Phase conjugation in $BaTiO_3$ by use of the indirect photorefractive coupling of orthogonally polarized light waves

P. Jullien, P. Mathey, and P. Lompré

Laboratoire de Physique, Equipe des Matériaux pour l'Optique Non Linéaire, Université de Bourgogne, Bâtiment Mirande, B.P. 400, 21011 Dijon Cédex, France

S. Odoulov

Institute of Physics, National Academy of Sciences, 265 650, Kiev-22, Ukraine

B. Sturman

Institute of Automation and Electrometry, Russian Academy of Sciences, 630 090 Novosibirsk, Russian Federation

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A phase-conjugate wave is generated when an ordinary (extraordinary) signal wave is mixed with two counterpropagating extraordinary (ordinary) waves in the plane normal to the $BaTiO_3$ polar axis. The photorefractive grating that couples the ordinary and the extraordinary waves appears if the incident waves induce a noticeable conical parametric scattering; this grating is a difference grating of many noisy scattering gratings recorded by means of the usual diffusion-mediated charge transport. For comparable intensities of signal and pump waves this type of nonlinear wave mixing is much more efficient than that which is due to the circular bulk photovoltaic effect. © 1998 Optical Society of America [S0740-3224(98)01507-0]

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1. INTRODUCTION

Frequency-degenerate four-wave mixing is known to be an efficient tool for generation of phase-conjugate light waves.[1,2] This technique was shown to be especially attractive with photorefractive crystals for conjugation and self-conjugation of low-power cw laser beams.[3,4]

Because the interacting waves have the same temporal frequency, in nonlinear materials they form standing refractive-index gratings, and the phase-conjugate wave appears because of diffraction of the pump waves from these gratings.[5] In this paper we study the phase conjugation in a $BaTiO_3$ crystal in one of the so-called forbidden configurations[6,7] in which the refractive-index grating cannot be recorded directly but can appear through high-order mixing processes with scattered light. The waves with orthogonal polarization indirectly record a grating, which couples these waves[7] and produces the phase-conjugate replica when it is read out by the counterpropagating pump wave. This process of vectorial wave mixing is especially pronounced in $BaTiO_3$ because of the important electro-optic constant r_{42} that is responsible for the so-called anisotropic diffraction.[7]

Exactly the same geometry of backward-wave four-wave mixing was used previously for phase conjugation in media with pronounced photovoltaic charge transport.[8,9] One purpose of this study is to develop criteria for distinguishing the phase conjugation that is due to the bulk photovoltaic effect[9] from that studied in this paper, which is due to difference grating recording. Apart from its discussion fundamental principles, this study is important in that it proposes a correct evaluation of the photovoltaic constants, especially if the coupling strength is rather small.

2. EXPERIMENT

The experimental setup is shown schematically in Fig. 1. An expanded light beam from a single-mode single-frequency Ar^+ laser (with a Gaussian beam waist of $\sim$1 mm) is split into three waves to form two counterpropagating pump beams, 1 and 2, and a signal beam, 3; the last signal beam is incident at an angle 2θ with respect to the pump beams. At first glance this arrangement is the same as that usually used for backward-wave four-wave mixing.[1–3] However, there are two important distinctions: (i) the polarization of the signal wave is orthogonal to that of the pump waves (an ordinary signal beam and extraordinary pump beams are shown in Fig. 1 as an example) and (ii) the wave vectors of all three waves lie in the plane normal to the crystal c axis.

A $\lambda/2$ phase retarder is used to rotate the polarization of the signal wave; another phase retarder, put in front of beam splitter BS2 together with polarizer P1, serves to control the overall intensity of waves 1 and 3. The intensity ratios of waves 1–3 are adjusted with phase retarder $\lambda/2$, polarizer P2, and neutral-density filters ND.

