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Defect-deformation mechanism of the size effect in the laser-induced formation of microstructures of the brass surface relief in liquid

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***Abstract*. It is shown that the model of the defect-deformation instability describes the recently found linear dependence of the period of ordered surface relief microstructures, produced on brass in liquid upon multipulse laser ablation, on the radius of the spot on the target surface.**

Keywords: *laser action, brass, formation of surface microstructures, size effect.*

1. An interesting size effect was first observed in [1]: the period $\Lambda_{\rm exp}$ of ordered surface relief microstructures produced on a brass surface (an alloy of copper with zinc) in liquid upon multipulse ablation by scanning laser radiation (Fig. 1a) linearly increases with increasing the laser spot radius r_0 on the target surface (Fig. 2): $\Lambda_{\rm exp} = (\partial\Lambda/\partial r)_{\rm exp} r_0 = 0.9 r_0$.

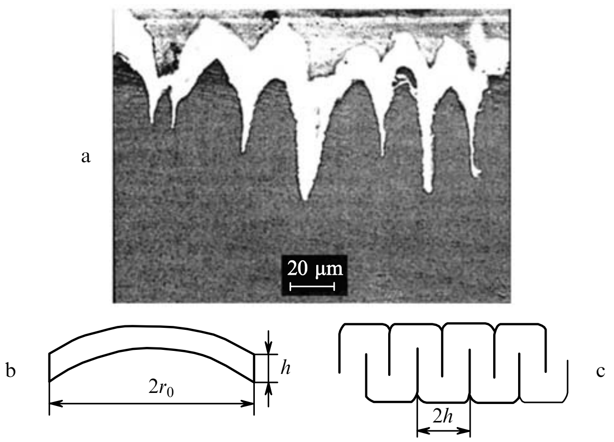

Figure 1. Scanning electron microscope image of the cross section of a brass sample with a two-dimensional surface relief structure produced by irradiation by a train of nanosecond pulses from a copper vapour laser with the fluence of 16 J cm^{-2} [1] (for clearness, colours in the photograph are reversed compared to the photograph in [1]) (a), a film in the state corresponding to the first bending mode from the spectra of bending modes [2] (b) and a film in the state corresponding to the limiting bending mode (c).

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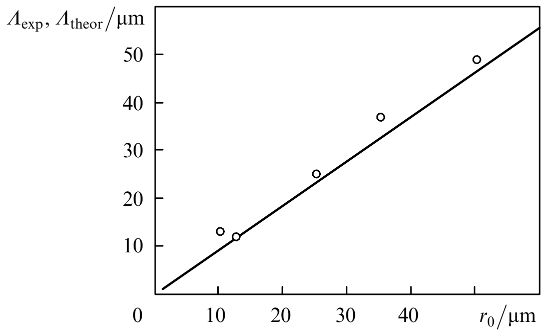

Figure 2. Experimental dependence of the period $\Lambda_{\rm exp}$ of the surface structure of brass on the laser spot radius r_0 (points) [1] and theoretical linear dependence of the DD model $\Lambda_{\rm theor}(r_0)$ (straight line) calculated by (1).

2. In this paper, the defect-deformation (DD) mechanism of the size effect in brass is proposed, which qualitatively and quantitatively describes experimental data of Ref. [1]. Upon multipulse laser irradiation at high temperatures, large concentrations $n_{\rm v}$ of mobile vacancies, which are initially distributed uniformly along the surface, are produced in the surface layer of brass of thickness h. We will consider this layer as a surface 'film' with elastic properties which differ from the properties of the 'substrate' – the rest part of the crystal. As was shown earlier in our review [2], a defect-deformation instability is developed in a film with mobile defects if their critical concentration ($n_{\rm v} > n_{\rm v\,cr}$) is exceeded. As a result, the relief modulation grating

$$\zeta(\boldsymbol{r}) = \zeta_q \cos[\boldsymbol{qr} + \phi(\boldsymbol{q})] \exp(\lambda_q t)$$

