Coupling of orthogonally polarized waves in barium titanate by parametric scattering

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We study the mechanism by which parametric scattering couples orthogonally polarized light waves in photorefractive $BaTiO_3$. We obtain explicit relationships that link the efficiency of diffraction by the fundamental lattice to the energy exchange in terms of the scattering characteristics. Finally, we obtain good qualitative agreement between theory and experiment. The results can be applied to many forbidden optical configurations in which direct photorefractive coupling is impossible. © *1996 American Institute of Physics.*
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1. INTRODUCTION

Intensive nonlinear scattering in photorefractive crystals of barium titanate was discovered and studied in Refs. 1–3. The scattering was induced by a pair of waves having equal frequency ω and the wave vectors $\mathbf{k}_p^o$ and $\mathbf{k}_p^e$, one of them being ordinary (o) and the other extraordinary (e). The scattered waves are ordinary; their wave vectors $\mathbf{k}$ and $\mathbf{k}'$ lie on the surface of a circular cone and correspond to the condition of synchronism:

$$\mathbf{k}_p^o+\mathbf{k}_p^e=\mathbf{k}+\mathbf{k}'. \tag{1}$$

The vectors of the conjugate parametric waves, $\mathbf{k}$ and $\mathbf{k}'$, correspond to the scattering angles φ and $\varphi'=\varphi+\pi$, respectively (Fig. 1).

As demonstrated in Refs. 1–3, parametric scattering is related to the formation of two spatial lattices of the refractive index (for each φ) with vectors $\mathbf{q}_1$ and $\mathbf{q}_2$,

$$\mathbf{q}_1=\mathbf{k}-\mathbf{k}_p^e\equiv\mathbf{k}_p^o-\mathbf{k}', \quad \mathbf{q}_2=\mathbf{k}-\mathbf{k}_p^o\equiv\mathbf{k}_p^e-\mathbf{k}', \tag{2}$$

(Fig. 2), and the diffraction of the extraordinary pump wave by these lattices.

The standard model of photorefractive nonlinearity[4,5] gives a fairly complete description of the properties of such scattering: the geometric and polarization properties, the sharp increase in scattering intensity resulting from an increase in the angle $2\theta_p$ between the pump beams, nontrivial kinetic characteristics, etc.

Intensive parametric scattering requires modifying the ideas, traditional for photorefractive crystals, about the mechanisms for the coupling of a pair of waves. Usually such coupling is caused by direct writing of the fundamental lattice at the difference spatial frequency (in our case this is the lattice with the vector $\mathbf{q}=\mathbf{k}_p^o-\mathbf{k}_p^e$). Direct writing means that light generates a spatially oscillating current with a period $2\pi/q$ and a corresponding electric field. However, as noted in Ref. 6, in the presence of parametric scattering there appears an additional contribution to the fundamental lattice. This contribution is caused by the nonlinearity of the material equations describing the field $\mathbf{E}$ of the spatial charge or, more precisely, by the presence of a contribution to the photocurrent $\kappa I\mathbf{E}$, where I is the light intensity, and κ is the photoconductivity constant. When lattices with vectors $\mathbf{q}_1$ and $\mathbf{q}_2$ are present in the crystal, because of nonlinearity there are sure to be lattices with spatial frequencies $2\mathbf{q}_1$, $2\mathbf{q}_2$, $\mathbf{q}_1\pm\mathbf{q}_2$, etc. Here, as Eqs. (2) and Fig. 2 imply, $\mathbf{q}_1-\mathbf{q}_2\equiv\mathbf{q}$ for any pair of parametric waves. In other words, the contributions from the pairs of parametric waves corresponding to different azimuthal angles φ build up in the fundamental q-lattice.

Another possibility of wave coupling by parametric scattering can be made to manifest itself in measurements of the efficiency of diffraction by the fundamental lattice by blocking one of the pump beams for a short time.[6] In this case, in addition to direct Bragg diffraction by the $\mathbf{q}$-lattice, there is successive two-stage diffraction by the $\mathbf{q}_1$- and $\mathbf{q}_2$-lattice (see Fig. 2). As a result, energy flows into the blocked beam. Thus, because of parametric scattering there appears a fictitious contribution to the efficiency of diffraction by the fundamental lattices.

Of course, the above coupling mechanisms are important only when the intensity of parametric scattering is high and the direct coupling of the pump waves by the fundamental lattice is weak. This is the situation with $BaTiO_3$. On the one hand, the geometry of the experiment depicted in Fig. 1 is optimal for parametric scattering. On the other hand, because of the orthogonality of polarizations of the pump waves, direct writing of the fundamental $\mathbf{q}$-lattice by drift and diffusion of photoexcited carriers is impossible. Direct writing becomes possible if we employ the polarization-dependent photovoltaic effect.[7–9] However, in $BaTiO_3$ this effect is weak.[6,7,10–12]

The aim of the present study is to develop a theory of wave coupling by parametric scattering and to use the theory in analyzing the experimental data. In Sec. 2 we derive the general relationships for the field of the space charge at the spatial sum and difference frequencies in photorefractive crystals. In Sec. 3 these relationships are used to calculate the

