Low-frequency noise and photoinduced light scattering in photorefractive crystals

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(Submitted 19 April 1991)
Zh. Eksp. Teor. Fiz. **100**, 1071–1076 (September 1991)

A new approach is proposed for finding an explanation of the anomalously large photoinduced scattering of light in photorefractive crystals. The approach starts from the amplification of low-frequency noise of the laser and the medium. Some specific mechanisms for this amplification are considered. Some predictions for experiments are discussed.

1. INTRODUCTION

Photorefractive crystals ($LiNbO_3$, $BaTiO_3$, etc.) exhibit a huge optical nonlinearity. Even at laser power levels in the milliwatt range, the rate of exponential signal growth (the gain) can reach values $\Gamma = (10^2–10^3)$ cm^{-1} (Refs. 1 and 2). Photorefractive crystals hold promise for many optical applications, e.g., amplification and correction of light beams and phase conjugation.[2]

To some extent, all photorefractive crystals exhibit photoinduced scattering of light. This is the amplification of weak seed scattering, which is always present in a crystal. It frequently leads to strong noise, which is imposed on the useful signal.[2,3] This photoinduced scattering is a serious obstacle to various applications of photorefractive crystals. On the other hand, data on this scattering contain useful information about the processes which underlie the photorefractive nonlinearity. Despite the several studies which have been carried out in an effort to interpret this photoinduced scattering (e.g., Refs. 3–5), the factors which determine the magnitude of the effect and whether it can be suppressed remain largely a mystery.

2. QUALITATIVE DISCUSSION

We suggest a new interpretation of the photoinduced scattering, in which the factor primarily responsible for the rapid growth of seed scattering is the low-frequency noise of the elements of the optical system: noise in the laser intensity, noise in the refractive index, oscillations of the crystal surface, and so forth. The interpretation is based on the known fact that the gain Γ is a strong function of the frequency difference ("detuning") $\Omega = \omega_0 - \omega$ between the pump and probe waves.[2] This dependence is shown schematically in Fig. 1. The maximum value Γ_m is reached at $\Omega \approx \gamma$, where $\gamma \approx 4\pi\sigma/\varepsilon$ is the reciprocal of the dielectric relaxation time, σ is the conductivity, and ε is the permittivity. As a rule, γ is determined by the light intensity and is less than (1–10) s^{-1}.

The quantity Γ_m is a known function of the electrooptic constants of the material and the electric field (either an external field or a field induced by a photogalvanic effect).[2,6] The value Γ_0, which corresponds to zero detuning ($\Omega = 0$), is associated with the so-called nonlocal nonlinear response.[2] Depending on the scattering direction, it can take on negative values. The quantity Γ_m is often much larger than Γ_0; it is this case which we examine below. In $LiNbO_3$ and $LiTaO_3$, for example, the ratio Γ_m/Γ_0 reaches a value of 10^2. The strong $\Gamma(\Omega)$ dependence described here is a direct consequence of the inertial of the photorefractive nonlinearity.

In the standard interpretation of photoinduced scattering it is assumed that the pump wave is monochromatic and that the seed scattering is elastic. In this case, the intense scattering should, after a transitional stage, cause bleaching of the crystal. At times $\gamma t \gg 1$, the increase in the intensity I^s of the scattered light, with the layer thickness x_0 should be described by $\exp(\Gamma_0 x_0)$. Since this behavior is actually not seen experimentally, and since there is an anomalously strong, steady-state, symmetric scattering, it is reasonable to suggest that in the steady state we would have

$$I^s = I_0^s \exp(\Gamma_m x_0), \tag{1}$$

and that the preexponential coefficient I_0^s is governed by the properties of the low-frequency ($\Omega \lesssim \gamma$) noise. We wish to stress that numerical values of several tens are typical for the argument of the exponential function, $\Gamma_m x_0$. At such values, even very weak low-frequency seeds could lead to strong scattering. Further effort should be made to determine the coefficient I_0^s.