The $BaTiO_3$ crystal with dimensions $3.6\ \text{mm} \times 6.1\ \text{mm} \times 6.0\ \text{mm}$ along the X, Y, and Z axes is nominally undoped. It is colorless and has absorption constants $\alpha_o = 0.86\ \mathrm{cm}^{-1}$ and $\alpha_e = 0.38\ \mathrm{cm}^{-1}$ at $\lambda = 0.514\ \mu\mathrm{m}$. When the $BaTiO_3$ crystal is illuminated by two copropagating orthogonally polarized light waves (1 and 3) the nonlinear scattering in the cone of the ordinarily polarized waves[10] is induced. This type of nonlinear scattering is attributed to the parametric amplification of weak ordinary waves that meet the phase-matching condition

$$\mathbf{k}_1{}^e + \mathbf{k}_3{}^o = \mathbf{k}_s{}^o + \mathbf{k}_i{}^o, \qquad (1)$$

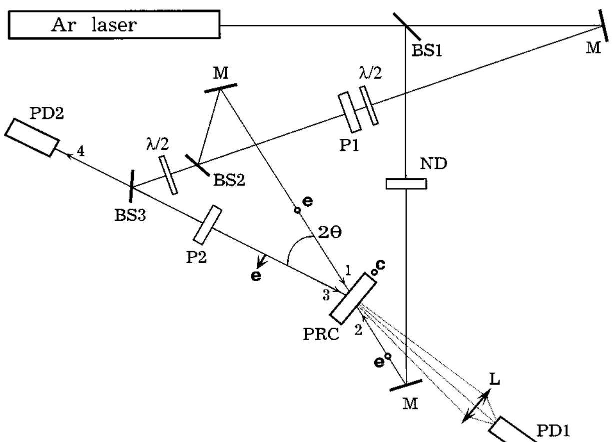

Fig. 1. Schematic representation of the experimental setup: M's, mirrors; PRC, photorefractive crystal; other abbreviations are defined in text.

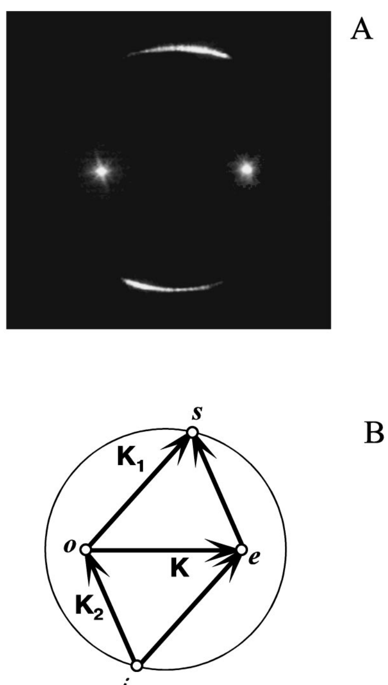

Fig. 2. A, Intensity distribution on the screen placed 20 cm behind the sample in the direction of the propagation of waves 1 and 3. Two bright dots mark the positions of the transmitted incident waves. B, Grating vector diagram to explain the parametric conical scattering. The tips of the wave vectors of the signal (3) and the pump (1) waves are labeled o and e; the tips of the wave vectors of the scattered waves are labeled s and i.

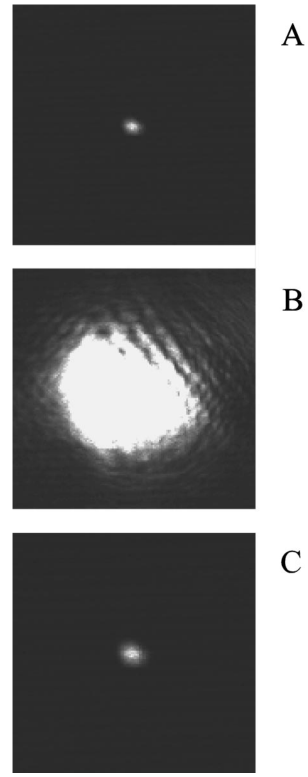

Fig. 3. Far-field intensity distributions, A, for two pump waves and an initial signal wave; B, for a distorted signal wave; C, for a conjugate beam passing back through the distorter (lens). The divergence of the beam in B is ~0.5°.

where $\mathbf{k}_{1,3}$ are the wave vectors of the incident waves, whereas $\mathbf{k}_s$ and $\mathbf{k}_i$ are the wave vectors of an arbitrary pair of scattered waves with the tips of wave vectors pointing to the diameter of the scattering ring; the subscripts o and e denote the ordinary and extraordinary polarization states, respectively. According to the classification given in Refs. 11 and 12, this type of nonlinear wave mixing is labeled an $A{:}oe \rightarrow oo$ process. The scattered light, which is shown in Fig. 1 by dotted lines, is collected with lens L and sent to detector PD1.

