

Photovoltaic effect, diffusion and drift of nonequilibrium electrons having finite mean free paths

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The photovoltaic effect, nonequilibrium diffusion, and nonequilibrium photoconductivity are investigated for an arbitrary relation between the period of the photoinduced grating and the diffusion length of energy-relaxing photoelectrons. It is shown that under rather lax conditions the nonequilibrium photoelectric phenomena prevail over effects connected with thermalized electrons. Manifestations of nonequilibrium effects in the optical properties of crystals are investigated.

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INTRODUCTION

In a considerable number of noncentrosymmetric dielectric crystals, such as LiNbO₃, LiTaO₃, BSN (strontium barium niobate), and others, the optically induced change of the refractive indices, i.e., the photorefractive (PR) effect, is uniquely determined by the photoinduced field

$$\Delta\epsilon = \hat{\alpha}E, \quad (1)$$

where $\hat{\alpha}$ is the tensor of the linear electro-optical effect. The optical properties of such crystals are closely related with the photoelectric phenomena that take place in them. It is customarily assumed that the main charge-separation mechanisms in a crystal, i.e., the main nonlinearity mechanisms, are: photoelectron diffusion, their drift in an external electric field, and the photovoltaic effect (PVE).^{1,2} It is just these mechanisms to which nonlinear interactions of light waves, photoinduced scattering, and holographic recording are attributed.³⁻⁵

The photoelectron lifetime τ , as a rule, exceeds considerably the momentum and energy relaxation times Γ^{-1} and γ^{-1} . The bulk of the photoelectrons is therefore in a thermalized state, i.e., they are described by a Boltzmann distribution function.⁶ The smallness of the number of nonequilibrium electrons makes it possible in many cases to neglect their contribution to the photoconductivity and to the diffusion. In this case the current density is

$$\mathbf{j} = e\mu_T n_T \mathbf{E} + eD_T \nabla n_T + \kappa G \mathbf{J}, \quad (2)$$

where n_T is the density of the thermalized electrons,¹⁾ μ_T and D_T are their mobility and diffusion coefficient, J is the light intensity, κ is the absorption coefficient, and G is the Glass coefficient⁷ that characterizes the PVE.

It was concluded in Ref. 8, on the basis of an analysis of the experimental data on the PVE and on the photoconductivity of PR crystals that account must be taken of the nonequilibrium contribution to the photoconductivity. In addition, the following charge-transport model was corroborated: thermalized electrons with energies $\epsilon \sim T$ have an exponentially small hopping conductivity. The smallness of μ_T is attributed either to large-scale fluctuations of the potential in a real crystal, or to the polaron effect. The nonequilibrium superthermal electrons with en-

ergy $T \lesssim \Delta \lesssim \epsilon \lesssim \epsilon_0$ have a high band-type mobility and correspondingly large mean free paths, $l \gg a$. Here a is the unit-cell dimension, $\epsilon_0 \gg T$ is the energy with which the electrons are ejected by the light into the conduction band, and Δ is the percolation level (at $\epsilon > \Delta$ one can speak of an ordinary band-type charge transport). Within the framework of this model the kinetic coefficients are sums of two contributions, equilibrium and nonequilibrium. The small number of the nonequilibrium electrons may be offset by their high mobility. As shown in Ref. 8, in crystals having anomalously large photoinduced fields, $E \gtrsim (10^4 - 10^5)$ V/cm, the nonequilibrium contribution to the photoconductivity becomes dominant.

The authors of Refs. 9 and 10 have recently observed independently a strong influence of a magnetic field on the PVE in LiNbO₃ and ZnS crystals. The data obtained indicate high mobilities of the nonequilibrium electrons, $\mu \approx 10^3$ cm²/V·sec, $\mu/\mu_T \gtrsim 10^3$, and large mean free paths, $l \sim 10^{-5}$ cm. These data are a direct confirmation of the proposed model. The suppression of the contribution to the transport of the thermalized electrons uncovers a possibility of investigating by optical methods directly the band properties of by excluding the difficulty controllable features due to the presence of defects.