It is clear from general considerations that this coefficient may be determined by several independent factors. First, there is the low-frequency noise of the laser intensity. In the pertinent frequency range, $\Omega \lesssim \gamma$, this noise is not usually monitored. It is largely determined by the individual characteristics of the particular apparatus and experiment. Second, there are the low-frequency fluctuations of the refractive index. From statistical physics[7] we know that in an equilibrium system with a relaxation rate γ the frequency spectrum of fluctuations in physical quantities is Lorentzian, i.e., proportional to $(\Omega^2 + \gamma^2)^{-1}$. In such a system, fluctuations with $\Omega \approx \gamma$ are comparable in magnitude to static fluctuations ($\Omega = 0$), which cause elastic scattering of

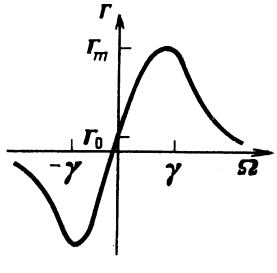

FIG. 1. Gain versus the frequency detuning.

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light. Third, a real crystal will contain a multitude of static structural defects, which may be thought of as nonequilibrium fluctuations with an unlimited relaxation time. The inelastic component of the bulk seed scattering may thus be relatively small. It may depend on the particular procedure used to prepare the sample and on the history of the sample in general. The inelastic component of the seed scattering may also be caused by low-frequency surface oscillations.

Finally, it is important to bear in mind that any real physical system will have a nonequilibrium low-frequency $1/f$ noise.[9] This noise is manifested in fluctuations of the laser intensity and fluctuations of the refractive index. The $1/f$ noise may be particularly important for photoinduced light scattering.

3. INITIAL EQUATIONS

We turn now to a model for photoinduced scattering. We can start with the following system of equations:

$$\left(\frac{\partial}{\partial x}+i\varkappa\right)a_{\mathbf{k}}=ik_0a_0(P_{\mathbf{q}}+P_{\mathbf{q}}^0),$$
$$\left(\frac{\partial}{\partial t}+\gamma\right)P_{\mathbf{q}}=\beta_{\mathbf{k}}a_{\mathbf{k}}a_0^*. \tag{2}$$

Here $a_{\mathbf{k}}(x,t)$ and $a_0(t)$ are the amplitudes of the scattered and pump waves, $\mathbf{k}$ and $\mathbf{k}_0$ are the corresponding wave vectors, $\varkappa=k-k_0$, $\mathbf{q}=\mathbf{k}-\mathbf{k}_0$, $P_{\mathbf{q}}$ and $P_{\mathbf{q}}^0$ are Fourier components of the induced and fluctuational relative changes in the refractive index, $\Delta n/n$, and $\beta_{\mathbf{k}}$ is a coupling coefficient which characterizes the photorefractive nonlinearity and which depends on the polarizations of the waves.[1,2,6] The x axis runs along the normal to the entrance surface and is parallel to the wave vector $\mathbf{k}_0$ (Fig. 2). If the nonlocal response is ignored, the parameter $\beta_{\mathbf{k}}$ is real. Equations (2) essentially describe diffraction of the pump wave by a dynamic three-dimensional grating of the refractive index, $\Delta n_{\mathbf{q}}e^{i\mathbf{qr}}+\text{c.c.}$

In the first of these equations we have ignored the time derivative of $a_{\mathbf{k}}$ in comparison with the spatial derivative. This is legitimate if the crystal thickness is small in comparison with the laser coherence length; this condition is usually satisfied well. The time evolution of the complex amplitudes a_0 and $a_{\mathbf{k}}$ thus includes not only intensity fluctuations but also effects of phase modulation and the finite width of the laser line. The relaxation parameter γ in (2) is a function of $|a_0|^2$ and also fluctuates in time. The approximation $\gamma\propto|a_0|^2$, which is the most typical one for photorefractive crystals, corresponds to a negligibly small intrinsic conductivity of the crystal and to a photoconductivity which is linear in the intensity. The boundary condition on $a_{\mathbf{k}}(x,t)$ can include scattering by the entrance surface ($x=0$).

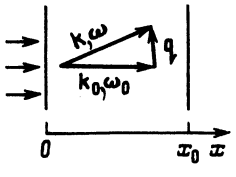

FIG. 2. Geometry of the problem.

