Theory of stimulated light scattering by trap-charging waves

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A theory of stimulated scattering of light by trap-charging waves is proposed. Scattering is possible in electrooptic crystals in the presence of a constant electric field E_0 maintained by a voltage source or by the photovoltaic effect. The role of the stimulating force that builds up the charge-exchange wave is played by the inhomogeneous light intensity that modulates, in the course of the interference, the trap-photoionization rate. It is shown that the intensity I of this effect does not have the high threshold typical of other forms of stimulated scattering, since the scattered-wave gain is independent of I. When the field E_0 exceeds a critical value E_{0c} the stationary regime of the spatial gain in a sample without a cavity should give way to a nonstationary emission regime that is not realizable in other types of stimulated scattering. As a result, the intensity of the anti-Stokes wave increases to values comparable with the intensity of the incident light, independently of the number of priming fluctuations. The theory explains a recently observed effect, that of self-waves of photoinduced scattered light, which does not agree with the heretofore considered scattering models.

1. INTRODUCTION

Nonlinear optical effects in photorefracting crystals, due to energy exchange between crossing light beams, are under active study in dynamic holography. The energy exchange is the result of self-diffraction, or diffraction of light by a holographic phase grating formed in a crystal by interference of light waves.[1] The grating is formed via the linear electrooptic effect by the field of the space charge produced when photoexcited electrons are trapped by impurity centers. An example of such effects is photoinduced scattering of light[2–6] and, in particular, photoinduced reflection,[7,8] whereby a light beam incident on an initially transparent crystal is almost completely converted into a reflected beam.

It is well known that stationary energy exchange is possible only in the case of a nonlocal response of the medium, i.e., in the presence of a $\pi/2$ phase shift between the light-intensity distribution and the grating. Physical nonlocal-response mechanisms are attributed to diffusion of electrons inhomogeneously excited by light[1,3,4,7] or to a circular photovoltaic current. [In the latter case (see Refs. 9 and 10) the energy exchange takes place at a special geometry of the incidence and polarization of the light, which is not considered in the present paper.] In theoretical investigations of these effects it is assumed that the diffraction gratings produced in the crystal are immobile. It has been recently shown, however (see Refs. 11 and 12) that in the presence of a constant electric field E_0 an important role is played in a number of effects observed in photorefracting crystals by weakly damped trap-charging waves, which were investigated earlier in compensated semiconductors. In typical experiments, the field E_0 is applied to a crystal with the aid of a voltage source, or is produced as a result of the linear photovoltaic effect. It is remarkable that the trap-charging wave constitutes a diffraction grating moving along the field E_0, since the low-frequency field of the wave modulates the refractive index of the crystal.

We propose in this paper a theory of stimulated scattering of light by trap-charging waves (see also Ref. 14). The role of the driving force that launches a trap-charging wave is the interference pattern of an incident light wave and a scattered one; this pattern modulates the rate of photoionization of the traps. We show that this process should not have the high threshold of the light-intensity (I) typical of other types of stimulated scattering (e.g., Brillouin scattering), since the gain α of the scattered wave, just as in photoinduced scattering by immobile gratings, does not depend on I. At sufficiently high values of the field E_0, when the drift length of the free electrons exceeds the diffusion length, scattering by trap-charging waves should be much more effective than scattering by immobile gratings.

We show that the linear photovoltaic current accompanying the trap photoionization can increase the scattering substantially, since this current, according to Ref. 12, decreases the damping of the trap-charging waves.

If the gain α exceeds the reciprocal damping length of the trap-charging wave (if the field E_0 exceeds a critical value E_{0c}), the stationary-amplification regime should give way to a nonstationary emission regime, in which the back-scattered anti-Stokes wave and the trap-charging wave increase exponentially with time until the incident light is essentially depleted. Note that a similar regime is possible theoretically also for stimulated Brillouin scattering,[15,16] but it requires a light intensity higher by many orders than the amplification regime, and is therefore not realized in experiment. The positive sign of the frequency shift of the scattered light in the emission regime is due to the specific feature of the dispersion law $\omega \propto K^{-1}$ of a trap-charging wave with a wave vector $\mathbf{K}$, wherein the group velocity is directed opposite to the phase velocity.