It will be shown in this paper that nonequilibrium effects manifest themselves to a great degree in the presence of strong spatial inhomogeneity. This is precisely the situation in holographic recording and in nonlinear wave interaction. To calculate the optical properties in this case it is necessary to solve the following kinetic problem. Let the light intensity in the crystal be determined by a superposition of two plane waves with vectors $\mathbf{q}_{1,2}$:

$$J = J_0(1 + M \cos \mathbf{q}\mathbf{r}), \quad \mathbf{q} = \mathbf{q}_1 - \mathbf{q}_2. \quad (3)$$

It is required to determine the spatially oscillating part of the current \mathbf{j} and of the field $\tilde{\mathbf{E}}$. Up to now, this problem was solved on the basis of Eq. (1), i.e., neglecting the nonequilibrium electrons. At sufficiently large mean free paths l of the nonequilibrium electrons, such a description becomes unsatisfactory for two reasons. It is clear, first, that with increasing l there is absolutely an increase of the contribution of the nonequilibrium electrons to the photoconductivity, the

PVE, and the diffusion. Second, at large mean free paths of the nonequilibrium electrons the contribution of the thermalized photoelectrons to \bar{j} decreases sharply. This important circumstance can be explained in the following manner. The spatial distribution of the electrons immediately after ejection by the light into the conduction band and having them an energy ε_0 practically duplicates the intensity profile (3). The electron, losing energy by phonon emission, diffuses away from the point of its production. The spatial inhomogeneity is then gradually smoothed out. Within the thermalization time the electron diffuses over a distance

$$l_{\text{dif}} \approx lN^{1/2}, \quad (4)$$

where $N = \Gamma / \gamma$ is the number of scattering acts during the thermalization time. If the diffusion length exceeds the grating spacing

$$l_{\text{dif}} \gg \Lambda \equiv 2\pi q^{-1},$$

The thermalized electrons cease in practice to feel the modulation of the light intensity and their contribution to \bar{j} decreases. We emphasize that owing to the diffuse character of the spreading of the spatial inhomogeneity the decrease of the equilibrium contribution to \bar{j} at large q^2/l_{dif}^2 is exponential, i.e., very abrupt. Equation (2) for the current under conditions of strong spatial inhomogeneity becomes meaningless and \bar{j} is due entirely to the nonequilibrium electrons. In experiment usually $q \approx (10^4 - 10^5) \text{ cm}^{-1}$. Putting by way of estimate $l = 10^{-6} \text{ cm}$ and $N = 10^2$, we see that both low and high spatial frequencies are realistic ($q^2/l_{\text{dif}}^2 \gg 1$).

The spatially oscillating part of the current \bar{j} contains, generally speaking, a set of Fourier harmonics

$$\bar{j} = \frac{1}{2} \sum_{n=1}^{\infty} \bar{j}_n e^{inqz} + \text{c.c.} \quad (5)$$

A similar expansion is valid for the field \bar{E} and for other spatial characteristics of the crystal. We shall be interested in what follows mainly in the first Fourier component \bar{j}_1 of the current, which in fact is responsible for the nonlinear interaction.^{3,11} Its real part \bar{j}'_1 describes a current contribution proportional to $\cos \mathbf{q} \cdot \mathbf{r}$, i.e., one duplicating the intensity profile, while the imaginary part \bar{j}''_1 describes the so-called displacement part of the current, proportional to $\sin \mathbf{q} \cdot \mathbf{r}$.

To determine the nonequilibrium contribution to the current it is necessary to start from the kinetic equation for the electron distribution function f_k . The properties of this equation and the approximation employed are discussed in §1. In §2 is determined the current \bar{j} in the absence of electric fields. In this case there exist two independent contributions to the current, due to the PVE and to the diffusion of the nonequilibrium electrons. It was shown that independently of the electron mean free path the PVE is responsible for the undisplaced part \bar{j}'_1 of the current, and the diffusion for the displaced part \bar{j}''_1 . In the limit of small spatial frequencies, $q^2 l_{\text{dif}}^2 < 1$, the alternating part of the photovoltaic effect is described by the usual expression (the third term of (2)). At high frequencies the contribution to \bar{j} from the PGE becomes exponentially small, i.e., the photovoltaic mechanism of the nonlinearity becomes strongly suppressed. At weak inhomogeneity,

the diffusion contribution to the current $J_{\text{dif}} \propto q$, just as J_{dif}^T . With increasing q , J_{dif} reaches a maximum and then at $ql_{\text{dif}} \gg 1$ it decreases in proportion to q^{-1} . The conditions for the predominance of nonequilibrium diffusion over the equilibrium, $J_{\text{dif}} > J_{\text{dif}}^T$, turns out to be less stringent than the condition given in Ref. 8 of predominance of the nonequilibrium photoconductivity, $\sigma_{\text{ph}} > \sigma_{\text{ph}}^T$. One might expect the photoelectron diffusion in crystals characterized by large photoinduced fields to have a nonequilibrium character.

