Stimulated diffusion backscattering of nanosecond pulses under exposure accumulation conditions

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A theoretical analysis is made of the process of energy exchange between counterpropagating waves due to stimulated diffusion scattering by photorefractive gratings, which grow as a result of a periodic pulsed interaction. The conditions ensuring exposure accumulation are found. Experiments on a $LiNbO_3$ crystal, made using $\lambda = 0.53\,\mu$m radiation, largely confirm the theory and demonstrate the possibility of phase conjugation.

Various new methods for the transformation of optical beams in photorefractive crystals have been proposed recently.[1] From the practical point of view there is considerable interest in developing those methods in which low-power cw lasers have been used earlier so as to replace them with compact and convenient pulse-periodic solid-state lasers. This possibility arises because of two circumstances. On the one hand, efficient formation of photorefractive gratings is quite readily attainable even when short single pulses are used.[2–4] On the other, if the dark conductivity in the intervals between the pulses is negligible, then the gratings already formed in a crystal are not removed.[1] Consequently, a series of short pulses may ensure that the grating grows to an efficiency comparable with the efficiency of gratings formed using cw radiation.[5,6] Stimulated diffusion backscattering in $LiNbO_3$ has been investigated theoretically[7] and also experimentally employing radiation from a pulse-periodic copper laser[8] and from a cw He–Cd laser.[9]

In the present study a $LiNbO_3$:Fe crystal was subjected to a series of nanosecond pulses of the second harmonic ($\lambda = 0.53\,\mu$m) of a YAG:Nd laser and stimulated diffusion backscattering (STDS) was observed under conditions of exposure buildup and with phase conjugation.

We first compare the efficiency of formation of diffusion gratings in two different cases of the optical interaction: using radiation from a cw laser and from a pulse-periodic laser when the average power is the same in both cases. We do this using the standard energy band model[1,2,7] and also its simplified variant:[10] with one type of photoexcited free carriers (electrons) of density $n(\mathbf{R})$ and one type of donor levels with a concentration N_D, assuming that these donors are initially partly ionized by compensating acceptor impurities whose concentration is N_A. In this model the zeroth and first spatial harmonics of the electron density

$$n(\mathbf{R}) = n_0 + n_1 e^{-i\mathbf{qR}} + \text{c.c.},$$

of the illumination intensity

$$I(\mathbf{R}) = I_0 + I_1 e^{-i\mathbf{qR}} + \text{c.c.},$$

and of the induced space charge field

$$\mathscr{E}(\mathbf{R}) = \mathscr{E}_1 e^{-i\mathbf{qR}} + \text{c.c.},$$

we can derive the following system of equations

$$\frac{\partial n_0}{\partial t} + \gamma n_0 (N_A + n_0) = \xi I_0(t),$$

$$\frac{\partial \mathscr{E}_1}{\partial t} + \omega \mathscr{E}_1 + i\frac{4\pi e D q}{\varepsilon_{st}} n_1 = 0,$$

$$\frac{\partial n_1}{\partial t} + [\gamma(N_A + 2n_0) + Dq^2] n_1 - iq\mu n_0 (1-\varphi)\mathscr{E}_1 = \xi I_1. \qquad (1)$$

Here, γ is a constant of the linear carrier recombination process; $N_A + n_0$ is the spatial average of the concentration of the ionized donors; $\omega(t) = 4\pi e\mu n_0(t)/\varepsilon_{st}$ is the instantaneous value of the Maxwellian relaxation time; D and μ are, respectively, the diffusion coefficient and mobility of conduction electrons; ε_{st} is the static permittivity; $\varphi = \varepsilon_{st}\gamma/4\pi e\mu = (r_D/L_D)^2$ is the dimensionless characteristic of the medium, equal to the ratio of the squares of the Debye radius and of the diffusion length. The system (1) describes the response of a pure diffusion type characterized by $\mathscr{E}_0 = 0$. The quantity $\xi = \alpha\eta$ is the product of the absorption coefficient of light α and the quantum efficiency η governing the fraction of the absorption processes accompanied by the formation of electrons in the conduction band. It is assumed in the system of equations (1) that φ is constant in space, i.e., the saturation of the donor levels[11] is neglected on the assumption that $N_D \gg \delta N_D$ and $N_D \gg N_a$, and the dependence of η on the radiation intensity is ignored.[5]

