A thermal model of phase explosion for high power laser ablation

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ABSTRACT

Although laser ablation of solid materials is finding applications in a growing number of fields, the basic mechanisms underlying laser ablation processes have not been fully understood. One fundamental parameter for high-power laser ablation applications is the ablation depth resulting from the interaction of individual laser pulses. The ablation depth for laser ablation of single-crystal silicon shows a dramatic increase at a laser intensity threshold of approximately 20 GW/cm^2. Above this threshold, micron-sized particulates have been observed to eject from the target surface. We present an analysis of this threshold phenomenon and demonstrate that thermal diffusion and subsequent explosive boiling *after* the completion of laser irradiation is a possible mechanism to describe the observed dramatic increase of the ablation depth. Calculations based on this delayed phase explosion model provide a satisfactory estimate of the measurements. In addition, we find that the shielding of an expanding mass plasma during laser irradiation plays an important role on this threshold phenomenon.

Keywords: laser ablation, phase explosion, plasma, theoretical model.

1. INTRODUCTION

High power laser ablation is finding an increasing number of applications, such as deposition of metal and dielectric films, and laser ablation chemical analysis[1]. Nevertheless, the fundamental mechanisms underlying laser ablation processes are not fully understood, especially when high-power laser pulses are utilized and superheating of target material occurs. It has been suggested that when the laser irradiance is sufficiently high, explosive boiling[2] is involved such that homogeneous bubble nucleation occurs when the target material reaches ~ 0.9 T_{tc} (T_{tc} is the thermodynamic critical temperature). As a consequence, the target material makes an abrupt transformation from superheated liquid into a mixture of liquid droplets and vapor, which are then violently ejected from the target.

Previously, we measured the mass ablation[3,4] from polished single-crystal silicon with single pulse laser irradiance ~ 10^9 – 10^{10} W/cm^2. A Nd:YAG laser with 266 nm wavelength and 3 ns pulse duration was focused to approximately ~ 35 μm diameter spot on the silicon target. The data showed that the ablation depth increased dramatically at the laser irradiance threshold of about 2 x 10^{10} W/cm^2 (figure 1). In measuring the ablation crater depth, a Zygo NewView-200 surface structure analyzing system was employed. The system uses scanning white light interferometry to image and measure the microstructures and topography of targets in three dimensions. Below the threshold, the ablation depth increased gradually from 0.6 μm to 1.5 μm as the laser irradiance increased from 3.0 x 10^9 W/cm^2 to 2 x 10^{10} W/cm^2. At the laser irradiance threshold of 2 x 10^{10} W/cm^2, the ablation depth abruptly increased from 1.5 μm to 6.3 μm, and reached 20 μm at 1.5 x 10^{11} W/cm^2. Below and above the threshold, a shock wave, which lasts about several tens of ns after the laser pulse, was formed due to the pressure difference between a dense plasma plume and the ambient. When the laser irradiance exceeds the threshold, there are large size particulates ejected about 300-400 ns after the development of the shock wave. More details about the experimental system and results can be found in reference 3 and 4. Similar results have been reported by other groups[5,6] using different laser irradiances and pulse duration.

The abrupt increase of the ablation depth at the threshold of 2 x 10^{10} W/cm^2 was speculated to result from explosive boiling *during* nanosecond laser irradiation, as a laser-induced transparent layer could form when the temperature approached the critical temperature. However, such a transparent layer during pulsed laser ablation of solid materials was never verified by experiments. In this article, we demonstrate that thermal diffusion and subsequent explosive boiling *after* the completion of laser irradiation may be a primary source of the measured threshold phenomenon. Calculations of the ablation depth based on a proposed delayed explosive boiling model will be presented. In contrast to previous theoretical investigations of laser-induced phase explosion, we included in the calculation the effect of an expanding mass plasma during high power laser irradiation of the target.

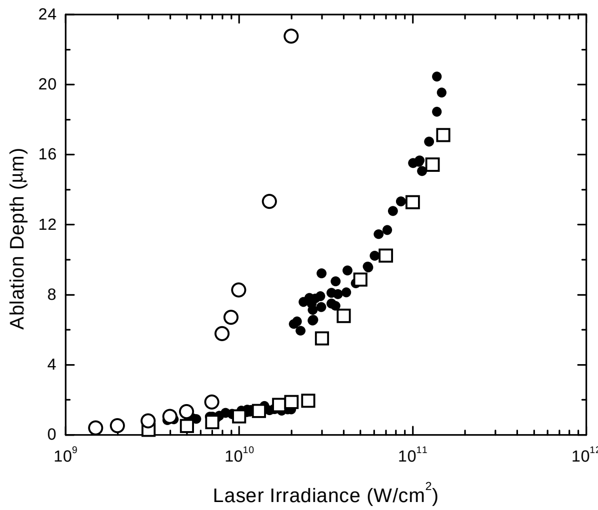

Fig. 1 Ablation depths (●: experimental measurements, □: computational data with plasma shielding, ○: computational data without plasma shielding).

