Degenerate Parametric Light Scattering in Periodically Poled $LiNbO_3$:Y:Fe

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The first observation of parametric light scattering patterns (rings, lines, and dots) in bulk periodically poled nonlinear media is reported. Development of the scattering patterns proves efficient photorefractive grating recording and considerable parametric gain for seed radiation in this new nonlinear material. Several novel phase-matched parametric processes, caused by the periodicity of the domain structure, are revealed.

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In many photorefractive crystals, propagation of one or two coherent light beams leads to a vast variety of parametric scattering processes [1,2] that manifest themselves as specific light patterns—rings, lines, and dots of scattered light. Even within a cw intensity range, a considerable part of the incident beams can be transformed into scattered light owing to the very high efficiencies of the relevant parametric processes (the rate of exponential spatial amplification can be of the order of 10^2 cm^{-1}). These efficiencies are typically higher than those of the photorefractive nonlinear processes responsible for wide-angle light-induced scattering [2].

These parametric processes, degenerate (or almost degenerate) in frequency, can be interpreted as a cycle of the following events: Initially, refractive index gratings are recorded by the incident pump wave(s) and weak seed waves scattered from the surface and bulk optical imperfections. This results in an enhancement of the seed waves because of Bragg diffraction of the pump wave(s) from the recorded gratings. Then, amplification of the scattered waves increases the amplitudes of the index gratings. As a consequence, a nonlinear growth of the scattered light occurs. Two main conditions are necessary to achieve strong nonlinear scattering: *phase matching and a sufficiently strong photorefractive nonlinear response*. The first condition defines the propagation directions for parametric waves, whereas the second one ensures an efficient grating recording.

The studies of parametric processes are important from the fundamental point of view because the scattering patterns are the fingerprints of various charge transport phenomena (responsible for grating recording) in photorefractive crystals [1,2]. These phenomena can be polarization sensitive, which is, e.g., the case for $LiNbO_3$ crystals [2,3]. These studies are also promising for such applications as image amplification [4], phase conjugation [2,5], and development of sensors based on the controllable changes of the crystal birefringence [6]. Lastly, they shed light on the problem of seed scattering, as parametric processes are initiated by noise through the effect commonly known as "fanning" [7]. In short, the above parametric processes exhibit a wide range of characteristic features, from fundamental and general to special, related to particular material properties.

Until now, starting from the first publication [8], the photorefractive parametric scattering has been studied exclusively in single-domain crystals which provide the best conditions for recording of the index gratings. However, a serious drawback of single-domain crystals is that their photorefractive nonlinearity remains very strong even in the limit of low spatial frequencies, causing strong distortions of light beams known as "optical damage" [2].

Recently, it has been shown [9–11] that periodically poled lithium niobate (PPLN), which consists of domains with opposite directions of spontaneous polarization, is a new photorefractive material combining an efficient grating recording with inhibited optical damage. Initially PPLNs have been attracting much interest because of their high efficiency in quasi-phase-matched processes, e.g., frequency doubling and parametric conversion [12]. They proved to be of great interest also as a nonlinear photonic band-gap material [13].

In this Letter, we report on the first observation and experimental study of parametric photorefractive scattering in periodically poled $LiNbO_3$:Y:Fe crystals. The detection of pronounced parametric processes is direct proof of the efficiency of this engineered nonlinear material. Furthermore, the presence of the periodic domain structure leads to a new set of phase-matched parametric processes and allows for the observation of many new scattering processes in bulk PPLN:Y:Fe. They can run not only for identically but also for orthogonally polarized pump beams.