The theory of the generation regime explains qualitatively the effect of scattering self-waves[17] recently observed in lithium niobate crystals and not interpreted theoretically so far. What makes the effect unusual is that the scattered light appears in a direction opposite to that expected theoret-

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ically when immobile gratings are considered, and also that it is nonstationary. The conclusions of the present paper are in good agreement with the experimental results.

2. PRINCIPAL EQUATIONS

Let a noncentrosymmetric crystal be subject to coherent illumination in the presence of a constant electric field $\mathbf{E}_0$. We assume for the sake of argument that the crystal is uniaxial and that the field $\mathbf{E}_0$ is directed along the optical axis. The field $\mathbf{E}_0$ can be either the result of the photovoltaic effect,[18] or due to connecting the crystal to a voltage source. We begin with the second of these variants, neglecting the photovoltaic current. We confine ourselves for simplicity to normal incidence of the light (see Fig. 1) through a transparent (negative) electrode and neglect the reflection from the crystal boundary. Assume that photoionization of impurity centers, electron transport in the conduction band, and their recapture by the free centers which act as traps, all take place in the crystal. Under these conditions there can propagate along the field $\mathbf{E}_0$ weakly damped trap-charging waves. These waves are manifested by oscillations of the degree f of occupancy of the traps by electrons, the density n of the electrons in the conduction band, and of the electric field E:

$$(f-f_0)/\tilde{f}=(n-n_0)/\tilde{n}=(E-E_0)/\tilde{E}=\exp(iKz-i\omega t).$$

Let f_0, n_0, E_0, $\tilde{f}$, $\tilde{n}$, and $\tilde{E}$ be the mean values and the amplitudes of the oscillations of the corresponding quantities, and let the z axis be chosen along the direction of the field $\mathbf{E}_0$.

In the case of uniform illumination, the electric fluctuations of the crystal contain trap-charging waves with a continuous set of wave vectors $\mathbf{K}$. The alternating field of each wave modulates the refractive index of the crystal n, with an amplitude

$$\tilde{n}_r=rn_r^3\tilde{E}/2, \tag{1}$$

where r is the component of the electrooptic-coefficients tensor and is determined by the direction of the applied field $\mathbf{E}_0$. The trap-charging waves are thus phase gratings from which the light propagating in the crystal is scattered. In the geometry considered (see the figure) the scattering is backward and in view of the Doppler effect the scattered wave is anti-Stokes, $\omega_2=\omega_1+\omega$, where ω, ω_1, and ω_2 are respectively the frequencies of the trap-charging wave and of the incident and scattered light. Obviously, the main contribution to the scattering is made by trap-charging waves that satisfy the Bragg condition $\mathbf{K}=\mathbf{k}_2-\mathbf{k}_1$, where $\mathbf{k}_1$ and $\mathbf{k}_2$ are the wave vectors of the incident and scattered light. Since $\omega\ll\omega_1$, ω_2, we have $k_2=k_1=k=\omega_1 n_r/c$, $K=2k$.

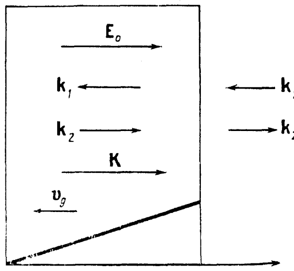

FIG. 1. Diagram of stimulated scattering of light by charge-trapping waves. The arrows show the directions of constant field $\mathbf{E}_0$, of the wave vectors $\mathbf{k}_1$ of the incident waves, $\mathbf{k}_2$ of the scattered light, and $\mathbf{K}$ of the trap-charging wave, and also the direction of its group velocity $\mathbf{v}_g$. The thick line shows the distribution of the intensity of the scattered light during the earlier stage of the process.

From the Maxwell equations we get for the smoothly varying amplitudes $\mathscr{E}_1$ and $\mathscr{E}_2$ of the incident and scattered light, with absorption disregarded (see Ref. 15)

$$\partial\mathscr{E}_1/\partial z=-i(\tilde{n}_r^*/2n_r)k\mathscr{E}_2, \tag{2}$$

$$\frac{\partial\mathscr{E}_2}{\partial z}=i(\tilde{n}_r/2n_r)k\mathscr{E}_1. \tag{3}$$

The quantity $\tilde{n}_r\propto\tilde{E}$ in these equations depends in turn on the distributions of $\mathscr{E}_1$ and $\mathscr{E}_2$, for in the case of interference between the incident and scattered waves the inhomogeneous light intensity modulates the rate of electron emission into the conduction band and excites the trap-charging wave. It is this feedback which leads to the stimulated scattering.