In §3 is calculated the field contribution $\delta \bar{j}_E$ to the current. Whereas in a uniform field \bar{E} there exist only the zeroth and first harmonic of the current, the presence of an oscillating component \bar{E} (with spatial frequency q) leads to generation of higher current harmonics. A solution of the kinetic equation can be obtained only in weak electric fields. In this limit we have

$$\bar{j} = \bar{\sigma} \bar{E} + \delta \bar{E}; \quad (6)$$

here $\bar{\sigma}$ has the meaning of the conductivity in the alternating field \bar{E} under uniform illumination, and δ is the oscillating part of the conductivity in a uniform field under inhomogeneous illumination (3). In the case of weak inhomogeneity $\bar{\sigma}$ and δ do not depend on q and constitute the constant and alternating parts of one quantity, the photoconductivity $\sigma_{\text{ph}}(J(\mathbf{r}))$ obtain in Ref. 8; they are connected by the obvious relation $\delta = M\bar{\sigma}$. In the limit $q^2 l_{\text{dif}}^2 > 1$ we land in the region of strong spatial dispersion; here $\bar{\sigma}$ and δ are independent quantities. The response δ of the medium to a constant field turns out to be exponentially small, whereas the decrease of $\bar{\sigma}$ with increasing q is smooth and follows a power law.

In §4 are discussed manifestations of nonequilibrium effects in the optical properties of crystals. The main conclusions are the following: 1) a considerable increase of diffraction efficiencies of the shifted gratings and a decrease of the lengths of the stationary unidirectional energy exchange compared with the prevailing notions³; 2) weakening of the influence of external electric fields in the PGE on the recording sensitivity at large spatial frequencies.

This section deals also with the experimental data that point to the nonequilibrium character of the diffusion of photoelectrons in LiNbO₃ crystals.

§1. BASIC EQUATIONS

As a rule the characteristic times in which J and E change are much longer than the photoelectron relaxation times. In this case stationary solutions of the kinetic equation suffice. Neglecting the crystal anisotropy and assuming a parabolic electron-dispersion law $\varepsilon_k = k^2 \hbar^2 / 2m$, this equation takes the form²⁾

$$\frac{eE}{\hbar} \frac{\partial}{\partial k_z} f_k + \frac{\hbar k_z}{m} \frac{\partial f}{\partial z} = \hat{I} f_k + Q_k \delta_{\varepsilon_0} (1 + M \cos qz). \quad (7)$$

We have assumed in (7) that the energy width of the electron source $\Delta \varepsilon < \varepsilon_0$; this made it possible to approximate its energy dependence by a δ function. In addition, we neglect depletion of the impurity levels and assume Q to be specified. The value of Q is connected with the light intensity J_0 by the relation

$$\int Q_k \delta_{\varepsilon_0} d\mathbf{k} = \beta \kappa J_0 / \hbar \omega,$$

where β is the quantum yield.

The z axis in (7) is directed along \mathbf{q} . The collision integral that describes scattering by phonons and by static defects can be substantially simplified by recognizing that the energy relaxation time γ^{-1} greatly exceeds the momentum relaxation time Γ^{-1} (Ref. 6). This allows us to retain only the first two terms of the expansion of f_k in Legendre polynomials,

$$f_k = f^0(\varepsilon, z) + f^1(\varepsilon, z) \cos \theta, \quad \theta = \angle \mathbf{k}, \mathbf{q}, \quad (8)$$

if the condition $q^2 l^2 \ll 1$ is satisfied. The action of the operator I on the first two harmonics $f_k^{0,1}$ is given by (see, e.g., Refs. 12 and 13)

$$\hat{I}f^0 = \hat{I}_\varepsilon f^0 = \varepsilon^{-1/2} \frac{\partial}{\partial \varepsilon} \varepsilon^{3/2} \gamma(\varepsilon) f^0, \quad \hat{I}f^1 \cos \theta = -\Gamma(\varepsilon) f^1 \cos \theta. \quad (9)$$

The first of them describes the energy relaxation via phonon emission and is valid at $\varepsilon \gg T$ and $\varepsilon \gg \Omega$. The second equation in (9) describes momentum relaxation; $\Gamma^{-1}(\varepsilon)$ is the usual transport time connected with the electron mean free path $l \approx v_k \Gamma_k^{-1} = \hbar k / \Gamma m$. The electron source Q_k in (7) can be chosen, without loss of generality, in the form

$$Q_k = Q_0 (1 + \xi \cos \theta). \quad (10)$$

The second term describes the asymmetry of the photoexcitation, i.e., it is responsible for the PVE, and $\xi < 1$ is the asymmetry parameter.⁷ Taking the foregoing into account, we can rewrite (7) in the form of a system of two partial differential equations

$$\begin{aligned} \frac{eE}{3\hbar k^2} \frac{\partial}{\partial k} k^2 f^1 + \frac{\hbar k}{3m} \frac{\partial f^1}{\partial z} &= \hat{I}_\varepsilon f^0 + Q_0 \delta_{\varepsilon_0} (1 + M \cos qz), \\ \frac{eE}{\hbar} \frac{\partial f^0}{\partial k} + \frac{\hbar k}{m} \frac{\partial f^0}{\partial z} &= -\Gamma f^1 + \xi Q_0 \delta_{\varepsilon_0} (1 + M \cos qz). \end{aligned} \quad (11)$$

