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ON THE STABILITY OF METAL EVAPORATION PRODUCED BY AN ULTRASHORT LASER PULSE

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The growth of corrugation perturbations of a plane phase boundary between a metal and its vapor is considered in the case of nonstationary metal evaporation caused by ultrashort laser pulse absorption. It is shown, that the number of e-foldings of the perturbation is less than unity, i.e., the phase boundary is stable. This result is in agreement with recent experiments on picosecond metal ablation.

Pulsed laser ablation is widely used as a tool for surface processing. Most of the laser ablation experiments are performed using nanosecond laser pulses. Recently, however, picosecond and subpicosecond pulses have received much attention. Such pulses make possible to diminish the thermal effects which limit the resolution and quality of the laser processing [1]. It will be shown in the present paper, that further advantage of ultrashort laser pulses is the absence of corrugation instability of the ablation front. This instability is characteristic for the surface vaporization of solids (or liquids) caused by volume energy absorption [2].

A laser beam, incident on a metal surface, is absorbed mainly by electrons through inverse Bremsstrahlung. The laser energy is released in the skin layer whose thickness is of the order of $10^{-5}-10^{-6}$ cm. To produce surface evaporation, the energy absorbed must be transferred from hot skin layer electrons to the lattice at the metal surface. This implies, that the temperature gradient at the surface is directed towards the condensed phase, i.e., the vaporization front moves in the direction of temperature gradient. In this situation, a plane stationary vaporization front appears to be unstable. Corrugation perturbations of a plane phase boundary exponentially grow, if the laser intensity exceeds a certain threshold [2].

Usually, the stationary evaporation regime is not reached in ultrashort pulse experiments. Moreover, for picosecond and subpicosecond pulses, lattice heating and evaporation take place, when the laser pulse was completed, hence, the stability conditions should not depend on the laser pulse shape. In what follows we will show that over a wide range of parameters, the plane evaporation front is stable in this case.

The two-temperature model is employed to describe metal vaporization. The energy exchange between electrons and the lattice is described by equations [3]

$$c_i \frac{\partial T_i}{\partial t} = -c_e \frac{\partial T_e}{\partial t} = \alpha (T_e - T_i) \ , \qquad (1)$$

where T_e, T_i, $c_e = \beta T_e$, c_i are the temperatures and specific heats (per unit volume) of the electrons and lattice, respectively, and α is the energy exchange rate between the two subsystems. According to [4],

$$\alpha = \frac{3\hbar\lambda < \omega^2 > \beta}{\pi k_B}$$

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where λ is the electron-phonon coupling constant and $<\omega^2>$ is the second moment of the phonon spectrum. Equations (1) define two characteristic times: $\tau_i = c_i/\alpha \sim 10^{-10} - 10^{-12}$ s is the time of lattice heating , and $\tau_e = c_e/\alpha \sim 10^{-12} - 10^{-14}$ s is the time of electron cooling due to the electron–lattice energy exchange. If the laser pulse length τ_l is smaller than τ_i, the lattice remains "cold" during the laser pulse, while electrons reach the temperature of the order of $T_e \approx \sqrt{Q\mu/\beta}$, where Q is the laser fluence absorbed and μ is the absorption coefficient (inverse skin layer thickness). During the time of the order of τ_i, the equilibrium between the lattice and electrons is established, and both temperatures become on the order of $T_e \approx T_i \approx Q/\delta c_i$. Here $\delta \approx \max\left[1/\mu, \sqrt{\kappa_0 c_i/\alpha\beta T_i}\right]$ is the thickness of the heated surface layer where κ_0 is the heat conductivity of equilibrium metal. Numerical calculation [5] gives for silver target and picosecond laser pulse the value of $\delta \sim 10^{-5}$ cm.

It is clear that, in the time scale of the lattice heating, τ_i, the evaporation may be neglected. Thus, to estimate the mass evaporated, we can solve the heat conduction equation with $T_i = T_e = T$ and standard boundary conditions at the evaporation front [6]. Taking the first two moments of the heat conduction equation and assuming the initial temperature and initial heated layer thickness to be equal to $T_0 = Q/\delta c_i$ and δ, respectively, we obtain the following estimate for the mass of evaporated material (per unit area):

$$\Delta M = \frac{Q}{L}\frac{\phi}{1+\phi}\ , \quad \phi = \frac{Q V_0}{3U\kappa_0}\left(\frac{U}{T_0}+3.2\right)\exp\left(-\frac{U}{T_0}\right)\ , \tag{2}$$

where L is the latent heat of vaporization, and U and V_0 are the constants in the equation for the vaporization front velocity [6]: $V = V_0 \exp(-U/T)$. It is seen from (2) that the evaporated mass is proportional to a small factor $\phi \ll 1$. Thus, the vaporization produced by ultrashort laser pulse absorption is energetically non-efficient. This is caused by the strong temperature dependence of the vaporization front velocity. The evaporation takes place only during a short initial period of time, and then stops due to surface cooling. Note that the cooling of the hot surface layer is governed for the most part by heat conduction. Evaporation energy loss is, usually, less important.