2. THE THEORY

The theory of explosive boiling may be considered from either a thermodynamic or kinetic point of view. The former provides a rigorous method to predict the thermodynamic critical temperature, while the latter mechanism models the rate of formation of vapor bubble growth at any temperature. According to thermodynamic theory of explosive boiling[7], the liquid begins to be superheated and becomes metastable when it exceeds a temperature limitation of about $0.8T_{tc}$. Above this temperature, homogeneous bubble nucleation may occur and the "liquid" is essentially a mixture of liquid droplets and vapor that can facilitate explosive boiling. It has been argued that explosive boiling may be a dominant mechanism during the interaction of high-power laser and materials, especially when the laser pulse is sufficiently short (e.g., shorter than a few hundreds of nanoseconds[8,9]).

Although explosive boiling may be an inevitable process when the liquid is superheated, there are limitations according to kinetic theory[10,11]. When the liquid is superheated, homogeneous bubble nucleation occurs and the liquid experiences large density fluctuation. Only if these bubbles reach a critical radius r_c, will they grow spontaneously. Bubbles with radius less than r_c are likely to collapse, and it takes the bubble a time of τ_c to grow to the critical radius. The expression of r_c and τ_c are[12]:

$$r_c = \frac{2\sigma}{p_{sat}(T_l)\exp\{v_l[p_l - p_{sat}(T_l)]/R_v T_l\} - p_l} \quad , \qquad (1)$$

$$\tau_c = r_c\{\frac{2}{3}[\frac{T_l - T_{sat}(p_l)}{T_{sat}(p_l)}]\frac{L_{ev}\rho_v}{\rho_l}\}^{-\frac{1}{2}} \quad , \qquad (2)$$

where σ is the surface tension of the liquid, L_{ev} and R_v are latent heat of vaporization and gas constant respectively. ρ_l and ρ_v are the densities of superheated liquid and vapor, with $v_l = 1/\rho_l$. T_l is the temperature of the superheated liquid, which can be taken as $0.85T_{tc}$ when explosive boiling occurs. Using a method suggested by Martynyuk[7], we calculate that the thermodynamic critical temperature of silicon is approximately 5200K. p_{sat} and T_{sat} are the saturation pressure and temperature at the superheated liquid temperature, which can be obtained from the Clausius-

Clayperon relation. P_l is the pressure of the superheated liquid, and can be approximated by the recoil pressure of the evaporating vapor, which is $0.54p_{sat}(T_l)$ [13]. According to the power law relation of surface tension σ for liquid metal[14], the surface tension drops about 80% at the assumed T_l. Using these parameters, we estimate r_c and τ_c to be approximately 0.6 μm and 70 ns, respectively.

These calculations indicate that it would take bubbles about 70 ns to grow to the critical radius of 0.6 μm. Subsequently, the superheated liquid will undergo a transition into a mixture of vapor and liquid droplets, followed by explosive boiling of the liquid-vapor mix. However, our laser pulse duration is only 3 ns; the bubble doesn't have enough time to reach the critical radius during the laser pulse. As a result, without efficient energy dissipation, the liquid temperature can exceed the critical temperature if the laser irradiance is sufficiently high. The value of τ_c is consistent with our experimental results; in our experiments, detectable micron-sized droplet ejection occurred 300 ~ 400 ns after the completion of the laser irradiation.

The thermal penetration depth $x_{th} = 0.969[(k\tau)^{1/2}]$ during a laser pulse of duration τ is much larger than the optical (266 nm) penetration $1/\alpha$ in our case[15]. In the expression for x_{th}, k is the thermal diffusivity of the liquid silicon, which is about 0.75 cm^2/s, and α is the absorption coefficient. The thermal penetration depth is calculated to be about 0.47 μm. The critical diameter of the bubble is $d_c = 2r_c$, or 1.2 μm, which is larger than the thermal penetration depth; the bubble cannot grow to its critical radius during the laser pulse. Experimental evidence suggests that explosive boiling occur only if the superheated layer is thick enough[16].

