# The formation of laser-induced periodic structures on surfaces of solids

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The influence of solid surface irregularities on the formation of laser-induced periodic structures is studied both experimentally and theoretically.

A large number of recent papers discuss (see, for example, Refs. 1–4) the various aspects of physical processes involved in the creation of laser-induced periodic structures on surfaces of solids. The emphasis of theoretical analysis carried out in this work was on the study of critical conditions for the formation of the above structures for the case when an initial periodic perturbation is present on the surface.

However, first of all, an experiment measures not the critical conditions required for the structure formation but a particular surface profile.[3,5–8] Second, a dominant role in a real situation can be played by surface imperfections which, far from having any periodic character, are local.[8] For these reasons it is interesting to carry out a theoretical and experimental investigation that would allow, within the framework of a particular model, a more complete comparison between theory and experiment. Such an attempt is undertaken in this work.

## 1. EXPERIMENT

We used a transversely excited pulsed $CO_2$ laser. The resonator consisted of a spherical ($R = 20$ m) totally reflecting mirror, two flat totally reflective rotating mirrors which provided seven beam passes through the active medium, and a GaAs plane-parallel plate used as an exit mirror. The resonator length was 4 m. The beam profile was Gaussian mode, and the divergence was close to the diffraction limit when the resonator iris had a diameter of 10 mm. The radiation was linearly polarized by a GaAs plate placed in the resonator at the Brewster angle. The wavelength was determined by an additional interference filter introduced into the resonator. The pulse length could be varied by an order of magnitude by changing the composition and pressure of the working mixture: from 180 ns at half-maximum with a 1-$\mu$sec tail, containing 5% of the energy ($CO_2$:$N_2$:He = 1:0:3 mixture at pressure $p = 0.5$ atm) up to 2 $\mu$s (1:1:3 mixture, $p = 0.3$ atm).

The radiation was focused on the surface of a target by a spherical mirror ($R = 4$ m); the radius of the area illuminated at the $e^{-1}$ level was 0.61 mm. As targets we used flat plates made from fused KU quartz with a polished surface. After repeated exposure to radiation, the quartz surface usually formed the characteristic periodic relief structures described in Ref. 9; the number of pulses required to form this relief was inversely proportional to the excess of energy density in a single pulse over a threshold value. This threshold value corresponds to the onset of evaporation, since simultaneously with the relief formation one can observe plasma emission near the target surface due to breakdown in the vapor with a low threshold value. At lower intensities (0.9 times the threshold value) the relief was not formed even with $10^3$ or more pulses. The periodic structures originate near small irregularities, possibly dust particles and microcracks. Typically these periodic structures are localized on a scale of approximately 100 $\mu$m; they fill a crater by developing near smaller and smaller irregularities until all the separate regions merge into one continuous structure.

In the Pater experiments the irregularities on the quartz surface were created artificially. A narrow groove with a reproducible (to within 10%) width was cut with a diamond saw. After a single exposure to radiation with an intensity above the threshold value, the periodic structures formed in the vicinity of the scratch. The period of the structures exhibited a weak dependence on groove width, provided the width did not exceed 2.5 $\mu$m; for this reason the width used in subsequent experiments was 2 $\mu$m. In Fig. 1 are shown the structures which formed by normally incident radiation, when the scratch was oriented perpendicular to the vibration plane of the light wave. The transverse size of the area within which the periodic structure was localized was significantly smaller than its length and practically independent of intensity. This fact is demonstrated in Fig. 2, which shows the measured distances, at which the amplitude of a periodic

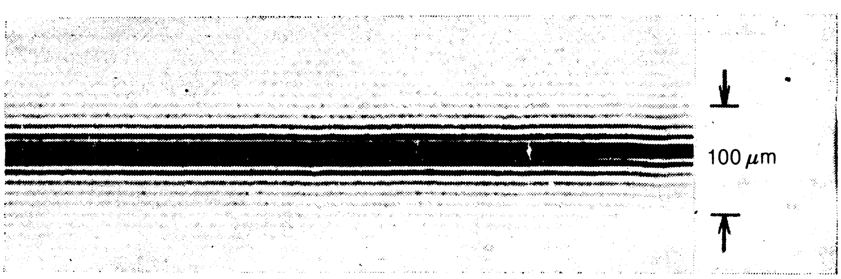


FIG. 1. Microphotograph of the surface structures formed by radiation with $\lambda = 10.6\mu$ at normal incidence. The arrows show an orientation of the electric field vector and indicate a linear scale.