We analyze the system of equations assuming a low level of photoionization $n_0 \ll N_A$. Under steady-state conditions in the case of cw radiation we can readily obtain the familiar result for the amplitude of a grating of the diffusion type:

$$\mathscr{E}_1(t) = \bar{\mathscr{E}}_1[1 - \exp(-t/\bar{\tau}_M)], \quad \bar{\tau}_M = \frac{(1+L_D^2q^2)}{(1+r_D^2q^2)}\frac{\varepsilon_{cr}\gamma N_A}{4\pi e\mu\xi I_0},$$

$$\bar{\mathscr{E}}_1 = -im\frac{Dq}{\mu(1+r_D^2q^2)}, \quad m = I_1/I_0. \qquad (2)$$

Since we shall be discussing pulse-periodic commercial lasers, we must consider the process of formation of a grating by a series of short pulses separated by an interval $\Delta t \gtrsim 10^{-3}$ s considerably greater than the electron lifetime in the conduction band $(\gamma N_A)^{-1} \lesssim 10^{-5}\,\text{s} \ll \Delta t$. It is convenient to introduce the increment $\delta\mathscr{E}_1$ of the grating amplitude $\mathscr{E}_1$ produced by one pulse:

$$\delta\mathscr{E}_1 = \mathscr{E}_1(t_2) - \mathscr{E}_1(t_1);$$

here, t_1 is a time before the application of the next pulse and t_2 is a time after this pulse when, firstly, the illumination has ceased so that $I_{0;1}(t_1) = I_{0;1}(t_2) = 0$ and, secondly, free

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carriers have recombined $n_{0;1}(t_2) = n_{0;1}(t_1) = 0$, i.e., the redistribution of the charges has been completed. Then, $\delta\mathscr{E}_1$ is described by the relationship

$$\delta\mathscr{E}_1 + \int_{t_1}^{t_2} \tilde{\omega}(t)\mathscr{E}_1(t)\,dt = -i\frac{4\pi eDq}{\varepsilon_{cr}\Gamma} m\int_{t_1}^{t_2} \xi I_0(t)\,dt. \qquad (3)$$

We have introduced here the instantaneous rate of dielectric relaxation of the grating

$$\tilde{\omega}(t) = \tau_M^{-1} = \omega(t)(1+r_D^2q^2)/(1+L_D^2q^2),$$

modified to allow for the finite diffusion length and the Debye radius; $\Gamma = \gamma N_A(1 + L_D^2q^2)$. The relationship (3) is obtained by integration of the second and third equations of the system (1) and subsequent algebraic transformations.

An important parameter in the task of recording the gratings in a pulse-periodic regime is the dimensionless quantity

$$M = \int_{t_1}^{t_2} \tilde{\omega}\,dt,$$

representing the relationship between the pulse length τ and the dielectric (Maxwellian) relaxation time τ_M during a pulse: $M \approx \tau/\tau_M$. In our limit of $n_0 \ll N_A$, it follows from the first equation of the system (1) that the rigorous definition of M is

$$M = \frac{1+r_D^2q^2}{1+L_D^2q^2}\frac{4\pi e\mu}{\varepsilon_{cr}\gamma N_A}\int_{t_1}^{t_2} \xi I_0\,dt. \qquad (4)$$

If $M \gtrsim 1$ and $\tau \gtrsim \tau_M$, a grating may be formed by one pulse. Therefore, we consider in greater detail the limit of low values of this dimensionless quantity when $M \ll 1$.

In this case one pulse changes the grating slightly ($\delta\mathscr{E}_1 \ll \mathscr{E}_1$), so that we have $\int\tilde{\omega}\mathscr{E}_1\,dt \approx M\mathscr{E}_1$ and Eq. (3) readily yields the dependence of the amplitude of the electric field grating on the serial number n of the interaction pulse:

$$\frac{d\mathscr{E}_1}{dn} + M\mathscr{E}_1 = M\bar{\mathscr{E}}_1, \qquad \mathscr{E}_1(n) = \bar{\mathscr{E}}_1(1-e^{-Mn}), \qquad (5)$$

where $\bar{\mathscr{E}}_1$ is the steady-state value of the grating amplitude corresponding to the case of continuous (cw) illumination described by Eq. (2).