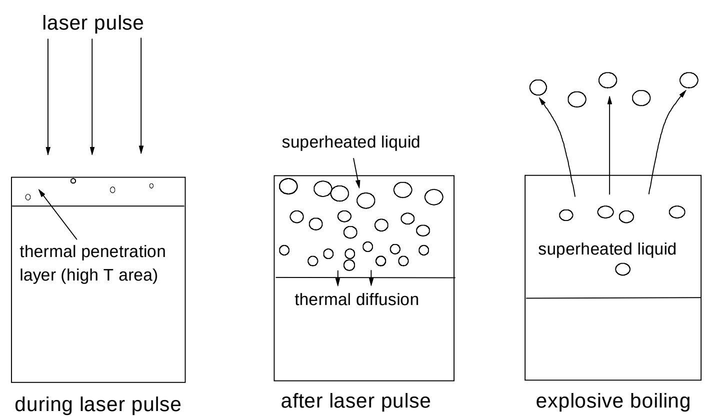

Fig. 2 The kinetic process of laser ablation - explosive boiling.

The rate of homogeneous nucleation is governed by,

$$I_n \approx 1.5\times10^{32}\exp(-\Delta G_n / k_B T)\exp(-\tau_{hn}/t) \quad \text{nuclei/cm}^3\text{s}. \tag{3}$$

Here ΔG_n is the free energy for formation of a stable homogeneous nucleus and τ_{hn} is the relevant time constant[17]. Martynyuk argued that I_n is numerically significant (i.e., $I_n > 1$ nucleus/cm^{3}s) only near T_{tc}. As an example, the value for metal Cs is, $I_n = 1$ nucleus/cm^{3}s at $T = 0.874T_{tc}$ and $I_n = 10^{26}$ nuclei/cm^{3}s at $T = 0.905T_{tc}$. The number of homogeneous nuclei which would be generated during the laser pulse is $I_n V^* \tau$, where V^* is the heated volume during the laser pulse, and $V^* = \pi x_{th} D^2_{laser}/4$. D_{laser} is the width of the laser pulse spot size (35 μm in our experiments), so $V^* = 4.5 \times 10^{-10}$ cm^3. If we take $I_n = 10^{26}$ nuclei/cm^{3}s and $\tau_{hn} = 50$ ns, the homogeneous nuclei generated during the laser pulse equals 5. Therefore, we cannot expect that explosive boiling will occur for such a low generation ratio of nuclei.

From the above analysis, we have shown that there are very few bubbles generated near the surface of the target *during* the 3ns laser pulse, and the bubbles do not have enough time to grow to a critical size. As a consequence, explosive boiling will not occur during the laser pulse. However, without significant bubble formation, a high temperature layer will form below the target surface during the laser pulse with a depth equal to about the thermal penetration depth. At the same time, the target undergoes normal vaporization from the extreme outer surface. Mass ablation below the laser irradiance threshold (2 x 10^{10} W/cm^2) is generated by this normal vaporization mechanism. The vaporization flux is governed by the Hertz-Knudsen equations, and the velocity of the surface recession can be calculated as[2],

$$\left.\frac{\partial x}{\partial t}\right|_{x=0} = \beta p_b \frac{m}{\rho}(2\pi m k_B T)^{-1/2} \exp[\frac{L_{ev} m}{k_B}(\frac{1}{T_b} - \frac{1}{T})] \quad \text{cm/s.} \tag{4}$$

Here, β is the vaporization coefficient, p_b is the boiling pressure (normally similar to 0.1 MPa), and T_b is the boiling temperature. At high laser irradiance, after the laser pulse is completed, the high temperature liquid layer will propagate into the target with thermal diffusion. Part of the liquid layer in the target may approach the critical temperature and therefore, new bubbles will emerge inside the superheated liquid, eventually leaving the target (figure 2).

3. NUMERICAL RESULTS

A numerical model based on the above diffusion-phase explosion mechanism has been established to estimate the depth of laser ablation. Using the heat conduction equation, the temperature distribution was calculated according to,

$$\rho C \frac{\partial T}{\partial t} = \frac{\partial}{\partial x}(k \frac{\partial T}{\partial x}) + \alpha I_{laser} \exp(-\alpha x) \ , \tag{5}$$

where T is temperature, C is specific heat, and I_{laser} is the laser irradiance that reaches the surface of the silicon target. We include in the model the absorption of laser-generated vapor plasma from the target surface. Such a plasma has been frequently observed during high power laser ablation of solids. However, it has previously not been included for modeling laser ablation in the explosive boiling regime.