The scattering pattern from waves 1 and 3, which impinge upon the sample at an angle $2\theta \approx 35°$ (in air), is shown in Fig. 2A. Note that the scattered light intensity can be comparable with the intensity of transmitted waves 1 and 3. Figure 2B is a grating vector diagram for this type of parametric scattering; it is considered during the discussion in Section 3.

If in addition pump wave 2 is sent to the sample with the help of the beam splitter BS1 and mirror M (see Fig. 1), one can observe the appearance of wave 4 propagating in the backward direction to signal wave 3. A part of its intensity is measured through beam splitter BS3 with detector PD2 (Fig. 1). The polarization of this wave is the same as that of the signal wave; i.e., it is ordinarily polarized.

To prove that wave 4 is phase conjugate to wave 4 we put a converging lens (focal length $F = 40$ cm) into the signal beam in front of the sample. Figure 3A represents the far field of the initial signal beam; Fig. 3B, the far field of the incident beam when the lens is added; Fig. 3C, that of the generated wave 4 after it has passed back through the lens. Note that the angular divergence of the signal wave is ~0.5°; i.e., the light beam is conjugated with the divergence much larger than the angular window imposed by the phase-matching condition [Eq. (1)] for parametric conical scattering.

Figure 4 shows the temporal dependence of the scattered light intensity (signal from photodetector PD1) and of the intensity of wave 4 (signal from photodetector PD2). An obvious correlation of these two curves (except at a short initial time interval) suggests the conclusion that the relevant processes are not independent.

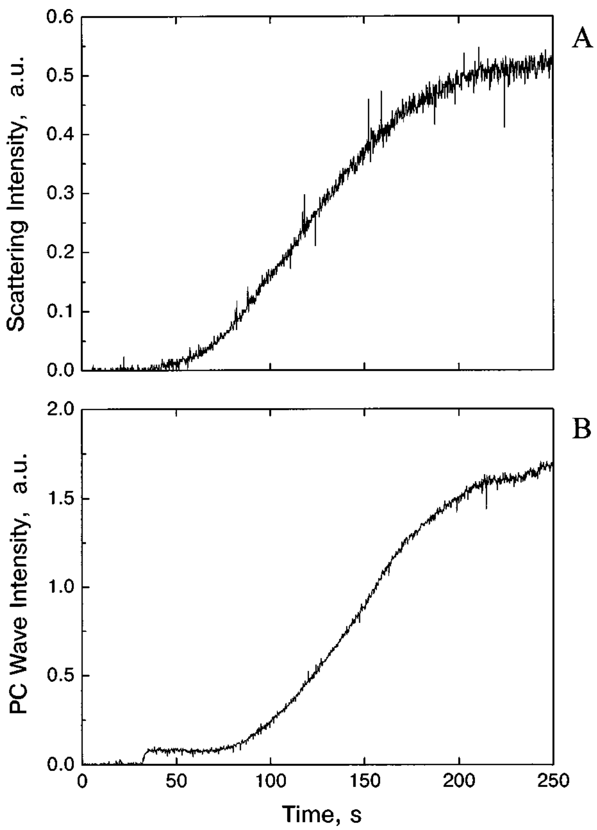

Fig. 4. Temporal development of A, the total intensity of the conical scattering, and of B, the phase-conjugate (PC) beam intensity. Two extraordinary pump waves permanently illuminate the sample while the signal wave is switched on at $t = 30$ s.

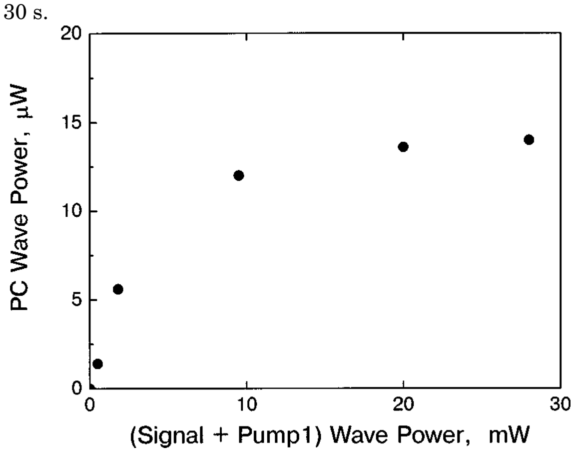

Fig. 5. Steady-state intensity of the phase-conjugate (PC) wave as a function of the overall intensity of copropagating waves 1 and 3. The power of the readout wave 2 is 0.5 mW.

