Stimulated thermal scattering of light into a surface electromagnetic wave

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A stimulated scattering theory is used in an investigation of the process of nonlinear scattering into a surface electromagnetic wave at a metal–dielectric interface and formation of periodic surface structures. It is shown that stimulated thermal scattering into a surface electromagnetic wave is due to a permittivity grating. Experimental evidence is given to demonstrate the spatially cumulative instability which gives rise to periodic structures on a surface exposed to laser radiation.

1. INTRODUCTION

Extensive experimental and theoretical investigations have been made of the formation of periodic structures as a result of interaction of laser radiation with surfaces of metals and semiconductors (for a review and a bibliography see Ref. 1). Periodic surface structures are usually formed as a result of interference of laser radiation incident on a sample with a nonlinearly excited surface electromagnetic wave. In the initial stage of the process a surface electromagnetic wave is excited linearly because of the scattering of laser radiation by natural irregularities of the surface of a sample. The subsequent interference with a laser beam results in an inhomogeneous distribution of the optical field and a consequent spatially modulated heat release along a surface. The resultant modulation of the temperature of surface layers may create, by a variety of mechanisms, a surface-relief grating which in turn enhances the efficiency of scattering into a surface electromagnetic wave. When certain conditions are satisfied by the energy characteristics of the radiation, the amplitude of such a surface relief can increase with time. It has been mentioned frequently that the process is analogous to stimulated scattering in nonlinear optics.

Formation of periodic surface structures is possible not only on the surfaces of bulk samples, but also when thin metal films are subjected to laser damage.[2,3] Nonlinear scattering into a surface electromagnetic wave may occur before the onset of damage. Therefore, in contrast to other experiments in which the formation of periodic surface structures is accompanied by damage, it was reported in Ref. 2 that scattering into a surface electromagnetic wave by transient periodic structures can occur without a change in the surface microstructure. Since in a study of the mechanism of formation of such transient periodic surface structures we have to ignore the processes involving melting or evaporation of surface layers, it is logical to consider the following two alternative mechanisms. The first was investigated theoretically before[1] and it is related to the formation of a surface relief as a result of inhomogeneous thermal expansion of the surface layer of a sample. The second is due to a thermally induced change in the permittivity ε of a sample and was considered in Ref. 4 where a theoretical study of stimulated scattering from one surface wave into another was reported. It was found in Refs. 4 and 5 that the efficiency of nonlinear interactions involving surface electromagnetic waves can be enhanced greatly by employing a metal–liquid interface and this enhancement is due to large values of the temperature derivative of the permittivity $\partial\varepsilon/\partial T$ exhibited by various liquids. Heat conduction transfers a temperature distribution from a metal into a liquid where a permittivity grating is also formed.

We investigated nonlinear excitation of a surface electromagnetic wave at a silver–ethanol interface. Enhancement of the recorded signal made it possible to carry out more investigations of the process of formation of transient periodic surface structures, which were more detailed than in a study of nonlinear excitation of a surface electromagnetic wave at a silver–air interface.[2] It was demonstrated in Ref. 2 that this effect is due to a permittivity grating formed in ethanol. The transient nature of periodic surface structures and the actual mechanism of formation of a grating led to the conclusion of a complete analogy between the investigated effect and stimulated thermal scattering of volume waves (for reviews see Refs. 6 and 7). It has been found that a theoretical description of the effect can be provided using the approximation of slowly varying amplitudes which is employed extensively in studies of stimulated scattering of volume waves. This strengthens the similarity between the two effects and makes it possible to describe various relationships governing the process of formation of periodic surface structures using a theory of stimulated scattering developed in the last two decades.

2. PRINCIPAL EQUATIONS

We shall consider a metal film of thickness l on a glass substrate. The outer surface of the film ($z = 0$) is covered by a dielectric (liquid) and a p-polarized volume electromagnetic wave is incident on this side of the film at an angle θ. We write down the magnetic field of this wave in the form

$$H_L \exp(-i\omega t + i\mathbf{k}_L\boldsymbol{\rho} + ik_\perp z)$$

(the z axis is normal to the surface and directed into the film), where

$$|\mathbf{k}_L| = k_L = (\omega/c)\varepsilon_D^{1/2}\sin\theta, \qquad k_\perp = (\omega/c)\varepsilon_D^{1/2}\cos\theta,$$
$$\boldsymbol{\rho} = (x, y),$$

ε_D is the permittivity of the dielectric. A surface electromagnetic signal wave is traveling in the direction of the x axis along the film–dielectric interface, and the magnetic field of this wave in the metal and dielectric will be found from

$$\mathscr{H}_S(x, z, t)\exp(-i\omega t + ik_S x \mp Q_{SM,SD} z),$$

where

$$Q_{SM,SD} = [k_S^2 - (\omega^2/c^2)\varepsilon_{M,D}]^{1/2},$$

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ε_M is the permittivity of the metal, k_S is a real quantity, $\mathscr{H}_S$ is a function of coordinates and time varying slowly on the scale of ω^{-1}, k_S^{-1} and Q^{-1}. We shall avoid details causing major complications by assuming that the film thickness is sufficient to solve the electrodynamic part of the problem ignoring the existence of an interface between the metal and the substrate. The problem can be solved also allowing for the finite film thickness, but this does not give rise to any qualitatively new results.

We shall assume that the scattering into a signal wave occurs in a permittivity grating formed in the dielectric and characterized by

$$\Delta\varepsilon_D=\Delta(x,\ z,\ t)\ \exp\ (ik_Sx-i\mathbf{k}_L\rho)+\text{c.c}, \tag{1}$$

where Δ is the grating amplitude varying slowly with time and with the coordinate x. We shall regard the amplitude of the magnetic field $\mathscr{H}_S$ to be low and we shall ignore the scattering of the signal into the laser pump wave, which is analogous to the approximation postulating that the pump waves are inexhaustible in the bulk case. We can then find the distribution of the laser pump field in all three media. The distribution of the field in the metal is

$$\tau H_L \exp\ (-i\omega t+i\mathbf{k}_L\rho+Q_Lz),$$

and the field of the wave reflected by the surface is

$$rH_L \exp\ (-i\omega t+i\mathbf{k}_L\rho-ik_\perp z),$$

where

$$\tau=2/(1-\beta_L),\quad r=(1+\beta_L)/(1-\beta_L) \tag{2}$$

are the amplitude "transmission" and reflection coefficients, and

$$\beta_L=\varepsilon_DQ_L/ik_\perp\varepsilon_M,\quad Q_L=[k_L^2-(\omega^2/c^2)\varepsilon_M]^{1/2}.$$

The source of the signal wave is nonlinear polarization in the dielectric ($z<0$) of the type

$$\mathbf{P}_{NL}=(4\pi)^{-1}\Delta(x,\ z,\ t)\theta(-z)[\mathbf{E}_L\exp(ik_\perp z)+\mathbf{E}_{LR}\exp\ (-ik_\perp z)]\ \exp\ (-i\omega t+ik_Sx), \tag{3}$$

where $\mathbf{E}_L$ and $\mathbf{E}_{LR}$ are the amplitudes of the electric fields of the incident and reflected waves and $\theta(-z)$ is the Heaviside function.