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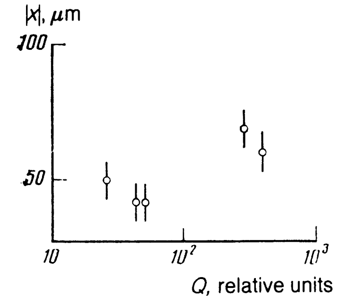


FIG. 2. Distance at which a periodic structure's amplitude decreases by a factor of two as a function of radiation intensity.

profile decreased by a factor of two for pulses differing in intensity by an order of magnitude.

If the radiation is not incident normally, the periodic structures become asymmetric relative to the scratch (Fig. 3). Figure 4 shows how the value of the period changes when the vector **E** in the incident wave is parallel to the plane of incidence.

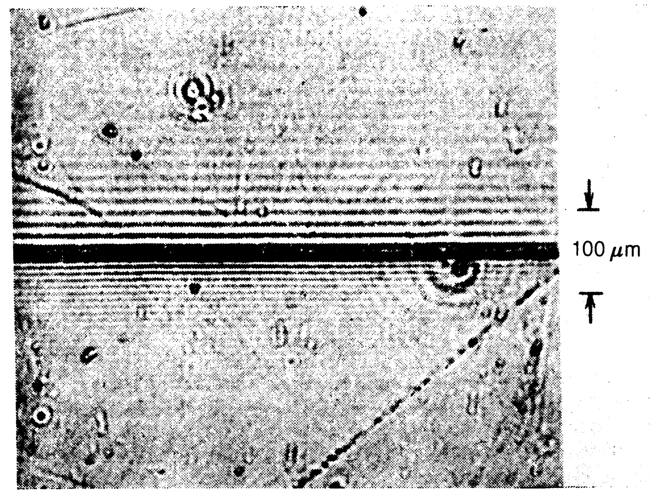


FIG. 3. Microphotograph of structures for the incidence angle of radiation in $\psi = 15°$ from a normal.

## 2. INTERPRETATION OF EXPERIMENTAL RESULTS

In order to provide a theoretical interpretation for the experimental results, we consider a model which, on the one hand, reflects the physics of the observed effects and explains the experiment, and, on the other hand, allows one to obtain simple and transparent results.

We will assume that the profile observed experimentally is a superposition of the initial roughness of the surface and a perturbation of this roughness due to the nonuniform energy absorption caused by the presence of the profile. Since from the experimental data it follows that the area of the structure is localized does not change with time, it is natural to assume that we are dealing with a quasistationary situation. Such a situation, as will be shown later, can be realized under certain conditions if one takes into account evaporation of the material (which actually occurs in the experiment).

Let us consider a half-space filled with a material, with an initial surface profile given by the function $z = \zeta_0(x)$, and assume that an electromagnetic wave

$$\mathbf{E}^{(0)} = \mathbf{E}_0 \exp(ikx \sin\psi + ikz\cos\psi),$$

is incident on this surface from vacuum; here the axis $z$ is assumed to be directed towards the vacuum, $k$ is the wave number, $\psi$ is the angle between the vector **k** and the negative $z$ axis. The profile amplitude is assumed to be sufficiently small that the correction to the field on the surface separating two media can be calculated by perturbation theory.

Taking into account the surface texture, we seek a solution of the temperature distribution problem in the material in the form

$$T(x, z) = T_0(z) + T_1(x, z),$$

where $T_0(z)$ is the solution of the evaporation problem with a flat boundary. In the coordinate system moving with a surface with the velocity $v$, we obtain for $T_0(z)$

$$\frac{\partial^2 T_0}{\partial \xi^2} - \frac{v}{\chi}\frac{\partial T_0}{\partial \xi} + f_0(\xi) = 0,$$
$$T_0(\xi = -\infty) = 0, \quad \lambda \left.\frac{\partial T_0}{\partial \xi}\right|_{\xi=0} = -v\Delta. \qquad (1)$$