Periodically poled crystals of $LiNbO_3$ are grown in Moscow University by the Czochralski technique from congruently melted composition [14], and contain 0.74 wt. % of Y and 0.006 wt. % of Fe. The period of the domain structure, L, is $\simeq 7$ μm and the domain walls are normal to the growth direction (OX crystallographic

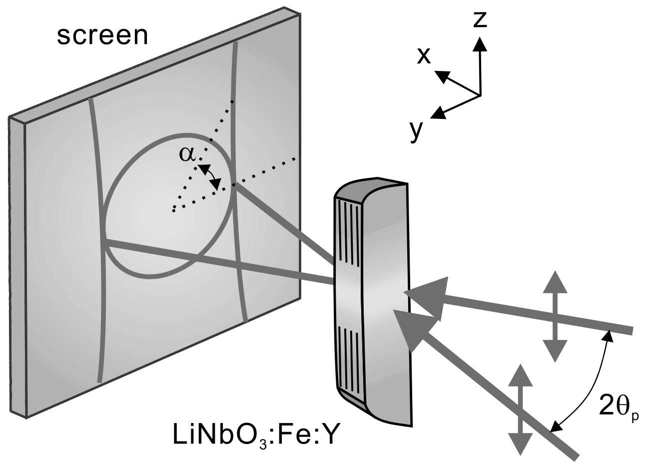

FIG. 1. Schematic of experiment.

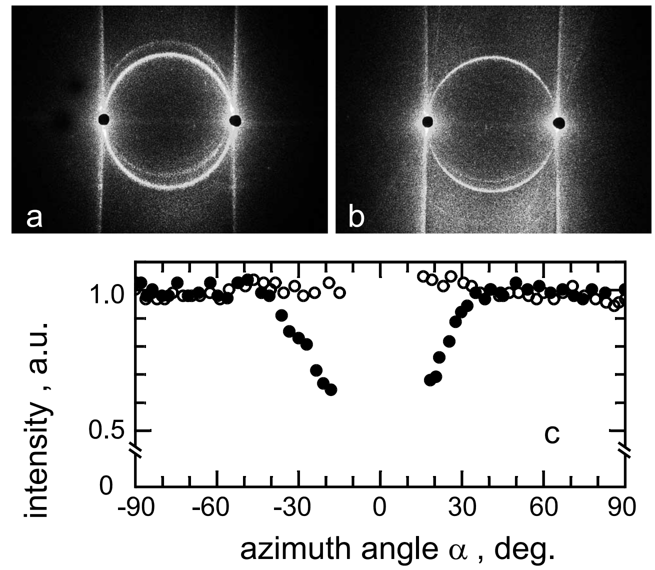

FIG. 2. Far-field distributions of scattered light for single-domain (a) and PPLN (b) parts of the sample. The ghost images are caused by reflection from the output face. Graph (c) gives the azimuth dependences of the light intensity scattered into the ring for the subfigures (a) (open dots) and (b) (filled dots).

axis). The vector of spontaneous polarization is parallel to the domain walls. Our 4-mm-thick sample is cut in such a way that the polished input/output faces are also parallel to the domain walls (see Fig. 1). A remarkable feature of this sample is that it consists of two periodically poled parts separated by a central 2-mm single-domain area (near the boule axis). This is useful for comparison of the data, obtained for PPLN, with those related to the single-domain part: all the parameters of the sample, except its domain structure are the same.

An unexpanded Ar^+ laser beam (514 nm wavelength, single frequency, TEM_{00} mode, diameter of about 1.2 mm) is used to form two pump beams impinging symmetrically upon the sample (Fig. 1). The scattering patterns are recorded from a white screen, placed 20 cm behind the crystal, with the help of a digital camera. Additionally, the light intensity scattered at a certain solid angle was measured with a photodiode.

Figures 2a and 2b display two typical examples of the scattering patterns obtained under the same conditions in the single-domain and PPLN parts of the sample, respectively. The polar (optical) axis is perpendicular to the plane of incidence, and the pump beams are extraordinarily (e) polarized. The black spots mark the positions of pump beams shadowed at the output by small opaque disks to prevent saturation of the camera. The angle (in air) between the pump beams is $2\theta_p \simeq 15°$.