The distributions of the signal wave field and of the nonlinear polarization depend in different ways on z and this complicates the solution of the electrodynamic problem. Similar difficulties are encountered in studies of other nonlinear interactions involving a surface electromagnetic wave (see, for example, Ref. 8). By analogy with Ref. 8, we shall first solve the Maxwell equations for all three media allowing for the boundary conditions at $z=0$ and l and assuming a nonlinear polarization of the type

$$\mathbf{P}_{NL}=(4\pi)^{-1}\delta(z-z')[\mathbf{E}_L+\mathbf{E}_{LR}]\exp(-i\omega t+ik_Sx), \tag{4}$$

where $\delta(z-z')$ is the Dirac delta function. Ignoring the second derivatives of the slowly varying amplitudes, we obtain the following equation for the field at the $z=0$ interface:

$$\frac{1}{k''}\frac{\partial\mathscr{H}_{S0}}{\partial x}+\mathscr{H}_{S0}=H_L\frac{\alpha_S}{\alpha_S+1}\frac{1}{\varepsilon_DQ_{SD}}[(k_Sk_L+ik_\perp Q_{SD}\cos\varphi)+r(k_Sk_L-ik_\perp Q_{SD}\cos\varphi)]\exp(Q_{SD}z). \tag{5}$$

Next, summing contributions of all the "layers" of the grating [which is a procedure analogous to a search for the solution in the form of a convolution of the fundamental solution with the right-hand side of Eq. (3)], we obtain

$$\frac{1}{k''}\frac{\partial\mathscr{H}_{S0}}{\partial x}+\mathscr{H}_{S0}=H_L\frac{\alpha_S}{\alpha_S+1}\frac{1}{\varepsilon_DQ_{SD}}\Big\{(k_Sk_L+ik_\perp Q_{SD}\cos\varphi)\times\int_{-\infty}^{0}\Delta(x,z',t)\exp[(Q_{SD}+ik_\perp)z']dz'+r(k_Sk_L-ik_\perp Q_{SD}\cos\varphi)\times\int_{-\infty}^{0}\Delta(x,z',t)\exp[(Q_{SD}-ik_\perp)z']dz'\Big\}. \tag{6}$$

Here,

$$\mathscr{H}_{S0}(x,\ t)=\mathscr{H}_S(x,\ 0,\ t),\quad \alpha_S=\varepsilon_MQ_{SD}/\varepsilon_DQ_{SM},$$

and φ is the angle between $\mathbf{k}_L$ and the x axis. The quantity k'' determines the distance $L=1/2k''$ traveled by a surface electromagnetic wave in the absence of a pump wave. In the case of a system with two interfaces, the expression for k'' is cumbersome and it will not be given here. It should be pointed out that the value of k'' for thick films can be determined from the familiar dispersion equation

$$k''=\mathrm{Im}\left(\frac{\omega}{c}\left[\frac{\varepsilon_M\varepsilon_D}{\varepsilon_M+\varepsilon_D}\right]^{1/2}\right).$$

The right-hand side of Eq. (6) simplifies greatly if we allow for the smallness of the depth d to which a liquid is heated by the transfer of energy dissipated in the metal film. In fact, in the case of nanosecond laser pulses we have $d\ll|Q_{SD}ik_\perp|^{-1}$ and then Eq. (6) becomes

$$\frac{1}{k''}\frac{\partial\mathscr{H}_{S0}}{\partial x}+\mathscr{H}_{S0}=a\tau H_L\Delta_{\text{eff}}(x,t),$$

$$a=\frac{\alpha_S}{\alpha_S+1}\left(\frac{k_Sk_L}{\varepsilon_DQ_{SD}}-\frac{Q_L}{\varepsilon_M}\cos\varphi\right), \tag{7}$$

$$\Delta_{\text{eff}}(x,t)=\int_{-\infty}^{0}\Delta(x,z',t)dz'.$$

We shall select the dimensions of H_L so that the laser beam intensity can be expressed in the form I_L [W/cm^2] $=\varepsilon_D^{-1/2}|H_L|^2$. In the case of a thermal source in a metal, we can use the expression q[W/cm^3] $=(\omega/c)$, where E is the distribution of the electric field in a metal and $\varepsilon_M''=\mathrm{Im}\varepsilon_M$. The increase in the temperature responsible for the formation of a grating will be sought in the form $T(x,z,t)\exp(ik_sx-i\mathbf{k}_L\rho)$, where T is the amplitude varying slowly with time and with the coordinate x. If we retain q only those terms which are proportional to $\exp(ik_sx-i\mathbf{k}_L\rho)$, we can write down the heat conduction equation for glass, metal, and dielectric (liquid):

$$\frac{\partial T_j}{\partial t}=\chi_j\left(\frac{\partial^2T_j}{\partial z^2}-\sigma^2T_j\right)+\frac{1}{(\rho c_p)_j}q_{0j}\tau^*H_L^*\mathscr{H}_S\exp[-(Q_L+Q_{SM})z], \tag{8}$$

$$q_{0D}=q_{0G}=0,\quad q_{0M}=\frac{\varepsilon_M''}{\varepsilon_M}\frac{c}{\omega\varepsilon_M}(k_Sk_L+Q_{SM}Q_L\cos\varphi),$$

where χ_j, ρ_j, and c_{pj} are, respectively, the thermal diffusivity, the density, and the specific heat of the medium; σ is the wave number of the grating; the index G refers to glass. We can write down the obvious equality

$$\Delta_{\text{eff}}(x,t)=\frac{\partial\varepsilon_D}{\partial T}\int\limits_{-\infty}^{0}T_D(x,z',t)\,dz', \tag{9}$$

which thus closes the system of equations (7)–(9) and the solution of this system will enable us to find the dependence of $\mathscr{H}_{S0}$ on time and on the coordinate x.