Here $\xi = z + vt$; $\chi$ is the thermal diffusivity of the medium; $\lambda$ is the coefficient of thermal conductivity;

$$f_0(\xi) = [8kn\varkappa Q/\lambda(n^2+\varkappa^2)]\exp(2k\varkappa\xi);$$

$Q$ is the power density of the incident radiation; $n$ and $\chi$ are the refractive coefficient and absorption coefficients of the medium, respectively; and $\Delta$ is the heat of evaporation per unit volume of the material. As is well known (Ref. 10), the vapor speed $v$ is related to the surface temperature by

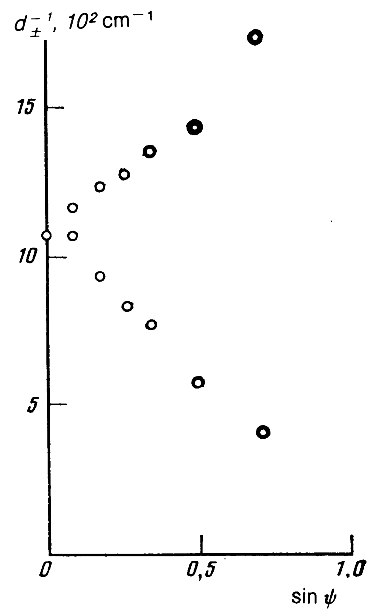


FIG. 4. Dependence of the structure period $d_\pm$ on the incidence angle $\psi$ for the case when the vector **E** in the incident lightwave is parallel to the incidence plane.

$$v = S\exp[-T^*/T_0(\xi=0)],\tag{2}$$

where $S$ is a quantity of the same order of magnitude as the sound velocity in the material, $T^* = M\Delta/R\rho$, $M$ is the molar weight of the material, $\rho$ is the density, and $R$ is the gas constant. For relatively low surface temperatures, when the inequality $(\lambda/\chi)\,T_0(\xi=0) \ll \Delta$ holds, it follows from (1) that

$$v \approx 4nQ/(n^2+\varkappa^2)\Delta.\tag{3}$$

In order to find the quantity $T_1(x, z)$ and the corresponding surface deformation, we will use a coordinate system moving with the velocity $v$. We will reduce our problem to that of the deformation of a flat surface with heat sources distributed nonuniformly as a function of $x$. Under the condition

$$T^*T_1(\xi=0)/T_0^2(\xi=0) \ll 1$$

the thermal conduction equation for $T_1(\xi)$ has the form

$$\frac{\partial^2 T_1}{\partial \xi^2} + \frac{\partial^2 T_1}{\partial x^2} - \frac{v}{\chi}\frac{\partial T_1}{\partial \xi} + f_1(x,\xi) = 0,$$

$$T_1(\xi=-\infty)=0,\quad \lambda\left.\frac{\partial T_1}{\partial \xi}\right|_{\xi=0} = -v\Delta\frac{T^*T_1(\xi=0)}{T_0^2(\xi=0)}.\tag{4}$$

Here we have neglected the derivative $\partial T_1/\partial t$, what corresponds to assuming that the change in profile caused by evaporation is small compared with the surface deformation, i.e., the surface texture is not produced by removal of a material. This is the condition for the situation to be quasistationary. The heat generation function is

$$f_1(x,\xi) = \frac{\omega\,\mathrm{Im}\,\varepsilon}{8\pi\lambda}(E_{1x}E_{0x}^* + E_{1x}^*E_{0x}),$$

where $\omega$ is the frequency of the incident radiation, $\varepsilon$ is the complex dielectric permittivity, $E_{0x} = 2\varepsilon^{-1/2}E_0$, and $E_{1x}(x)$ is a correction to the field on the plane $\xi = 0$, caused by the roughness of the real surface.

In order to calculate $E_{1x}$, we use an approach developed in Ref. 11. For the Fourier-representation $\tilde{E}_{1x}(q)$ we obtain

$$\tilde{E}_{1x}(q) \approx \varepsilon^{-1/2}E_0\tilde{\xi}(q)D(q),\tag{5}$$

where

$$D(q) = \frac{q+k\sin\psi}{k}B(q) + \frac{2i\varepsilon^{1/2}}{k}[q(q+k\sin\psi) - G^2(q)],$$

$$B(q) = \frac{2\varepsilon^{1/2}[G^2(q)\sin\psi - kq]}{\varepsilon^{1/2}G - ik},$$