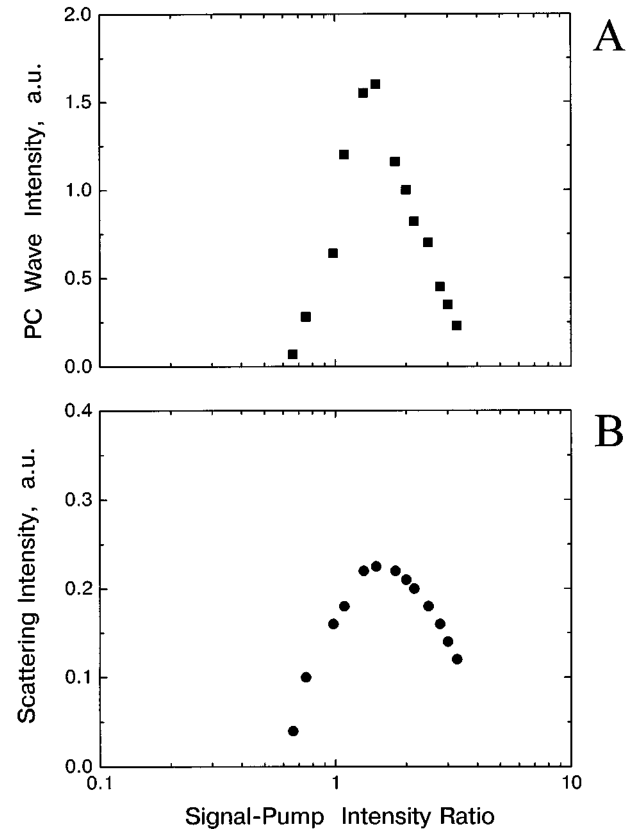

Fig. 6. A, Phase conjugate (PC) wave intensity and B, scattered light intensity versus the intensity ratio of the signal wave to the copropagating pump wave. The intensity of the counterpropagating pump wave is 10 times smaller than that of the copropagating pump wave.

Note that the buildup time of the light-induced scattering is much longer than the characteristic buildup time of grating recording with the two identically polarized waves (0.3 s for the same total intensity). Therefore the slow development of the diffracted signal points out the unusual origin of the grating that couples orthogonally polarized waves 1 and 3 and diffracts wave 2. Thus we conclude that the appearance and growth of the intensity of wave 4 are related to the development of the light-induced scattering rather than to the direct recording of the photorefractive grating by the two incident waves (which would be a much faster process).

By measuring the steady-state intensity of wave 4 (from the curves similar to that shown in Fig. 4B) we plot various intensity-ratio dependences. Figure 5 shows the intensity of wave 4 as a function of total intensity of waves 1 and 3, provided that the ratio $I_1 : I_3 \approx 1:1$ is kept constant. As one can see, the larger the intensity of the recording waves (1 and 3), the larger is the intensity of phase-conjugate wave 4.

With the known powers of the signal and pump waves (14.6, 5.2, and 13.5 mW for waves 1, 2, and 3, respectively), the phase-conjugate reflectivity (R_{PC}) and the diffraction efficiency (η) of the grating that couples orthogonally polarized waves 1 and 3 can be evaluated to be $R_{\mathrm{PC}} \approx 10^{-3}$ and $\eta \approx 10^{-2}$.

The intensity of the phase-conjugate wave can also be plotted versus the intensity ratio of the signal wave and the copropagating pump wave. In fact, the intensities of the two pump waves were kept constant during the entire experiment (20 and 2.1 mW for pumps 1 and pump 2, respectively) while the intensity of the signal wave was varied. Figure 6 demonstrates the result: The largest intensity of the phase-conjugate wave is reached for nearly (but not exactly) equal intensities of the signal and the pump waves. Figure 6B shows the dependence of the scattered intensity on the intensity ratio of the signal wave and the counterpropagating pump wave. Again, a good correlation between the curves for the intensity of the phase-conjugate wave and for the scattered light intensity is obvious.