3. APPROXIMATE SOLUTION

The approximate solution of the system (7)–(9) allows us to find the main relationships governing stimulated thermal scattering into a surface electromagnetic wave and to demonstrate its full analogy with transient stimulated scattering of volume waves. We shall assume that the bulk of the dissipated heat is in the dielectrics (glass and liquid) and ignore the heat in the metal film. We shall also ignore the transfer of heat along the surface, which is justified in the case of large periods of the temperature grating. In this case the effective grating amplitude Δ_{eff}, proportional to the amount of heat in the dielectric (liquid), grows with time:

$$\Delta_{\text{eff}}=\frac{1}{(\rho c_p)_D}\xi\frac{\partial\varepsilon_D}{\partial T}\frac{q_{0M}\tau^*}{Q_{SM}+Q_L}\int\limits_0^t H_L{}^*\mathscr{H}_{S0}(t')\,dt', \tag{10}$$

$$\xi=(1+\lambda_G\chi_D^{1/2}/\lambda_D\chi_G^{1/2})^{-1},$$

where λ_j is the thermal conductivity of the medium. Time is measured from the beginning of a laser pulse. Substituting the above expression into Eq. (7) and differentiating the resultant equation with respect to time, we obtain

$$\frac{\partial^2\mathscr{H}_{S0}}{\partial x\,\partial t}+k''\frac{\partial\mathscr{H}_{S0}}{\partial t}=GI_L\mathscr{H}_{S0},$$

$$G=\xi\frac{\varepsilon_D^{1/2}}{(\rho c_p)_D}\frac{\partial\varepsilon_D}{\partial T}\frac{aq_{0M}k''}{Q_{SM}+Q_L}|\tau|^2. \tag{11}$$

Simple transformations can reduce Eq. (11) to the familiar telegraph equation.

We shall assume that a laser beam illuminates a surface zone of the film defined by $0<x<D$. The signal wave originates from a linearly excited (at surface irregularities of the sample) surface electromagnetic wave amplitude

$$H_0\int\limits_0^x\gamma(x')\exp[-k''(x-x')]\,dx',$$

where $\gamma(x)$ represents the distribution of the scattering centers on the surface. Then, the solution of Eq. (11) becomes

$$\mathscr{H}_{S0}(x,t)=H_L\int\limits_0^x\gamma(x')I_0(2[GW(x-x')]^{1/2})\times\exp[-k''(x-x')]\,dx', \tag{12}$$

where I_0 is a Bessel function and $W=I_Lt$ is the running value of the energy density in a laser beam. As a rule, such a "seed" signal appears as a result of the scattering of bulk radiation by the largest defects. We shall consider one of such defects at the point $x=0$ and assume that $\gamma(x)=(H_S/H_L)\delta(x)$. In an analysis of the solution (12) we shall use the familiar asymptote which is valid for high values of the argument of the Bessel function:

$$\mathscr{H}_{S0}(x,t)=H_S\exp[2(GWx)^{1/2}-k''x]. \tag{13}$$

When the energy density is sufficiently high so that $W(t)>k''/2(\text{Re}\cdot G^{1/2})^2$, the maximum signal intensity, proportional to $|\mathscr{H}_{S0}|^2$, is reached at a point

$$x_0=\left(\frac{\text{Re}\,G^{1/2}}{k''}\right)^2W. \tag{14}$$

The intensity can then be many times greater than the intensity of the signal at $x=0$:

$$\frac{|\mathscr{H}_{S0}(x_0)|^2}{|H_S|^2}=\exp\left[\frac{2(\text{Re}\,G^{1/2})^2}{k''}W(t)\right]. \tag{15}$$

This instability is cumulative and increases in time and space. This is particularly noticeable when the illuminated zone is small: $D\ll x_0$. In this case the intensity of the signal wave does not reach the values typical of large beams. If $D\gg x_0$, the use of a "seed" which is delta-function-like with respect to x yields a paradoxical result: the signal is attenuated in space. Therefore, if $D\gg x_0$, we have to allow for the real distribution of $\gamma(x)$, which complicates greatly the discussion without yielding any qualitatively new results.

4. ALLOWANCE FOR THE SPECIFIC HEAT OF A FILM AND FOR HEAT EXCHANGE ALONG ITS SURFACE

We shall seek the general solution of the system of equations (7)–(9) by the operator method. For the Laplace transforms $\tilde{\mathscr{H}}_{S0}(x)$, $\tilde{\Delta}_{\text{eff}}(x)$, and $\tilde{T}_j(x,z)$ this system of equations is

$$\frac{1}{k''}\frac{d\tilde{\mathscr{H}}_{S0}}{dx}+\tilde{\mathscr{H}}_{S0}=a\tau H_L\tilde{\Delta}_{\text{eff}},$$

$$p\tilde{T}_j=\chi_j\left(\frac{\partial^2\tilde{T}_j}{\partial z^2}-\sigma^2\tilde{T}_j\right)+\frac{1}{(\rho c_p)_j}q_{0j}\tau^*H_L{}^*\mathscr{H}_S\exp[-(Q_L+Q_{SM})z], \tag{16}$$

$$\tilde{\Delta}_{\text{eff}}=\frac{\partial\varepsilon_D}{\partial T}\int\limits_{-\infty}^{0}\tilde{T}_D(x,z')\,dz'.$$

In determination of the Laplace transform of the required amplitude $\tilde{T}_D(x,z)$ we have to allow for the following circumstances. The time for equalization of the temperature across the thickness of the metal film does not exceed 10^{-11} s, which is much less than the duration of a laser pulse. This allows us to assume that the temperature of the metal film T_M is practically the same across the thickness. The thermal diffusivity of the metal is three orders of magnitude higher than the thermal diffusivity of dielectrics, so that we need to allow only for heat transfer along the surface inside the film. Then, assuming the usual conditions of the equality of temperatures in heat fluxes at $z=0$ and $z=l$, and allowing for the slow changes in the amplitudes of T_j along the coordinate x, we obtain the following expression:

$$\tilde{T}_D=\frac{\xi q_{0M}\tau^*}{\chi_D^{1/2}(\rho c_p)_D(Q_{SM}+Q_L)}\frac{\exp[(p/\chi_D)^{1/2}z]}{p^{1/2}+(p+\chi_M\sigma^2)\tau_0^{1/2}}H_L{}^*\tilde{\mathscr{H}}_{S0}, \tag{17}$$

where

$$\tau_0=\left[\frac{(\rho c_p)_Ml}{\chi_D^{1/2}(\rho c_p)_D+\chi_G^{1/2}(\rho c_p)_G}\right]^2.$$

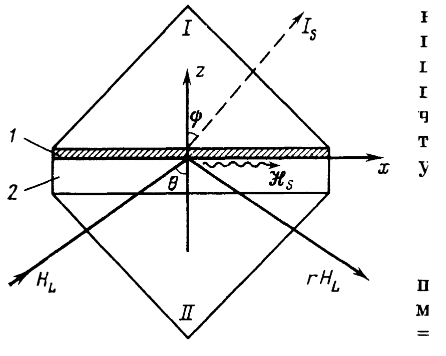

FIG. 1. Optical experimental setup (1—metal film; 2—liquid).