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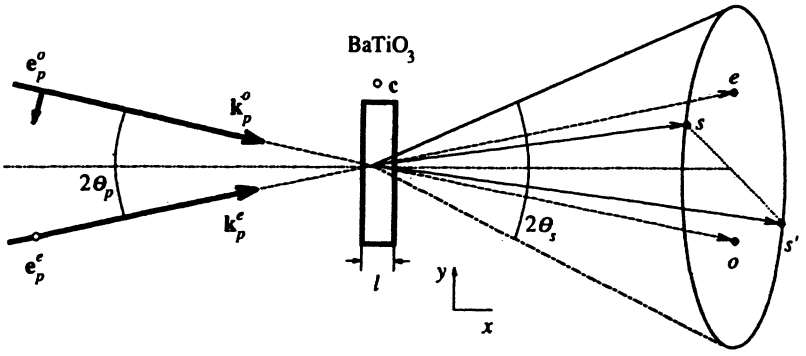

FIG. 1. The geometry of the experiment. Points s and s' on the scattered light cone correspond to a parametrically conjugate pair of waves, and points o and e correspond to the pump beams.

parametric-scattering contribution to the amplitude of the fundamental lattice. What is important is that this contribution can be expressed in terms of an easily determined scattering characteristic, the total scattering intensity. In Sec. 4 we calculate the main observable characteristics of two-beam coupling, the efficiency of diffraction by the fundamental lattice and the energy exchange. These characteristics allow for the possibility of weak direct coupling by the photovoltaic effect.[7] In Sec. 5 we find the two-stage contribution to the diffraction efficiency and show it to be small in $BaTiO_3$. Section 6 is devoted to a comparison of the theoretical ideas developed here and the experimental data for the barium titanate crystal. In the Conclusion we formulate the main results of our work.

2. THE SPACE CHARGE FIELD AT SPATIAL COMBINATION FREQUENCIES

Let the intensity of the light in the crystal be

$$I=I_0+[I_1\exp(i\mathbf{q}_1\mathbf{r})+I_2\exp(i\mathbf{q}_2\mathbf{r})+\text{c.c.}], \tag{3}$$

where I_0 is the spatially homogeneous part of the intensity. Such a spatial distribution of light is formed, in particular, by orthogonally polarized pump waves (o and e) and a pair of weak parametric waves with wave vectors $\mathbf{k}$ and $\mathbf{k}'$ [see Eqs. (1) and (2)]. The electric current density induced by the light can be written as

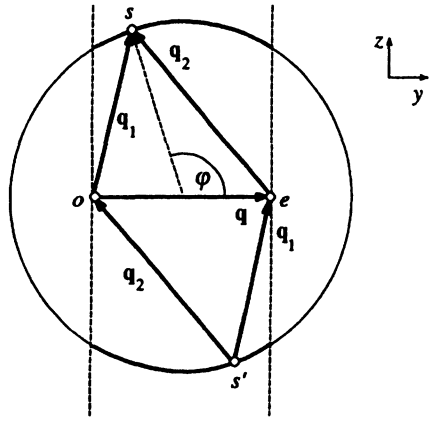

FIG. 2. Arrangement of lattice vectors. The points o, e, s, and s' denote the tips of the vectors $\mathbf{k}_p^o$, $\mathbf{k}_p^e$, $\mathbf{k}$, and $\mathbf{k}'$. The "heavy" parts of the ring correspond to the gain region, where $\Gamma(\varphi)>0$.

$$\mathbf{j}_\Sigma=\kappa I\mathbf{E}+\mathbf{j}, \tag{4}$$

where $\mathbf{j}$ is the current density not related to carrier drift in the field $\mathbf{E}$ of the spatial charge. This current density may contain contributions related to the photovoltaic effect,[7] the diffusion of photoexcited carriers, and the drift of these carriers in an externally applied uniform electric field. We assume that $\mathbf{j}$ can be represented in the form (3) with coefficients $\mathbf{j}_0$, $\mathbf{j}_1$, and $\mathbf{j}_2$.

Assuming that $\mathbf{E}=-\nabla\psi$, we write the steady-state condition $\operatorname{div}\mathbf{j}_\Sigma=0$ in the form

$$I\Delta\psi+(\nabla\psi\nabla I)=\kappa^{-1}\operatorname{div}\mathbf{j}. \tag{5}$$

If we also assume that $I_{1,2}\ll I_0$, we can employ the following expansion to find the potential ψ:

$$\psi=\psi^{(1)}+\psi^{(2)}+\cdots, \tag{6}$$

where $\psi^{(1)}$ incorporates terms that are linear in $I_{1,2}$, $\psi^{(2)}$ incorporates quadratic terms, and the like. In first order we have

$$\psi^{(1)}=\psi_1\exp(i\mathbf{q}_1\mathbf{r})+\psi_2\exp(i\mathbf{q}_2\mathbf{r})+\text{c.c.},$$
$$\psi_{1,2}=-i\frac{(\mathbf{q}_{1,2}\mathbf{j}_{1,2})}{\kappa I_0 q_{1,2}^2}. \tag{7}$$

Accordingly, the amplitudes of the electric fields for the q_1- and q_2-lattice are

$$\mathbf{E}_{1,2}=\mathbf{n}_{1,2}E_{1,2},\quad E_{1,2}=-\frac{(\mathbf{n}_{1,2}\mathbf{j}_{1,2})}{\kappa I_0}, \tag{8}$$

where $\mathbf{n}_{1,2}=\mathbf{q}_{1/2}/q_{1,2}$ are the unit lattice vectors. Equations (8) are quite obvious.

