Laser excitation of coupled surface electromagnetic and acoustic waves and of static surface structures in solids

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A theory is developed for the induced generation of surface acoustic waves by the action of a single beam of laser radiation on absorbing condensed materials. The surface acoustic wave instability mechanism is the following. When a surface acoustic wave is caused by fluctuations, the laser wave excites a surface electromagnetic wave, and the interference of this wave with the laser wave leads to a spatially periodic heating of the surface and, correspondingly, to a periodic variation in the pressure which reinforces the fluctuational start of the surface acoustic waves. In addition to ordinary (Rayleigh) surface acoustic waves, the dispersion equation that is derived also describes the excitation of a new class of surface acoustic waves. These are quasistatic induced surface acoustic waves having a frequency determined by the intensity of the laser pumping. The dependences of the characteristics of the excited surface acoustic waves on frequency, polarization, and angle of incidence of the laser wave are studied. The formation of reversible and irreversible surface ripples which is observed in the action of laser radiation on absorbing solids is interpreted on the basis of the theory that is developed.

1. INTRODUCTION

It has been shown experimentally[1–9] that when intense laser radiation acts on strongly absorptive solid materials, (semiconductors, metals, and insulators), one- and two dimensional structures (ripples) are formed on their surfaces. The experimental data indicate that these structures arise because of the excitation of a surface electromagnetic wave[10] (SEW) and the interference of this wave with the incident wave.[5–7] There can be a variety of specific mechanisms for ripple formation that are associated with a spatially periodic distribution of light intensity produced on the surface. The mechanism that has gained the widest currency is the vaporization mechanism,[5–7,11,12] which leads to the formation of only irreversible ripples. However the formation of reversible ripples as well has been observed,[8] these ripples appearing during the laser pulse and disappearing after the pulse stops. A theory must account for the formation of both irreversible and reversible ripples.

In this paper a theory is developed for the induced generation of surface acoustic waves (SAW)[13] by the action of laser radiation on absorptive solids. On the basis of this theory an interpretation is given of the formation of surface ripples, including both the irreversible and the reversible cases. The theory that is developed is also of intrinsic interest for acousto-optics as a new mechanism of laser excitation of coherent surface acoustic waves.

The surface acoustic wave instability mechanism considered in this investigation is the following. Fluctuational excitation of a surface acoustic wave gives rise to a surface-relief modulation that is periodic in space and time, and as a result of this modulation the external light wave $\mathbf{E}_i$ excites a surface electromagnetic wave. The interference of the latter with the light wave transmitted into the medium, together with optical absorption, produces a surface temperature wave and, as a result of thermal expansion, a corresponding pressure wave, which augments the fluctuational start of the surface acoustic wave. When a critical (threshold) value $|\mathbf{E}_i|^2_{\text{th}}$ of pumping is exceeded, this positive feedback leads to an exponential (in time) growth in the surface acoustic wave amplitude. The frequencies ω_α ($\alpha = s,a$) of the surface electromagnetic waves, the frequency $\Omega_\mathbf{q}$ of the surface acoustic wave and the frequency ω of the pumping wave are connected by the relations

$$\omega=\omega_s+\Omega_\mathbf{q}, \quad \omega=\omega_a-\Omega_\mathbf{q}. \tag{1}$$

In addition, the equalities

$$\mathbf{k}_t=\mathbf{k}_s+\mathbf{q}, \quad \mathbf{k}_t=\mathbf{k}_a-\mathbf{q}, \tag{2}$$

are satisfied, where $\mathbf{k}_t$ is the pumping wave-vector component tangent to the plane surface, and $\mathbf{k}_\alpha$ and $\mathbf{q}$ are, respectively, the wave vectors of the surface electromagnetic wave and surface acoustic wave lying in the plane of the surface.

Conditions (1) and (2) define a class of coherent surface acoustic waves that in principle can be excited via the mechanism under consideration. Of these waves, those for which the generation threshold is minimal will grow the fastest. The values of $\Omega_\mathbf{q}$ and $\mathbf{q}$ for these waves depend substantially on the frequency, polarization, and angle of incidence of the pumping wave. Standing as well as travelling surface acoustic waves can be excited.

The excitation of standing surface acoustic waves under conditions where Hooke's law has not yet broken down may be responsible for the formation of the reversible ripples; otherwise, irreversible ripples are formed.

Besides the generation of ordinary (Rayleigh) surface acoustic waves, the mechanism that we are investigating leads to the formation of a new class of surface acoustic waves. These are quasistatic surface acoustic waves, with frequencies determined by the intensity of the laser pump-

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ing. The excitation of these waves constitutes a nonequilibrium phase transition of the soft mode (relaxation) type. The excitation of quasistatic surface acoustic waves also can be responsible for the formation of reversible and irreversible surface ripples.

2. EXCITATION OF A SURFACE ELECTROMAGNETIC WAVE BY AN EXTERNAL LIGHT WAVE WITH SPACE-TIME MODULATION OF THE SURFACE RELIEF

Let us consider a medium that occupies the half-space $z \geqslant \xi(\mathbf{r},t)$:

$$\xi(\mathbf{r},t)=\xi_{\mathbf{q}}(t)\exp\{-i\mathbf{qr}+i\Omega_{\mathbf{q}}t\}+\text{c.c.},\tag{3}$$

where $\mathbf{r}=\{x,y\}$ is a vector lying in the plane $z=0$ and $\xi_{\mathbf{q}}(t)$ is a slow modulation of the surface relief in the presence of a surface acoustic wave. A plane electromagnetic wave

$$\mathbf{E}=\mathbf{E}_i(\omega)e^{-i\omega t}+\text{c.c.},\qquad \mathbf{E}_i(\omega)=\mathbf{E}_i\exp\{i\mathbf{k}_t\mathbf{r}+ik_zz\}\tag{4}$$

is incident from the vacuum $[z<\xi(\mathbf{r},t)]$ onto the interface. We shall seek to express the field outside the medium as a superposition of incident (i) and reflected (r) waves at the frequency ω and two diffracted waves with Raman frequencies ω_α

$$\mathbf{E}=[\mathbf{E}_i\exp(ik_zz)+\mathbf{E}_r\exp(-i\mathbf{k}_zz)]\exp\{i\mathbf{k}_t\mathbf{r}-i\omega t\}+\sum_{\alpha=s,a}\mathbf{E}_\alpha\exp\{i\mathbf{k}_\alpha\mathbf{r}+\Gamma_\alpha z-i\omega_\alpha t\}+\text{c.c.},\tag{5}$$

where ω_s, ω_a, $\mathbf{k}_s$, and $\mathbf{k}_a$ are given by formulas (1) and (2).