The total current is expressed in terms of $f^1(\varepsilon, z)$

$$j = \frac{8\pi m}{3\hbar^3} \int_{\Delta} f^1(\varepsilon) \varepsilon d\varepsilon. \quad (12)$$

It will be shown below that a contribution to the integral (12) is made by the energy region $\varepsilon \sim \varepsilon_0$. This means that the non-equilibrium contribution to the current depends little on the position of the percolation level Δ and the integration can be carried out from $\Delta = 0$.

From (7) we can obtain directly a useful general relation for the current \bar{j} . We recognize that the quantity $\hat{I}_\varepsilon f_k$ can be represented as the divergence of the particle-number flux P_k :

$$I_\varepsilon f = -\frac{1}{k^2} \frac{\partial}{\partial k} k^2 P_k, \quad P_k = \frac{1}{2} k \gamma_k f_k.$$

The particle flux is directed towards lower energies: it contains a spatially-oscillating part \bar{P} and a constant part \bar{P} . Integrating (7) with respect to \mathbf{k} we obtain a general expression for the particle-number conservation law:

$$\frac{1}{e} \frac{\partial}{\partial z} \bar{j} = M \frac{\beta \kappa J_0}{\hbar \omega} \cos qz + 4\pi [k^2 \bar{P}_k]_{\hbar=0}. \quad (13)$$

Thus, to find the current \bar{j} it suffices to know the alternating part \bar{f} of the distribution function in the low-energy region.

To carry out concrete calculations it is necessary to specify the forms of the functions $\gamma(\varepsilon)$ and $\Gamma(\varepsilon)$. In most cases these are power-law functions of the energy:

$$\gamma(\varepsilon) = \gamma_0 (\varepsilon_0 / \varepsilon)^\alpha, \quad \Gamma(\varepsilon) = \Gamma_0 (\varepsilon_0 / \varepsilon)^\beta. \quad (14)$$

For the Fröhlich electron-phonon interaction, which is typical of ionic crystals, we have

$$\gamma = c_0 \Omega (\hbar \Omega / \varepsilon)^{3/2} \ln(4\varepsilon / \hbar \Omega), \quad \Gamma = c_0 \Omega (2N_f + 1) (\hbar \Omega / \varepsilon)^{1/2}. \quad (15)$$

Here c_0 is the interaction constant, Ω is the frequency of the longitudinal optical phonon, and $N_f(\Omega / T)$ is the equilibrium distribution of the phonons. We have thus with logarithmic accuracy $\alpha = 3/2$ and $\beta = 1/2$.

§2. SPATIALLY OSCILLATING CURRENT DUE TO THE PGE AND TO DIFFUSION

We put $E = 0$ in (11). It can be seen then that the distribution function contains only the zeroth and first Fourier harmonics:

$$f^s(\varepsilon, z) = \bar{f}^s(\varepsilon) + A^s(\varepsilon) \cos qz + B^s(\varepsilon) \sin qz, \quad s=0, 1. \quad (16)$$

Equation (11) breaks up in this case into three independent systems of equations. One system describes the spatially homogeneous \bar{f} part of the distribution function. The second and third systems determine the alternating part \bar{f} . The second and third systems determine the alternating part \bar{f} . The second system contains only the asymmetric (odd) part of Q_k responsible for the PGE, and determines A^1 and B^0 :

$$-\frac{\hbar q k}{3m} A^1 = \hat{I}_\varepsilon B^0, \quad \frac{\hbar q k}{m} B^0 = -\Gamma A^1 + \xi M Q_0 \delta_{\varepsilon_0}. \quad (17)$$

The third system of equations, containing only the symmetric (even) part of Q_k , describes A^0 and B^1 :

$$\frac{\hbar q k}{m} A^0 = \Gamma B^1, \quad \frac{\hbar q k}{3m} B^1 = \hat{I}_\varepsilon A^0 + M Q_0 \delta_{\varepsilon_0}. \quad (18)$$