To second order in $I_{1,2}/I_0$ Eq. (5) yields

$$\Delta\psi^{(2)}=-\frac{1}{\kappa I_0^2}[\tilde{I}\operatorname{div}\mathbf{j}+\kappa I_0(\nabla\psi^{(1)}\nabla\tilde{I})], \tag{9}$$

where $\tilde{I}=I-I_0$ is the spatially oscillating part of the intensity. As Eqs. (7)–(9) show, $\psi^{(2)}$ contains contributions corresponding to the spatial frequencies $\mathbf{q}_1\pm\mathbf{q}_2$, $2\mathbf{q}_1$, and $2\mathbf{q}_2$. For the amplitudes of the first two lattices, $\mathbf{E}_\pm$, Eqs. (7)–(9) readily yield $\mathbf{E}_\pm=\mathbf{n}_\pm E_\pm$, where $\mathbf{n}_\pm=(\mathbf{q}\pm\mathbf{q}_2)/|\mathbf{q}_1\pm\mathbf{q}_2|$, and

$$\mathbf{E}_+=-I_0^{-1}[E_1(\mathbf{n}_1\mathbf{n}_+)I_2+E_2(\mathbf{n}_2\mathbf{n}_+)I_1],$$
$$E_-=-I_0^{-1}[E_1(\mathbf{n}_1\mathbf{n}_-)I_2^*+E_2^*(\mathbf{n}_2\mathbf{n}_-)I_1]. \tag{10}$$

These relationships express in general form the amplitudes of combination lattices in terms of the amplitudes $E_{1,2}$ and the angles between the vectors $\mathbf{n}_{1,2}$ and $\mathbf{n}_{\pm}$.

Now let us examine the important case where the main mechanism of spatial lattice formation is the diffusion of photoexcited electrons or holes. Because of the orthogonality of polarizations of the pump beams there can be no diffusion current $\mathbf{j}$ at the spatial frequency $\mathbf{q}$, i.e., direct writing of the fundamental lattice by diffusion is impossible. But, owing to the presence of the frequencies $\mathbf{q}_{1,2}$ in the intensity distribution (3), lattices with amplitudes

$$E_{1,2}=\pm iE_{1,2}^{D}\frac{I_{1,2}}{I_0}, \tag{11}$$

are written in the crystal (here $E_{1,2}^{D}=q_{1,2}T/e$ is what is known as the diffusion field, T is the temperature, and e is the absolute value of the electron charge). The "plus" in (11) corresponds to holes and the "minus" to electrons. The presence of the unit imaginary number in (11) means that the q_1- and q_2-lattice are "shifted," i.e., the field distribution in each is shifted by one-quarter of the period in relation to the corresponding intensity distribution. As is known,[4,5] this fact plays an important role in describing nonlinear optics phenomena in photorefractive crystals.

3. A FUNDAMENTAL LATTICE INDUCED BY PARAMETRIC WAVES

We write the total electric field of the light waves in a crystal as follows:

$$\mathscr{E}=\left[a_p^o\mathbf{e}_p^o\exp(i\mathbf{k}_p^o\mathbf{r})+a_p^e\mathbf{e}_p^e\exp(i\mathbf{k}_p^e\mathbf{r})+\int_0^{\pi}(a_k\mathbf{e}_k^o\exp(i\mathbf{kr})+a_{k'}\mathbf{e}_{k'}^o\exp(i\mathbf{k}'\mathbf{r}))d\varphi\right]\times\exp(i\omega t)+\text{c.c.}, \tag{12}$$

where $a_p^{o,e}$ and $a_{k,k'}$ are the amplitudes of the pump waves and the conjugate parametric waves, and $\mathbf{e}_p^{o,e}$ and $\mathbf{e}_{k,k'}^o$ are the corresponding real unit polarization vectors. The degree of modulation of the intensity at the spatial frequencies $\mathbf{q}_1$ and $\mathbf{q}_2$ in (10) is determined in our case by o-waves, since $\mathbf{e}_p^e\mathbf{e}_p^e=0$ and $\mathbf{e}_p^e\mathbf{e}_k^o\approx\theta_p^2\ll 1$. Hence for the $\mathbf{q}_1$- and $\mathbf{q}_2$-lattice corresponding to the azimuthal angle φ (Fig. 2) we must put

$$\frac{I_1}{I_0}=\frac{a_p^o a_{k'}^*}{|a|_\Sigma^2},\quad \frac{I_2}{I_0}=\frac{a_k a_p^{o*}}{|a|_\Sigma^2}, \tag{13}$$

where $|a|_\Sigma^2=|a_p^o|^2+|a_p^e|^2$.