We write the field inside the medium as a sum of a transmitted (t) wave and two diffracted waves with Raman frequencies ω_α:

$$\mathbf{E}=\mathbf{E}_t\exp\{i\mathbf{k}_t\mathbf{r}-\gamma z-i\omega t\}+\sum_{\alpha=s,a}\mathbf{E}_\alpha'\exp\{i\mathbf{k}_\alpha\mathbf{r}-\gamma_\alpha z-i\omega_\alpha t\}+\text{c.c.}\tag{6}$$

From Maxwell's equations and (5) and (6) we obtain for the damping constant

$$\gamma^2=k_t^2-k_0^2\varepsilon(\omega),\qquad \operatorname{Re}\gamma\geqslant 0;$$
$$\gamma_\alpha^2=k_\alpha^2-k_{0\alpha}^2\varepsilon(\omega_\alpha),\qquad \operatorname{Re}\gamma_\alpha\geqslant 0;$$
$$\Gamma_\alpha^2=k_\alpha^2-k_{0\alpha}^2,\quad \operatorname{Re}\Gamma_\alpha\geqslant 0;\quad k_0=\omega/c,\quad k_{0\alpha}=\omega_\alpha/c,\tag{7}$$

where $\varepsilon(\omega)$ is the dielectric constant. The amplitudes of the fields in (5) and (6) are determined from the conditions that the components of the vector $\mathbf{E}$ and of the magnetic field $\mathbf{H}$ tangent to the surface $z=\xi(\mathbf{r},t)$ are continuous and that $\operatorname{div}\mathbf{E}=0$, the last condition being satisfied everywhere. Introducing the unit vector $\hat{e}_3$, pointing along the z axis into the medium and the unit vectors $\hat{e}_2=\mathbf{k}_t/|\mathbf{k}_t|=\hat{k}_t$ and $\hat{e}_1=\hat{e}_2\times\hat{e}_3$, we have the following expression for the corresponding components of the fields inside the medium:

$$E_{t1}=\frac{2k_z}{k_z+i\gamma}E_{i1},\qquad E_{t2}=\frac{-2i\gamma k_z}{k_t[k_z\varepsilon(\omega)+i\gamma]}E_{i3},$$
$$E_{t3}=\frac{2k_z}{k_z\varepsilon(\omega)+i\gamma}E_{i3},\tag{8}$$

$$E_{\alpha1}'=\frac{2k_0k_z(1-\varepsilon(\omega))}{k_{0\alpha}(k_z+i\gamma)}\frac{k_\alpha^2(\hat{k}_t\hat{k}_\alpha)^2-\gamma_\alpha\Gamma_\alpha}{\varepsilon(\omega_\alpha)\Gamma_\alpha+\gamma_\alpha}\xi_\alpha E_{i1}-\frac{2ik_zk_\alpha(1-\varepsilon(\omega))[k_0^2k_\alpha\gamma(\hat{k}_t\hat{k}_\alpha)-k_{0\alpha}^2k_t\gamma_\alpha]}{k_0k_{0\alpha}k_t(k_z\varepsilon(\omega)+i\gamma)(\varepsilon(\omega_\alpha)\Gamma_\alpha+\gamma_\alpha)}[\hat{k}_t\hat{k}_\alpha]_z\xi_\alpha E_{i3},\tag{9}$$

$$E_{\alpha2}'=\frac{2k_0k_zk_\alpha^2(1-\varepsilon(\omega))}{k_{0\alpha}(k_z+i\gamma)[\varepsilon(\omega_\alpha)\Gamma_\alpha+\gamma_\alpha]}(\hat{k}_t\hat{k}_\alpha)[\hat{k}_t\hat{k}_\alpha]_z\xi_\alpha E_{i1}+\frac{2ik_z(1-\varepsilon(\omega))[k_0^2\gamma\gamma_\alpha\Gamma_\alpha-k_0^2\gamma k_\alpha^2[\hat{k}_t\hat{k}_\alpha]_z^2-k_{0\alpha}^2k_\alpha k_t\gamma_\alpha(\hat{k}_t\hat{k}_\alpha)]}{k_0k_{0\alpha}k_t(k_z\varepsilon(\omega)+i\gamma)[\varepsilon(\omega_\alpha)\Gamma_\alpha+\gamma_\alpha]}\xi_\alpha E_{i3},\tag{10}$$

$$E_{\alpha3}'=\frac{2ik_zk_\alpha k_0\Gamma_\alpha(1-\varepsilon(\omega))[\hat{k}_t\hat{k}_\alpha]_z}{k_{0\alpha}(k_z+i\gamma)[\varepsilon(\omega_\alpha)\Gamma_\alpha+\gamma_\alpha]}\xi_\alpha E_{i1}+\frac{2k_zk_\alpha(1-\varepsilon(\omega))[k_{0\alpha}^2k_tk_\alpha-k_0^2\gamma\Gamma_\alpha(\hat{k}_t\hat{k}_\alpha)]}{k_0k_{0\alpha}k_t(k_z\varepsilon(\omega)+i\gamma)[\varepsilon(\omega_\alpha)\Gamma_\alpha+\gamma_\alpha]}\xi_\alpha E_{i3},\tag{11}$$

where $\xi_s=\xi_q(t)$, $\xi_a=\xi_q^*(t)$. Analogous expressions hold for the fields outside the medium.