Let us determine the missing relation between n_r, $\mathscr{E}_1$, and $\mathscr{E}_2$, confining ourselves to weak scattering, $\mathscr{E}_2\ll\mathscr{E}_1$. The trap-occupancy balance equation takes in the considered model the form

$$\partial(Nf)/\partial t=\Gamma N(1-f)n-SINf-\hat{g}, \tag{4}$$

where N is the density of the impurity centers, S the photoionization cross section, Γ the coefficient of electron capture by a trap, and $\hat{g}$ a random function that describes the fluctuation of the photoionization and capture rates. We neglect thermal emission, assuming the impurity level to be deep enough. For the mean values we have

$$n_0=SI_0f_0\Gamma^{-1}(1-f_0)^{-1}.$$

After linearization, in the case

$$\delta f=f-f_0\ll f_0,\quad \delta n=n-n_0\ll n_0,\quad \delta I=I-I_0\ll I_0$$

we obtain

$$\partial(N\delta f)/\partial t=\delta n/\tau-N\delta f/\tau_1-SNf_0\delta I-\hat{g}, \tag{5}$$

where $\tau=[\Gamma N(1-f_0)]^{-1}$ is the average electron lifetime in the conduction band and $\tau_1=f_0(\Gamma n_0)^{-1}$ is the characteristic trap-occupancy relaxation time. Expression (5) must be supplemented by the continuity and Poisson equations:

$$\frac{\partial\delta n}{\partial t}-\frac{1}{e}\operatorname{div}\mathbf{j}=0,\quad \mathbf{j}=e\mu(n_0\delta\mathbf{E}+\mathbf{E}_0\delta n)+eD\nabla\delta n, \tag{6}$$

$$\operatorname{div}\delta\mathbf{E}=-\frac{\varepsilon e}{4\pi}(N\delta f+\delta n). \tag{7}$$

where $\delta\mathbf{E}=\mathbf{E}-\mathbf{E}_0$, ε the low-frequency dielectric constant of the crystal, e the absolute value of the electron charge, μ the electron mobility, and D the diffusion coefficient.

We assume that the intensity I_0 is not too high: $I_0\ll N\Gamma(1-f_0)^2S^{-1}$. In this case $n_0\ll N$ and the condition $\tau\ll\tau_1$ necessary for the existence of trap-charging waves is met.[13] Since the frequency of these waves $\omega\ll\tau^{-1}$, we can neglect n compared with $N\delta f$ in Eq. (7), as is clear from (5), i.e., the bulk of the space charge is bound to the traps.

To facilitate the analysis we transform to dimensionless variables, using as the units the Maxwell relaxation time $\tau_M=\varepsilon(4\pi e\mu n_0)^{-1}$, and the drift length $l_0=\mu\tau E_0$, leaving the rest of the notation unchanged. Eliminating from Eqs. (5)–(7) the quantities δn and δf we get

$$\left(a\frac{\partial^3}{\partial z^2\,\partial t}+\frac{\partial^2}{\partial z\,\partial t}+ab\frac{\partial^2}{\partial z^2}+b\frac{\partial}{\partial z}-\frac{\partial}{\partial t}-1\right)\frac{\delta E}{E_0}=\left(1+a\frac{\partial}{\partial z}\right)\left(\frac{\delta I}{I_0}+\frac{\tau g}{n_0}\right), \qquad (8)$$

where

$$b=\tau_M/\tau_1=\varepsilon\Gamma(4\pi e\mu f_0)^{-1},\ a=(l_D/l_0)^2,\ l_D=(D\tau)^{1/2},$$

and l_D is the diffusion length. For most photorefracting crystals we have $b\ll 1$.