By substituting (11) and (12) into (10) we can easily see that the partial contribution to the amplitude of the fundamental $\mathbf{q}$-lattice, corresponding to the parametric pair $\mathbf{k}$ and $\mathbf{k}'$, is expressed in terms of the diffusion field $E_q^D=qT/e$. More than that, we must allow for the fact that the sum of phases of the partial waves is equal to the sum of phases of the pump waves: $\Phi_k+\Phi_k'=\Phi_p^o+\Phi_p^e$. This follows directly from the formulas of Refs. 1 and 2 and is typical of parametric wave processes. As a result, the total contribution of the partial waves to the fundamental lattice amplitude, $E_q^s=\int E_-(\varphi)d\varphi$, assumes the form

$$E_q^s=\pm iE_q^D\frac{I_p^o}{I_\Sigma^2}\exp[i(\Phi_p^o-\Phi_p^e)]\times\int_0^{\pi}\sqrt{I_s(\varphi)I_s(\varphi+\pi)}d\varphi, \tag{14}$$

where $I_s(\varphi)$ is the intensity of parametric scattering for the angle φ, and $I_\Sigma=I_p^o+I_p^e$.

As Eq. (14) implies, the fundamental lattice induced by parametric waves is shifted. The direction of the spatial shift in relation to the isophasal planes, $\mathbf{qr}+\Phi_p^o-\Phi_p^e=0, 2\pi, 4\pi, \ldots$, depends on the sign of the charge carriers. The upper sign corresponds to holes, as it does in (11).

Equation (14) allows for various generalizations. First we note that it does not change if the anisotropy in photoconductivity is taken into account. The reason is that in the diffusion charge-separation mechanism considered here the induced electric field is determined by the ratio of the carrier diffusion coefficient to the carrier mobility, $D/\mu=T/e$, which is isotropic. Allowing for anisotropy in light absorption results in a modification of Eq. (14), which amounts to the substitution $I_\Sigma\equiv I_p^o+I_p^e\to I_p^o+\alpha_e I_p^e/\alpha_o$, where $\alpha_{o,e}$ are the absorption coefficients for the ordinary and extraordinary waves. In the $BaTiO_3$ sample used in our experiments and by Holtmann *et al.*,[6] $\alpha_o/\alpha_e\simeq 2$ at 514 nm.

Further simplification of Eq. (14) can be achieved by allowing for the fact that for $I_s\ll I_\Sigma$ the intensity of parametric scattering increases exponentially with the distance x from the entrance surface with a growth rate[1,2]

$$\Gamma(\varphi)=\frac{4\pi^2 n_o^3 r_{51}T}{e\lambda^2}\frac{\sqrt{I_p^o I_p^e}}{I_\Sigma}\sqrt{\theta_s^2\sin^2\varphi-n_o\Delta n}, \tag{15}$$

where n_o is the ordinary refractive index, $\Delta n=n_o-n_e$ is the double-refraction index, r_{51} is the electro-optical constant, θ_s is the polar scattering angle in air, and λ is the wavelength. According to (15), the growth rate Γ is the same for all angles φ and $\pi\pm\varphi$; it reaches its maximum value Γ_m at $\varphi=\frac{1}{2}\pi$. Hence the main contribution to the lattice amplitude E_q^2 is provided by the nearest neighborhood of point $\varphi=\frac{1}{2}\pi$, and the integral in (14) can be calculated by the saddle-point method.[13] As a result, E_q^s can be expressed in terms of the total parametric-scattering intensity I_Σ^s as follows:

$$E_q^s\simeq-\frac{i}{2}E_q^D\frac{I_p^o I_\Sigma^s}{I_\Sigma^2}\exp[i(\Phi_p^o-\Phi_p^e)]. \tag{16}$$

Note that $I_\Sigma^s\propto\exp(\Gamma_m x)$, and for this reason the parametric contribution to the fundamental lattice is concentrated primarily at the rear face of the crystal.

4. DIFFRACTION BY THE FUNDAMENTAL LATTICE

Let us calculate the main observable characteristics of the fundamental lattice. These are the energy exchanged between the pump beams and the diffraction efficiency η, i.e., the fraction of the radiation that has been diffracted with a change in polarization in the direction $\mathbf{k}^o$ (or $\mathbf{k}^e$) with only

one incident test wave with a wave vector $\mathbf{k}^e$ (or $\mathbf{k}^o$) satisfying the Bragg condition $\mathbf{k}^o-\mathbf{k}^e=\mathbf{q}$. In the simplest case, η is measured by blocking one pump beam (in our case $\mathbf{k}^{o,e}=\mathbf{k}_p^{o,e}$) for a short time.