Expressions (8)–(11) are valid for media with arbitrary values of $\varepsilon(\omega)=\varepsilon'(\omega)+i\varepsilon''(\omega)$. If $\varepsilon'(\omega)<0$, with $|\varepsilon'(\omega)|\gg\varepsilon''(\omega)$ and $|\varepsilon'(\omega)|\gg 1$, then for $\Omega_q\ll\omega$ we have

$$-\gamma_\alpha(1-\varepsilon(\omega))[\varepsilon(\omega_\alpha)\Gamma_\alpha+\gamma_\alpha]^{-1}\approx k_0(\Delta k_\alpha-i\Gamma_p)^{-1},\tag{12}$$

where

$$\Delta k_\alpha=k_\alpha\{(|\varepsilon'|-1)|\varepsilon'|^{-1}\}^{1/2}-k_{0\alpha},\tag{13}$$

$$\Gamma_p={}^1/_2k_0\varepsilon''(\omega)|\varepsilon'(\omega)|^{-2}\ll k_0.\tag{14}$$

From (12) it can be seen that the amplitudes of the scattered waves (9)–(11) undergo resonant growth when their wave vectors k_α coincide with the wave vectors of the free surface electromagnetic waves[10]:

$$k_\alpha=k_{0\alpha}\{|\varepsilon'|(|\varepsilon'|-1)^{-1}\}^{1/2}.\tag{15}$$

In the derivation of (8)–(11) it was assumed that $\xi_\alpha(t)=\text{const}$. This assumption is true if the characteristic time of variation of the slow amplitude of the surface acoustic wave ($\xi_{\mathbf{q}}(t)$), which is equal to $\gamma_{\mathbf{q}}^{-1}$, is much larger than the time $\tau_{\text{SEW}}=(c\Gamma_p)^{-1}$ for establishing the steady-state amplitudes of the surface electromagnetic waves.

3. EXCITATION OF SURFACE TEMPERATURE WAVE BY THE INTERFERENCE OF THE SURFACE ELECTROMAGNETIC WAVE AND THE EXTERNAL LIGHT WAVE

In order to describe the heating of the surface of the medium it is necessary to add the term $(4\pi)^{-1}\mathbf{E}\partial\mathbf{D}/\partial t$ to the right-hand side of the heat conduction equation, where $\mathbf{E}$ is defined by formula (6) and the displacement vector $\mathbf{D}$ is written in the form

$$\mathbf{D}=\varepsilon(\omega)\mathbf{E}_t\exp\{i\mathbf{k}_t\mathbf{r}-\gamma z-i\omega t\}+\sum_{\alpha=s,a}\varepsilon(\omega_\alpha)\mathbf{E}_\alpha'\exp\{i\mathbf{k}_\alpha\mathbf{r}-\gamma_\alpha z-i\omega_\alpha t\}.\tag{16}$$

The heat conductivity equation for a solid has the form

$$\frac{\partial T}{\partial t}=\chi\Delta T+\frac{1}{4\pi c_V}\mathbf{E}\frac{\partial\mathbf{D}}{\partial t},\qquad(17)$$

where $\chi=\varkappa/c_v$, χ is the thermal diffusivity, c_v is the specific heat per unit volume, and $\varkappa$ is the heat conductivity.

In the subsequent discussion we shall not be interested in the equilibrium heating and shall consider only the space-time modulation of the surface temperature associated with the product of $\mathbf{E}_t$ and $\mathbf{E}'_\alpha$ on the right hand side of (17) (Ref. 14). Using (16), we have in (17)

$$\mathbf{E}\frac{\partial\mathbf{D}}{\partial t}=\left\{2\omega\varepsilon''(\omega)+{}^1/_2\Omega_{\mathbf{q}}{}^2\frac{\partial\varepsilon''(\omega)}{\partial\omega}+i\omega\Omega_{\mathbf{q}}\frac{\partial\varepsilon'(\omega)}{\partial\omega}+i\Omega_{\mathbf{q}}\varepsilon'(\omega)\right\}$$

$$\times\{\mathbf{E}_t{}^*\mathbf{E}_s{}'\exp[-(\gamma_s+\gamma^*)z]+\mathbf{E}_t\mathbf{E}_a{}'^*\exp[-(\gamma_a{}^*+\gamma)z]\}$$

$$\times\exp(i\Omega_{\mathbf{q}}t-i\mathbf{qr})+\text{c.c.}$$

Let us limit ourselves to considering the case where the inequality $|\omega\varepsilon''(\omega)|\gg\Omega_{\mathbf{q}}\varepsilon'(\omega)|$ holds. Assuming, in addition, that the conditions $\Omega_{\mathbf{q}}\ll\omega$, $k_a\sim q\sim k_0\ll\gamma_0$ are satisfied and using formulas (8), (9)–(11), and (16) we obtain

$$(4\pi c_V)^{-1}\mathbf{E}\partial\mathbf{D}/\partial t\approx a_{\mathbf{q}}\xi_{\mathbf{q}}(t)\exp\{-i\mathbf{qr}-\gamma_0 z+i\Omega_{\mathbf{q}}t\}+\text{c.c.},\qquad(18)$$

where in the case of s-polarized pumping wave (the vector $\mathbf{E}_i$ is perpendicular to the plane of incidence)

$$a_{\mathbf{q}}=\frac{2\gamma_0\omega|\mathbf{E}_i|^2\cos^2\theta}{\pi c_V}\left\{\frac{\Gamma_p\sin^2\varphi_s}{\Delta k_s-i\Gamma_p}+\frac{\Gamma_p\sin^2\varphi_a}{\Delta k_a+i\Gamma_p}\right\},\qquad(19)$$

and in the case of p-polarized pumping wave ($\mathbf{E}_i$ lies in the plane of incidence)

$$a_{\mathbf{q}}=\frac{2\gamma_0\omega|\mathbf{E}_i|^2\cos^2\theta}{\pi c_V(\cos^2\theta+|\varepsilon'|^{-1})}\left\{\frac{\Gamma_p f_s}{\Delta k_s-i\Gamma_p}+\frac{\Gamma_p f_a}{\Delta k_a+i\Gamma_p}\right\};$$

$$f_a=\cos^2\varphi_a-\sin\theta\cos\varphi_a\qquad(20)$$

Here $\gamma_a+\gamma^*\approx\gamma_0=2k_0|\varepsilon'|^{1/2}$, $\cos\varphi_a=(k_t k_a)$, θ is the angle between $\mathbf{k}_0$ and the normal to the surface $z=0$ i.e., it is the angle of incidence of the pumping wave, and Δk_α and Γ_p are defined in (13) and (14).

