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Mechanism of self-organized light-induced scattering in periodically poled lithium niobate

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It is shown that the photorefractive grating produced by a pair of plane waves in periodically poled lithium niobate includes an additional set of spatial harmonics related to the periodic domain structure. This results in new schemes for photorefractive wave coupling. Using the modified phase matching conditions and the concept of optical oscillation we accurately describe the position of the diffraction peaks and explain the main characteristics of self-organized photoinduced scattering reported recently. © *1996 American Institute of Physics.* [S0003-6951(96)01636-1]

Recently, an unusual and exciting photorefractive phenomenon was detected in periodically poled lithium niobate (PPLN).[1] Its main features were as follows. Two clear symmetric diffraction orders appeared in the far field during propagation of a laser beam ($\lambda=532$ nm and diameter $d=40$ μm) along the x axis, perpendicular to the periodic layers (see Fig. 1). This effect showed a clearly defined threshold in the pump power, $P_{th}=10$ mW at $T=20$ °C; the above-threshold part of the power was transformed into the diffraction orders. The external angles, $\theta^{(1)}$ and $\theta^{(2)}$, were dependent on the domain pitch, x_0. For samples A and B with pitches $x_0^A=6.8$ and $x_0^B=9$ μm these angles were $\theta_A^{(1)}=12°$, $\theta_A^{(2)}=26°$ and $\theta_B^{(1)}=9.5°$, $\theta_B^{(2)}=20°$. The length l and the thickness a of both samples were ~4 and 0.2 mm, respectively. All waves were extraordinarily polarized. Below threshold only a broad spread of scattering angles was observed which disappeared with decreasing pump power. In the regions of samples A and B where no domain inversion was present the described effect was absent.

In this letter we propose a mechanism for the above effect. This includes a quantitative explanation for the scattering angles and an interpretation of the other observed behavior based on the concept of optical generation and the photovoltaic transport model.[2,3]

The photorefractive nonlinearity is usually characterized[4] by the relation $E_K=mf_K$ between the complex amplitude E_K of the space-charge field, induced by a pair of plane light waves, and the contrast m of the corresponding interference pattern with a period $\Lambda=2\pi/K$. The term f_K (generally complex) depends on the mechanism of light-induced charge transport. The space-charge field modulates the refraction index due to the linear electro-optic effect. For the case of extraordinary (e) waves and the interference fringes approximately perpendicular to the optical z-axis the amplitude of the spatial oscillations of the refraction index is $n_K=\pi m n_e^3 f_K r_{33}/\lambda$, where n_e is the extraordinary refractive index, and r_{33} the relevant electro-optic coefficient.

In the reported experiment, the only transport mechanism capable of producing appreciable nonlinear optical effects is the photovoltaic effect which consists in the generation of photocurrents directed along the polar c axis.[2,3] In single domain $LiNbO_3$crystals the quantity f_K is comparable to the characteristic photovoltaic field, $E_{pv}=(10^1-10^2)$ kV/cm.

Let us now consider the distinctive features of the photovoltaic transport in PPLN. Under quasiuniform illumination of the sample by a light beam of a diameter $d\gg x_0$, see, e.g., Fig. 1(a), the average electric field inside the illuminated region is considerably less than E_{pv} because of very frequent spatial alternation of (+) and (−) charges along the outer boundary of the beam. This leads to reduced photorefractive effect with regard to a single domain crystal[1] and also permits the use of the closed circuit approximation for calculations of the photorefractive nonlinear response. If the light intensity has a spatially oscillating part proportional to $m\cos Kz$ and $Kx_0\gg1$, the space-charge field E inside a given domain practically coincides with the field induced in a single domain crystal of the same orientation [see Fig. 2(a)]. Consequently, the gratings in neighboring (±) domains are shifted with respect to each other by 180°. Near the domain boundaries the component E_z, responsible for the change of the refractive index, becomes very small.

Now we take into account the fact that the inversion of the polar c axis alters not only the sign of the photovoltaic current but also the sign of the electro-optic coefficient r_{33}.

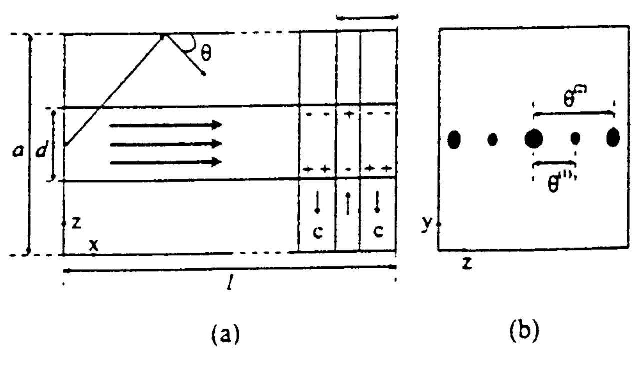

FIG. 1. Geometrical schemes. (a) Characteristics of the sample: l is the length, a the thickness, d the diameter of the pump beam, x_0 the domain pitch, c the polar axis, θ the scattering angle. (b) Scattering pattern, the central spot corresponding to the pump beam.