$$G(q) = [(q+k\sin\psi)^2 + (-ik)^2]^{1/2},$$

Here $\varepsilon^{1/2} = n + i\chi$ with $|\varepsilon^{1/2}| \gg 1$, $\tilde{\xi}$ is the Fourier transform of the function $\xi(x) = \xi_0(x) + \xi_1(x)$, and $\xi_1(x)$ is the deformation profile of the flat surface which is to be determined. As a result, from Eqs. (4) and (5) we obtain for $T_1(x,\xi)$

$$T_1(x,\xi) \approx \frac{A}{(2\pi)^{1/2}}\int_{-\infty}^{+\infty} dq\, e^{ixq}\frac{\tilde{\xi}(q)[D(q)+D^*(-q)]}{p(q)+r}e^{p(q)\xi},$$

$$A = \frac{2nQ}{(n^2+\varkappa^2)\lambda},\quad p(q) = \frac{v}{2\chi} + \left[\left(\frac{v}{2\chi}\right)^2 + q^2\right]^{1/2},$$

$$r = \frac{v}{\lambda}\frac{T^*\Delta}{T_0^2(\xi=0)}.\tag{6}$$

Using familiar results from elasticity theory[12], we calculate the component of the deformation vector in the $\xi$ direction. Differentiating the result with respect to $x$ to eliminate displacements that do not depend on $x$, and assuming $\xi = 0$, we obtain

$$\frac{\partial \xi_1(x)}{\partial x} = -\frac{2\alpha(1-\sigma^2)}{3\pi(1-2\sigma)}\int_{-\infty}^{+\infty} dx'\,\frac{T_1(x',\xi=0)}{x-x'}$$

$$+\frac{4\alpha(1+\sigma)}{3\pi}\int_{-\infty}^{+\infty} dx'\int_{-\infty}^{0} d\xi'\,\frac{(x-x')\xi'}{[(x-x')^2+\xi'^2]^2}T_1(x',\xi').\tag{7}$$

Here $\alpha$ is the volume expansion coefficient and $\sigma$ is the Poisson coefficient.

In support of the model we have used here to describe the formation of surface structures, we can put forward the following arguments. First of all, estimates show that because the thermal conductivity of the material in our experiments increases sharply due to ionization during heating to high temperatures, the melted layer on the quartz surface can reach a depth of about $10^{-3}$ cm. It is quite realistic to assume that the local temperature "jump" of approximately $10^3$ K in the $x$ direction can lead to a spatial variation in the broadening of this layer of the same order of magnitude as that observed during the experiment for the value of the profile amplitude, i.e., about $10^{-4}$ cm.

Secondly, even though we are dealing with a liquid, this liquid can be sufficiently viscous to be considered as a solid. The characteristic interaction frequencies in our case are $\tau_l^{-1} \sim 10^6$–$10^7$ sec$^{-1}$, where $\tau_l$ is the laser radiation pulse length. The relaxation time $\tau$ in fused quartz at the temperatures of several thousand degrees estimated by the extrapolation of viscosity data is $\tau \sim 10^{-5}$ s.[12-14] Thus, the condition $\tau/\tau_l \gg 1$ for treating the liquid as a solid state is satisfied.

In order to solve Eq. (7), we apply a Fourier transformation to both sides. As a result we obtain for quantity $\tilde{\xi}_1(q)$ an algebraic equation. By solving the equation and performing the inverse Fourier transformation, we obtain

$$\xi_1(x) = \frac{1}{(2\pi)^{1/2}}\int_{-\infty}^{+\infty} dq\,e^{ixq}\tilde{\xi}_0(q)\frac{\Lambda[D(q)+D^*(-q)]}{P(q)+\Lambda[D(q)+D^*(q)]},\tag{8}$$

where

$$P(q) \approx |q|(|q|+r),\quad \Lambda = \frac{\alpha(1+\sigma)(3-4\sigma)}{3(1-2\sigma)}A.$$