Since the medium abuts the vacuum there is no heat exchange at the interface. Thus, with an accuracy to terms proportional to $\xi^2_{\mathbf{q}}$ the initial and boundary conditions can be written in the form

$$T(\mathbf{r},z,t=0)=T_0,\qquad\left.\frac{\partial T(\mathbf{r},z,t)}{\partial z}\right|_{z=0}=0,$$

$$T(\mathbf{r},z=\infty,t)=T_0.\qquad(21)$$

For times $t>\tau_{\text{SEW}}$ we shall set

$$\xi_{\mathbf{q}}(t)=\xi_0 e^{\gamma_{\mathbf{q}}t}\qquad(22)$$

in (18), and introduce the complex frequency

$$\Omega=\Omega_{\mathbf{q}}-i\gamma_{\mathbf{q}}.\qquad(23)$$

The temperature wave is established in a time

$$t\gtrsim t_T=1/2q^2\chi+(1+z^2q^2)^{1/2}/2q^2\chi\gg\tau_{\text{SEW}},$$

so that the solution of the problem (17), (21), taking into account (18) and (22) may be represented by the formula

$$T(\mathbf{r},z,t)-T_0\approx-\frac{\gamma_0}{\delta}\frac{a_{\mathbf{q}}}{i\Omega-\chi\gamma_0{}^2}\xi_0\exp\{-i\mathbf{qr}-\delta z+i\Omega t\}+\text{c.c.},\qquad(24)$$

where $\delta^2=q^2+i\Omega/\chi$, $\text{Re}\delta>0$, and the quantity $a_{\mathbf{q}}$ is defined in (19) and (22). Expression (24) is valid for the conditions q, $|\delta|\ll\gamma_0$.

4. GENERATION OF COHERENT SURFACE ACOUSTIC WAVES AND STATIC RIPPLES BY LASER PUMPING

The temperature wave (24) gives rise to a driving force in the equation for the deformation vector $\mathbf{u}$ of the medium[15]:

$$\rho\ddot{\mathbf{u}}=\rho c_t{}^2\Delta\mathbf{u}+\rho(c_l{}^2-c_t{}^2)\,\text{grad div}\,\mathbf{u}+\eta\Delta\dot{\mathbf{u}}+(\eta/3+\zeta)\,\text{grad div}\,\dot{\mathbf{u}}-K\alpha\,\text{grad}\,T,\qquad(25)$$

where ρ is the density of the medium, c_l and c_t are the longitudinal and transverse velocities of sound, η and ζ are the first and second coefficients of viscosity, K is the bulk modulus, and α is the thermal expansion coefficient.

At the free surface, neglecting ponderomotive forces, to the approximation linear in $\xi_{\mathbf{q}}$ the boundary conditions at $z=0$ are written in the form

$$\frac{\partial u_x}{\partial z}+\frac{\partial u_z}{\partial x}=0,\qquad\frac{\partial u_y}{\partial z}+\frac{\partial u_z}{\partial y}=0,$$

$$-\frac{(T-T_0)K\alpha}{\rho}+(c_l{}^2-2c_t{}^2)\left(\frac{\partial u_x}{\partial x}+\frac{\partial u_y}{\partial y}\right)+c_l{}^2\frac{\partial u_z}{\partial z}=0.\qquad(26)$$

Let us take the x axis along the direction of the vector $\mathbf{q}$. Then in (24), the value of $T-T_0$ is independent of y. Therefore $u_y=0$ and u_x and u_z are independent of y. Consequently, the second of the conditions (26) is satisfied automatically.

The solution of the problem (24), (25), and (26) has the form

$$u_x=\left(-\varkappa_t Ae^{-\varkappa_t z}-qBe^{-\varkappa_l z}+\frac{iqR\xi_0 e^{-\delta z}}{\Omega^2+c_l{}^2(\delta^2-q^2)+i\Omega\eta'(\delta^2-q^2)/\rho}\right)\times e^{-iqx+i\Omega t}+\text{c.c.},$$

$$u_z=\left(iqAe^{-\varkappa_t z}+i\varkappa_l Be^{-\varkappa_l z}+\frac{\delta R\xi_0 e^{-\delta z}}{\Omega^2+c_l{}^2(\delta^2-q^2)+i\Omega\eta'(\delta^2-q^2)/\rho}\right)\times e^{-iqx+i\Omega t}+\text{c.c.}\qquad(27)$$

Here

$$R=\frac{\gamma_0 K\alpha}{\rho\delta}\frac{a_{\mathbf{q}}}{i\Omega-\chi\gamma_0{}^2},\qquad\varkappa_t{}^2=q^2-\frac{\Omega^2}{c_t^2+i\Omega\eta/\rho},\qquad(28)$$

and $\varkappa_l$ is obtained from $\varkappa_t$ by interchanging $c_t\to c_l$, $\eta\to\eta'=4/3\eta+\zeta$; while A and B are constants.

Since the modulation of the surface relief (3) and (22) is produced by the surface acoustic wave, $[\xi(\mathbf{r},t)=u_z(\mathbf{r},z=0,t)]$, we have, from (3), (22), and (27),

$$\xi_0=iqA+i\varkappa_l B+\delta R[\Omega^2+c_l{}^2(\delta^2-q^2)+i\Omega\eta'(\delta^2-q^2)/\rho]^{-1}\xi_0.\qquad(29)$$

Now substituting (27) into the boundary conditions (26), we obtain, together with (29), a system of three homogeneous linear algebraic equations in the constants A, B, and ξ_0. The condition (det = 0) that this system of equations have a solution leads to a dispersion equation in Ω. Neglecting in this equation small terms proportional to $\eta^2|\mathbf{E}|^2$, we write it in the form

$$c_l^2(q^2-\varkappa_l^2)(q^2+\varkappa_t^2)-2c_t^2(q^2+\varkappa_t^2)q^2+4c_t^2q^2\varkappa_l\varkappa_t = \frac{c_t^2}{c_l^2}\frac{q^4-\varkappa_{0t}^4}{\delta+\varkappa_{0l}}R, \tag{30}$$

where

$$\varkappa_{0l}^2=q^2-\Omega^2/c_l^2, \qquad \varkappa_{0t}^2=q^2-\Omega^2/c_t^2. \tag{31}$$

Substituting (23), (28), and (31) into (30) and setting the real and imaginary parts of this equation equal to zero, we obtain two equations for determining $\Omega_\mathbf{q}$ and $\gamma_\mathbf{q}$. In the absence of an external field ($R = 0$) and of dissipation ($\eta = \eta' = 0$) we obtain from (30) two solutions.