To calculate these characteristics, we use the equations for the amplitudes of the ordinary and extraordinary pump waves under the conditions of Bragg diffraction:[7,9]

$$\frac{\partial a_p^o}{\partial x}=-i\frac{\pi n_o^3 r_{51}}{\lambda}(\mathbf{ne}_p^o)a_p^e E_q,$$
$$\frac{\partial a_p^e}{\partial x}=-i\frac{\pi n_o^3 r_{51}}{\lambda}(\mathbf{ne}_p^o)a_p^o E_q^*, \tag{17}$$

where $\mathbf{n}=\mathbf{q}/q$. In addition to the parametric contribution E_q^s discussed in Sec. 3, the amplitude E_q of the fundamental lattice can contain an independent contribution E_q^{pv} caused by the photovoltaic effect,[6,7]

$$E_q^{pv}=i(\mathbf{ne}_p^o)(E_{12}^C+iE_{15}^L)\frac{a_p^o a_p^{e*}}{|a_-|_\Sigma^2}, \tag{18}$$

where E_{12}^C and E_{15}^L are the electric fields characterizing the transverse photovoltaic effect in class $4mm$ crystals (these fields are expressed in terms of the corresponding components of the photovoltaic tensor). If pump depletion is ignored, E_q^{pv} is independent of x.

Note that Eq. (18) describes direct writing of the $\mathbf{q}$-lattice by orthogonally polarized waves, which is impossible by the mechanism of drift and diffusion of photoexcited carriers. In $BaTiO_3$ the contribution (18) is definitely weak, but its detection and study are of fundamental interest.[6,7,11,12,14]

In what follows, the diffraction by the $\mathbf{q}$-lattice is assumed weak. The assumption is justified by the experimental results and further estimates.

Let us first find the diffraction efficiency η measured with one of the pump beams being blocked for a short time. Allowing for the fact that $I_\Sigma^s\propto\exp(\Gamma_m x)$ and using Eq. (15) for Γ, we obtain from (17) the following:

$$\eta=|\sqrt{\eta^{pv}}\exp(i\Phi_{pv})\mp\sqrt{\eta^s}|^2, \tag{19}$$

where η^{pv} and η^s are the diffraction efficiencies caused, respectively, by the photovoltaic effect and parametric scattering,

$$\sqrt{\eta^{pv}}=\frac{\pi n_o^3 r_{51} l}{\lambda}(\mathbf{ne}_p^o)^2\frac{\sqrt{I_p^o I_p^e}}{I_\Sigma}|E_{12}^C+iE_{15}^L|,$$
$$\sqrt{\eta^s}=\frac{1}{2}\sqrt{\frac{I_p^o}{I_p^e}}\frac{I_\Sigma^s(l)}{I_\Sigma}. \tag{20}$$

Here l is the crystal thickness, and $\Phi_{pv}=\arg(E_{12}^C+iE_{15}^L)$. Under these assumptions, $\eta\ll 1$.

Note that the efficiency η^s can be expressed very simply in terms of an observable characteristic, the intensity of parametric scattering at the exit from the crystal. As Eq. (19) implies, the result of adding the contributions to η strongly depends on the phase Φ_{pv}, i.e., on the relationship between the characteristic fields E_{12}^C and E_{15}^L.

Now let us study the energy exchange between the pump waves caused by self-diffusion by the fundamental lattice. Using Eqs. (17), for small intensity variations we find that

$$\delta I_p^{o,e}=\pm 2\sqrt{I_p^o I_p^e}(\sqrt{\eta^{pv}}\cos\Phi_{pv}-\sqrt{\eta^s}). \tag{21}$$

The "minus" in front of $\sqrt{\eta^s}$ refers to hole photoconductivity, which is most typical of $BaTiO_3$ (photoelectrons correspond to the "plus" sign). The chosen sign corresponds to the direction of energy exchange $o\to e$ caused by parametric scattering. The first term on the right-hand side of Eq. (21) describes the well-known effect of unidirectional energy exchange caused by a circular photovoltaic effect, i.e., the contribution E_{12}^C in Eq. (18).

Separating the contribution to energy exchange caused by the difference lattice,

$$\delta I_p^{o,e}=\mp I_\Sigma^s(l)\left[1+\left(\frac{I_p^e}{I_p^o}\right)\right]^{-1}, \tag{22}$$

we note that the fraction of the intensity passed from one beam to the other is comparable in magnitude to the total intensity of parametric scattering (it is approximately half of the intensity at $I_p^o/I_p^e\approx 1$).

5. TWO-STAGE DIFFRACTION

Now suppose that after steady-state parametric scattering has been achieved we block the ordinary pump beam. Let us find the contribution to the amplitude of the o-wave with the wave vector $\mathbf{k}_p^o$ caused by two- stage diffraction of the pump e-wave. At the first stage, waves with the wave vectors $\mathbf{k}$ and $\mathbf{k}_1$ appear as a result of diffraction by the $\mathbf{q}_1$- and $\mathbf{q}_2$-lattice (see Fig. 2). The amplitudes of these waves, a_k and a_k', are practically equal to the amplitudes of the parametric waves in the presence of both pump beams. This fact manifests itself most vividly in experiments and is related to the anomalously high value of the electro-optical constant r_{51} in $BaTiO_3$ (see Ref. 4). This constant responsible for diffraction accompanied by a change in polarization type ($e\to o$ or $o\to e$) exceeds the constants r_{13} and r_{33} responsible for diffraction of the $o\to o$ and $e\to e$ types, by a factor of at least 15. The intensities of the diffracted waves here are proportional to the square of the electro-optical constant [see, e.g., Eq. (17)]. Thus, because of the anomalously large value of r_{51}, the parametric waves draw their energy from the extraordinary pump wave, while the ordinary pump wave is needed to form the $\mathbf{q}_1$- and $\mathbf{q}_2$-lattice.