To describe stimulated scattering it is necessary to insert in (8) the intensity distribution produced by interference of the incident $\mathscr{E}_1\exp(-ikz-i\omega_1 t)$ and scattered $\mathscr{E}_2\exp(ikz-i\omega_2 t)$ light waves:

$$I=I_0[1+\tilde{I}\exp(iKz-i\omega t)], \qquad (9)$$

where

$$I_0=\mathscr{E}_1^2,\quad \tilde{I}=2\mathscr{E}_2/\mathscr{E}_1\ll 1,\quad K=2k,\quad \omega=\omega_2-\omega_1.$$

Accordingly, we seek the solution of (8) in the form

$$\delta E=\tilde{E}\exp(iKz-i\omega t), \qquad (10)$$

assuming that the characteristic scale of the spatial and temporal variation of the amplitudes $\tilde{E}$ and $\tilde{I}$ is much larger than that of K^{-1} and ω^{-1}. Since we assume that $\mathscr{E}_2\ll\mathscr{E}_1$, the amplitude $\mathscr{E}_1$ in (3) and (9) should be regarded as a constant. Thus, equations (3) and (8) form a close system upon substitution of (1), (9), and (10).

3. STATIONARY AMPLIFICATION REGIME

Let us analyze Eqs. (3) and (8), assuming hereafter satisfaction of the condition $1\ll K\ll a^{-1}$, b^{-1} which determines the existence of weakly damped trap-charging waves.[13] We assume to start with that the amplitudes of $\tilde{E}$ and $\mathscr{E}_2$ vary so slowly that all the derivatives of these quantities can be neglected in (8). Using the boundary condition $\mathscr{E}_2=0$ at $z=0$ (see the figure), we get

$$\tilde{E}=\tilde{E}_0\exp(\alpha z),\quad \mathscr{E}_2=\mathscr{E}_0[\exp(\alpha z)-1],\quad \mathscr{E}_0=irn_r^2\mathscr{E}_1K\tilde{E}_0/8\alpha, \qquad (11)$$

$$\alpha={}^1/_4 rn_r^2E_0iK(1+iaK)\,[\omega K-1+i(\omega+bK+a\omega K^2)]^{-1}. \qquad (12)$$

The amplitude of the priming trap-charging wave E_0 is determined by the noise level in the crystal and can be expressed in terms of the spectral density of the random function $\hat{g}_{\omega K}$. Note that in addition to g it would be necessary to introduce in (6) a Johnson-noise source, but at $a\ll 1$ its contribution can be shown to be small compared with the generation-recombination contribution. For a consistent calculation of $\tilde{E}_0$, account must be taken of small deviations of the wave vector from the value corresponding to the Bragg condition. We eschew such a calculation, for besides the noise due to the random generation and recombination acts at $I=\text{const}$, the quantity $\hat{g}_{\omega K}$ is determined also by the fluctuations of the intensity I, which are produced by the light source and whose level depends strongly on the experimental conditions.

It follows from (12) that α', the real part of α, which determines the gain of the scattered light, has a clearly pronounced maximum

$$\alpha'=\alpha_m={}^1/_4 rn_r^2E_0\gamma^{-1},\quad \gamma=(1+aK^2+bK^2)K^{-2} \qquad (13)$$

at $\omega=\Omega=K^{-1}$. The imaginary part is $\alpha''=0$ at $\omega=\Omega$. This effect is physically due to resonant excitation of the charge-trapping waves: the relation $\Omega(K)=K^{-1}$ coincides with the dispersion law for these waves.[13] The fact that $\alpha(\Omega)$ is real means that at resonance the charge-trapping wave, propagating at a phase velocity $v_f=K^{-2}$, lags by $\pi/2$ the moving interference pattern that plays the role of the periodic driving force.

In the more general case the dispersion law of the trap-charging waves, with allowance for their damping, can be obtained by equating to zero the denominator of (12) and assuming ω to be complex. Substituting $\omega=\Omega+i\omega''$, we get $\Omega=K^{-1}$ and $\omega''=\gamma$. The quantity γ [see Eq. (13)] is thus the damping rate of the trap-charging wave. If the condition $1\ll K\ll a^{-1}$, b^{-1} used above is met, the characteristic distance negotiated by trap-charging wave during its damping time

$$\varkappa=\gamma^{-1}v_f=(1+aK^2+bK^2)^{-1}, \qquad (14)$$

turns out to be much greater than its length $\Lambda=2\pi K^{-1}$.

It follows from (12) and (13) that the gain α is independent of the light intensity I. This is true if, as we assume, the photoconductivity exceeds substantially the conductivity in the dark. The result has a simple physical meaning. The amplitude of the trap-charging wave is proportional to the driving force I and also to the wave damping time $\gamma^{-1}\propto I^{-1}$. Therefore the amplitude of the oscillations that build up, and hence the gain, does not depend on I. On the contrary, for ordinary forms of stimulated scattering such as Brillouin scattering, the relaxation time of the natural oscillations of the scattering medium is practically independent of I, therefore the gain is proportional to I.