1) A dynamic solution (surface acoustic waves). Taking into account the relation $c_l^2(q^2 - \varkappa_{0l}^2) = c_t^2(q^2 - \varkappa_{0t}^2)$, which follows from (31), we reduce (30) to the following form:

$$(\varkappa_t^2+q^2)^2=4q^2\varkappa_\perp\varkappa_\|,$$

$$\varkappa_\|^2=q^2-\Omega_\mathbf{q}^2/c_l^2, \quad \varkappa_\perp^2=q^2-\Omega_\mathbf{q}^2/c_t^2. \tag{32}$$

The solution of (32) is the usual formula for a Rayleigh surface acoustic wave[15]:

$$\Omega_\mathbf{q}=qc_t\beta, \tag{33}$$

where, as can be seen from (32) the coefficient β is a function of c_t/c_l. For various materials $0.874\leqslant\beta\leqslant 0.955$.[13,15]

2) A static solution: $\Omega_\mathbf{q} = 0$ and $\gamma_\mathbf{q} = 0$ for all $\mathbf{q}$.

Let us now investigate how both of these solutions change when $R \neq 0$ and when $\eta\neq 0$ and $\eta'\neq 0$.

a) We substitute (23), (28), and (31) into (30). Taking into account that $|\gamma_\mathbf{q}|\ll\Omega_\mathbf{q}$, we linearize (30) with respect to $\gamma_\mathbf{q}$, η, and η'. The real part of the linearized equation gives once again Eq. (32), whose solution is the relation (33). Since $\Omega_\mathbf{q}$ is large ($\Omega_\mathbf{q}\gg|\gamma_\mathbf{q}|$), we have neglected the effect of the pumping field on $\Omega_\mathbf{q}$ in the derivation of (32). The imaginary part of the linearized equation defines an expression for $\gamma_\mathbf{q}$ which, taking into account (32), (33), and (28), can be written in the form

$$\gamma_\mathbf{q}=I/\sigma_0-\gamma_\eta, \tag{34}$$

where

$$\sigma_0=\frac{\varkappa_\|}{\varkappa_\perp}+\frac{\varkappa_\perp c_t^2}{\varkappa_\| c_l^2}-\frac{q^2+\varkappa_\perp^2}{q^2}\approx\frac{\varkappa_\|}{\varkappa_\perp}\leqslant 1, \tag{35}$$

and the damping constant of the surface acoustic wave due to the viscosity of the medium is

$$\gamma_\eta=\frac{\eta\beta^2q^2}{4\sigma_0\rho}\left(\frac{2\varkappa_\|}{\varkappa_\perp}-\frac{q^2+\varkappa_\perp^2}{q^2}\right)+\frac{\eta'\beta^2q^2c_t^2}{4\sigma_0\rho c_l^2}\left(\frac{2\varkappa_\perp c_t^2}{\varkappa_\| c_l^2}-\frac{q^2+\varkappa_\perp^2}{q^2}\right)\approx\frac{\eta\beta^2q^2}{2\rho}.$$

In addition,

$$I=-\frac{c_t^2}{4c_l^2}\frac{q^4-\varkappa_\perp^4}{q^2\Omega_\mathbf{q}}\frac{K\alpha}{\rho\chi\gamma_0}\operatorname{Im}a_\mathbf{q}[(1-i\Omega/\chi\gamma_0^2)\delta(\delta+\varkappa_\|)]^{-1}, \tag{36}$$

where the quantity $a_\mathbf{q}$ is defined in (19) and (20).

As can be seen from (34) and (36), the growth rate is expressed by the imaginary part of the product of two factors, one of them resulting from the surface electromagnetic wave (proportional to $a_\mathbf{q}$) and the other a temperature factor.

Depending on the pumping wavelength and on the parameters of the material, two limiting cases should be distinguished. If $\chi\gamma_0^2\gg|\Omega|$ and $\chi q\ll\beta c_t$, then in (36)

$$\left(1-\frac{i\Omega}{\chi\gamma_0^2}\right)\delta(\delta+\varkappa_\|)\approx\frac{i\Omega_q}{\chi}=\frac{i\beta c_tq}{\chi}$$

and the expression for the increment is written in the form

$$\gamma_\mathbf{q}=\frac{K\alpha\varkappa_\perp}{4\beta^2c_l^2\rho\gamma_0\varkappa_\|}\operatorname{Re}a_\mathbf{q}-\frac{\eta\beta^2q^2}{2\rho}. \tag{37}$$

However, if $\chi\gamma_0^2\gg|\Omega|$ and $\chi q\gg\beta c_t$, then

$$\gamma_\mathbf{q}=-\frac{c_t^2}{c_l^2}\frac{K\alpha\varkappa_\perp}{8\Omega_\mathbf{q}\rho\chi\gamma_0\varkappa_\|}\operatorname{Im}a_\mathbf{q}-\frac{\eta\beta^2q^2}{2\rho}.$$

Let us now estimate the critical intensity for the excitation of surface acoustic waves. For normal incidence $\mathbf{k}_i = 0$ in (2) and we find that for resonance surface acoustic waves $q = k_\alpha \sim k_0$ (see Ref. 15). From (37) and (19) we have, with $\sin^2\varphi_s = \sin^2\varphi_a = 1$

$$\max(\operatorname{Re}a_\mathbf{q})=2\gamma_0\omega|\mathbf{E}_i|^2/\pi c_V.$$

Then from (7) and the condition $\gamma_\mathbf{q} = 0$ we obtain an expression for the critical intensity

$$|\mathbf{E}_i|^2_{\text{th}}=\gamma_\eta\frac{2\pi\beta^2c_l^2\rho c_V}{K\alpha\omega}\left(\frac{1-\beta^2c_t^2/c_l^2}{1-\beta^2}\right)^{1/2}. \tag{38}$$