Thus, the photovoltaic current can contain only an even number of spatial derivatives of the light intensity. In other words, the PVE is responsible for the undisplaced part of the current, and the diffusion for its displaced component. It is easily seen that this property follows directly from (7) (all that is needed is linearity of \hat{I} with respect to f_k), i.e., is independent of the relaxation mechanism and is valid at arbitrary electron mean free paths^{4) l}. Using (9), we easily obtain from (17) and (18)

$$A^0 = \frac{M Q_0 \varepsilon_0^{1/2}}{\gamma \varepsilon^{3/2}} e^{\varphi - \varphi_0}, \quad B^0 = \frac{\xi}{3} q l_0 A^0, \quad (19)$$

where

$$\varphi = \frac{2q^2}{3m} \int_0^\varepsilon \frac{d\varepsilon}{\gamma \Gamma}, \quad \varphi_0 = \varphi(\varepsilon_0). \quad (20)$$

These equations are valid at $\varepsilon \leq \varepsilon_0$, and at $\varepsilon > \varepsilon_0$ we have $A^0 = 0$ and $B^0 = 0$. In the case of a power-law dependence on the energy

$$\varphi = \frac{q^2 l_{dif}^2}{3\nu} \left(\frac{\varepsilon}{\varepsilon_0} \right)^\nu, \quad \varphi_0 = \frac{q^2 l_{dif}^2}{3\nu}, \quad (21)$$

where $\nu = \alpha + \beta + 1$ and $l_{dif} = l_0 (\Gamma_0 / \gamma_0)^{1/2}$. For the Fröhlich interaction $\nu = 3$. Relations (19)–(21) show how rapidly

the inhomogeneity of the electron spatial distribution decreases with decreasing energy. It can be seen that this decrease is very abrupt. At $\varphi_0 \gg 1$ the low-energy electrons with $\varepsilon \ll \varepsilon_0$, and with them also the thermalized electrons with $\varepsilon \sim T$, hardly feel the intensity modulation. The exponential decrease of A_0 and B_0 is a universal law due to the diffuse spreading of the spatial inhomogeneity.

Determining A^1 and B^1 from (17)–(19) and calculating the nonequilibrium contribution to the current, we obtain

$$\vec{j} = j_{\text{ph}}^0 M e^{-\varphi_0} \cos qz - e \frac{\kappa \nabla J}{\hbar \omega} \frac{1}{q^2} (1 - e^{-\varphi_0}), \quad (22)$$

where $j_{\text{ph}}^0 = \kappa \mathbf{G} J_0$ is the photovoltaic current under uniform illumination. In the case of weak inhomogeneity j_{ph} duplicates the intensity distribution. In this case the nonequilibrium contribution to the current can be written in a form similar to (2), provided that we introduce for the nonequilibrium electrons an effective density n and a diffusion coefficient D :

$$n = \frac{2}{3\nu} \frac{\beta \kappa J}{\hbar \omega \gamma_0}, \quad D = \frac{1}{2} \Gamma_0 l_0^2 = \frac{\varepsilon_0}{m \Gamma_0}. \quad (23)$$

It can be easily seen that $D = \mu \varepsilon_0 / e$, where $\mu = e / m \Gamma_0$ is the mobility of the nonequilibrium carriers. This formula is similar to the Einstein relation $D_T = \mu_T T / e$ for equilibrium particles; the average energy of the thermalized electrons is replaced by the characteristic energy ε_0 of the nonequilibrium electrons.

On going into the region of spatial frequencies the PVE, as can be seen from (22), is very strongly suppressed: at $q^2 l_{\text{dif}}^2 > 1$ it is exponentially small. The dependence of the diffusion current on the grating spacing is much weaker. At $\psi_0 \gg 1$ the current j_{dif} decreases with decreasing Λ and is independent of the electron mean free paths:

$$j_{\text{dif}} = \frac{M}{2\pi} e \frac{\beta \kappa J}{\hbar \omega} \Lambda \sin qz. \quad (24)$$

we point out that the laws governing the behavior of \vec{j} at $q^2 l_{\text{dif}}^2 > 1$ follow directly from the general relation (13). Since \vec{j}_k is exponentially small at low energies, owing to the diffuse spreading of the inhomogeneity, \vec{j} is practically completely determined by the first term of the right-hand side of (13), i.e., by the diffusion process.