For $\omega=0$, Eq. (12) leads to a known result[1,7] that describes the amplification of scattering light interacting with an immobile grating produced by diffusion of photoexcited electrons. In dimensional units we have

$$\alpha={}^1/_4 rn_r^2K^2Te^{-1}, \qquad (15)$$

where T is the crystal temperature in energy units. Comparing Eqs. (13) and (15), we conclude that for $a\ll 1$, i.e., for $l_0\gg l_D$, the trap-charging waves make the predominant contribution to the light scattering.

We emphasize that, in the discussed light-incidence geometry, amplification is possible if $r>0$. On the contrary, if the directions of k_1 and E_0 coincide, the condition $\alpha_m>0$ is met at $r<0$ and the backscattered light is a Stokes wave.

Note that an expression similar to (12) was derived earlier[19] for a description of the experimentally observed parametric amplification of the weak optical signal applied to a crystal simultaneously with a pump wave shifted in frequency (see also Ref. 20). However, the connection between this effect and the natural oscillations of the crystal—the trap-charging waves—were not discussed. The possibility of light scattering when dynamic noise is amplified in the crystal was not considered in these references.

Let us determine the region in which Eqs. (11)–(13) are valid. Substituting (11) in (8) we can verify that it is bounded by the inequality $\alpha\ll\varkappa^{-1}$.

4. EMISSION REGIME

The approximation used in the preceding section presupposes a local character of the connection between the trap-charging wave amplitude $\tilde{E}$ and the light intensity $\tilde{I} \propto \mathscr{E}_2$. Let us examine qualitatively the results that should ensue from absence of locality at $\alpha \sim \varkappa^{-1}$ or $\alpha > \varkappa^{-1}$

It is well known under inhomogeneous excitation the distribution of the amplitude (of the smooth envelope) of the wave is shifted relative to the distribution of the driving force in a direction determined by the group velocity of the wave. In our problem the amplitude $\mathscr{E}_2$, which plays the role of the driving force, is zero near the rear face of the crystal (at $z = 0$) and increases along the z axis (see the figure). In accordance with the dispersion law, the group velocity $v_g = K^{-2} = -v_f$ of the trap-charging wave is directed counter to the field $\mathbf{E}_0$ (along the electron drift). Thus, the distribution of the amplitude $\tilde{E}$ should be shifted relative to the distribution of the positive electrode—into the region of minimum intensity of the scattered light. Increasing the amplitude $\tilde{E}$ near $z = 0$ causes the scattered-light intensity to increase in the entire volume of the crystal. This leads in turn to further increase of $\tilde{E}$ near $z = 0$, owing to transfer of the amplitude, etc. As a result, if this transfer is effective enough, the gain turns out to be unstable and an emission regime is produced in which the backscattered anti-Stokes wave and the trap-charging wave increase exponentially with time.

The conditions for the appearance of emission can be easily understood from the following considerations. Assume that when the light is turned on there is present in the crystal an extremely small trap-charging wave produced by the fluctuations. The scattered line has here a distribution $\mathscr{E}_2 \propto z$ (see the figure, thick line). According to the local theory, the priming wave should be damped in the region of small z and grow in the region of large z, where a sufficiently strong driving force $\mathscr{E}_2$ acts. The plane separating these regions is located at $z = z' \sim \alpha_m^{-1}$. Obviously, if the distance z is shorter than the characteristic transport length $\varkappa = |v_g|\gamma^{-1}$, see (14), the transfer of the amplitude $\tilde{E}$ from the region $z > z'$ into the region $z < z'$ causes the oscillations to grow in the entire crystal. The emission condition takes thus, apart from a numerical factor, the form $\alpha_m \varkappa > 1$.