Integrating Eq. (17) with respect to the coordinate z, using the definition of $\Delta_{\rm eff}$, and substituting the resultant expression for $\tilde{\Delta}_{\rm eff}$ into the first equation of the system (16), we obtain

$$\frac{d\tilde{\mathscr{H}}_{s0}}{dx}+k''\tilde{\mathscr{H}}_{s0}=\frac{1}{p+(p\tau_0)^{1/2}(p+\chi_M\sigma^2)}GI_L\tilde{\mathscr{H}}_{s0}. \qquad (18)$$

If we still assume that the amplitude of the "seed" surface electromagnetic wave is $\mathscr{H}_{S0}(0,t) = H_S$, which corresponds to $\tilde{\mathscr{H}}_{S0}(0) = p^{-1}H_S$, we can find the Laplace transform of the amplitude of the signal wave $\tilde{\mathscr{H}}_{S0}$:

$$\tilde{\mathscr{H}}_{s0}=\frac{H_s}{p}\exp\left[\frac{GI_Lx}{p+(p\tau_0)^{1/2}(p+\tau_1^{-1})}-k''x\right], \qquad (19)$$

where $\tau_1 = (\chi_M\sigma^2)^{-1}$. Eq. (19) makes it possible to find the original function $\mathscr{H}_{S0}(x,t)$, but the expression describing it is cumbersome and will not be given here. The value of τ_0 for silver films used in experiments is about 5 ns, which is much less than the duration of a laser pulse τ_p. This makes it possible to write down the expression for $\mathscr{H}_{S0}$ in the case when $\tau_0 \ll \tau_p \ll \tau_1$ and for large values of $\mathrm{Re}(GWx)^{1/2}$ this expression is

$$\mathscr{H}_{s0}(x,t)=H_sI_0(2(GWx)^{1/2})\exp(-k''x)\,[1-(GWx)^{3/4}(\tau_0/t)^{1/2}-(GWx)^{1/4}(t\tau_0/\tau_1)^{1/2}]. \qquad (20)$$

It follows that allowance for heat exchange along the surface and for the specific heat of the film reduces the signal intensity. However, if the influence of these effects is slight, there should be no qualitative changes. In the case of short grating periods the exchange of heat along the film surface increases considerably and this results in smearing out of the grating by heat conduction and reduces strongly the amplitude of the signal.

5. EXPERIMENTS

We used the experimental setup shown in Fig. 1. Radiation from a single-mode neodymium laser ($\tau_p = 30$ ns) polarized in the plane of incidence reached a silver film which was evaporated on a diagonal face of a rectangular glass prism I at an angle θ. The film was in contact with air or with the dielectric (liquid). In the latter case an auxiliary prism II was used to ensure grazing incidence of the beam on the film. The diameter of the beam at the entry to the prism was 2.5 mm. The attention was concentrated on nonlinear excitation of a surface electromagnetic wave at the silver–ethanol interface. A surface wave excited in the process of stimulated thermal scattering was partly re-emitted into the interior of the prism and this volume radiation was detected experimentally. The silver film thickness was varied in these experiments within the range 450–550 Å. The experimental results reported below were obtained for a film of thickness $l = 480$ Å and the permittivity of silver was assumed to be $\varepsilon_M = -50 + i1.1$. These values were found experimentally by a method described in Ref. 5.

In the case of a laser beam with a large transverse dimension the maximum gain was experienced by a signal wave which had the highest value of Re $G^{1/2}$. Equation (11) made it possible to establish that Re $G^{1/2}$ was maximal for $\varphi = 0$ and $k_S = k_{\rm SEW}$, where SEW refers to a surface electromagnetic wave and

$$k_{\rm SEW}=\mathrm{Re}\left(\frac{\omega}{c}\left[\frac{\varepsilon_M\varepsilon_D}{\varepsilon_M+\varepsilon_D}\right]^{1/2}\right).$$

In fact, the experimentally detected volume wave propagated in the plane of incidence of the original laser beam, which corresponded to $\varphi = 0$. The angle of observation ψ (Fig. 1) could be calculated quite accurately from $\psi = \sin^{-1}(ck_{\rm SEW}/\omega n_G)$, where n_G is the refractive index of the glass prism. Figure 2b shows the angular distribution of

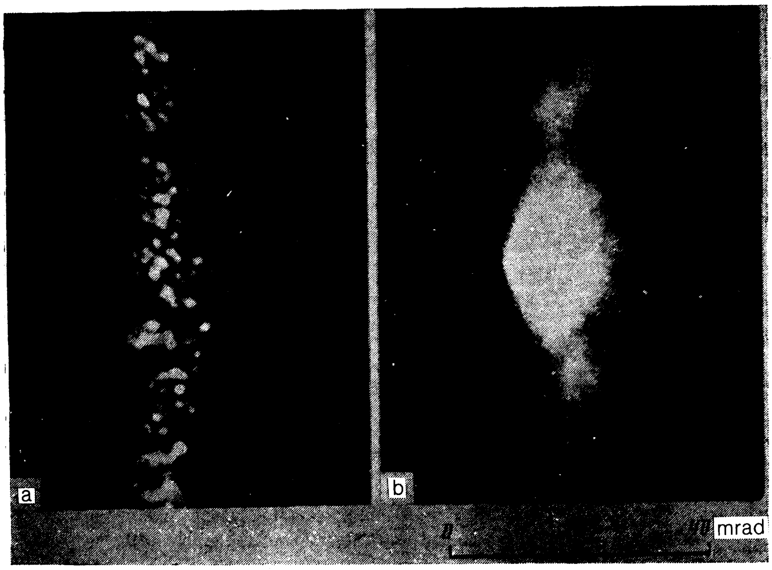

FIG. 2. Angular distribution of the linear noise field (a) and of the volume signal of stimulated thermal scattering (b).

