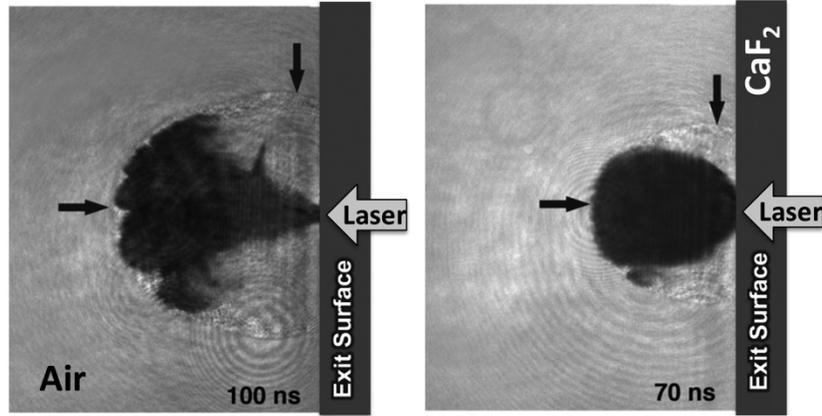


Figure 3. Visualization of the gaseous material jet ejection during exit surface damage in  $\text{CaF}_2$ .

The point explosion (blast) theory is good for description of this aspect of the experiment [9]. It relates the shock radius  $R$  to the released energy  $E$  by the relation  $R = \left(\frac{2.4E}{\rho_0}\right)^{1/5} t^{2/5}$ , where  $\rho_0$  is air density. Experimentally the extent of the damage zone where the energy was deposited was  $a \sim 10 \mu\text{m}$ . For 3w light, the plasma absorbs almost all of the incident energy and  $E \sim a^2/S$  [ $S$  is the beam area] of total laser pulse energy  $\sim 0.1$  mJ. The radius of the shock, calculated when the ejecta stops is about  $800 \mu\text{m}$ , consistent with experiment. The time to stop is  $\sim R/s$  [10] where  $s$  is the sound speed in air  $\sim 300$  m/sec so the time to stop  $\sim 2.4 \mu\text{sec}$  consistent with experimental data.

Visualization of ejected non-ionized gas is a difficult problem. Our multi-material investigation revealed that  $\text{CaF}_2$  produces a highly visible gaseous material [8]. These experiments were performed using 1064 nm laser pulses, 10 ns in duration at FWHM, with a peak fluence on the order of  $1 \text{ KJ}/\text{cm}^2$ . Typical experimental results are shown in Fig. 3. These results allow measurement of the kinetics and comparison with theory. After the pulse termination, we have a dense, high pressure cloud expanding into the surrounding gas with much lower pressure, practically vacuum. The maximal velocity of gas  $u$  expanding into the vacuum, the boundary velocity, is given by the expression [10]  $u = \frac{2}{\gamma-1} s = \frac{2}{\gamma-1} \sqrt{\frac{\gamma P}{\rho}}$ . Using pressure value derived above, solid state density for  $\text{CaF}_2$   $\rho = 3.2 \text{ g}/\text{cm}^3$  and the adiabatic constant for a monatomic gas  $\gamma = 5/3$  we find velocity  $u \sim 7 \text{ km}/\text{sec}$ . Ionization increases the number of degrees of freedom, which alters the adiabatic index closer to unity, thus increasing the expansion velocity. This result is consistent with the experimental value  $u \sim 10 \text{ km}/\text{sec}$ . [8]

The surrounding air decelerates the expansion. In this situation, the contact surface between the ejecta and ambient gas is unstable similar to the Rayleigh-Taylor instability [11]. The instability of the contact surface is clearly visible in Fig.3. We observed that the instability saturates at a low level of perturbations and does not produce noticeable mixing, probably, due to the compressibility effects and energy depletion.

The localized jet ejected from the damage spot is subject to the Kelvin-Helmholtz (tangential jump) instability on the jet boundary. The instability results in mixing and rapid particle density depletion on the jet periphery. This may explain the visible narrowing of the jet one sees in Fig.3. The vortices formed as a result of the instability development present an efficient mechanism for particle return to the surface and re-deposition [12]

Ejection of particles and slow moving flakes well after laser pulse termination are seen in Fig.4. One mechanism of particle formation is condensation in the cooling down ejecta [10]. For conventional ablation, condensation usually forms submicron clusters [13]. In our case, due to the plasma confinement, the expanding plasma is much denser and produces clusters of micron size.