The value of $\mathscr{E}_1$ obtained in this way should be substituted into the Maxwell equations for a signal wave E_S in the presence of a laser pump wave E_L, where

$$m = E_L^* E_S/(|E_L|^2 + |E_S|^2).$$

A suitable procedure (see, for example, Ref. 1) does not need to have any special features for pulses of $\tau \sim 10^{-8}$ s duration, because the time taken to travel across a medium ~ 1 cm long is $nL/c \sim 10^{-10}$ s. In other words, the problem of propagation of light during illumination of the medium is solved in the quasistatic approximation for a given grating amplitude $\mathscr{E}_1$ which is gradually accumulated from one pulse to the next. We recall also that the factor $(-i)$ on the right-hand side of Eq. (3) corresponds to shifted gratings, i.e., it represents the amplification of the signal E_S due to the scattering in its direction of the energy of the laser pump wave E_L.

We now consider the results obtained. Firstly, in the case of a low level of ionization considered here, when $n_0 \ll N_A$, the steady-state amplitude of a grating formed in either regime is the same. Secondly, the functional law describing the change in $\mathscr{E}_1$ in both regimes is the same and, since $Mn = M\nu t \sim t/\bar{\tau}_M$ (ν is the pulse repetition frequency), the running value of the amplitude $\mathscr{E}_1$ depends only on the running exposure density

$$\omega = \int^t I_0(t')\,dt'.$$

Moreover, if we assume that the parameter $\xi = \alpha\eta$ is independent of the intensity of light and, therefore, the same for both recording regimes, we find that $M\nu t = t/\bar{\tau}_M$, and the dynamics of pulse-periodic recording of gratings is governed by the dielectric relaxation time $\bar{\tau}_M$ from Eq. (2), which is calculated from the average power of a laser source:

$$I_0 = \nu\int_{t_1}^{t_2} I_0(t')\,dt'.$$

We must stress an important point: the validity of these conclusions is independent, within the framework of the assumptions made above, of the details of the time profile of the pulses $I_0(t)$ or of the relationships between the pulse duration τ_p and the times $\tilde{\omega}^{-1}$, $(\gamma N_A)^{-1}$, and $(Dq^2)^{-1}$.

If we do not assume that ξ is constant, then since

$$M\nu t \approx (t/\bar{\tau}_M) \quad (\xi_{pp}/\xi_{cw}),$$

we can conclude that the ratio of the sensitivities of recording under cw and pulse-periodic (pp) regimes is governed by the ratio of the efficiencies of carrier photogeneration per incident photon at the radiation intensities in these two regimes.

An experimental investigation of the efficiency of pulse-periodic formation of gratings was carried out for the process of backward STDS, representing two-wave amplification in the field of a weak counterpropagating signal beam created in the crystal itself by spontaneous light scattering of the laser pump bear on defects in the crystal. We used the second harmonic of a YAG:Nd laser operating at a pulse repetition frequency $\nu \leqslant 0.5$ Hz and a crystal of $LiNbO_3$:Fe. The same process accompanied by phase conjugation had been observed earlier[12] in the same crystals using cw pump radiation from a copper vapor or an He–Cd laser.[9]

An optical beam representing the second harmonic ($\lambda = 0.53\,\mu$m) passed through a stop with a diameter of 1.1 mm and its divergence was 6×10^{-4} rad; it reached a lens with a focal length of $f = 5.5$ cm located at a distance of 13 cm from the stop and was focused inside the crystal which it entered at a small angle relative to the optic axis **c** (this angle was $\sim 22°$ in air). When this focusing geometry was used, the minimal diameter of the focal constriction was $\approx 15\,\mu$m and the length of the constriction was ≈ 1.5 mm, so that this constriction was readily accomodated inside the crystal of thickness $L \approx 6$ mm. The reflection from the rear face of the crystal did not return radiation to the constriction of the pump beam and could not serve as a "seed" for the counterpropagating wave. We determined the exact dependence of