We proceed now to describe the emission regime on the basis of Eqs. (3) and (8). Substituting (1), (9), and (10) and using the condition $1 \ll K \ll a^{-1}, b^{-1}$, we obtain at resonance (at $\omega = K^{-1}$) the simplified equations

$$\gamma^{-1}\partial\tilde{E}/\partial t - \varkappa\partial\tilde{E}/\partial z + \tilde{E} - \tilde{E}_0 = \alpha_m \mathscr{E}_2', \qquad (16)$$

$$\partial\mathscr{E}_2'/\partial z = \tilde{E}, \qquad (17)$$

where

$$\mathscr{E}_2' = 8\alpha_m (irn_r^2 K\mathscr{E}_1)^{-1}\mathscr{E}_2.$$

We choose the initial and boundary conditions in (16) and (17) in the form

$$\mathscr{E}_2'(0, t) = 0, \quad \tilde{E}(z, 0) = \tilde{E}_0, \quad \tilde{E}(L, t) = \tilde{E}_0,$$

where L is the crystal thickness. The meaning of the first two conditions is obvious. The reason for the third is that growth of a trap-charging wave at a given direction of its group velocity is possible only at a finite distance from the front face $z = L$ of the crystal, where the driving force can act on it.

Expressions (16) and (17) are formally analogous to the equations that describe stimulated Brillouin scattering, see Refs. 15 and 16. The solution can be obtained by taking a Laplace transform with respect to time. For $\alpha_m L \gg 1$ the transformed solution has a pole

$$s = |v_g|\varkappa^{-1/2}(2\alpha_m^{1/2} - \varkappa^{-1/2}).$$

In the case $s > 0$ the solution turns out to be unstable and emission sets in. The conditions for the transition to the emission regime take thus the form $\alpha_m \varkappa > 1/4$, the result obtained above from qualitative considerations. On substituting (13) and (14), this condition takes the form

$$rn_r^2 E_0 K^2 (1 + aK^2 + bK^2)^{-2} > 1. \qquad (18)$$

Expression (18) determines the critical field E_{0c}. For $E_0 > E_{0c}$ [if (18) is satisfied] the scattered-light intensity increases like

$$\mathscr{E}_2 \propto \exp\left[\alpha_m^{1/2}\varkappa^{-1/2} z + (2\alpha_m^{1/2} - \varkappa^{-1/2})\varkappa^{-1/2}|v_g|t\right] \qquad (19)$$

until the incident wave is depleted. As a result, the light-reflection coefficient should increase to a value on the order of unity independently of the level of the priming fluctuations.

We note that, in contrast to the amplification regime, emission is possible only when $\mathbf{k}_1$ and $\mathbf{E}_0$ are antiparallel and $r > 0$, since its onset requires, besides the condition $\alpha_m > 0$, also that the group velocity $\mathbf{v}_g$ be antiparallel to the gradient of the intensity $\tilde{I}$. The Doppler shift of the scattered light, determined by the phase velocity v_f, is then positive.

5. FEATURES OF LIGHT SCATTERING UNDER PHOTOVOLTAIC-EFFECT CONDITIONS

In a crystal without an inversion center the photoionization of the impurity centers is accompanied by a photovoltaic current $J = GSINf$, where G is the Glass constant.[18] In an isolated crystal this current leads to photovoltaic effects and maintains the field E_0 needed for trap-charging waves to exist in the absence of an external voltage source. This, however, is not the only role played by the current J. When the trap-charging wave propagates this current becomes nonuniform because of the oscillations of the occupancy factor f and causes redistribution of the space charge. The current J can therefore decrease the damping rate of the trap-charging waves, and under certain conditions it can even amplify them.[12] We confine ourselves in the present paper to weakly damped waves.

To describe light scattering under conditions of the photovoltaic effect, the current $\mathbf{J}$ must enter in the continuity equation (6) with a minus sign, since $\mathbf{J}$ is antiparallel to the field $\mathbf{E}_0$. It can be shown that allowance for this current reduces to the substitutions

$$E_0 \to (E_0 - E_s), \quad b \to b\left[1 - (1 - f_0)E_s/E_0\right] \qquad (20)$$

in Eqs. (13)–(19). Here

$$E_s = GSI_0 N f_0 (e\mu n_0)^{-1}$$

is the field acting in an isolated crystal under conditions of mutual cancellation of the current J and the conduction current. Since the electric insulation of the crystal is usually not perfect, the field E_0 turns out to be somewhat weaker than its possible maximum E_s.