At the second stage the parametric waves $\mathbf{k}$ and $\mathbf{k}'$ are diffracted by lattices with vectors $\mathbf{q}_2$ and $\mathbf{q}_1$, respectively, into an ordinary wave with the wave vector $\mathbf{k}_p^o$ (see Fig. 2). This process does not change the type of polarization; it is characterized by the electro-optical constant r_{13} and is described by an equation for the amplitude $a^o\equiv a_{k_p^o}$:

$$\frac{da^o}{dx}=-i\frac{\pi n_o^3 r_{13}}{\lambda}(a_k E_2^* n_{2z}+a_{k'}E_1 n_{1z}). \tag{23}$$

Using (11), we can easily see that the entire dependence of the right-hand side of Eq. (23) on the azimuthal scattering angle φ is contained in the factor $\sin\varphi(I_k^2-I_{k'}^s)$. Because of

the symmetry of the problem with respect to the xy plane, $I^s_{k'}=I^s(\pi-\varphi)$, and hence the total contribution to a^o over all parametric wave pairs vanishes. Actually, cancellation of the contributions occurs for each pair in two pairs corresponding to the angles φ and $\pi-\varphi$. Thus, the diffraction mechanism of charge separation does require allowing for two-stage diffraction in $BaTiO_3$. This is also true when the blocked pump beam is extraordinary rather than ordinary.

6. COMPARISON OF RESULTS OF CALCULATIONS WITH THE EXPERIMENTAL DATA AND A DISCUSSION

The experiments were carried out using the same $BaTiO_3$ sample as in the studies of nonlinear light scattering.[1-3,6] The sample was cut along the crystallographic axes, and its thickness l was 2.6 mm. Its photoconductivity increased linearly with light intensity, and the charge carriers were holes.

Using a telescope, we expanded the cross section of the beams incident on the crystal so that the entire sample volume was uniformly illuminated. Immediately behind the crystal we placed a diaphragm 2 mm in diameter, which made it possible to easily recalculate the powers of the transmitted, lattice-diffracted, and scattered waves measured by a detector into the respective intensities in the sample.

As for parametric scattering, our experiment (just as in Refs. 1–3) demonstrated good qualitative agreement and adequate quantitative agreement between the experimental data and the results of calculations. The polar angle θ_s and the polarization of the scattered waves are in strict quantitative agreement with the theoretical values. The azimuthal distribution of the parametric waves in steady-state conditions corresponds to the Γ vs φ dependence, i.e., steady-state scattering occurs within allowed angle intervals (where the growth rate $\Gamma(\varphi)$ is positive) and reaches its maximum at $\varphi=\pm\frac{1}{2}\pi$ [see Eq. (15)]. Within the forbidden angle intervals (where $\Gamma(\varphi)$ is imaginary), only transient parametric scattering is observed, which agrees with the theoretical results.[2,3] The intensity of steady-state scattering sharply increases with the angle $2\theta_p$ between the pump beams. For convergence angles $\theta_p\geqslant 14°$ considerable depletion of the extraordinary pump beam was observed. In accordance with the theoretical results,[2,3] the time it takes parametric scattering to develop exceeds the time of dielectric relaxation, i.e., the characteristic time of the photorefractive nonlinear response, by a factor of approximately ten.

Let us now examine the data directly characterizing the formation of a fundamental lattice by parametric waves. Figure 3 depicts the results of simultaneous measurements of the efficiency of diffusion by the fundamental lattice and the intensity of parametric scattering at $\theta_p=5°$ and $\lambda=514$ nm. The diffraction efficiency η was measured by blocking the incident extraordinary pump beam for a short time and measuring the intensity of the transmitted extraordinary wave with the wave vector $\mathbf{k}^e_p$. Two stages in the development of $\eta(t)$ can be clearly seen in Fig. 3. For small time intervals comparable with the dielectric relaxation time, the efficiency η becomes saturated at a relatively low level η_0; for all practical purposes at this stage there is still no parametric

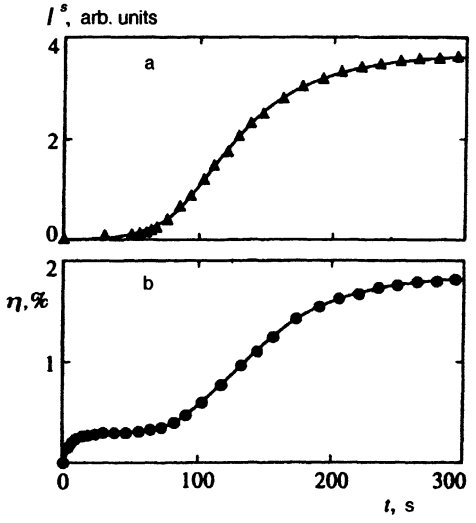

FIG. 3. Time dependence of the scattering intensity (a) and the diffraction efficiency (b) at $\theta_p=5°$, $I^o_p=5.5$ mW, and $I^e_p=2.3$ mW.

scattering. At the second stage the efficiency slowly grows; it becomes saturated at a higher level. The resumption of the growth in $\eta(t)$ at the second stage obviously correlates with the development of parametric scattering.