increases with the growth rate λ_q, where $\zeta(\boldsymbol{r})$ is the local surface displacement along the z axis (the z axis is directed from the surface into the bulk); $\boldsymbol{q}$ is the grating vector in the surface plane; $\boldsymbol{r} = \{x, y\}$; ζ_q is the real amplitude; and $\phi(\boldsymbol{q})$ is a random phase. The vacancies gather in the relief minima and form a surface DD grating. The period $\Lambda = 2\pi/q_{\rm m}$ of dominating (with the maximum growth rate $\lambda_{q_{\rm m}}$) DD gratings is proportional to the film thickness h and at a great enough concentration of defects is $\sim 2h$ [2]. In this case, the film has the state corresponding to the limiting bending mode (Fig. 1c). Apart from dominating DD gratings with wave vectors $\boldsymbol{q}_{\rm m}$ randomly distributed over different directions, the continuum of DD gratings with wave vectors also randomly distributed over different directions, whose moduli lie near $q_{\rm m}$ in the amplification band in which $\lambda_q > 0$, grows in time. The superposition of the continuum of DD gratings selected in this way in the amplification band gives a picture of a seed chaotic modulation of the surface relief (roughness) (Fig. 3) whose characteristic scale along the surface is $2h$. Laser ablation of this seed DD structure produces a resulting surface relief modulation whose periodicity and symmetry are determined by the seed DD structure.

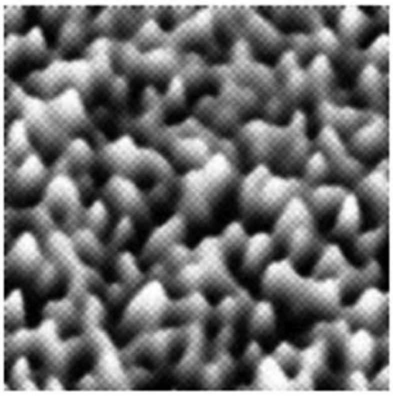

Figure 3. Seed surface relief obtained by summing all DD gratings of the relief with the wave vectors uniformly distributed within the angle 2π (see details in [3]).

3. Let us determine the thickness h of the defective layer under specific experimental conditions of Ref. [1]. Let a sequence N of laser pulses with the fluence $F_0 > F_m$ in each pulse (F_m is the threshold fluence of melting) and the beam radius r_0 irradiate the brass surface. A surface melted layer of thickness h_m is produced upon irradiation by each of these pulses. It is assumed that $h \gg h_m$ so that the main part of the layer remains in the solid phase. The laser heating of the medium, which is nonuniform along the surface, leads to the formation of a laterally deformed region with the characteristic size $2r_0$ along the surface at the melt–solid body interface. It follows from the elasticity theory that this deformation penetrates inside the elastic solid medium for the same distance $2r_0$. Deformation in this region is $\xi = \alpha_T \Delta T$, where α_T is the thermal expansion coefficient and ΔT is the temperature increase due to laser heating. The deformation gradient is $\partial\xi/\partial z \simeq -\xi/(2r_0) < 0$ along the z axis. The vacancy moving drift velocity from the thin melted layer into the bulk is $\upsilon = \theta_v(\partial\xi/\partial z)[D_v/(k_B T)]$, where θ_v is the deformation potential ($\theta_v < 0$); D_v is the diffusion coefficient of vacancies; $T = T_0 + \Delta T$; T_0 is the initial temperature of brass. We will show below that in the case under study the laser irradiation of brass in liquid can realise an efficient mechanism of lateral surface heat transfer due to which the condition $r_0^2/\chi_\parallel < h^2/\chi_\perp$ is fulfilled, where $\chi_\parallel$ and $\chi_\perp$ are the effective coefficients of lateral and normal thermal diffusivity in the surface layer, respectively. Therefore, the drift time of the vacancies into the bulk (taking place during the existence of the temperature-induced deformation) can be estimated as $\tau_{\rm drift} = r_0^2/\chi_\parallel$. Vacancies penetrate into the volume at a depth $\upsilon\tau_{\rm drift} = \theta_v(\partial\xi/\partial z)\times$ $[D_v/(k_B T)](r_0^2/\chi_\parallel)$ within this time. After irradiation of brass by N pulses, the doubled thickness of the layer (film) enriched with vacancies sets the characteristic size Λ of the grain in the seed DD structure under the condition $\Delta T \gg T_0$:

$$\Lambda = 2h = 2N\upsilon\tau_{\rm drift} = (\partial\Lambda/\partial r)_{\rm theor} r_0,$$
$$(\partial\Lambda/\partial r)_{\rm theor} = N[\theta_v \alpha_T D_v/(\chi_\parallel k_B)]. \quad (1)$$

The scale of the roughness Λ linearly increases with increasing the radius r_0 of the laser spot (size effect) and the number N of pulses (this increase saturated for $h \sim 2r_0$).

4. The estimate according to expression (1) for $\theta_v =$ 50 eV, $D_v = 10^{-5}$ cm^2 s^{-1} (the diffusion coefficient of Zn in Cu for the melting temperature of brass), $\chi_\parallel = 1 - 2$ cm^2 s^{-1} (see the estimate $\chi_\parallel$ below), $N = 10^4$ and $\alpha_T = 2 \times 10^{-5}$ K^{-1} yields $(\partial\lambda/\partial r)_{\rm theor} \sim 0.6 - 1.2$, which corresponds to the experimental value. The condition $r_0^2/\chi_\parallel < h^2/\chi_\perp$ of applicability of Eqn (1) to describe the results of experiments in [1] can be realised due to the inclusion of the surface mechanism of the heat removal from the spot centre to its periphery due to the evaporation and convection under the pressure of the liquid vapours with a simultaneous decrease in $\chi_\perp$ in the surface layer because of generation of vacancies. Indeed, the evaporation of the mobile and volatile component (zinc) from the brass surface upon laser heating leads both to generation of vacancies in the surface layer and to the liquid heating (vapour). The surface deformation structure and, hence, the volume deformation and deformation-induced drift of vacancies into the bulk will exist during the time $\tau_T = r_0/V$ of equalisation of the Gaussian temperature distribution in the laser spot on the surface. Here $V = [H^2/(12\eta)](\partial p/\partial x) \sim [H^2/(12\eta)](p/r_0)$ is the lateral velocity of saturated viscous vapour (liquid) in the evaporated layer of thickness H adjoining the brass surface; η is the liquid viscosity; $p = p(\boldsymbol{r})$ is the pressure in liquid [4]. By using the estimate $p(\boldsymbol{r}) = nk_B T(\boldsymbol{r})$ for the pressure, where n is the concentration of molecules in the vapour, we obtain $\tau_T = r_0^2/\chi_\parallel$, where the effective lateral thermal diffusivity is $\chi_\parallel = H^2 nk_B T/(12\eta)$. For $H = 10^{-4}$ cm, $n = 10^{20}$ cm^{-3}, $T = 10^3$ K, $\eta = 10^{-2}$ g s^{-1} cm^{-1}, we have $\chi_\parallel \sim 1$ cm^2 s^{-1}. In this case, $\tau_T = \tau_{\rm drift} \sim 10^{-5}$ s. The view of the cross section of the sample (Fig. 1a) shows that the surface relief in accordance with the film DD model (see clause 2) can in fact be considered to be formed by a periodically bent surface defective film (periodically bent light regions), which is in the state close to the limiting bending mode (cf. Figs 1a and c). Note that the DD interpretation allows one to explain also the fact that in air (in the absence of a liquid layer) the relief is not formed on the brass surface [1]. The liquid layer prevents the evaporation of zinc from the sample and, therefore, prevents the increase in the concentration of vacancies so that the state of the limiting bending mode is not achieved [2] (Fig. 1a). In air, due to a more intense evaporation of zinc, the concentration of vacancies becomes so high that the state of the limiting bending mode is achieved (Fig. 1c) and as the concentration of defects further increases, the surface relief is smoothed.

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