The laser irradiance at the target surface I_{laser} can be written as:

$$I_{laser} = I_0(t)\exp(-Hk_1) \ , \tag{6}$$

where I_0 is the laser irradiance, and H is the thickness of the plasma. k_1 is the absorption coefficient of the plasma; in this model only the inverse Bremsstrahlung process is considered. The details of the model for plasma shielding can be found in reference 18, and its validity has been confirmed by comparison with experiments[19].

For solving equation (5), boundary conditions are required at x = 0 and L, where L is the length of the computational domain. At $x = L$, the temperature of the material is assumed to be unaffected by the laser irradiation, i.e. $T(L, t > 0) = T_0$, T_0 is the initial temperature of the solid. For x = 0 boundary condition, we use a method recommended by Miotello and Kelly[2], including energy loss due to evaporating vapor.

The ablation depth due to evaporation was calculated by integrating equation (4). During the laser pulse, a high temperature layer is formed at and beneath the surface of the target; this layer subsequently propagates into the target by thermal diffusion. We regard the liquid whose temperature is larger than $0.85T_{tc}$ as superheated liquid, and in such a metastable state, homogeneous bubble nucleation will take place.

Ablation for laser irradiances below threshold is governed by normal evaporation according to equation (4). Ablation for irradiances larger than the threshold is caused by both normal evaporation and explosive boiling. The calculated ablation depths based on the present delayed phase explosion model are compared with experimental data in figure 1. The model predicts that the laser irradiance threshold for explosive boiling is about 3 x 10^{10} W/cm^2, in close agreement with experimental findings. In our model, the superheated liquid reaches its maximum depth several hundred nanoseconds after the laser pulse is completed, which also agrees with experimental results.

The computational ablation depths without plasma shielding are also given in figure 1. Plasma shielding plays an important role in determining the laser irradiance threshold for explosive boiling. The effect of plasma shielding can be illustrated by plotting the transmitted laser temporal profile through the plasma (figure 3). When the laser irradiance is relatively low, the laser pulse retains its original profile with little attenuation by the plasma.

However, when the laser irradiance is larger than 2 x 10^{10} W/cm^2, the trailing part of the laser pulse is truncated, as clearly seen in figure 3 (c and d).

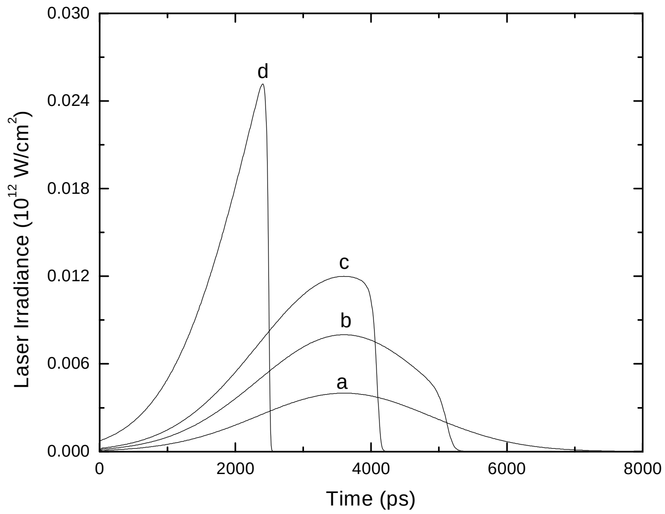

Fig. 3 Temporal profiles of laser intensities on target surface for different initial peak laser irradiances, I_{peak}, before the interaction with a mass plasma. The values of I_{peak} are, **a**: 10^{10}, **b**: 2 x 10^{10}, **c**: 3 x 10^{10}, and **d**: 1 x 10^{11} W/cm^2.

4. CONCLUSIONS

To conclude, we have analyzed the dramatic ablation depth growth during high-power UV nanosecond laser ablation of silicon. We developed a model to describe this threshold phenomenon and demonstrated that thermal diffusion and subsequent explosive boiling *after* the completion of laser irradiation could be a potential mechanism. Plasma shielding during laser irradiation was found to have a significant effect on this threshold phenomenon, and our calculations provide a satisfactory estimate of the experimental results. The model developed here should be applicable for a broad range of pulse duration, and we are working both experimentally and theoretically along this direction.

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