If we assume a frequency shift $a_{\mathbf{k}}\propto a_0\exp(i\Omega t)$ in (2) and set $|a_0|^2=\text{const}$, $P_{\mathbf{q}}^0=0$, and $\varkappa=0$, we immediately find the known frequency dependence of the gain:

$$\Gamma(\Omega)=\frac{2\beta k_0|a_0|^2\Omega}{\gamma^2+\Omega^2}. \tag{3}$$

Obviously, $\Gamma_m=\beta k_0|a_0|^2/\gamma$.

Curiously, expression (3) is like system (2) in that it can be rewritten in a form which describes the temporal instability of a monochromatic plane wave $\mathbf{k}_0$ with respect to spatially uniform perturbations. The instability growth rate then has a singularity at $k=k_0$. The singularity problem does not arise in problems with boundary conditions.

4. FLUCTUATIONS IN THE PUMP INTENSITY

Let us consider the situation in which the photoinduced scattering results from fluctuations of the pump intensity. Without any loss of generality, we can set $P_{\mathbf{q}}^0=0$ in this case, and we can assume that the seed scattering is elastic and occurs at the surface:

$$a_{\mathbf{k}}(0,t)=z_{\mathbf{k}}a_0(t). \tag{4}$$

The coefficient $z_{\mathbf{k}}$ depends only on the direction of the scattering. In addition, in describing the diffraction by the self-consistent grating $P_{\mathbf{q}}$ we should set $\varkappa=0$ in (2). We then easily find from (2) and (3) that the case $\gamma\propto|a_0|^2$ is degenerate: Fluctuations in $|a_0|^2$ do not give rise to amplification of the seed scattering in this case.[1)] However, a deviation of $\gamma(|a_0|^2)$ from linearity causes an increase in $a_{\mathbf{k}}$. Such a deviation might arise when either the intrinsic conductivity σ_0 of the crystal, or the sublinear (or superlinear) intensity dependence of the photoconductivity is taken into account.

For simplicity we consider the case of a large intrinsic conductivity, $\sigma\approx\sigma_0$, with $\gamma\approx\text{const}$. Solving system (2) under the boundary condition (4), and with a homogeneous initial condition for the grating amplitude $P_{\mathbf{q}}$, and assuming that the fluctuations δI in the pump intensity around the mean value I_0 are small, we find the following result for the time-average intensity of the light scattered into a solid angle do in the direction of $\mathbf{k}$:

$$\frac{dI_{\mathbf{k}}^s}{I_0}\approx|z_{\mathbf{k}}|^2\gamma\Phi_\gamma\left(\frac{\pi}{\Gamma_m x_0}\right)^{1/2}\exp(\Gamma_m x_0)\,do. \tag{5}$$

Here Φ_γ is the Fourier transform of the correlation function of the fluctuations, and

$$\Phi(\tau)=\frac{\langle\delta I(t+\tau)\delta I(t)\rangle}{I_0^2}. \tag{6}$$

The angle brackets mean a time average. We used the method of steepest descent of Ref. 9 in evaluating the integral over Ω; that method is valid under the condition $\Gamma_m x_0\gg 1$.

Expression (5) gives the intensity of the photoinduced scattering in terms of the parameters of the crystal and a basic characteristic of the fluctuations, the correlation function Φ. Expressions with a corresponding meaning can be derived for the case of bulk elastic scattering and the case of sublinear (or superlinear) I_0 dependence of the photoconductivity.

5. LOW-FREQUENCY FLUCTUATIONS OF THE REFRACTIVE INDEX

We turn now to the case in which the photoinduced scattering involves low-frequency fluctuations of $P_{\mathbf{q}}^0$. We assume $|a_0|^2 = \text{const}$, and we ignore surface scattering. Using (2), we can express $dI_{\mathbf{k}}^s$ in terms of the Fourier transform $K_{\mathbf{q}\Omega}$ of the correlation function of the fluctuations in the refractive index:

$$K(\boldsymbol{\rho},\tau) = \frac{\langle \Delta n(\mathbf{r}+\boldsymbol{\rho},t+\tau)\Delta n(\mathbf{r},t)\rangle}{n^2}. \tag{7}$$