We compare now the equilibrium and nonequilibrium diffusion currents. As already noted, j_{dif}^T is exponentially small at high spatial frequencies; we therefore have here practically always $j_{\text{dif}} \gg j_{\text{dif}}^T$. In the weak-inhomogeneity region we obtain as the condition for the predominance of nonequilibrium diffusion the inequality

$$\gamma_0^{-1} \gg \tau \frac{\mu_T}{\mu} \frac{T}{\varepsilon_0}. \quad (25)$$

Let us compare (25) with the condition $\sigma_{\text{ph}} > \sigma_{\text{ph}}^T$, which is necessary for the existence of anomalous photoinduced fields in crystals.⁷ The latter condition, obviously, is

$$\gamma_0^{-1} \gg \tau \mu_T / \mu. \quad (26)$$

Clearly, the condition (25) is much less stringent than (26). Thus, with increasing parameter $\mu_T \tau$ which characterizes the thermalized photoelectrons; the first to dominate is non-

equilibrium diffusion, and nonequilibrium photoconductivity sets in only later. Assuming by way of estimate an energy relaxation time $\gamma_0^{-1} = 10^{11}$ sec and $\mu_T T / \mu \varepsilon_0 = 10^{-4}$, we obtain from (25) a rather low condition $\tau < 10^{-7}$ sec for the electron lifetime in the band. This estimate is by far not the limit. One can expect nonequilibrium diffusion to be a typical phenomenon for PR crystals even at low spatial frequencies.

To conclude this section, we note the following nontrivial circumstance. Until recently the PGE effect was identified with asymmetry of the distribution function f_k , an asymmetry due to the asymmetry of the elementary electronic processes (ionization, recombination, scattering).⁷ Our analysis started out precisely from such premises. Recently, however, the existence of the so-called shift photovoltaic effect (SPGE) was demonstrated,¹⁵ which is not connected with asymmetry of the electron velocity distribution. The SPGE is due to the shift of the electrons in coordinate space in quantum transitions. The shift per absorbed photon is of the order of the crystal lattice constant a and does not depend on the electron mean free path. Therefore the influence of the inhomogeneity on the SPGE is determined by the small parameter a/Λ and not by the parameter L_{dif}/Λ as before. It can be shown, however, that a correction $\delta j_{\text{ph}}^{\text{shift}}$ to the photocurrent appears even in first order in the intensity gradient. It might seem that this circumstance could be used to distinguish between the PGE and the SPGE. The difficulties in this case are due to the presence of the diffusion contribution, which is phenomenologically similar to $\delta j_{\text{ph}}^{\text{shift}}$, but is larger by a factor $(l_{\text{dif}}/a)^2 \gg 1$. Thus, the SPGE turns out to be inessential for the study of the considered group of phenomena.

§3. ALLOWANCE FOR THE ELECTRIC FIELD

A spatially oscillating current \vec{j} can result also from the action of electric fields. The dc component \vec{E} of the field, as seen from (7), produces besides a direct current also a current \vec{j} with spatial frequency q . The alternating field \vec{E} due to the charge separation inside the crystal, generates higher current harmonics $q, 2q, 3q, \dots$. To determine all the fields produced in the crystal we must solve an infinite chain of equations for the Fourier harmonics of the distribution function. At an arbitrary value of E this problem can apparently not be solved analytically. We confine ourselves below to the case of weak fields

$$\vec{E}, E \ll E_0(q) = m \gamma_0 \Gamma_0 / e q, \quad (27)$$

when the kinetic equation can be solved by iteration. The condition (27) means that the characteristic drift length of a nonequilibrium photoelectron during the thermalization time should be less than the lattice period. In the approximation linear in the field E , the relation (6) is valid for the current. When calculating the correction δf_E to the distribution function, the asymmetry of the photoexcitation can be neglected by virtue of the smallness of the parameter ξ . Calculations for the field contribution $\delta \vec{j}_E$ to the current yield

$$\delta \tilde{f}_E = \frac{2}{3} \frac{e^2}{m} \frac{\beta \kappa J_0}{\hbar \omega} \left\{ E \int_0^\infty \frac{1}{\gamma e^{\gamma/2}} \frac{d}{d\epsilon} \left(\frac{\epsilon^{\gamma/2}}{\Gamma} e^{-\epsilon} \right) d\epsilon \right. \\ \left. + M \bar{E} \cos qz e^{-\varphi_0} \int_0^\infty \left[\frac{1}{\gamma e^{\gamma/2}} \frac{d}{d\epsilon} \left(\frac{\epsilon^{\gamma/2}}{\Gamma} \right) - \frac{4q^2}{3m} \frac{1}{\gamma^2 \Gamma^2} \right] d\epsilon \right\}. \quad (28)$$

In the limit of small spatial frequencies we arrive at the natural result:

$$\delta j_E = \sigma_{ph}(J(z)) E(z), \quad (29)$$

$$\sigma_{ph} = \frac{2}{3} \frac{e^2}{m} \frac{\beta \kappa J}{\hbar \omega} \int_0^\infty \frac{1}{\gamma e^{\gamma/2}} \frac{d}{d\epsilon} \left(\frac{\epsilon^{\gamma/2}}{\Gamma} \right) d\epsilon.$$