At the very late stage of damage, we observed ejection of slow moving flakes. A likely explanation for this is related to post pulse thermoelastic material evolution. The deposited laser energy produces hot material in the damage spot and its cooling time is long. At  $2 \mu\text{sec}$  time, the last frame in Fig.4, the heat diffusion length is less than  $2 \mu\text{m}$ . Gradual material cooling eliminates evaporation, the material becomes more brittle. Residual thermal stresses can result in flakes chipping out as visible in Fig.4

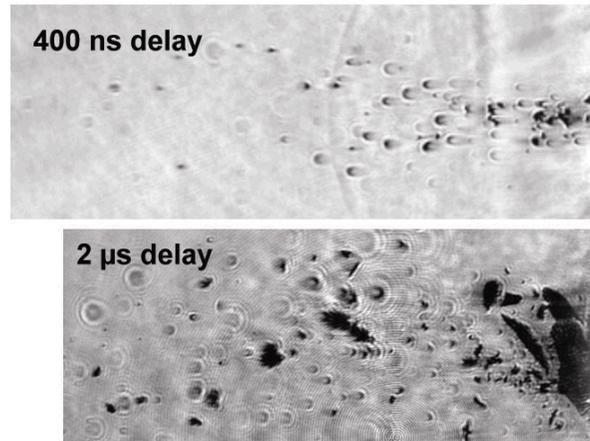


Figure 4. Particles and flakes ejection associated with SiO<sub>2</sub> exit-surface damage.

#### 4. SUMMARY

The effect of plasma confinement is important for the understanding of complex phenomena associated with laser damage of transparent dielectrics. We demonstrated that the model of confined plasma interaction with laser light developed for laser peening provides reasonable estimates for the plasma pressure and temperature, which are difficult to measure directly. From these results, we estimated the velocity of the ejected particles, the velocities of ejecta and found good agreement between model predictions and experimental data.

#### 5. ACKNOWLEDGEMENTS

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#### REFERENCES

- 1 S. G. Demos, M. Staggs, "Application of fluorescence microscopy for noninvasive detection of surface contamination and precursors to laser-induced damage" *Appl. Opt.*, **41**, 1977-1983, 2002
- 2 L. Hongjie, H. Jin, W. Fengrui, Z. Xinda, Y. Xin, Z. Xiaoyan, S. Laixi, J. Xiaodong, S. Zhan, Z. Wanguo, "Subsurface defects of fused silica optics and laser induced damage at 351 nm" *Opt. Express*, **21**, 12204 (2013)
- 3 B. Bertussi, JY Natoli, M. Commandre, "High-resolution photothermal microscope: a sensitive tool for the detection of isolated absorbing defects in optical coatings", *Appl. Opt.*, **45**, 1410-(2006)
- 4 S. G. Demos, R. A. Negres, R. N. Raman, A. M. Rubenchik, M. D. Feit, "Material response during nanosecond laser induced breakdown inside of the exit surface of fused silica", *Laser and Photonics Reviews*, **7**, 444-452, 2013
- 5 H. Maezawa and H. Miyauchi, Rigorous expressions for the Fresnel equations at interfaces between absorbing media, *J. Opt. Soc. Am. A* **26**, 330 (2009).
- 6 L. Berthe, R. Fabbro, P. Peyre, L. Toller and E. Bartinski, "Shock waves from a water-confined laser-generated plasma", *J. Appl. Phys.* **82**, 2826, 1997
- 7 R. Fabbro, J. Fournier, P. Ballard, D. Devaux, J. Virmont, "Physical study of laser-produced plasma in confined geometry", *J. Appl. Phys.* **68**, 775, 1990.
- 8 Stavros G. Demos, Raluca A. Negres, Alexander M. Rubenchik, "Dynamics of gaseous material ejection following laser breakdown on the surface of CaF<sub>2</sub>", Submitted for publication
- 9 A. Salleo et al Energy deposition at front and rear surfaces during the picosecond laser interaction with fused silica. *Appl. Phys. Lett.* **78**.2840.2001
- 10 Ya. B. Zeldovich, Yu. P. Raizer, "Physics of Shock Waves and High Temperature Hydrodynamic Phenomena", Academic Press, New York, 1967
- 11 S.I. Anisimov, V.A. Khokhlov *Instabilities in laser-matter interaction* CRC Press 2000
- 12 R. N. Raman, S. Elhadj, R. A. Negres, M. J. Matthews, M. D. Feit, and S. G. Demos, Characterization of ejected fused silica particles following surface breakdown with nanosecond pulses, *Opt. Expr.*, **20**, 174194, 2012
- 13 D. Bauerle, "Laser Processing and Chemistry", Springer, Berlin, 2000