3. DISCUSSION

The results described can be explained when we take into account the space-charge gratings that appear as a consequence of nonlinear mixing of a large number of noisy gratings that are responsible for the strong conical light-induced scattering.[13,14]

Let us assume that the cone of scattered waves with ordinary polarization is already developed. Then a certain scattered wave (with wave vector $\mathbf{k}_s{}^o$) records, together with ordinary signal wave 3, a grating with grating vector $\mathbf{K}_1 = \mathbf{k}_s{}^o - \mathbf{k}_3{}^o$ (see Fig. 2B). In the same manner the idler parametric wave with wave vector $\mathbf{k}_i{}^o$ records the other grating with grating vector $\mathbf{K}_2 = \mathbf{k}_3{}^o - \mathbf{k}_i{}^o$. Because of the inherent nonlinearity of the diffusion-mediated charge transport (both the diffusion current and the photoconductivity are spatially modulated, which results in deviation of the spatial distribution of the space-charge field from sinusoidal), the higher spatial harmonics ($2\mathbf{K}_1$, $2\mathbf{K}_2$, $3\mathbf{K}_1$, $3\mathbf{K}_2$, etc.) and also the sum and the difference gratings (with grating vectors $\mathbf{K}_1 \pm \mathbf{K}_2$) appear in the sample in addition to gratings $\mathbf{K}_{1,2}$ recorded directly. It is easy to verify (Fig. 2B) that for the difference grating with the grating vector $\mathbf{K} = \mathbf{K}_1 - \mathbf{K}_2$ all three incident waves and the generated phase-conjugate wave meet the Bragg diffraction condition, i.e.,

$$\mathbf{K} = \mathbf{k}_1{}^e - \mathbf{k}_3{}^o = \mathbf{k}_4{}^o - \mathbf{k}_2{}^e. \tag{2}$$

Note that Eq. (2) is valid for an arbitrary component of the conical scattering; therefore all the scattered light waves will contribute to the space-charge field with spatial frequency $\mathbf{K}$.[7,13]

In such a way, a difference grating can appear that couples two orthogonally polarized waves incident upon the crystal, even with purely diffusion-mediated charge transport. This grating may be efficiently read out by counterpropagating pump wave 2, giving rise to wave 4, which is phase conjugate with respect to signal wave 3 (the so-called anisotropic diffraction related to the largest for $BaTiO_3$ electro-optic constant r_{42}). Waves 2 and 4 will enhance the noisy gratings responsible for the conical scattering; a new cone of scattered light will appear with the opposite direction of propagation (e.g., waves with wave vectors $\mathbf{k}_s{}'$ and $\mathbf{k}_i{}'$ conjugate to those with wave vectors $\mathbf{k}_s$ and $\mathbf{k}_i$). Thus the buildup of wave 3 results from the coherent superposition of many elementary processes of nonlinear mixing of eight waves $(1, 2, 3, 4, s, s', i, i')$.

The scattered waves are of primary importance for this mixing process; the larger the intensity of the scattered waves, the stronger the initial noisy gratings with grating vectors $\mathbf{K}_1$ and $\mathbf{K}_2$, and, consequently, the stronger the difference grating with grating vector $\mathbf{K} = \mathbf{K}_1 - \mathbf{K}_2$. The theory of indirect coupling of two incident light waves (1 and 3 in our case) caused by the development of the difference gratings is given in Refs. 7 and 11. The expression for the diffraction efficiency of the difference grating, η_s, derived in the undepleated-pump approximation is

$$\eta_s = (I_3{}^o/I_1{}^e)[I_\Sigma{}^s(l)/2I_\Sigma]^2, \tag{3}$$

where $I_3{}^o$ and $I_1{}^e$ are the intensities of the recording waves and $I_\Sigma{}^s(l)$ is the integral intensity of conical scattering, where I_Σ is the total intensity of light inside the sample and l is the sample thickness.

Equation (3) can be used for a qualitative analysis of our results. The diffraction efficiency of the difference grating in our case is defined as the intensity ratio of phase-conjugate wave 4 to readout wave 2. [In fact, the efficiency of the difference grating related to waves 1 and 3 is affected by the presence of readout wave 2 and also by the generated waves, 4, s', and i'; this is why only a qualitative comparison with Eq. (3) is possible.]

First, in accordance with the theory the grating efficiency increases nonlinearly with the intensity of the scattered light. This result is in agreement with the experimental data. At the initial stage of exposure the temporal development of the scattered intensity is smoother than that of the phase-conjugate wave intensity (see Fig. 4), which can be expected because $\eta_s \propto [I_\Sigma{}^s(l)]^2$. Comparing the signal-to-pump ratio dependences for the intensities of the phase-conjugate wave and the scattered wave, we can see that the half-width for the scattering intensity curve is larger than that for phase-conjugate wave intensity (see Fig. 6), also in agreement with Eq. (3).