Two similar scattering patterns, a ring plus two lines passing through the pump spots, can be seen in both pictures, with nearly the same angular "thickness" of rings and lines about 0.9°, all e polarized. The light scattered within the same small angular aperture (positioned, e.g., at the points of the ring located symmetrically to the pump spots) differs in intensity by no more than 20% for the single-domain and PPLN areas.

The observed ring and lines correspond to two different parametric processes involving only extraordinary (pump and scattered) waves. According to the accepted classification [1], these processes are $A{:}ee \rightarrow ee$ and $B{:}ee \rightarrow ee$, respectively. The relevant wave vectors obey the phase-matching conditions

$$\mathbf{k}_{p1}^{e} \pm \mathbf{k}_{p2}^{e} = \mathbf{k}_{s1}^{e} \pm \mathbf{k}_{s2}^{e}, \qquad (1)$$

where the subscripts $p1, 2$ and $s1, 2$ mark two pump waves and two parametrically conjugate scattered waves, and the plus and minus signs refer to A (ring) and B (lines) processes, respectively.

We see that the considered parametric processes remain almost unaffected by the periodic alternation of the spontaneous polarization vector despite the fact that the mechanisms of the photorefractive nonlinearity of ferroelectrics are caused by their polarity [2,3]. This property is similar to the one predicted in [10] and confirmed in [11] for the efficiency of recorded gratings in crystals with dominating photovoltaic charge transport.

Theory predicts decreasing efficiency of grating recording in PPLN at spatial frequencies comparable or smaller than the inverse period of the domain structure [10]. This should result in decreasing brightness of the light ring near the pump spots. A close examination of Figs. 2a and 2b clearly shows this expected feature. The relevant quantitative data are shown in Fig. 2c. One sees that the light intensity does not depend on the azimuth scatter angle α in the single-domain part (open dots), whereas in the PPLN part this intensity decreases considerably for $\alpha < 45°$ (filled dots). The data for $|\alpha| < 15°$ (strongly affected by the stray light) are not shown in Fig. 2c.

By varying the pump beams' orientation and polarization we additionally observed ten known strong parametric processes in both single-domain and PPLN parts of the sample. These processes are $A, B{:}oo \to oo$, $A, B{:}oo \to oe$, $A, B{:}ee \to eo$, $A, B{:}eo \to eo$, $A{:}ee \to oo$, and $B{:}oo \to ee$, where the sign o refers to the ordinarily polarized waves [1]. They involve the tensorial nature of the photorefractive response in lithium niobate [1,3]; generally, the pump waves, as well as the scattered waves, differ in polarization from each other.

The presence of the periodic domain structure gives rise to a wealth of new parametric processes and, correspondingly, to new scattering patterns. For example, the following phase-matching condition, involving not only the grating vector of the light pattern $\mathbf{K} = \mathbf{k}^e_{p2} - \mathbf{k}^e_{p1}$ but also the vector of the periodic structure $\mathbf{G} = (2\pi/L)\mathbf{x}$, is fulfilled for a certain pump half-angle θ_p:

$$\mathbf{k}^e_{p1} - \mathbf{k}^o_{s1} - \mathbf{G} = \mathbf{k}^o_{s2} + \mathbf{G} - \mathbf{k}^e_{p2} = \mathbf{K} \qquad (2)$$

(see also Fig. 3a). The scattered waves are ordinary here, i.e., anisotropic diffraction [2,15] (with the change of polarization) is involved in this nonlinear process.

Figure 3b shows the relevant experimental scattering pattern. Two new symmetric ordinary light beams, indicated by the arrows, appear in the far field. The pump half-angle (in air) is here $\theta_p \simeq 17.4°$. The corresponding theoretical relation for θ_p found from Eq. (2) is $\sin\theta_p = 0.5(n\Delta n + L^{-1}\lambda n)^{1/2}$. With the birefringence of lithium niobate $\Delta n \simeq 0.1$, the refractive index $n \simeq 2.3$, and the period $L \simeq 7\ \mu$m we obtain from here $\theta_p \simeq 17.6°$. This agrees well with the above experimental value.