In the case of copper we have $\alpha = 7\cdot10^{-5}$ deg^{-1}, $K = 1.4\cdot10^{12}$ dyn/cm^2, $c_v = 4\cdot10^7$ erg/cm^3-deg, $\beta = 0.93$, $c_t = 2.3\cdot10^5$ cm/sec, $c_t^2/c_l^2 = 0.237$, and $\rho = 9$ g/cm^3 (Ref. 16). For a pumping wavelength $\lambda = 1$ μm, ($\omega = 2\cdot10^{15}$ sec^{-1}), for $\eta = 3.6\cdot10^{-2}$ dyn-sec/cm^2, we have from (35) $\gamma_\eta = 6.8\cdot10^6$ sec^{-1}. Then from (38) we obtain for the threshold intensity for the excitation of surface acoustic waves

$$(I_i)_{\text{th}}=\frac{c|\mathbf{E}_i|^2_{\text{th}}}{2\pi}=1.9\cdot10^7\ \text{W/cm}^2$$

b) Let us now consider the quasistatic case ($\Omega\approx 0$). We substitute (23), (28), and (31) into (30) and shall assume

$$|\Omega|/\chi q^2, \quad |\Omega|/qc_t, \quad |\Omega|/\chi\gamma_0^2, \quad \eta|\Omega|/\rho c_t^2\ll 1.$$

Then, from (30) we obtain

$$\gamma_\mathbf{q}=-\frac{2}{3}\left[\frac{K\alpha\operatorname{Re}a_\mathbf{q}}{\gamma_0\rho(c_l^2-c_t^2)}+2\chi q^2\right], \quad \Omega_\mathbf{q}=-\frac{2}{3}\frac{K\alpha\operatorname{Im}a_\mathbf{q}}{\gamma_0\rho(c_l^2-c_t^2)}. \tag{39}$$

For normal incidence, $\operatorname{Im}a_\mathbf{q} = 0$ and $\Omega_\mathbf{q} = 0$. In (39), as can be seen from (19) or (20), $\operatorname{Re}a_\mathbf{q} < 0$ and $|\operatorname{Re}a_\mathbf{q}|$ is maximal for $\Delta k_s = \Delta k_a = -\Gamma_p$. Here the relaxation mode of $\gamma_\mathbf{q}$ is soft:

$$\gamma_\mathbf{q}=\frac{2}{3}\left[\frac{2\omega K\alpha|\mathbf{E}_i|^2}{\pi c_V\rho(c_l^2-c_t^2)}-2\chi q^2\right].$$

The critical intensity (at which $\gamma_{\mathbf{q}} = 0$) for copper for $\lambda = 10.6\,\mu$m is $I_{\text{th}} = 2\cdot 10^8$ W/cm². If $I_i > I_{\text{th}}$, then a static sinusoidal network builds up on the surface with a time increment $\gamma_{\mathbf{q}}$ and having a period which, as follows from (2), where $\mathbf{k}_t = 0$, is equal to the pumping wavelength $\lambda = 2\pi/k_0$ and the vector $\mathbf{q}$ parallel to $\mathbf{E}_i$.

5. CHARACTERISTICS OF SURFACE ACOUSTIC WAVES AND THEIR DEPENDENCE ON THE PUMPING PARAMETERS

We shall limit this investigation to the case (37), which is realized in solid materials. From (37), with (19) and (20), it can be seen that the increment is maximal for those surface acoustic waves for which the resonance condition $\Delta k_\alpha = \Gamma_p$ is satisfied, i.e., according to (13)

$$k_\alpha = (|\varepsilon'|/(|\varepsilon'|-1))^{1/2}(k_{0\alpha}+\Gamma_p)\approx k_0. \tag{40}$$

Thus, in (2) the value of $|\mathbf{k}_\alpha|$ is fixed and the direction of the vector $\mathbf{k}_\alpha$ remains arbitrary. This means that the possible vectors $\mathbf{k}_\alpha$ terminate on a circle of radius k_0 (see Fig. 1). Then from (2) the modulus and wave vector of the coupled surface acoustic wave are determined as functions of the orientation of the vector $\mathbf{k}_\alpha$ of the surface electromagnetic wave:

$$q = k_0(1+\sin^2\theta - 2\sin\theta\cos\varphi_\alpha)^{1/2},\quad \cos\delta_\alpha = k_0(\cos\varphi_\alpha - \sin\theta)/q, \tag{41}$$

where $\cos\varphi_\alpha = (\hat{k}_t\hat{k}_\alpha)$.

All possible cases of mutual orientation of the vectors $\mathbf{q}$ and $\mathbf{k}_t$ are divided into two classes.

a) Double resonance of the surface electromagnetic waves: the vectors $\mathbf{q}$ and $\mathbf{k}_t$ are such that the resonance conditions (40) are satisfied simultaneously for $\mathbf{k}_s$ and $\mathbf{k}_a$. In this case both terms with $\alpha = s$ and $\alpha = a$ give the same contributions in (19), (20), and (37). For s-polarization pumping and sufficiently small angles of incidence θ double resonance is realized for any orientation of $\mathbf{q}$ with respect to $\mathbf{k}_t$ (here $|\mathbf{k}_s| \sim |\mathbf{k}_a| \sim q$). For sufficiently large θ it is realized only for those orientations shown in Fig. 2. In this case $\cos\varphi_s = \cos\varphi_a = \sin\theta$. In the case of p polarization, double resonance occurs only for quite small angles θ (with the orientation shown in Fig. 2 it does not occur, since here $\cos\varphi_a = \sin\theta$ and $f_\alpha = 0$, so that $\gamma_{\mathbf{q}} \leqslant 0$).

b) The single-resonance case: The resonance conditions are satisfied only for one of the vectors, $\mathbf{k}_s$ or $\mathbf{k}_a$ (see Fig. 1). In this case the main contribution to (19), (20), and (37) will be given by only one of the terms, that with $\alpha = s$ or $\alpha = a$. For s polarization the single-resonance case occurs for $\theta \neq 0$

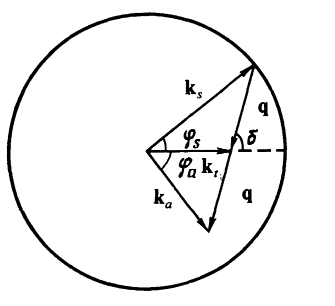

FIG. 1. Single-resonance case of orientation of the vector $\mathbf{q}$ relative to the vector $\mathbf{k}_t$. Radius of the circle is k_0. Here $|k_s| \neq |k_a|$.