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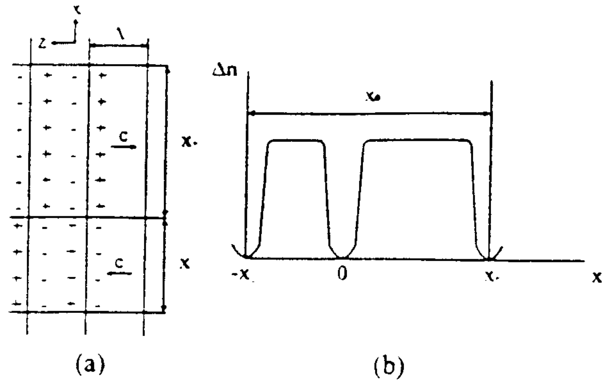

FIG. 2. (a) Scheme of charge separation for a light interference pattern. Intensity maxima are shown by vertical lines. (b) Schematic dependence of $\Delta n(x)$.

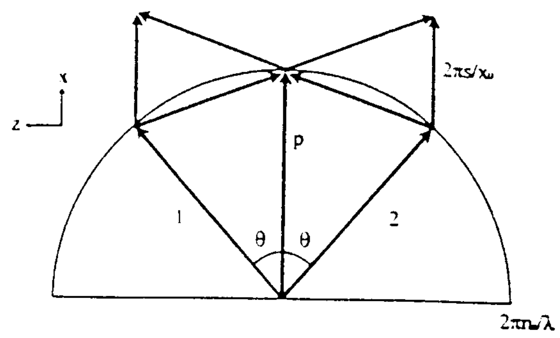

FIG. 3. Wave vector diagram for the phase-matching condition (3).

This means that except for the narrow transition regions between domains the change of refractive index $\Delta n(x,z)$ in PPLN is the same as for a single domain crystal. In the transition regions the periodic function $\Delta n(x)$ has deep dips [Fig. 2(b)]. Therefore, the following expansion for $\Delta n(x,z)$ is valid for PPLN

$$\Delta n = e^{iKz} \sum_{s=-\infty}^{\infty} n_s(K) e^{is\kappa x} + c.c., \tag{1}$$

where $\kappa = 2\pi/x_0$. The effect of periodicity is described by the terms with $|s| \geq 1$. In the limit $\Lambda \ll x_0$ the spatial harmonics with $|s| \geq 1$ are small in comparison with the main component $n_0 \simeq n_K$. With increasing Λ the main component, n_0, decreases and the higher harmonics become more important. In the experiment, the ratio $r = \Lambda/x_0$ is 0.3–0.4. The photovoltaic model gives in the steady state: $n_0 \simeq n_K$

$$\frac{n_s}{n_0} \simeq -\frac{2r \cos \varphi_n}{\pi(1+s^2r^2)} e^{-i\varphi_n}, \tag{2}$$

and $\varphi_n = \pi n(1+\zeta)/2$, where $\zeta = (x_+ - x_-)/x_0$, and $x_\pm$ are the sizes of the $(\pm)$ domains, see Fig. 2. In the experiment the asymmetry parameter ζ is about 0.4, $n_1/n_0 \simeq 0.1-0.2$, and $n_1/n_2 \simeq 2-3$.

The presence of new spatial frequencies in the distribution $\Delta n(x,z)$ gives additional prospects for nonlinear wave coupling. Suppose the wave vectors of a pair of extraordinary waves, $\mathbf{k}_1$ and $\mathbf{k}_2$, meet the following phase-matching condition

$$\mathbf{k}_1 + \mathbf{k}_2 + s\boldsymbol{\kappa} = 2\mathbf{k}_p, \tag{3}$$

where $\mathbf{k}_p$ is the pump wave vector. Then the grating vector $\mathbf{k}_p - \mathbf{k}_1$ of the main grating induced by the wave pair p, 1 coincides with the sth harmonic produced by another pair 2, p, see Fig. 3. The same is valid with respect to the main component of the grating formed by the pair 2, p, and the sth harmonic from the pair p, 1. In such a way, Eq. (3) means that two different wave pairs can contribute to the formation of the same grating. This condition also permits additional Bragg-diffraction processes. As seen from Fig. 3, the pump wave p can diffract into the wave 1 (2) from the grating produced by the pair p, 2 (1, p).