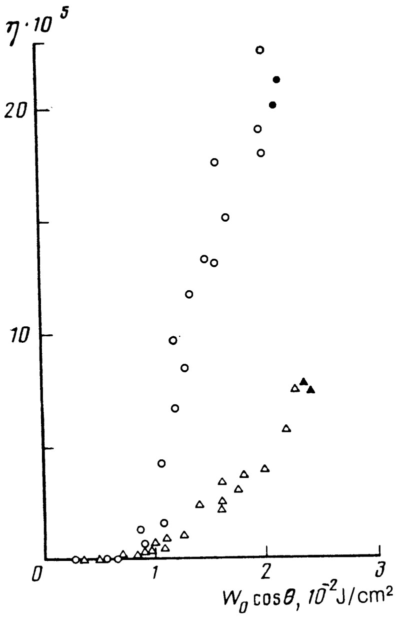

FIG. 3. Energy dependence of the efficiency of stimulated thermal scattering at the silver–ethanol interface; the triangles correspond to $\theta = 76°$ and the circles to $\theta = 81.6°$. The dark symbols correspond to damage to the film surface.

the volume stimulated thermal scattering signal obtained with the aid of an image converter. For comparison, Fig. 2a gives the angular distribution of the linear noise (at pump energies below the threshold of stimulated thermal scattering); the amplitude of this noise was practically independent of the angle φ. In contrast to the linear noise with a strong speckle structure, the nonlinear signal did not exhibit a small-scale spatial inhomogeneity and was highly directional. The range of the values of φ and k_s used in our experiments was determined from the angular divergence of the volume signal.

Figure 3 shows the energy dependences of the efficiency η of stimulated thermal scattering obtained for different angles of incidence θ. The efficiency was defined as the ratio of the volume signal energy E_S (after subtraction of the linear noise) to the energy of the laser radiation E_L incident on the film. The value of E_L was determined allowing for the Fresnel losses on the faces of the auxiliary prism. The density $W_0 \times \cos\theta$ of the energy delivered to the film surface throughout a laser pulse was calculated allowing for changes in the transverse size of the beam when it became refracted in the prism. The investigated effect had a clear threshold. When the threshold was exceeded, the efficiency η rose steeply on increase in the energy density of the laser beam. Attainment of high values of η was prevented by damage to the silver film by the laser beam. The damage threshold exceeded the threshold of stimulated thermal scattering by a factor of about 2.5. At the silver–air interface this ratio was considerably less and did not exceed 1.2 (Ref. 2). Moreover, at the silver–ethanol interface there was a major reduction in the absolute value of the energy threshold of stimulated scattering compared with the silver–air interface. The maximum value of η obtained experimentally for the investigated film was $\eta_{\max} \approx 2 \times 10^{-4}$, which was an order of magnitude higher than $\eta_{\max}$ for stimulated thermal scattering at the silver–air interface. The relatively high efficiency of the scattering process made it possible to carry out a more detailed (compared with Ref. 2) investigation of stimulated thermal scattering and, in particular, to study the influence of the angle of incidence θ on this type of scattering.

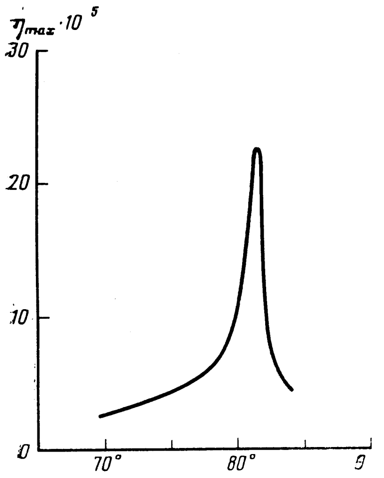

FIG. 4. Dependence of the maximum efficiency $\eta_{\max}$ of stimulated thermal scattering on the angle of incidence θ.

Figure 4 shows the angular dependence of the maximum efficiency $\eta_{\max}$. Clearly, the investigated process was characterized by an optimal angle of incidence θ_{opt} corresponding to high values of the efficiency η. In the case of the investigated film we found that $\theta_{\text{opt}} \approx 82°$. An increase in the film thickness reduced θ_{opt}. The angle θ influenced also the stimulated thermal scattering threshold and the film damage threshold. The observed dependence of the investigated effect on the angle of incidence can be explained as follows. The reduction in $\eta_{\max}$ in the range $\theta > \theta_{\text{opt}}$ is due to the angular dependence of Re $G^{1/2}$, particularly due to a reduction in $|\tau|^2$, which occurs as a factor in the expression for G given by Eq. (11). If $\theta < \theta_{\text{opt}}$, there is a reduction in the temperature grating period and this increases heat exchange along the surface and thus reduces the efficiency η.

A considerable increase in the efficiency and a reduction in the absolute energy threshold of stimulated thermal scattering at the silver–ethanol interface allows us to conclude that the investigated effect is determined by the permittivity grating formed in ethanol. We checked further this hypothesis by investigating also the nonlinear scattering into

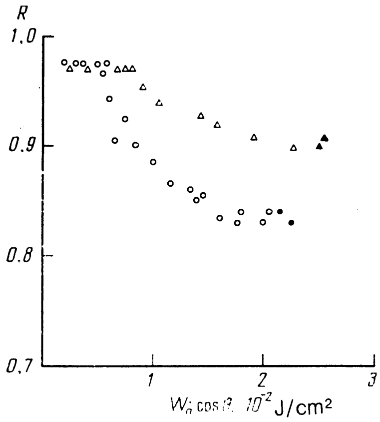

FIG. 5. Energy dependence of the reflection coefficient R of the film under stimulated thermal scattering conditions; the notation is the same as in Fig. 3.

a surface electromagnetic wave at the interface of silver and water, which is known to have optical and thermophysical constants similar to those of ethanol, but a much smaller temperature coefficient of the permittivity. As expected, the maximum efficiency attained in this way was even less than at the silver–air interface. In this case the effect was probably governed solely by the metal and the permittivity grating in water did not have a significant influence. Unfortunately, because of the strong temperature dependence of the distance traveled by a surface electromagnetic wave it was not possible to compare quantitatively the observed threshold of stimulated thermal scattering with theoretical predictions.

As demonstrated in Ref. 2, the process of stimulated thermal scattering was accompanied by additional dissipation of the energy in the film. The specular reflection coefficient R of the laser radiation incident on the film decreased. Figure 5 shows the energy dependences of R for different values of the angle of incidence θ. Clearly, when the stimulated thermal scattering threshold was exceeded, the reflection coefficient decreased reversibly by 13–15% for $\theta = \theta_{\rm opt}$. For other angles of incidence the reduction in R was somewhat less. Such a strong increase in the energy deposited in the film undoubtedly affected the laser damage threshold. In particular, for $\theta = \theta_{\rm opt}$ the damage threshold of the film in contact with ethanol could decrease, because of stimulated thermal scattering, more than threefold compared with the damage threshold of the film in contact with the water.

As pointed out in Ref. 3, the process of stimulated thermal scattering into a surface electromagnetic wave could occur also in the geometry of frustrated total internal reflection, when the laser beam was incident on the film from the glass side across the prism I (Fig. 1). In fact, in this geometry we found that in a wide range of angles of incidence the reflection coefficient R decreased when the energy density of the laser beam exceeded a certain threshold value and this was clearly a consequence of stimulated thermal scattering. However, direct experimental confirmation of the occurrence of stimulated thermal scattering, which would be detection of the signal transferred to the interior of glass, was not obtained for this case. This was due to a strong increase in the linear noise (not attenuated by the passage across the film), which was due to the scattering of the original laser beam by the surface irregularities of the film.