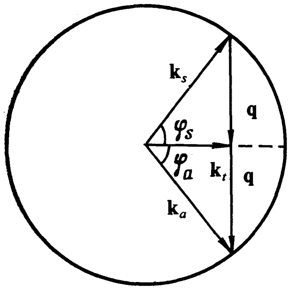

FIG. 2. Orientation of the vector $\mathbf{q}$ relative to the vector $\mathbf{k}_t$ in the double resonance case. Radius of the circle is k_0. Here $|k_s| = |k_a|$ and $\cos\varphi_s = \cos\varphi_a$.

and $\cos\varphi_a \neq \sin\theta$. For p polarization it occurs when $\theta \neq 0$.

Let us consider first the case of an s-polarized pumping wave. We consider the resonance conditions (40) to be satisfied only for $\mathbf{k}_s$. In this single-resonance case ($\theta \neq 0$ and $\cos\varphi_s \neq \sin\theta$) according to (19) and (37) the increment $\gamma_{\mathbf{q}}$ can be written in the form

$$\frac{\gamma_{\mathbf{q}}}{b_s} = -\left(\cos\varphi_s - \frac{a}{b_s}\sin\theta\right)^2 + \left(1-\frac{a}{b_s}\right)\left(1-\frac{a}{b_s}\sin^2\theta\right), \tag{42}$$

where

$$b_s = \frac{K\alpha}{4\beta^2 c_t^2\rho}\,\frac{\varkappa_\perp}{\varkappa_\parallel}\,\frac{\omega|\mathbf{E}_i|^2}{\pi c_V}\cos^2\theta,\qquad a = \eta\frac{\beta^2 k_0^2}{2\rho}.$$

In the case of double resonance, when $\theta \neq 0$ and $\cos\varphi_a = \sin\theta$ (Fig. 2), we have from (19) and (37)

$$\gamma_{\mathbf{q}}/b_s = 2(1 - a/2b_s)(1-\sin^2\theta). \tag{43}$$

For small angles of incidence $\theta \approx 0$ and for any $\cos\varphi_s$ we have from (19) and (37)

$$\gamma_{\mathbf{q}}/b_s = -2\cos^2\varphi_s + 2(1 - a/2b_s). \tag{44}$$

Curves of $\gamma_{\mathbf{q}}/b_s$ as a function of $\cos\varphi_s$ according to formulas (42) to (44) are shown in Fig. 3.

An analysis of formulas (42)–(44) permits us to draw the following conclusions:

1) For s-polarization pumping incident at an angle θ generation ($\gamma_{\mathbf{q}} > 0$) of a continuum of surface acoustic waves is possible; their wave vectors are determined by formulas (41) and their frequencies by (33). The necessary condition

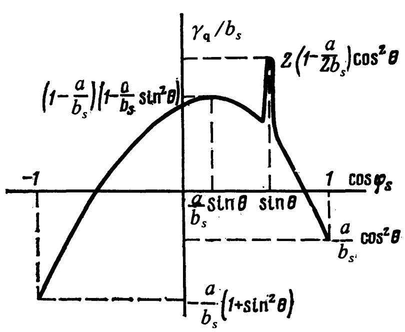

FIG. 3. Growth rate $\gamma_{\mathbf{q}}$, (42) and (43) as a function of $\cos\varphi_s$ in the case of non-normal incidence ($\theta \neq 0$) of s polarized pumping wave. For normal incidence ($\theta = 0$) the dependence of $\gamma_{\mathbf{q}}$ on $\cos\varphi_s$ is determined by formula (44).

for the excitation of these surface acoustic waves is the inequality $b_s > a/2$.

2) If $\theta \neq 0$ and

$$2\cos^2\theta\left(1-\frac{a}{2b_s}\right)>\left(1-\frac{a}{b_s}\right)\left(1-\frac{a}{b_s}\sin^2\theta\right)$$

(see Fig. 3) then there is an absolute maximum of the increment $\gamma_{\mathbf{q}}$ for a given θ when the vector $\mathbf{q}$ is oriented such that $\cos\varphi_s = \cos\varphi_a = \sin\theta$. In this case surface acoustic waves will be generated with a wave vector $\mathbf{q}$ which, from (41), is determined by the formulas

$$q\perp\mathbf{k}_t \quad (\text{т. е. } \mathbf{q}\|\mathbf{E}_i); \qquad q=k_0\cos\theta. \tag{45}$$

3) If

$$\left(1-\frac{a}{b_s}\right)\left(1-\frac{a}{b_s}\sin^2\theta\right)>2\left(1-\frac{a}{2b_s}\right)\cos^2\theta \text{ и } b_s>a$$

(see Fig. 3) then $\gamma_{\mathbf{q}}$ has an absolute maximum for $\cos\varphi_s = (a/b_s)\sin\theta$ and the waves generated most intensely will be two surface acoustic waves the directions of whose $\mathbf{q}$ vectors are determined by the angles δ and $2\pi - \delta$ (the angle δ is defined in Fig. 1), with

$$q=k_0\left(1+\left(1-\frac{2a}{b_s}\right)\sin^2\theta\right)^{1/2}, \quad \cos\delta=\frac{k_0(1-a/b_s)\sin\theta}{q}. \tag{46}$$

4) For $\theta = 0$ a continuum of surface acoustic waves is generated, with increments given by (44). The most intense of these are surface acoustic waves with parameters near to those of surface acoustic waves with $\cos\varphi_s = 0$. For these waves, $q \approx k_0$ and $\mathbf{q}\|\mathbf{E}_i$.

