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Influence of Langmuir layer on the development of instability of molten metal surface under the influence of laser torch plasma

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The problem of metal surface instability under the influence of laser plasma in the process of nanosecond laser ablation in vacuum has been considered. To solve the problem, a numerical model of the Langmuir layer is constructed, which is applicable for calculations in the case when the layer thickness is much smaller than the characteristic wavelength of the melt surface perturbation. Analysis of the calculation results allowed us to find linear approximations of the ion pressure and electric field distributions on the metal surface. The use of these approximations allowed us to obtain an analytical criterion for the transition of the surface into an unstable state. It is also shown that the influence of the Langmuir layer leads to a decrease in the action of capillary forces, which can be used to modify the criteria for the development of instability in other mechanisms.

Keywords: laser torch plasma, capillary instability, plasma-liquid interaction.

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Numerous theoretical and experimental studies of melt surface [1–7] dynamics during nanosecond ablation of metal in vacuum have shown that capillary instability can develop on the irradiated surface, culminating in the escape of micron and submicron-sized melt droplets into the plasma of the laser torch. Microdroplets of melt, falling into the plasma of the laser torch, are charged to the value of the floating potential, which is determined by the plasma parameters. Estimates of plasma parameters for the initial stage of plume dispersal after the laser pulse termination using analytical and numerical [8,9] models show that in the case of laser pulses with radiation intensity $I \sim 1-3\,\mathrm{GW/cm^2}$, pulse duration $\tau \sim 20-30\,\mathrm{ns}$ and radiation wavelength $\lambda_I \approx 1\,\mu\mathrm{m}$, the characteristic plasma density is in the range $n_{pl} \sim 10^{23}-10^{25}\,\mathrm{m^{-3}}$, and the characteristic temperature of plasma electrons takes values $T_e \sim 1-5\,\mathrm{eV}$. As shown in the works [5,10], at such plasma parameters the charge of microdroplets begins to exceed the Rayleigh limit charge, which can lead to the beginning of the process of cascade crushing of microdroplets up to nanometre sizes. Based on this effect, a [5,6,11] laser electrodispersion method was developed (Fig. 1, *a*), which makes it possible to obtain nanostructures consisting of amorphous nanoparticles of various metals (Cu, Ni, Au, Ag, W, etc.) possessing narrow size dispersion (Fig. 1, *b*). Experimental studies of these nanostructures have shown their wide potential applicability due to their special catalytic, magneto-optical and structural properties.

Inhomogeneous heating of the target surface [1], modulations of plasma pressure and vapor vapor [2], directional motion of the plasma relative to the melt surface [3], etc., were considered as possible effects causing the development of instability, which is the source of melt microdroplets in the plasma. However, in these works, the formation of the Langmuir layer (plasma layer) near the target surface and its electric field on the melt surface were not taken into account when describing the plasma. The effect of the Langmuir layer electric field on the dynamics of the liquid-metal surface was analyzed separately both in the analytical approximation [12] and using the numerical approach [13–15]. In these works, it was shown that the electric field pressure of the Langmuir layer can significantly influence the surface dynamics. However, the direct application of the developed numerical models in the considered range of plasma parameters leads to numerical instabilities, which are probably related to the fact that the main change of the electric field potential occurs in the thin boundary layer near the curvilinear boundary. The aim of this study is to develop an approach that allows numerical calculations of Langmuir layer parameters near a curvilinear surface for the case of a dense laser torch plasma, and to apply the calculation results to analyze the influence of the layer on the instability development.

In the range of plasma parameters under consideration ($n_{pl} \sim 10^{23}-10^{25}\,\mathrm{m^{-3}}$, $T_e \sim 1-5\,\mathrm{eV}$), the effect of secondary electron emission from the surface can be neglected, so the melt surface is negatively charged. The electric field potential u near the melt surface can be described by the Poisson [16] equation, in the right-hand side of which is the density difference between ions n_i and electrons n_e. In the present work, it is assumed that the electron density in the layer obeys the Boltzmann distribution, i. e. $n_e = n_{pl}\exp(-q_e u/k_B T_e)$, where q_e — the charge of the electron, and k_B — the Boltzmann constant. To solve this problem, it is convenient to introduce dimensionless coordinates $\xi = x/\lambda$, $\eta = y/\lambda$ (λ — wavelength at the melt surface), and dimensionless electric field potential in the layer $\phi = q_e u/(k_B T_e)$. In this case, the equation for the