6. STIMULATED SCATTERING INTO A SURFACE ELECTROMAGNETIC WAVE AT THE SILVER–AIR INTERFACE

Stimulated thermal scattering can be recorded also at the silver–air interface.[2] In this case we found that when the damage threshold of the film was exceeded, irreversible periodic structures visible under a microscope were formed. This happened because the damage occurred initially at maxima of the temperature grating. Visualization of the grating responsible for the effect provided a unique opportunity for the investigation of stimulated scattering, practically impossible in the case of scattering of volume waves. It should be pointed out that, in spite of the much greater amplitude of the signal due to stimulated thermal scattering at the silver–ethanol interface, we were unable to detect irreversible periodic surface structures. This was clearly due to the fact that liquid prevented free removal of the products of evaporation from the surface when damage to the metal film occurred. Since investigations of irreversible periodic surface structures could provide important additional information on the investigated effect, some of the experiments were carried out on the silver–air interface.

The most probable mechanism by which a transient periodic structure, responsible for stimulated thermal scattering at the silver–air interface, could form was the thermal change in the permittivity of the metal, although this was not confirmed by direct experiments. Since the alternative mechanism of formation of transient periodic surface structures had been investigated earlier,[1] we concentrated our attention on a grating of the permittivity of the metal. In determination of the Laplace transform of the amplitude of the signal wave $\tilde{\mathscr{H}}_{S0}$ we used the approach described above (see Secs. 2–4). It should be mentioned only that the amplitude of the permittivity grating in the metal was proportional not to the integral of the amplitude of the temperature grating, but to its value at $z = 0$ [see Eq. (17)]. Bearing all these points in mind, we can now describe $\tilde{\mathscr{H}}_{S0}$ by the following expression:

$$\tilde{\mathscr{H}}_{S0}(x) = \frac{H_S}{p} \exp\left[\frac{G I_L x}{(p/\tau_0)^{1/2} + p + \tau_1^{-1}} - k'' x\right], \tag{21}$$

where

$$G = \frac{1}{(\tau_0 \chi_G)^{1/2} (\rho c_p)_G} \left(\frac{\partial \varepsilon_M}{\partial T}\right) \frac{a q_{0M} k''}{Q_{SM} + Q_L} |\tau|^2,$$

$$a = \frac{1}{\alpha_S + 1} \frac{k_S k_L + Q_{SM} Q_L \cos\varphi}{\varepsilon_M Q_{SM} (Q_{SM} + Q_L)}, \qquad \tau_0 = \left[\frac{(\rho c_p)_M l}{(\rho c_p)_G \chi_G^{1/2}}\right]^2,$$

$\partial \varepsilon_M / \partial T$ is the temperature derivative of the permittivity of the metal and H_S is the amplitude of a "seed" wave at $x = 0$. In the case of large periods of the temperature grating the value of τ_1^{-1} is small and the expression for $\mathscr{H}_{S0}$ can be written in the form[9]

$$\mathscr{H}_{S0}(x,t) = \frac{H_S}{2\pi^{1/2}} \exp(-k'' x) \int_0^t \frac{t + t'}{\tau_0^{1/2} (t')^{3/2}} \times \exp\left[-\frac{(t - t')^2}{4\tau_0 t'}\right] I_0(2[G I_L x (t - t')]^{1/2})\, dt'. \tag{22}$$

At high values of $\tau_0 \gg \tau_p$, we have

$$\mathscr{H}_{S0}(x,t) = H_S I_0(2[G W x]^{1/2}) \exp(-k'' x).$$

However, in spite of some increase in the time τ_0 because of the absence of heat exchange with the dielectric, the value of τ_0 is still less than the duration of a laser pulse τ_p. Nevertheless, an analysis of Eq. (22) indicated that at high energy densities all the main features of the process were observed. When the laser beam was of considerable size, a single surface scattering defect could not enhance the signal over the whole area of the spot and there was a coordinate $x = x_0$ at which the signal intensity reached its maximum. The first to rise was the signal wave with k_s close to $k_{\rm SEW}$ corresponding to $\varphi = 0$ and the instability was of cumulative nature in space. As before, an increase in the rate of heat exchange across the surface reduced the signal.

We shall be interested primarily in the case of large periods of a temperature grating, because it is then possible to detect experimentally the nonlinear scattering signal. Under these circumstances, the process in question is analogous to transient stimulated scattering of volume waves, as demonstrated above. Equation (21) makes it possible to consider the opposite situation. We shall discuss the case when τ_1 is much less than the pulse duration, which is possible in the case of small-period gratings and long pulses. An analog of the process in question is steady-state stimulated scattering of volume waves. Following the formalism of stimulated scattering theory, we shall assume the existence of a small frequency shift Ω of a surface electromagnetic wave toward the Stokes region relative to the pump frequency. The initial system of equations was modified somewhat, namely $\chi_M\sigma^2$ in Eq. (8) was replaced with $\chi_M\sigma^2 + i\Omega$ and consequently in Eq. (21) and later τ_i was understood to be $\tau_1 = (\chi_M\sigma^2 + i\Omega)^{-1}$. The solution for $|\mathscr{H}_{S0}(x)|^2$ is as follows:

$$|\mathscr{H}_{S0}(x)|^2=|H_S|^2\exp\{2[\mathrm{Re}(G\tau_1(\Omega)I_L)-k'']x\}. \qquad (23)$$

Clearly, depending on the sign and magnitude of the quantity $\mathrm{Re}[G\tau_1(\Omega)I_L]$, we can expect either spatial amplification or attenuation of $\mathscr{H}_{S0}$. The largest gain increment is exhibited by a surface electromagnetic wave at Ω_0, corresponding to the maximum of $\mathrm{Re}[G\tau_1(\Omega)]$, which should increase when the threshold pump intensity is exceeded. The expressions for Ω_0, I_{th} are as follows:

$$\Omega_0=[u-(u^2+1)^{1/2}]\chi_M\sigma^2, \qquad I_{\mathrm{th}}=k''/\mathrm{Re}(G\tau_1(\Omega_0)) \qquad (24)$$

where

$$u=\mathrm{Im}\frac{\partial\varepsilon_M}{\partial T}\Big/\mathrm{Re}\frac{\partial\varepsilon_M}{\partial T}.$$

In the case of sufficiently large dimensions of the laser spot D, Eq. (23) corresponds to an increase in the signal intensity without limit in the range $I_L > I_{\mathrm{th}}$, but the following comments should be made here. We have assumed so far that the pump waves are given and we have ignored the influence of the signal on the reflection coefficient r and on the "transmission" coefficient τ. However, in the case of sufficiently large amplitudes $\mathscr{H}_S$ this approximation becomes invalid and we have to allow for the scattering of the signal itself by the permittivity grating. Allowance for the "depletion" of the pump wave shows that when the threshold I_{th} is exceeded significantly, the signal intensity becomes quite rapidly steady-state along the coordinate x and is independent of this coordinate over a large part of the spot. It should be pointed out that at high stimulated-thermal-scattering rates allowance for the pump depletion is essential also under transient conditions. It is sufficient to mention here that a reduction in the reflection coefficient $R = |r|^2$ of a film by more than 10% had been observed experimentally.