This case corresponds to complete neglect of the inhomogeneity in the left-hand side of the kinetic equation (7). In the strong-inhomogeneity region the response $\bar{\sigma}$ to a strong field is exponentially small. This result follows from the general formula (13). The reason for it is that a weak homogeneous field is incapable of influencing the oscillating part of the distribution function at low energies. Attention is called to the reversal of the sign of $\bar{\sigma}$ at $q^2 l_{dif}^2 \sim 1$. At first glance this result may seem paradoxical, since it means that in a constant field the total current j can be a minimum in regions where the light intensity is a maximum.⁵⁾ It must be borne in mind, however, that in the strong-inhomogeneity region (i.e., at strong spatial dispersion) the action of the electric field is different than in weak inhomogeneity. For weak inhomogeneity, δj_E is formed in accord with the usual scheme $f_0 \rightarrow f_1$, i.e., the field produces a current correction to the zeroth harmonic of the distribution function. If, however, the inhomogeneity is strong, a second channel for the onset of δj_E is also possible (see Eq. (11):

$$f_0 \xrightarrow{\nabla} f_1 \xrightarrow{\nabla} \delta f_0 \xrightarrow{\nabla} \delta f_1.$$

The contributions indicated are of opposite sign, become comparable in magnitude at $\varphi_0 \gg 1$, and cancel each other with high accuracy at $\varphi_0 \gg 1$ in accordance with (13).

It follows from (28) that in an alternating field \bar{E} and at high spatial frequencies the conductivity $\bar{\sigma}$ decreases not exponentially but in power-law fashion. Using (14) we obtain at $\varphi_0 \gg 1$

$$\bar{\sigma} = e \mu n \left(\frac{3}{2} - \alpha \right) \left(\frac{3\nu}{q^2 l_{dif}^2} \right)^{1-1/\nu} \Gamma \left(1 - \frac{1}{\nu} \right), \quad \nu = \alpha + \beta + 1 > 1, \quad (30)$$

where Γ is the Gamma function. It can be seen that the sign of $\bar{\sigma}$ and its decrease are determined by the singularities of the behavior of γ and Γ at $\epsilon < \epsilon_0$. In particular, for a Fröhlich interaction, neglecting the logarithmic dependence of γ on energy (see (15)), we have $\alpha = 3/2$ and consequently $\bar{\sigma}$ is exponentially small. When account is taken of the weak logarithmic dependence we have

$$\bar{\sigma}_{Fröhlich} \approx e \mu n \Gamma \left(\frac{2}{3} \right) \left\{ \frac{3}{q l_{dif} \ln(4\epsilon_0/\hbar\Omega)} \right\}^{1/3}. \quad (31)$$

The singularities in the behavior of $\bar{\sigma}(q)$ can again be connected with the general relation (13). To this end it must be

recognized that the influence of \bar{E} on the alternating part of the distribution function \tilde{f} is not connected at low energies with the diffuse spreading of the inhomogeneities with increasing energy relaxation. The field \bar{E} acts on the homogeneous part of \tilde{f} , which obviously is not exponentially small at low energies. It follows also from the foregoing that the contribution of the thermalized electrons to $\bar{\sigma}$ at large q does not contain an exponentially small quantity and can generally be substantial in the calculations.

§4. DISCUSSION OF RESULTS

Let us see how the investigated nonequilibrium effects manifest themselves in the nonlinear optic properties of crystals. From experiments on grating recordings of the refractive index one determines reliably the first Fourier harmonic of the spatially oscillating field $|E_1| = (E_1'^2 + E_1''^2)^{1/2}$ (Refs. 3 and 4). At the same time it is relatively easy to determine the phase shift between $J(z)$ and $E(z)$:

$$\psi = \text{arctg}(E_1''/E_1').$$

Thus, the aggregate of the measurements makes it possible to reconstruct completely the field $E_1(t)$ in the crystal. In sufficiently thick crystals an important role is assumed by energy exchange between the light beams that produce the grating (3). Data on the redistribution of the beam intensities at the exit from the crystal also contain complete information on the field E_1 (Refs. 3 and 11).

The main question that will interest us below is that of distinguishing, with the aid of the observed optical properties of the crystals, the photoelectric phenomena connected with the thermalized and nonequilibrium electrons.