On the basis of theoretical analysis and the above data it is natural to assume that the main contribution to the diffraction efficiency is related to indirect writing of the fundamental lattice by parametric waves, while the contribution η_0 corresponds to direct writing of the $\mathbf{q}$-lattice by the pump waves owing to the photovoltaic effect. This assumption is directly supported by the following additional experiments. Fairly strong and rapid oscillations of the crystal in the $\pm z$ direction completely suppress parametric scattering, and with it the second stage in the temporal evolution of η (see Ref. 6). At the same time, these oscillations have no effect on η_0. This feature can be given an elementary explanation by using the present theoretical ideas. Parametric scattering and the corresponding contribution η^s are related to the $\mathbf{q}_1$- and $\mathbf{q}_2$-lattice (see Fig. 2). At $\theta_p=5°$ these vectors are almost parallel to the z axis. Hence displacements of the crystal in the $\pm z$ direction easily suppress the writing of $\mathbf{q}_{1,2}$-lattice. At the same time, oscillations of the crystal cannot have a strong effect on direct photovoltaic writing of the fundamental lattice by the pump waves, since $\mathbf{q}\perp\mathbf{z}$.

The absence of a noticeable contribution of two-stage diffusion to the intensity of the diffracted wave was verified by reading the fundamental lattice with an additional test beam from a HeNe laser. The fact that this beam satisfied the conditions of Bragg diffraction by the fundamental lattice excludes, because of a change in λ, the possibility of simultaneous two-stage diffraction. At the same time, in our experiment we actually observed the same temporal evolution of η as the one depicted in Fig. 3.

The above data belong to diffraction of a light beam by the fundamental lattice. The energy exchange between the pump beams caused by parametric scattering becomes well-defined at angles $\theta_p>5°$. Figure 4 depicts the time dependence of $I^{o,e}_p(t)$ at the exit from the crystal and the depen-

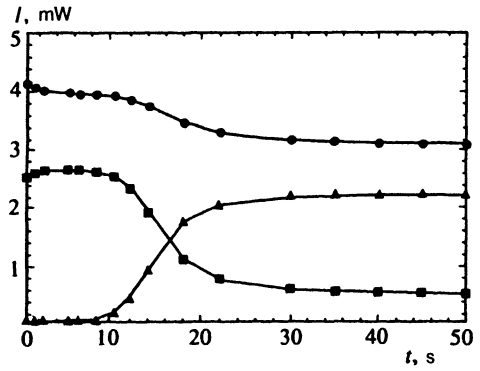

FIG. 4. Time dependence of the intensities I_p^o (dots), I_p^e (squares), and I_Σ^s (triangles) at the exit from the crystal for $\theta_p = 14°$.

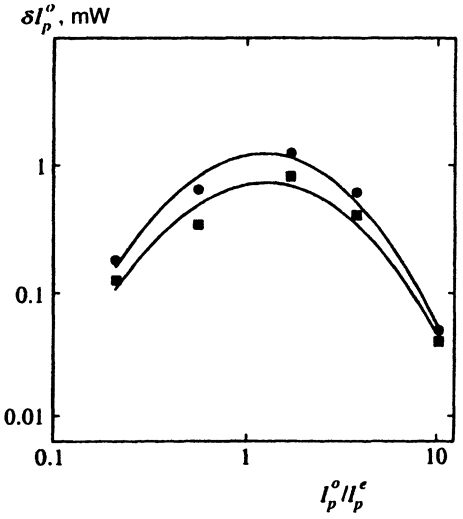

FIG. 5. The dependence of the intensity of an ordinary wave transformed into an extraordinary wave as a result of self-diffraction by the difference lattice, on the intensity ratio for the incident waves. The dots represent the results of calculations by formula (22) with the scattered light intensity taken from the experiment, and the squares represent the results of direct measurements.

dence of $I_\Sigma^s(t)$ at $\theta_p = 14°$. Again two stages in the temporal development are evident. For small time intervals there is a slight transfer of energy from the ordinary pump wave to the extraordinary, and parametric scattering is practically nil. As scattering develops, I_p^e decreases sharply and becomes smaller than I_Σ^s, with I_p^o experiencing a slight decrease. The total intensity at the exit, $I_p^o + I_p^e + I_\Sigma^s$, remains practically constant.