7. INFLUENCE OF THE SPATIAL DISTRIBUTION OF SPONTANEOUS SEEDS ON THE SIGNAL WAVE

To study the influence of the spatial distribution of "seed" surface electromagnetic waves on the investigated process, we concentrated our attention on irreversible periodic surface structures formed as a result of laser damage to a silver film. The radiation was incident on the film from the air at an angle $\theta = 68°$, which was optimal for stimulated thermal scattering at the silver–air interface.[2] The period of the surface structure agreed quite well with the hypothesis of the scattering of incident radiation into a surface wave, and it amounted to 12.8 μm. Figure 6a shows a photomicrograph of a damaged surface of silver. We can see that under natural conditions the excitation of seed surface electromagnetic waves occurs as a result of scattering of laser radiation by large point defects on the surface. In spite of the strong divergence of the linearly excited seed wave, nonlinear amplification is experienced by surface electromagnetic waves close to zero value of the angle φ. The weak divergence of the signal wave determines the morphology of the damaged surface. One can see also that the only point defect which cannot act as a nucleus of a single periodic surface structure occupying the whole area of the spot. As a rule, we observed several isolated structures in the direction transverse to $\mathbf{k}_L$. It should be pointed out that in the case of large dimensions of a laser beam, surface periodic structures were isolated also in the longitudinal direction relative to $\mathbf{k}_L$. Therefore, in the process of propagation the signal electromagnetic wave was first amplified and attenuated, in agreement with the theoretical results of Sec. 3. Although the phase of the signal wave was governed primarily by the initial seed, one could not exclude the possibility of some influence on the signal due to shallower surface defects.

The signal surface electromagnetic wave corresponding to the maximum value of $\mathrm{Re}\, G^{1/2}$ should have the largest amplitude only in the case of a relatively homogeneous angular distribution of the seed. Selection of the seed with a nonzero value of φ was essential to ensure a certain amplification

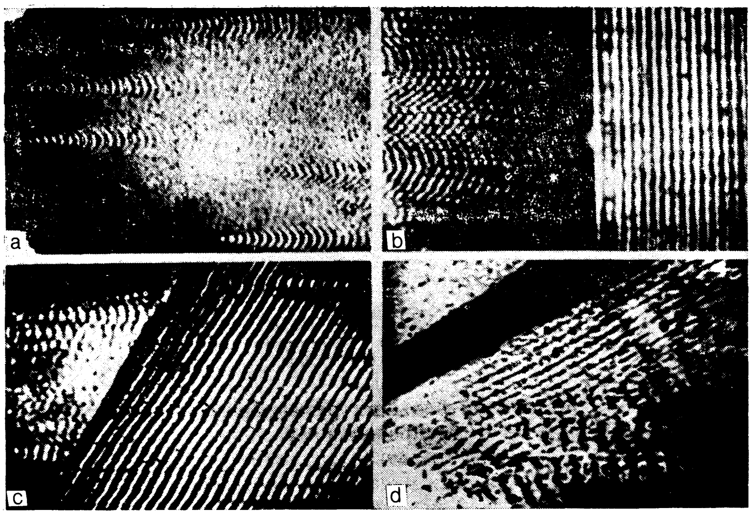

FIG. 6. Periodic structures formed by laser damage to the surface of a silver film: a) periodic surface structures in the case of natural roughness; b)–d) periodic surface structures at an incipient crack.

length, as a consequence of the nature of the instability growing in space. This maximun length could be quite considerable for a large amplitude of the seed and a small difference of Re $G^{1/2}$ from its maximum value corresponding to $\varphi = 0$.

The above discussion was checked in an experiment in which a seed surface electromagnetic wave was excited as a result of scattering of laser radiation by a scratch formed deliberately on the film surface. The transverse size of the scratch was a few microns and the angle of tilt Ω of the scratch relative to the vector $\mathbf{k}_L$ could be varied during an experiment. Figure 6b reproduces a photomicrograph of the damaged surface of the film observed for $\Omega = 90°$, corresponding to $\varphi = 0$. The asymmetry of the figure showed that the seed surface electromagnetic wave did have a significant influence on the signal wave. The phase-matched seed in the form of a scratch gave rise to a single, in contrast in the case of natural surface electromagnetic waves (on the left), periodic structure (to the right of the figure). For small values of φ, when Re $G^{1/2}$ differed slightly from its maximum value, there was no discrimination of the initial seed even over a long amplification length amounting to several hundreds of microns (Fig. 6c). The orientation of a surface periodic structure then corresponded to the direction of the scratch. However, a further increase in the angle φ (Fig. 6d) resulted in attenuation of the seed surface electromagnetic wave already at short distances, in spite of the large amplitude of the initial seed. Amplification of the seed surface electromagnetic waves was then competitive and they were due to the scattering by natural surface defects; they were also characterized by the maximum value Re $G^{1/2}$.

8. CUMULATIVE NATURE OF THE DEVELOPMENT OF AN INSTABILITY IN SPACE

The cumulative (in space) nature of the investigated instability was manifested most strikingly by stimulated Raman scattering into a surface electromagnetic wave in the case of samples of small size because of what happened to the surface of the film as a result of exposure to a laser beam. In fact, if $D \ll x_0$ [see Eq. (14)] the intensity of the signal and the amplitude of a temperature grating could not reach significant values during a pulse even at high energy densities W in the beam, because the amplification length was small. The laser damage threshold of the film W_{th}, which decreased when the process of stimulated thermal scattering was advanced and represented quantitatively the efficiency of formation of periodic structures, was found to be constant and the same as in the conventional damage to a film in the absence of the investigated effects.