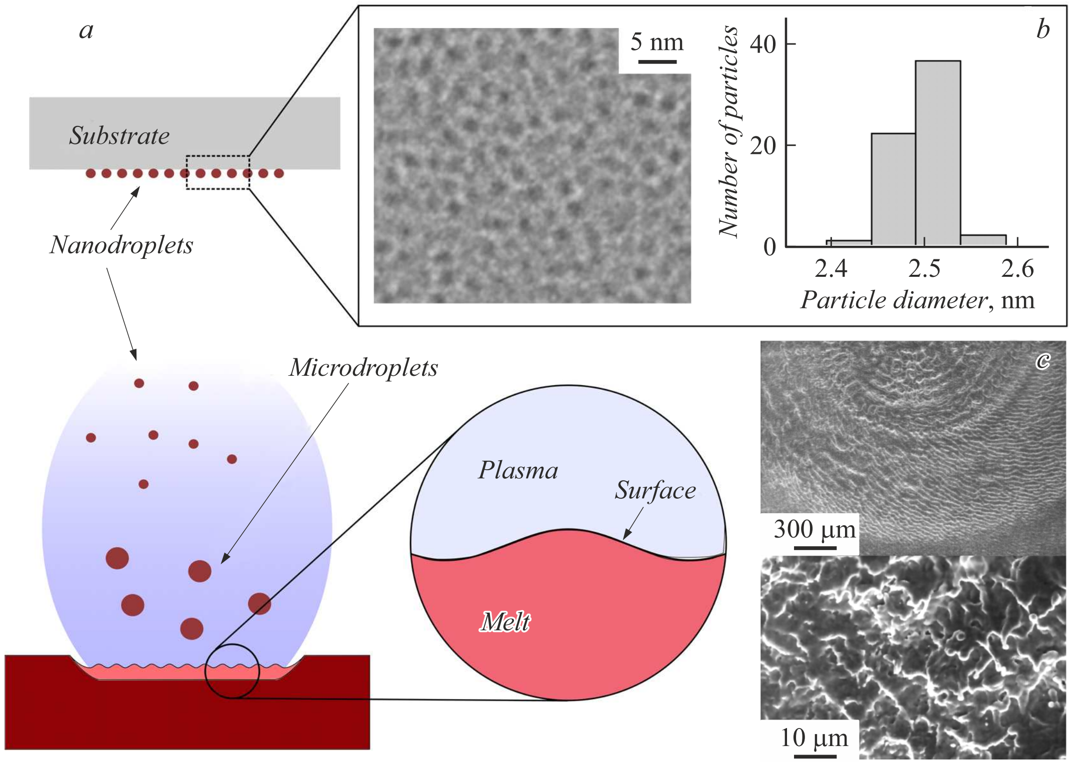

Figure 1. *a* — laser electrodispersion scheme; *b* — Ni nanoparticles obtained by laser electrodispersion [6]; *c* — microphotographs of the target surface after multiple irradiation.

potential has the form

$$\alpha^2\Delta\phi = \rho_i - \exp(-\phi), \tag{1}$$

where $\alpha = l_D/\lambda$ — parameter, l_D — Debye length, $\rho_i = n_i/n_{pl}$ — dimensionless value of the ion density in the layer. To find the density and velocity of ions (ρ_i and $\mathbf{v}_i$, respectively), we can use the system of equations for convective transport under the action of the electric field in the layer

$$\operatorname{div}(\rho_i \mathbf{v}) = 0,$$
$$\mathbf{v}(\nabla \cdot \mathbf{v}) = -\nabla\phi. \tag{2}$$

Here $\mathbf{v} = \mathbf{v}_i/c_i$ — the dimensionless ion velocity in the layer, and c_i — the ion-sonic velocity.

The system (1), (2) is solved in the region Ω, defined as follows (Fig. 2):

$$\Omega = \left\{(\xi, \eta) : -\frac{1}{2} \leqslant \xi \leqslant \frac{1}{2}, \qquad \Gamma_m \leqslant \eta \leqslant \Gamma_{sh}\right\}, \tag{3}$$

where Γ_m — the boundary of the region describing the metal surface, Γ_{sh} — the boundary of the Langmuir layer. The boundary to describe the perturbed metal surface in parametric form has the form

$$\Gamma_m = \left\{ (\xi_m, \eta_m) : \xi_m = t,\ \eta_m = \frac{A}{\lambda}\cos(2\pi t);\ t \in \left(-\frac{1}{2}; \frac{1}{2}\right) \right\}, \tag{4}$$

where A — the amplitude of the surface perturbation, t — the parameter. The boundary of the Langmuir layer with plasma Γ_{sh} in the present work is set in such a way that the normalized width of the $d = L_{sh}/\lambda$ layer along the normal to the surface takes the same value everywhere. In this case, the upper boundary of the layer in parametric form has the following form:

$$\Gamma_{sh} = \left\{ (\xi_{sh}, \eta_{sh}) : \xi_{sh} = \xi_m - \frac{(\eta_m)'_t d}{\sqrt{1 + \left((\eta_m)'_t\right)^2}}, \right.$$

$$\left. \eta_{sh} = \eta_m + \frac{d}{\sqrt{1 + \left((\eta_m)'_t\right)^2}} \right\}. \tag{5}$$

This choice of boundary ensures the preservation of the effect of screening of the electric field in the plasma and allows simplifying the system of equations by integrating the equation for the ion velocity in the system (2), but limits the applicability of the model only to cases in which the normals to the surface do not intersect, i.e. $AL_{sh}/\lambda^2 < 1$. The expression for the ion velocity in the approximation of motion along electric field lines is $\mathbf{v} = (1 + 2\phi)^{1/2}\nabla\phi/|\nabla\phi|$. The conditions for equation (1) at the boundaries are in the form of Dirichlet condition: $\phi|_{\Gamma_{sh}} = 0$, $\phi|_{\Gamma_m} = \phi_{fl}$ Here, ϕ_{fl} — floating potential, which satisfies the condition that