Furthermore we have found several parametric processes involving the **G** translation and waves of the same type of polarization (all ordinary or all extraordinary). Similar processes involving the double translation (2**G** instead of **G**) are detected as well. A distinctive feature of the mentioned processes is the necessity to adjust carefully the angle between two pump beams. The values of the pump angles, necessary for the appearance of parametric waves, are in good agreement with those calculated from the relevant phase-matching conditions.

Many other parametric processes involving the periodicity of the domain structure do not require any sharp adjustment of the pump angle. The corresponding scattering patterns can be observed in a wide range of θ_p. Let us consider, e.g., the parametric process defined by the phase-matching condition

$$\mathbf{k}^e_{p1} - \mathbf{k}^e_{s1} = \mathbf{k}^o_{s2} + \mathbf{G} - \mathbf{k}^e_{p2} \qquad (3)$$

(see also Fig. 4a). Figure 4b shows the expected dependence of the scattering angles θ_s on the pump half-angle θ_p (solid lines) calculated from Eq. (4) for $G = 2\pi/L \simeq 9 \times 10^3$ cm^3 and several experimental dots. To make sure that the periodic domain structure is really important, we have plotted (dashed lines) the dependence $\theta_s(\theta_p)$ for $\mathbf{G} = 0$. It is evident that the experimental dots are fitted much better by the solid lines.

Finally, we present a rather unusual pump-angle insensitive process defined by the phase-matching condition

$$\mathbf{k}^e_{p2} - \mathbf{k}^o_{p1} = \mathbf{k}^o_{s2} - \mathbf{k}^e_{p2} - \mathbf{G}\,. \qquad (4)$$

The left- and right-hand sides of this equation give the vector $\mathbf{K}_1$ of a grating recorded by two orthogonally

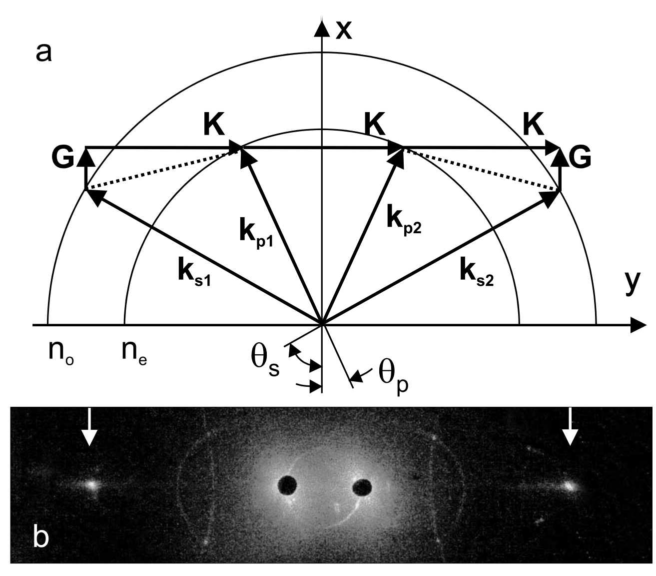

FIG. 3. Wave vector diagram (a) and far-field intensity distribution (b) for polarization-anisotropic scattering.