In the experiment the illuminated zone of the film surface was a long strip with a small transverse size, formed by focusing of radiation with the aid of a cylindrical lens of 10-cm focus. Figure 7 shows the dependence of W_{th} on the transverse size of such an illuminated strip D (deduced from the diffraction minima). The strip was oriented at right-angles to the direction $\mathbf{k}_L$ and the angle of incidence should still be $\theta = 68°$. As illustrated in Fig. 7, in the range $D < 300\ \mu m$ the short amplification length prevented attainment of a high signal intensity; lowering of W_{th} did not occur and formation of a periodic surface structure was not observed. Beginning from $D = 1600\ \mu m$, we exceeded the scale of x_0, the process of stimulated thermal scattering developed fully and further increase in the size of the strip D did not increase the efficiency of stimulated thermal scattering at the silver–air interface. The range $300\ \mu m < D < 1600\ \mu m$ is known as intermediate.

A different proof of the spatially cumulative nature of the investigated process was provided theoretically in Ref. 10 by the following reasoning. Let us assume that a laser spot is still a long narrow strip and that the orientation of the strip makes a nonzero angle φ_0 with the vector $\mathbf{k}_L$ (Fig. 8). Then,

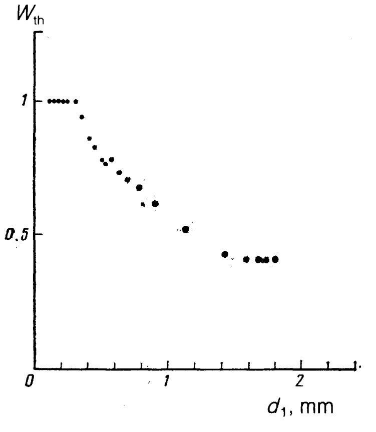

FIG. 7. Dependence of the laser damage threshold of a silver film (in relative units) on the transverse size of the illuminated strip.

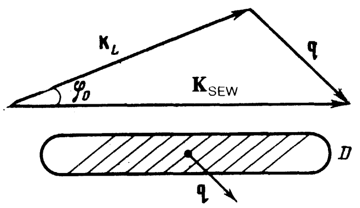

FIG. 8. Diagram of the wave vectors used in the formation of a periodic surface structure by a laser beam of small size ($\mathbf{q}$ is the vector of the periodic surface structures).

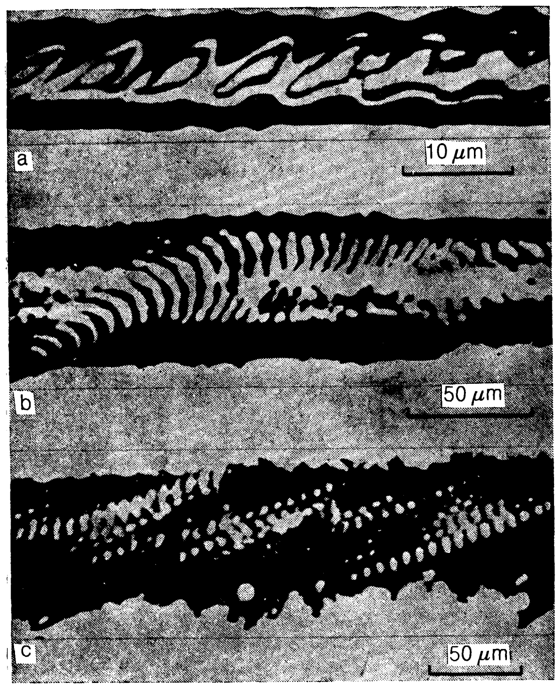

FIG. 9. Photomicrographs of surface periodic structures observed for different values of the transverse size of the beam.

the signal surface electromagnetic wave corresponding to $\varphi = 0$ does not reach significant amplitude because its amplification length is short. On the other hand, a surface wave traveling along the $\varphi = \varphi_0$ direction (along the strip) grows in space without hindrance in spite of the fact that the value of Re $G^{1/2}$ is somewhat smaller. The period and orientation of surface periodic structures which are formed in this way depend strongly on the angle φ_0 (Ref. 10). Such periodic structures have indeed been observed experimentally; Fig. 9 shows photomicrographs of the damage to the film surface in the case when $\varphi_0 = 15°$ and $\theta = 60°$ when the size of the strip D was varied. An investigation of irreversible periodic surface structures established that if $D < 200\ \mu$m (Fig. 9a) the direction of propagation of the signal surface electromagnetic wave should indeed coincide with the direction of the strip ($\varphi = \varphi_0$). It should be pointed out that for sufficiently large values of the angle φ_0 the damage threshold decreased below the threshold of stimulated thermal scattering and the structures in question were not observed. For a strip of size $D > 500\ \mu$m (Fig. 9c) the orientation and period of a surface structure corresponded to the maximum value of Re $G^{1/2}$ ($\varphi = 0$). For intermediate values in the range $200\ \mu\text{m} < D < 500\ \mu$m the periodic structures were of the combined nature (Fig. 9b).

Instead of the expected amplification of the signal surface electromagnetic wave with some fixed value of $0 < \varphi < \varphi_0$, the orientation of the periodic surface structure during the propagation of the signal did not change. At the edge of the spot the period and orientation of a surface structure corresponded to the maximum value of Re $G^{1/2}$ ($\varphi = 0$). However, in the process of amplification of the signal its wave vector $\mathbf{k}_s$ exhibited a smooth variation of its orientation without disturbing the phase of the signal surface electromagnetic waves and then the angle φ increased practically from zero to φ_0. This unexpected result could not be explained without additional investigations in which we would have to allow for competition between the amplification and attenuation of various angular components of the signal and also for the role of the seed surface electromagnetic waves distributed on the surface. In this range of transverse dimensions of the strip D the periodic surface structures of this type predominated, which prevented the influence of randomness of detection.

9. CONCLUSIONS

In view of the full analogy between the process of stimulated thermal scattering into a surface electromagnetic wave, on the one hand, and stimulated thermal scattering of volume waves, we used the latter in theoretical analyses involving approximation of slowly varying amplitudes, which made it possible to obtain all the results of the theory of stimulated scattering investigated before. The experimental data obtained in the present study confirmed the validity of the above theoretical ideas. It should be pointed out that an approach of this type is not used widely in the description of the process of formation of periodic surface structures. Nevertheless, the methods developed in the theory of formation of periodic surface structures[10–12] also make it possible to obtain similar results. A theoretical approach which ignores the dependence of the signal amplitude on the coordinate in the plane of the surface[1] is convenient for finding the period and orientation of the dominant structures and it can also be used to describe the processes similar to steady-state stimulated scattering, because in this case the amplitude of the signal wave was independent of the spatial coordinate over a large part of the laser spot.

It therefore follows that the selection of some particular theoretical approach is governed primarily by the conditions in a concrete problem and the convenience of its use. On the other hand, the main results of the theory of stimulated scattering developed so far can have a considerable influence on the development of the theory of formation of periodic surface structures.

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