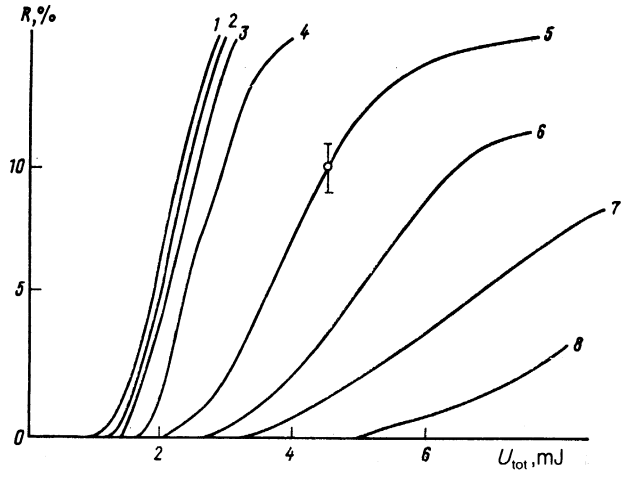

FIG. 1. Dependence of the back-reflection coefficient R (%) on the total energy U_{tot}, obtained for different values of the average energy carried by single pulses (μJ): *1)* 32; *2)* 44; *3)* 42; *4)* 22; *5)* 11; *6)* 6.5; *7)* 3; *8)* 2.

the nonlinear coefficient of reflection in the backward direction on the serial number of the incident pulse for different energies of the pulses. The pulse duration was ~ 20 ns and the energy was varied from 0.5 to 200 μJ.

Figure 1 shows the dependence of the retroreflection coefficient R on the total energy U obtained for different energies of a single pulse. Figure 2 gives the dependence of the threshold exposure U_{th} on the energy carried by one pulse U_1 plotted on the basis of the experimental data in Fig. 1 and those which are not included in Fig. 1.

The results obtained led us to the conclusion that, in the investigated range of parameters of the pulse-periodic irradiation of $LiNbO_3$:Fe crystals, the efficiency of grating formation depended strongly only on the running value of the exposure density W, but if the pulse energy exceeded 20–30 μJ, then the $R(U)$ curves in Fig. 1 practically merged. Then, a nonlinear reflection coefficient amounting to a few percent in the case of "seeding" by the spontaneous noise required a pump energy density of $\sim 10^3$ J/cm^2.

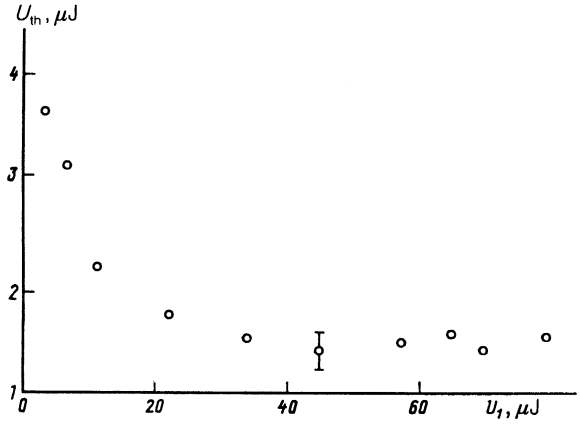

FIG. 2. Dependence of the threshold exposure U_{th} (for $R \approx 0.5\%$) on the energy carried by a single pulse U_1.

For $U_1 \leqslant 20\,\mu$J, corresponding to limitation of the pump intensity to $I_p \leqslant 5 \times 10^8$ W/cm^2, the dependence $R(U)$ ceased to be universal and the sensitivity of the crystal fell. In our opinion the reason for this deviation from the universal behavior at low intensities was due to the real dependence of both the absorption coefficient α and the quantum efficiency η on the illumination intensity. It was shown in Ref. 5 that this dependence could make the sensitivity of photorefractive recording in $LiNbO_3$:Fe approximately 8 times less at low intensities compared with high intensities.

Placing of a phase plate inside the pump beam and sharper focusing of the beam in the crystal resulted in backward STDS accompanied by phase conjugation. As in the case of cw radiation,[9] the phase conjugation quality reached its maximum in the unstable part of the dependence $R(U)$ and then fell, but the total reflection coefficient remained unaffected.

Our investigation thus demonstrated that in the case of pulse-periodic interaction the process of backward STDS could occur also under exposure accumulation conditions with a threshold exposure density $\approx 10^3$ J/cm^2.

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