Second, the diffraction efficiency depends on the intensity ratio of the recording waves, mainly because the intensity of the scattered light strongly depends on the ratio

$$I_\Sigma{}^s(l) \propto \exp(\Gamma l),$$

where $\Gamma = (8\pi^2 n_o{}^3 r_{42} k_B T/e\lambda^2)[(I_3{}^o I_1{}^e)^{1/2}/I_\Sigma](\theta_s{}^2 - n_o\Delta n)^{1/2}$ is the gain factor, $\Delta n = n_o - n_e$ is the crystal birefringence, $n_{o,e}$ are the refractive indices for the ordinary and the extraordinary waves, respectively, e is the electron charge, k_B is the Boltzmann constant, T is the absolute temperature, and θ_s is the apex angle of the scattering cone inside the sample.

The factor $[(I_3{}^o I_1{}^e)^{1/2}/I_\Sigma]$ that enters into the expression for the gain factor Γ is of main interest for us. The largest intensity of the scattered light is achieved at nearly equal intensities of waves 1 and 3. The small deviation (for a factor α_o/α_e) from exact equality $I_3{}^o = I_1{}^e$ is known to be due to the difference in photoconductivity for ordinary and extraordinary waves.[15] This deviation is clearly seen in Fig. 6.

Finally, the dependence presented in Fig. 5 can also be explained qualitatively. The smaller the intensity of readout wave 2 is in comparison with the total intensity of copropagating beams 1 and 3, the larger the efficiency of the difference grating becomes. When I_2 becomes so small that its contribution to the photoconductivity is negligible, the diffraction efficiency tends to saturation. Just for this range of power ratios the comparison with the calculation[13] is well justified, as wave 2 can be considered the probe wave, which does not affect the difference grating created by copropagating waves 1, 3, s, and i.

$BaTiO_3$ crystals belong to the $4mm$ group of spatial symmetry; therefore they exhibit the bulk photovoltaic effect.[16,17] One of the independent contributions to the photovoltaic current directed perpendicularly to the c axis can be excited only if an ordinary wave and an extraordinary wave are present simultaneously in the sample (so-called spatially oscillating photovoltaic currents[18]). This unusual charge transport process, which is sensitive to the light-field polarization, ensures the possibility of grating recording by two orthogonally polarized waves. While in doped $LiNbO_3$ and $LiTaO_3$ this process is highly efficient,[8] in $BaTiO_3$ the contribution of the photovoltaic grating is rather small.[10,13,19–21] In the case of optimized conditions for recording of the difference grating, the corresponding contribution to the intensity of the phase-conjugate wave is smaller than 10%. This fact is shown in Fig. 4, where the initial growth and saturation of the phase-conjugate wave intensity (exposure time from 30 to 70 s) are attributed to the direct recording of the grating with grating vector $\mathbf{K}$ by means of the bulk photovoltaic effect.

Note, in addition, that the strong coupling that is due to the difference grating buildup may lead to overestimation of the photovoltaic constant measured from the wave mixing experiments. A relatively large effect of coupling of ordinary and extraordinary components of one beam in Codoped $BaTiO_3$ (Ref. 20) is obviously misinterpreted as being caused by photovoltaic charge transport. To measure the photovoltaic constants correctly one should avoid the formation of a difference grating by suppressing the light-induced scattering (e.g., by using two writing waves with considerably different intensities or by vibrating the sample in the direction normal to the grating vector[10]).

In conclusion, we observed phase conjugation in a $BaTiO_3$ crystal in the forbidden configuration in which the incident light waves cannot record a grating by diffusion-mediated charge transport. The appearance of a phase-conjugate wave is interpreted as a result of anisotropic diffraction from the difference grating, self-developing from nonlinear interaction of the scattering noisy gratings, recorded by the usual diffusion process. In spite of the fact that the parametric conical scattering (with its rather severe angular selectivity because of phase-matching requirements) is involved in the wave mixing process that we have considered, phase conjugation of the light beam with considerable divergence is shown to be possible.

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Address correspondence to S. Odoulov (e-mail: odoulov@marion.iop.kiev.ua).

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