The decrease in intensity of the extraordinary pump beam is caused by strong diffraction into parametric o-waves. The diffraction of the ordinary pump beam into the parametric waves is small in the parameter $(r_{13}/r_{51})^2 \approx 10^{-3}$. Hence the decrease of $I_p^o(t)$ at the second stage must be interpreted as diffraction of the ordinary pump wave into the extraordinary by the fundamental lattice. This process is again controlled by the largest electro-optical constant r_{51}. The direction of energy exchange, $o \to e$, agrees with the theory, which predicts precisely this sign of the effect for crystals with hole conduction.

Now let us make some numerical estimate based on Eqs. (20) and (21) and the experimental data. In Ref. 6 it was noted that the maximum diffraction efficiency of the lattice, coupling the incident waves directly, amounts to several percentage points for a beam convergence angle θ_s of approximately 11°. Bearing in mind that according to the data of Fig. 4 the intensity of the scattered light normalized to the total intensity of the pump beams, $I_\Sigma^s(l)/I_\Sigma$, is approximately 0.4 at $\theta_s \approx 14°$ (which is close to the above-noted value) and $I_p^o/I_p^e = 1.7$, we conclude that the expected value of the maximum diffraction efficiency by formula (20),

$$\eta^s = \frac{1}{4}\left(\frac{I_p^o}{I_p^e}\right)\left(\frac{I_\Sigma^s(l)}{I_\Sigma}\right)^2 \approx 0.07, \tag{24}$$

is in good agreement with the experimental data.

Note that in all estimates of this section the intensity I_Σ has been correlated with the difference in the absorption coefficients of the extraordinary and ordinary waves: $I_\Sigma \equiv I_p^o + \alpha_e I_p^e/\alpha_o$.

By using the data of Figs. 3a and b we can compare the results of calculations with the experimental data. Assuming that the contributions of the photovoltaic lattice and the difference lattice ($\Phi_{pv} = 0$) are added coherently, we can extract the contribution of the difference lattice from the data of Fig. 3b by employing Eq. (19):

$$\sqrt{\eta^s} = \sqrt{\eta} - \sqrt{\eta^{pv}} = 0.026. \tag{25}$$

If we allow for the fact that the transmitted beams had an intensity of 2.3 mW and 5.5 mW, we find that $I_p^o/I_p^e \approx 2.4$, which according to (20) yields $\sqrt{\eta^s} \approx 0.01$.

Thus, the calculated value is approximately half the experimental one. For a theory without fitting parameters this result can be assumed adequate. The reason for this discrepancy could lie in the method of measuring the efficiency, in which the intensity of the diffracted signal is divided by the intensity of the transmitted beam rather than by the intensity of the incident beam. Another source of error could be the understating of the intensity of the scattered light in recording the divergent light beams by additional collimating optical devices.

Here is an estimate of the values of the energy exchange on the shifted difference lattice. Using the well-known data on the intensity of scattered light, I_Σ^s, and the initial ratios of the pump beam intensities (Fig. 4), we can calculate $(\delta I_p^o)_{\text{cal}}$ by employing (20) and (21) and compare the results with the experimental data in Fig. 4. For $(\delta I_p^o)_{\text{exp}}$ we took the difference of the intensities of this beam at $t = 5$ s (by which time the energy exchange on the shifted "photovoltaic" lattice has stabilized) and $t = 60$ s (by which time the formation of the "parametric" lattice has been completed). As a result we obtain $(\delta I_p^o)_{\text{exp}} = 0.8$ and $(\delta I_p^o)_{\text{cal}} = 1.24$, i.e., the experimentally measured value of energy exchange is smaller than the calculated value by a factor of approximately 1.5.

By making similar estimates for other ratios of incident beam intensities we can build the dependence $(\delta I_p^o) = f(I_p^o/I_p^e)$ depicted in Fig. 5. Clearly, in the entire interval with noticeable nonlinear scattering, qualitatively the

experimental points follow the calculated dependence, with the discrepancy amounting to a factor of 1–5.

We also note that according to the theoretical estimates (see the last part of Sec. 4), the fraction of intensity pumped from the ordinary wave to the extraordinary is comparable to the parametric scattering intensity.

Thus, we can speak of good qualitative and even semi-quantitative agreement between theory and experiment.

Up to this point we did not touch on the nature of the direct writing of the fundamental lattices by extraordinary and ordinary pump waves. The above results suggest that such writing can be interpreted as a manifestation of the transverse photovoltaic effect in $BaTiO_3$, with the characteristic fields E_{12}^C and E_{15}^L being roughly 1 V cm^{-1}.

7. CONCLUSIONS

In optical configurations in which direct photorefractive coupling is ineffective, two-beam coupling can be achieved by parametric scattering. The mechanism of such indirect coupling can be the addition of spatial combination lattices induced by parametric waves and two-stage diffraction. In $BaTiO_3$, due to its specific electro-optical properties, the first coupling mechanism is dominant. The efficiency of diffraction by the fundamental lattice and the energy exchange between orthogonally polarized waves in $BaTiO_3$ can be expressed in terms of observable scattering characteristics. For a broad spectrum of characteristics there is good qualitative agreement between theory and experiment.

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