Consider now the stability of the evaporation front. Since the front is non-stationary, the results obtained in [2] can not be directly applied to the stability analysis. We can use, however, the WKB-approximation, as it was suggested in [7], assuming that the instability growth rate is a slowly varying function of time. Thus, we consider two types of evaporation front motion: one-dimensional motion, associated with "slow" changes of temperature and velocity of the plane evaporation front, and two-dimensional motion, associated with "fast" perturbation growth. Under this condition we can introduce small perturbations to the temperature field and to the phase boundary position in the form

$$\delta T = f(\zeta)\exp\left(iky + \int_0^t \gamma\, dt\right) \qquad \delta Z = a\exp\left(iky + \int_0^t \gamma\, dt\right) \tag{3}$$

$$\zeta = z - \int_0^t V(t)\, dt \qquad \delta Z = Z(y,t) - \int_0^t V(t)\, dt$$

where k is the perturbation wavenumber, and the integration is performed over the "slow" time. Substituting (3) into the heat conduction equation and boundary conditions, and performing standard calculations (similar to those carried out in [2, 8]), we arrive at an eigenvalue problem for the perturbation growth rate $\gamma(k,t)$. The dispersion equation can be written in much the same way as in [2,7,8]. Examples of the dispersion curves for constant laser intensity can be found in [7].

It is conventional to consider the inequality $\mathrm{Re}\,\gamma > 0$ as the instability condition. For present purposes, this condition is inadequate because γ, generally, changes its sign with time. It is reasonable to characterize the growth of perturbations by the number of e-foldings

$$\Gamma(k) = \int_0^\infty \gamma(k,t)\,dt \ , \tag{4}$$

and assume the stability condition in the form: $\Gamma = \max[\Gamma(k)] \le C$, where C is a constant of the order of unity. Note that a major contribution to the integral (4) results from large values of $\gamma(k,t)$ which can be calculated by the WKB-method with a high degree of accuracy. As shown in [2,7,8], the growth rate γ as a function of k reaches its maximum at $k = k_m \sim \left(\frac{V(\rho L)^2}{\chi\sigma Q}\right)^{1/3}$. The maximum value of γ can be estimated as

$$\gamma_m \approx \frac{1}{3}\frac{V^2}{\chi}\left(\frac{U}{T}\right)^2 (1-S), \qquad S \approx 3\left(\frac{\chi\sigma}{V\delta^2\rho L}\frac{T}{U}\right)^{1/3}, \tag{5}$$

where the term S describes the stabilization of short-wavelength perturbations owing to the surface energy σ. Equation (5) was derived considering the temperature at the evaporation front T and the front propagation velocity V as slowly varying functions of time. To estimate Γ, we substitute γ_m defined by (5) into equation (4), transform the integration variable t into T and incorporate the fact, that $V(T)$ decreases rapidly when T decreases. Then the integration is performed easily and yields

$$\Gamma \approx \frac{T_0 F^2}{6U}\left[1 - 3.6\left(\frac{\Lambda}{F}\right)^{1/3}\right], \qquad F = \frac{V\delta U}{\chi T_0}\ , \qquad \Lambda = \frac{\sigma}{\rho L\delta}\ . \tag{6}$$

Recall that the boundary between stable and unstable evaporation regimes is defined by the condition $\Gamma \approx 1$. It is easy to check by direct calculation that no considerable growth of the corrugation perturbations is expected in physically realizable region of parameters: $V_0\delta/\chi < 10$, $T_0/U < 1$, that corresponds to typical short-pulse ablation experiments. This conclusion seems to be in agreement with recent experiments on metal ablation carried out using laser pulses of different lengths (see, e.g., [9,10]). In the case of nanosecond pulses, considerable disturbances of the ablated surface are observed. Disruptions of the surface diminish as the laser pulse length decreases. The structure of the ablated surface is strongly material dependent. The quality of the surface is higher for metals with higher saturated vapor pressure at the melting point. For subpicosecond laser pulses, very high quality of the surface can be reached for many metals.

Note that, in the calculation of instability growth rate, the WKB-method provides only qualitative results, when γ is small. The reason is that the change in the vaporization velocity during the period of time when vaporization occurs is

on the order of the velocity itself. The same is true for the instability growth rate whose relative change during this time is of the order of unity. The perturbations of temperature profile and phase boundary position can be presented in the form (3) if the change of the instability growth rate $\gamma(k,t)$ during the time of perturbation growth ($\sim \gamma^{-1}$) is smaller than γ. As noted above, surface cooling at $t > \tau_i$ is caused mainly by the heat flux to the bulk of metal. The characteristic time of surface cooling is of the order of δ^2/χ; therefore, the characteristic time of instability growth rate decrease is about $T_0\delta^2/U\chi$. This means that WKB-calculation of $\gamma(k,t)$ is accurate provided that $\gamma T_0\delta^2/U\chi > 1$. Substituting γ as defined by equation (5) into this inequality and comparing with equation (6), we conclude that the WKB-method allows one to calculate the boundary between stable and unstable regimes with reasonable accuracy.

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