We shall now consider the case of a p polarized pumping wave. From (20) and (37) it can be seen that for $f_\alpha > 0$ the increment is maximal for a surface acoustic wave having $\Delta k_\alpha = \Gamma_p$, and for $f_\alpha < 0$ it is maximal for the surface acoustic wave having $\Delta k_\alpha = -\Gamma_p$. It follows from these resonance conditions that for these surface acoustic waves, as in (40) also, $k_\alpha \approx k_0$. In this discussion we shall assume that this resonance condition is satisfied only for $\alpha = s$.

In the single-resonance case ($\theta \neq 0$) we have for the increment $\gamma_{\mathbf{q}}$, from (20) and (37),

$$\frac{\gamma_{\mathbf{q}}}{b_p}=|\cos^2\varphi_s-\sin\theta\cos\varphi_s|-\frac{a}{b_p}(1+\sin^2\theta-2\sin\theta\cos\varphi_s),$$

$$b_p=\frac{K\alpha}{4\beta^2c_l^2\rho}\frac{\varkappa_\perp}{\varkappa_\|}\frac{\omega|\mathbf{E}_i|^2\cos^2\theta}{\pi c_T(\cos^2\theta+|\varepsilon'|^{-1})}, \tag{47}$$

where the quantity a is defined in (42).

Curves of $\gamma_{\mathbf{q}}$ (47) as a function of $\cos\varphi_s$ in the case of sufficiently large pumping intensities ($b_p > 2a$) are shown in Fig. 4. The magnitude and direction of $\mathbf{q}$ for the corresponding surface acoustic waves are given by formulas (41).

In the double resonance case ($\theta \neq 0$, $\cos\varphi_s = -\cos\varphi_a$, and $k_s = k_a$) the increment $\gamma_{\mathbf{q}}$ has the form

$$\gamma_{\mathbf{q}}/b_p=2\cos^2\varphi_s-a/b_p. \tag{48}$$

An analysis of formulas (47) and (48) along with Fig. 4 permits us to draw the following conclusions:

1) For intensities $b_p > a(1-\sin\theta)$, a surface acoustic

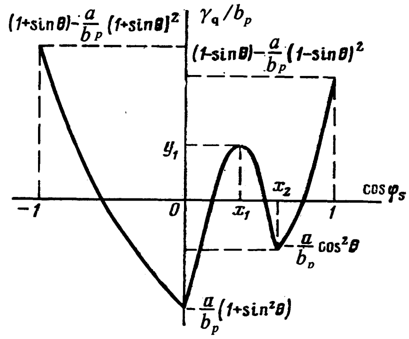

FIG. 4. Growth rate $\gamma_{\mathbf{q}}$ (47) as a function of $\cos\varphi_s$ in the case of nonnormal incidence ($\theta \neq 0$) of p polarized pumping wave. For normal incidence ($\theta = 0$) the dependence of $\gamma_{\mathbf{q}}$ on $\cos\varphi_s$ is determined by formula (48). $x_1 = \frac{1}{2}(1+2a/bp)\sin\theta$, $x_2 = \sin\theta$, $y_1 = \frac{1}{4}(1+4a^2/b_p^2)\sin^2\theta - a/b_p$.

wave is generated which corresponds to $\cos\varphi_s = 1$, with the $\mathbf{q}$ vector antiparallel to $\mathbf{k}_t$:

$$\mathbf{q}\uparrow\downarrow\mathbf{k}_t, \qquad q=k_0(1-\sin\theta). \tag{49}$$

With increasing intensity ($b_p > a(1+\sin\theta)$), in addition to the last-mentioned wave, another is excited having the $\mathbf{q}$ vector parallel to $\mathbf{k}_t$:

$$\mathbf{q}\uparrow\uparrow\mathbf{k}_t, \quad q=k_0(1+\sin\theta). \tag{50}$$

2) For large intensities ($b_p \gg a$) the growth increment of the surface acoustic wave corresponding to (50) is larger than that of the wave corresponding to (49).

We have investigated the process of self-excitation of a travelling surface acoustic wave with wave vector $\mathbf{q}$ given in the form (3). An analogous investigation shows that for a surface acoustic wave with wave vector $-\mathbf{q}$ the growth increment is $\gamma_{-\mathbf{q}} = \gamma_{\mathbf{q}}$. This means that two oppositely-directed surface acoustic waves are excited at once and their superposition can form a standing surface acoustic wave.

6. CONCLUSION

The process of laser excitation of coupled surface acoustic waves and surface electromagnetic waves that we have investigated here is analogous to the induced scattering of light caused by absorption in the bulk.[17] However, in contrast to the latter situation, the case $\gamma_\alpha \gg \gamma_\eta$ can occur at the surface so therefore the scattered wave (surface electromagnetic wave) is adiabatically tuned to the surface acoustic wave and the latter is also amplified (in the bulk absorption case, conversely, $\gamma_\alpha \ll \gamma_\eta$ and the acoustic phonon is tuned to the scattered electromagnetic wave).

It is important to note that the excitation of surface acoustic waves by this mechanism is possible not only in media having $\varepsilon' < -1$, but also in a medium with an arbitrary value of ε'. In the general case the role of the surface electromagnetic wave is taken by the fields diffracted by surface roughness [described by the general formulas (9)–(11)]. Investigation of these cases is beyond the scope of this paper. Let us only note that the calculations show that in this case only surface acoustic waves with wave vectors $\mathbf{q}\perp\mathbf{E}_i'$ can be generated.

Observation of the generation of surface acoustic wave by the mechanism that we have studied here may be facilitated by the use of a scheme of active spectroscopy,[14] i.e., the use of two laser beams, where the difference of the projections of their wave vectors gives the wave vector **q** of the surface acoustic wave.

At the present time a number of experiments have been performed in which the formation of reversible and irreversible networks have been reliably recorded on the surfaces of solids: nickel,[1] copper,[8] aluminum,[18] and silicon, germanium, and gallium arsenide,[2,3,5,7] sodium chloride, and fused quartz.[4,9] The experimentally determined periods and orientations of the surface networks as functions of frequency and angle of incidence and polarization of the pumping wave, as well as the characteristic intensities at which the formation of the networks is observed correspond to the regularities in the behavior of the excitation of surface acoustic waves found in this investigation.

We note that the vaporization mechanism leads to exactly the same dependences on the pumping parameters for the characteristics of the dominant surface networks as in the case of the excitation of surface acoustic waves.

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