The final expression, under the condition $\Gamma_m x_0 \gg 1$, is

$$\frac{dI_{\mathbf{k}}^s}{I_0} \approx \frac{\gamma k^4 K_{\mathbf{q}\Gamma}}{\Gamma_m}\left(\frac{\pi}{\Gamma_m x_0}\right)^{1/2}\exp(\Gamma_m x_0)\,do, \tag{8}$$

where $\mathbf{q} = \mathbf{k} - \mathbf{k}_0$ and $k = k_0$. The $\propto k^4$ dependence is typical of a scattering of light by fluctuations in the refractive index. To estimate the total scattering intensity, it is sufficient to replace do by $2\pi^2$.

The correlation function $K(\rho,\tau)$ can be approximated by the simple expression[7]

$$K = \left\langle\left(\frac{\Delta n}{n}\right)^2\right\rangle \exp(-\gamma|\tau| - \rho R^{-1}), \tag{9}$$

where $[\langle(\Delta n/n)^2\rangle]^{1/2}$ is the mean square fluctuation, and R the correlation radius. The approximation (9) is valid in the situation described in Sec. 2, in which the medium can be characterized by a single relaxation parameter γ. Using (8) and (9), we easily find, for the case $qR \ll 1$,

$$\frac{I^s}{I_0} \approx \frac{(2\pi n)^2\langle(\Delta n)^2\rangle}{\Gamma_m\lambda}\left(\frac{R}{\lambda}\right)^3 \exp(\Gamma_m x_0), \tag{10}$$

where λ is the wavelength of the light. Under the condition $qR \gg 1$, the scattering intensity falls off rapidly. Expression (10) does not contain the parameter γ. The properties of the steady-state scattering are thus independent of the light intensity. It is also obvious that the preexponential coefficient is sensitive to the value of the correlation radius R.

Detailed information on $\langle(\Delta n/n)^2\rangle$ and R could be found either from a microscopic theory or from a special experiment. However, it is clear even from general considerations that in a photorefractive crystal, in which the changes in the refractive index are related to photoinduced electric fields, the fluctuations of the photocurrent should be the ultimate cause of the seed scattering. We should focus on the photogalvanic current here. In the first place, it is the photogalvanic effect which leads to the anomalously strong photoinduced fields, $E_{\text{ph}} \approx (10^4$–$10^5)$ V/cm, in crystals such as $LiNbO_3$ and $LiTaO_3$. Second, the photogalvanic effect results not from thermalized electrons but from "hot" electrons, with energies $\varepsilon_0 \gg T$ (Ref. 6). This is an exceedingly favorable circumstance for the growth of fluctuations. If these arguments are correct, then we should treat as the correlation radius R the the diffusion length of a photoexcited carrier over the energy relaxation time.[10] Estimates of the preexponential coefficient in (10) turn out to be extremely sensitive to the choice of parameter values of the microscopic theory.

Low-frequency oscillations of the crystal surface also lead to general expressions of the type in (5) and (8).

6. CONCLUSION

Let us summarize the results of this study.

The general considerations and model calculations presented above demonstrate that low-frequency fluctuations play a key role in the problem of the photoinduced scattering of light. They also demonstrate that there are a large number of possible mechanisms for the anomalously strong steady-state scattering in photorefractive crystals.

A question which remains open is one of much general interest: the relationship between photoinduced scattering and the well-known low-frequency $1/f$ noise.

There should be a frequency shift $\Omega \approx \gamma$ between the scattered light and the pump wave.

It is possible that the photoinduced scattering could be reduced by stabilizing the pump intensity.

It is important to take low-frequency fluctuations into account not only in a description of wide-angle scattering of light but also in the analysis of any nonlinear-amplification process in a photorefractive crystal which requires seeds. In particular, we have in mind the extremely common parametric-scattering processes,[11,12] which are seen as bright rings and lines of scattered light.

Finally, these results are applicable in large part to any medium with a very slow nonlinearity.

[1)] This degeneracy does not, of course, contradict the amplification of a frequency-shifted light beam with the gain (3).

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Translated by D. Parsons