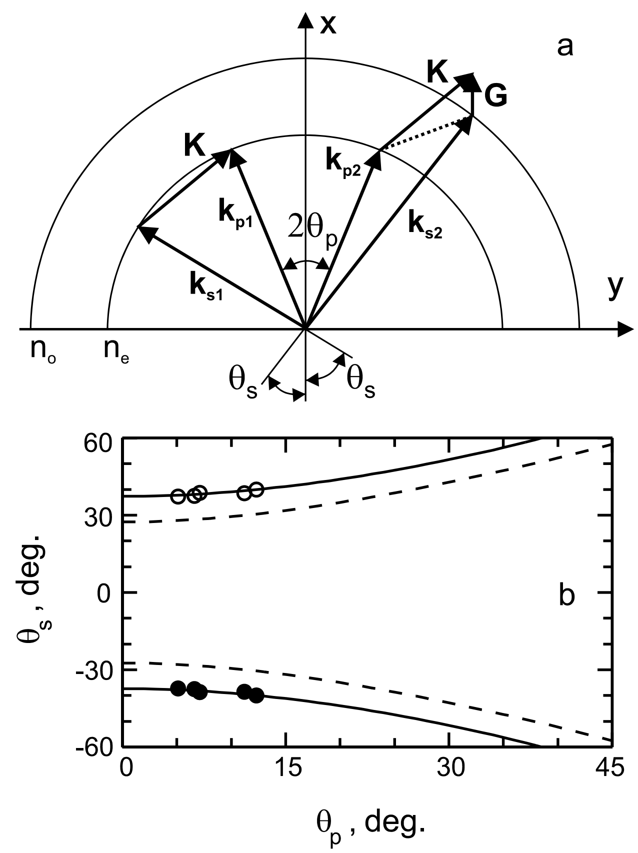

FIG. 4. Wave-vector diagram in the pump plane (a) and pump-angle dependence of the scattering angles (b) for the parametric process defined by Eq. (3).

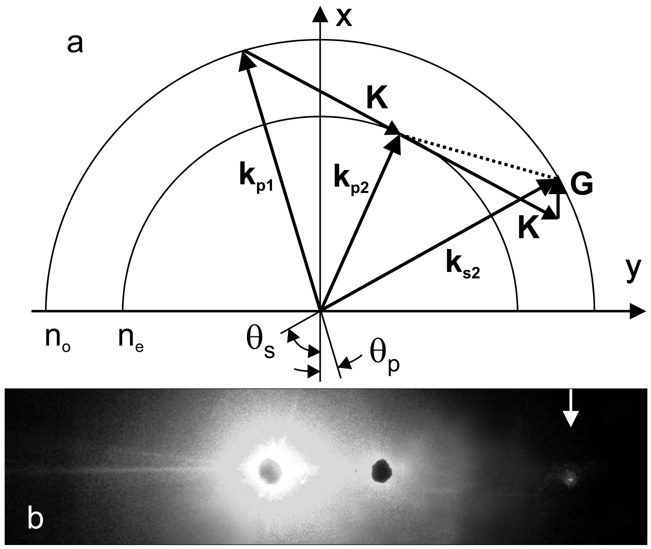

FIG. 5. Wave-vector diagram (a) and far-field intensity distribution (b) for the parametric scattering process defined by Eq. (4).

polarized pump waves and also supported by the pair of orthogonally polarized waves $p2, s2$ (see Fig. 5a). Such a recording occurs exclusively owing to the tensorial nature of the photovoltaic charge transport [1,3]. Figure 5b shows an experimental realization of this process. The arrow indicates the scattered ordinary wave $s2$.

Physically, all parametric processes involving the periodicity of the domain structure are due to the fact that an index grating recorded by a pair of light beams (possibly different in polarization) includes not only the fundamental $\mathbf{K}$ component but also the harmonics $\mathbf{K} + \mathbf{G}, \mathbf{K} + 2\mathbf{G}$, etc. At the same time, the periodic structure is not accompanied, by itself, by changes of the refractive index.

In summary, a large variety of pronounced frequency-degenerate parametric scattering processes is detected in bulk periodically poled lithium niobate codoped with Y and Fe. A part of these processes is known for single-domain crystals, the other, previously unknown, processes involve both the conventional grating recording and the periodicity of the domain structure. Even a richer variety of photorefractive parametric processes is expected in recently engineered 2D nonlinear photonic crystals [13]. The novel type of frequency conversion reported therein is similar in phase matching to new frequency-degenerate processes detected in PPLN. The results of our study show that PPLN:Y:Fe is a new promising nonlinear material that allows one to combine the strong suppression of large-scale distortions of refractive index (no optical damage) with numerous strong processes of spatial amplification (including polarization sensitive processes) owing to the photovoltaic charge transport.

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