Let us consider a simple situation by assuming $\xi_0(x) = (2\pi)^{1/2}h^2\delta(x)$. Then for large values of $x$ such that $k|x| \gg 1$, and also when the inequality $\Lambda|\varepsilon^{1/2}|/(k+r) \ll 1$ that limits the quantity $Q$ is satisfied, we obtain from (8)

$$\xi_1(x) = F_\pm(|x|)\theta(\pm x) + \Phi(|x|) + \text{c.c.}\tag{9}$$

Here

$$F_\pm(|x|)=-\frac{ih^2\eta\Lambda k}{q_\pm+r}\exp(i|x|q_\pm+ik|x|\eta^2)$$
$$\times\left[\Gamma\left(-\frac{1}{2},\ ik|x|\eta^2\right)+4\pi^{1/2}\theta(\varkappa-n)\right],$$
$$\mathrm{Re}[(i\eta^2)^{1/2}]>0,\quad q_\pm=k(1\mp\sin\psi).$$

The step function satisfies $\theta(y)=1$ when $y>0$ and $\theta(y)=0$ when $y<0$, $\Gamma$ is an incomplete gamma function, $\eta=i(2\varepsilon)^{-1/2}$,

$$\Phi(|x|)=h^2\beta[\mathrm{ci}(\beta|x|)\cos(\beta|x|)+\mathrm{si}(\beta|x|)\sin(\beta|x|),$$
$$\beta=4k\varkappa\cos^2\psi(\Lambda/r),\quad \beta/k\ll 1,$$

and ci and si are the cosine and sine integrals.

Two limiting cases are of special interest. The first, when $a\gg b$, where $a=\mathrm{Re}\,\eta$ and $b=\mathrm{Im}\,\eta$, corresponds to the existence of surface electromagnetic waves (SEW). When the inequalities $a^2k|x|\gg 1$ and $b^2k|x|\ll 1$ hold, we find

$$\mathrm{Re}\,F_\pm(|x|)\approx\frac{h^2\Lambda ka}{q_\pm+r}\left[4\pi^{1/2}\exp(-2k|x|ab)\sin(q_\pm|x|+a^2k|x|)-\frac{\sin(q_\pm|x|\pm\pi/4)}{(a^2k|x|)^{1/2}}\right]. \quad (10)$$

For the inverse situation, when $b\gg a$ and the inequalities $b^2k|x|\gg 1$ and $a^2k|x|\ll 1$ hold, we obtain

$$\mathrm{Re}\,F_\pm(|x|)\approx\frac{h^2\Lambda kb}{q_\pm+r}\frac{\sin(q_\pm|x|+\pi/4)}{(b^2k|x|)^{1/2}}. \quad (11)$$

The theoretical results thus obtained make it possible to explain the experimental results given above. First of all we note that, in agreement with experiment (see Fig. 1) and expressions (9) and (10), the structures which form are related to the excitation of SEW, since the period $d_\pm$ of a profile at small angles of incidence differs from the wavelength of the incident radiation. As the increase of the incidence angle the picture becomes asymmetric with respect to the initial perturbation (see Figs. 3 and 4), which is explained theoretically [see formula (9)] by the presence of the function $\theta(\pm x)$, multiplied by the function $F\pm(|x|)$, which oscillates with different periods on opposite sides of initial perturbation. Note that the function $\Phi(|x|)$ that enters Eq. (9) does not change significantly the type of structure, since it is a sufficiently slowly oscillating, weakly decaying function.

From Eq. (10) follows an interesting fact, namely, that the periodic structure is localized over distances $|x|\sim(2kab)^{-1}$, since at large values of the power density $Q$ the quantity $\Lambda/(q_\pm+r)$ depends weakly on $Q$ [$\Lambda\sim Q$, $r\sim Q$ (see Refs. 3, 6, and 8)]. This explains the experimental dependence shown in Fig. 2. The estimates obtained by comparing theory with experimental data give for the quantities $a$ and $b$ the values $a=0.3$ and $b=0.03$, what agrees well with the inequality $a\gg b$.

Let us note that, as follows from this theoretical analysis, excitation of SEW is impossible when $b>a$, but in this case the structures can develop with a period which is determined only by the wavelength and angle incidence of the laser radiation. These structures can be formed as a result of interference of volume electromagnetic waves.

We emphasize that all the experimental results given in this work can be explained within the framework of linear theory. We think that the fact that the periodic structures do not spread to a larger area with further repeated exposures to laser radiation also supports the above statement that linear theory describes the major effects in the formation of surface structures.

In conclusion, we note that the experimental and theoretical results obtained in this work demonstrate the important role of initial perturbations in the forming periodic surface structures.

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