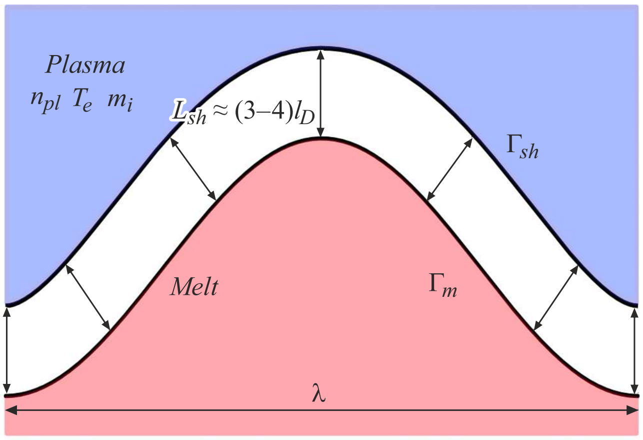

Figure 2. Schematic of the computational domain for the system of equations (1) − (6).

the total current along the surface is zero:

$$\int\limits_{\Gamma_m} \big(\rho_i v - \beta \exp(-\phi)\big) ds = 0, \quad \beta = \sqrt{\frac{m_i}{m_e}}. \tag{6}$$

The boundary condition for the ion density is $\rho_i\big|_{\Gamma_{sh}} = 1$, and periodic conditions are set at the $\xi = \pm 1/2$ boundaries. The solution of problem (1)−(6) was carried out in Comsol Multiphysics environment. The thickness of the Langmuir layer chosen for the calculations should be large enough to ensure the shielding effect of the electric field in the plasma ($E_m \gg E_{sh}$, where $E = |\nabla\phi|$), but also should not violate the convergence of the numerical methods. The results of preliminary calculations showed that the optimal choice in this case is the layer width $L_{sh} = (3{-}4)l_D$.

The curvature of the surface, as calculations have shown, does not affect the value of the floating potential ϕ_{fl}, given by formula (6), so in further calculations we used the value of the potential equal to the floating potential on a flat surface $\phi_{fl} = \ln\beta$.

The small perturbation of $A/\lambda \ll 1$ was analyzed in the linear approximation

$$\rho_i \approx \rho_{i,0} + \delta\rho_i\kappa, \quad E \approx E_0 + \delta E\kappa, \tag{7}$$

where κ — the curvature of the surface. The values of the coefficients $\rho_{i,0}$, $\delta\rho_i$, E_0, δE were determined by numerically solving the boundary value problem (1) − (6). The approximation of these quantities is described quite accurately as follows: $E_0 \approx \ln\beta/2.843\alpha$, $\delta E \approx 0.87\ln\beta$, $\delta\rho_i \approx 1.75\alpha v_{i,Me}^2$, where $v_{i,Me} = c_i(1 + 2\ln\beta)^{1/2}$ — ion velocity near the melt surface.

Due to the incompressibility of the fluid, the constant part of pressure along the surface does not affect its dynamics, so to determine the criterion of instability it is necessary to consider only the variation of pressure along the surface, which, taking into account $\kappa \ll 1$, has the following form

$$\delta P = \delta P_i + \delta P_\sigma - \delta P_E = \frac{m_i n_{pl} v_{i,Me}^2}{2}\delta\rho_i\kappa + \sigma\kappa - \frac{\varepsilon_0 T_e^2}{q_e^2\lambda^2}E_0\delta E\kappa. \tag{8}$$

Here δP, δP_i, δP_σ, δP_E — variations of total pressure, ion pressure, capillary pressure and electric field pressure at the surface, respectively, σ — melt surface tension coefficient. The total pressure variation is in the form of capillary pressure with some effective value of the surface tension coefficient σ_{eff}. The condition of surface instability development in this case will take the form $\sigma_{eff} \leqslant 0$ or

$$\sigma_{eff} = \sigma + \frac{m_i n_{pl} v_{i,Me}^2}{2}\delta\rho_i - \frac{\varepsilon_0 T_e^2}{q_e^2\lambda^2}E_0\delta E \leqslant 0. \tag{9}$$

The calculation results show that the plasma electron temperature $T_e \approx 20$ eV for plasma density $n_{pl} = 10^{25}$ m $^{-3}$ is required for the development of electrocapillary instability, but it increases rapidly as the density decreases. The reason for this effect is the rapid decrease in E_0 as the layer thickness increases. The electric field of the Langmuir layer can also reduce the effective surface tension coefficient, influencing the development of other types of instabilities. In particular, as was shown in [2], for the mechanism caused by plasma pressure modulation, the optimum wavelength for instability growth is proportional to the surface tension coefficient. Therefore, a decrease in this coefficient will lead to an increase in the growth rate of higher modes at the melt surface.

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Conflict of interest

The authors declare that they have no conflict of interest.

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