It follows from (13) and (20) that in the presence of the photovoltaic effect the amplification condition $\alpha_m > 0$ is met, in contrast to the case considered in the preceding sections, if $\mathbf{k}_1$ and $\mathbf{E}_0$ are antiparallel and $r < 0$, or if $\mathbf{k}_1$ and $\mathbf{E}_0$ are parallel and $r > 0$. Emission is possible only if $\mathbf{k}_1$ and $\mathbf{E}_0$ are antiparallel and $r < 0$. These equations show also that the current J can decrease the damping rate γ. The gain is therefore increased and satisfaction of the emission criterion (18) is facilitated.

6. COMPARISON OF THEORY WITH EXPERIMENT

Let us consider the feasibility of emission under experimental conditions.[17] In the cited study, a laser beam of wavelength $\lambda \approx 0.44$ μm was focused on an $LiNbO_3$:Fe crystal along the **c** optical axis.[1)] Periodic light scattering was observed in the form of cones directed towards the pump, and called by the authors of Ref. 17 scattering self-waves. The effect is critically dependent on the pump frequency and is unobservable when the pump wave vector $\mathbf{k}_1$ is directed counter to the **c** axis.

These results contradict drastically the theory of photoinduced scattering by immobile gratings.[1,4,7] According to these references, stationary backscattering that depends little on the pump frequency should take place if $\mathbf{k}_1$ and **c** are antiparallel, but if, on the contrary, $\mathbf{k}_1$ and **c** are parallel bleaching of the crystal is predicted. Indeed, in the latter case it follows from Eq. (15), obtained under the assumption $\omega = 0$, that $\alpha_m < 0$, since we must put in this equation $r < 0$ for parallel $\mathbf{k}_1$ and **c**.

Let us discuss now the possibility of scattering by trap-charging waves, recognizing that in the experiments of Ref. 17 a photovoltaic current **J** flowed through the crystal. It is remarkable that the wavelength of the effect in Ref. 17 corresponds to the maximum value of **J**, see Ref. 18. It is known that in $LiNbO_3$:Fe the current **J** is directed along **c** and consequently $\mathbf{E}_0$ and **c** are antiparallel. We see accordingly that the effect of Ref. 17 occurs only if $\mathbf{k}_1$ and $\mathbf{E}_0$ are antiparallel and $r < 0$, the same geometry as the emission in a crystal with a photovoltaic current (see Sec. 5).

Let us examine, finally, the feasibility of satisfying the quantitative criterion (18). Using typical parameters $r \sim 3\cdot 10^{-9}$ $\mathrm{V^{-1}\cdot cm}$, $N \approx 5\cdot 10^{19}$ $\mathrm{cm^{-3}}$, $f_0 \sim 1$, $n_r \sim 2{,}3$, $\varepsilon \sim 30$, $\mu\tau \sim 10^{-9}$ $\mathrm{V^{-1}\cdot cm^2}$ and $E_s \sim 10^5$ $\mathrm{V\cdot cm^{-1}}$ we find from (18) and (20) that the emission criterion can be satisfied at $E_0 \sim E_s$.

The effect observed in Ref. 17 can thus be interpreted as stimulated scattering of light by trap-charging waves in the emission regime. The sensitivity of the effect to the light wavelength λ is probably due to the fact that a change of λ decreases the current J and condition (18) no longer holds. The trap-charging-wave frequency ω which determines the frequency shift of the scattering light should be small under the conditions of Ref. 17, $\omega \lesssim 1$ Hz.

It should be noted that the proposed theory does not describe the case of strong scattering $\mathscr{E}_2 \sim \mathscr{E}_1$ and can therefore not explain the transition observed in Ref. 17 from a periodic to a stochastic regime when the pump intensity I is decreased. The theory yields only a criterion for the nonstationary scattering regime and can describe the initial stage of its development [Eq. (19)]. Nonetheless, since the time scale in the initial equations is $\tau_M \sim I^{-1}$, it can be concluded that the period of the scattered-light intensity fluctuations should also be proportional to I^{-1}, as indeed observed in Ref. 17.

We note also that to describe the scattered-light angular-distribution dynamics investigated in Ref. 17 it is necesary to take into account trap-charging waves propagating at different angles to the field $\mathbf{E}_0$. This, just as the case of $\mathscr{E}_2 \sim \mathscr{E}_1$, is outside the scope of the present article.

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1) As in Ref. 17, we define the positive **c**-axis direction such that a field applied along **c** leads to an increase of the refractive index of the crystal.

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