We consider first the case of small spatial frequencies. Over times shorter than Maxwellian, $t \lesssim t_M \equiv \bar{E}/4\pi\sigma_{ph}$ (\bar{E} is the static dielectric constant) we have $\tilde{E} \approx 4\pi j t / \bar{E}$. In this case

$$\text{ctg } \psi = (E_{ph} - \bar{E}) / E_{dif}, \quad (32)$$

where $E_{ph} = j_{ph}/\sigma_{ph}$ is the blocking field due to the PGE and can be easily determined from independent experiments, \bar{E} is the applied field, and $E_{dif} = c q \epsilon_0 / e$ is the characteristic field connected with the nonequilibrium diffusion. The number $c \sim 1$ is determined by the details of the relaxation mechanisms; for a Fröhlich interaction $c = 1/3$. If the diffusion has an equilibrium character, we can make in (32) the substitution $E_{dif} \rightarrow E_{dif}^T = qT/e$. Since E_{dif}^T is a well-defined quantity, it can be easily determined from optical experiments whether the diffusion has an equilibrium character or whether it is connected with nonequilibrium electrons.

It appears that until recently no purposeful simultaneous measurements were made of the diffraction efficiency η and of the phase ψ as function of the lattice periods. An exception, to some degree is Ref. 16, in which a thorough study was made of the initial stage of recording a lattice with $\Lambda \approx 10^{-4}$ cm in LiNbO₃ crystals. According to data of this paper, the phase ψ amounted to 10.4° in a blocking field $E_{ph} = 50$ kV/cm. This corresponds to a diffusion field $E_{dif} \approx 10$ kV/cm, six times larger than the field E_{dif}^T . These data serve as direct indication of the nonequilibrium character of the photoelectron diffusion.

After long times, $t \gg t_M$, the field E reaches a stationary value determined from the condition $j = 0$. Under stationary conditions E is determined by the diffusion and has only a shifted component ($E'_1 = 0$):

$$E_1^T = -i\Phi(M)E_{dif}^T, \quad E_1 = -i\Phi(M)E_{dif}; \quad (33a)$$

$$\Phi = M / [1 + (1 + M^2)^{1/2}]. \quad (33b)$$

Thus, owing to the nonequilibrium character of the diffusion, the degree of modulation of the refractive index is increased under stationary conditions by a factor $c\varepsilon_0/T$, and the efficiency of diffraction by the grating, η , is by increased $(c\varepsilon_0/T)^2$ times. It is important that the optical characteristics of the crystal in the equilibrium-diffusion model are expressed only in terms of its well known parameters, namely the refractive index, the electro-optic constants and the temperature. The formation of stationary purely shifted gratings of the refractive index with diffraction efficiency larger than specified by the first equation in (33a) is a direct indication that the diffusion is in disequilibrium.

In the stationary state the nonequilibrium diffusion leads also to a considerable enhancement of the nonlinear interaction of the waves, particularly to an enhancement of the unidirectional energy exchange.^{11,17} All the equations obtained in Ref. 11 for the nonlinear interaction of the light beams remain in force if the characteristic length of the energy exchange is decreased by a factor $c\varepsilon_0/T$.

We call attention to the fact that in the region $q^2 l_{dif}^2 \ll 1$ the condition $E_{dif} < E_0$, which is necessary for the use of Eqs. (28), is automatically satisfied.

We proceed now to the case of large spatial frequencies. It can be seen from (30) that $\tilde{E} \gg E_0$ under stationary conditions, i.e., the criterion for the applicability of our description is not satisfied. The equations obtained are valid only at short recording times.

The main features of the nonequilibrium effects here are the following. The phase is close to $\psi \approx \pm \pi/2$, notwithstanding the fact that the blocking field E_{ph} connected with the PGE, and the external field E , are not small compared with E_{dif}^T ; the sign of the phase can depend on the grating spacing. The recording sensitivity depends little on the applied field \tilde{E} and decreases with decreasing Λ .

Although the stationary state cannot be described by our equations in the case of strong inhomogeneity, we can estimate the limiting value of \tilde{E} . Since the displaced field component can contain only odd powers of q , under station-

ary conditions we have $\tilde{E} \sim E_{dif}$. We note here that the field E_{dif} is the largest of all the characteristic fields, $E_{dif} > E_0$ and $E_{dif} > E_{ph}$. Such fields can actually produce breakdown inside the crystal.

- ¹¹ All properties of the thermalized electrons will hereafter be labeled by the letter T .
- ¹² A characteristic feature of the crystals considered by us is impurity absorption of the light. Only one type of carrier need therefore be considered.
- ¹³ We point out the substantial difference between the kinetic properties of nonequilibrium and thermalized electrons. The region $N^{-1} \ll q^2 l^2 \ll 1$, which for nonequilibrium photoelectrons is the strong-inhomogeneity region, does not exist at all for thermalized electrons.
- ¹⁴ In Ref. 14, which is based on semi-intuitive premises concerning the influence of the inhomogeneity, it was erroneously concluded that a displaced current component due to the PVE exists.
- ¹⁵ The total current is directed in this case along the field E .

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