This nonlinear coupling of the waves 1 and 2 through a common grating is analogous to the parametric coupling in single domain photorefractive crystals involving waves of different polarizations. Numerous experimental and theoretical studies show, see, e.g., Refs. 5 and 6, that such a coupling results in an enhancement of the gain coefficients and, as a consequence, in a variety of bright rings, lines, and dots of scattered light.

Equation (3) defines the value of the angle θ_s between the pump beam and the side beams 1 and 2. Outside the sample we have

$$\sin \theta_s \sqrt{|s| n_e \lambda / x_0 - (|s|\lambda/2x_0)^2}. \tag{4}$$

Taking $n_e = 2.23$ and $|s| = 1$ we find that θ_1 approximately equals 25° and 21° for the samples A and B, respectively. This is very close to the experimental values of the angle $\theta^{(2)}$ for the outermost diffracted order in Fig. 1(b). Furthermore, it is easy to verify that the experimental values $\theta^{(1)}$ for the first diffraction order coincide with the calculated difference $\theta_2 - \theta_1$ (10.5° and 9° for samples A and B, respectively) to a good accuracy. In such a way, the detected diffraction orders are identified with the parametric processes of the first and second order in $|s|$ defined by the phase-matching condition (3).

Now we offer a qualitative explanation for the main experimental facts on the basis of the above phenomenological analysis. The second diffraction order, $\theta^{(2)}$, results from a generation process involving parametrically coupled waves with $|s| = 1$. The enhanced gain coefficient for the parametric waves 1 and 2 compared to that for other waves results in the lowest generation threshold.

We see two main possibilities for optical generation. The first one corresponds to an equivalent ring oscillator.[7,8] The parametric seed waves 1 and 2 are amplified during propagation to the rear face of the sample at the expense of the pump (see Fig. 1). Then, after reflection from this face, total internal reflections at the side faces, and, finally, reflection from the front surface, the waves 1 and 2 return to the input, thus providing the positive feedback. At the generation threshold the losses for one roundtrip must be compensated by the amplification process. To estimate the passive losses we assume that only $\sim 10^{-2}$ of the initial power returns to the input. The amplification factor may be estimated as $\exp(\Gamma l_{int})$, where $\Gamma \simeq \pi n_e^3 r_{33} E_{pv}/\lambda$ is the gain coefficient[3,4] and $l_{int} = ld/a$ is the interaction length. The latter is less than the length of the sample, l, for geometrical reasons, see Fig. 1. With the accepted values for n_e, r_{33}, and λ, the generation condition is $E_{pv} > 30$ kV/cm, which is realistic for $LiNbO_3$.

Note that the estimation for Γ implies a small (of the order of the reciprocal dielectric relaxation time) frequency detuning between the waves 1, 2 and the pump. Similar detunings (originated from different temporal fluctuations) are typical of many generation schemes.[7-9]

The second possibility corresponds to an oscillation with two counterpropagating pump beams.[7,10] An additional backward pump wave appears owing to reflection of the laser beam from the output face of the sample. The contrast of the pump interference pattern, m_p, here is about (0.5–0.6). In this case, the condition for optical oscillation via formation of a transmission grating has the form $m_p \Gamma l_{int} \gtrsim \pi$. It also may be fulfilled in experiment.

The power threshold observed in the experiment admits the following explanation: It has been pointed out[3,11-13] that in $LiNbO_3$ and $LiTaO_3$ crystals the photovoltaic field shows an increase with respect to light intensity at relatively high intensity values, $\gtrsim 10^3$ W cm^{-2}. The experimental threshold intensity falls just in this region and for these condition the values of E_{pv} may be as high as 10^2 kV/cm. Note that the waves at $\pm\theta_2$ are below the threshold of generation because of the smallness of the ratio n_2/n_1.

Our model also provides an explanation for the occurrence of the first diffraction order, $\theta^{(1)}$. Let us consider one of the generated beams, $\theta^{(2)}$, as a new pump. This beam, in contrast to the primary one, fills the whole sample. For this reason, we should put $l_{int}=l$ for the generation waves. In addition, the experiment indicates that this new pump can contain up to half of the threshold power, P_{th}. In such a situation, some decrease of the photovoltaic field through pump depletion can be compensated by the increased interaction length, by a factor $a/d \simeq 5$. We now turn to Fig. 3. A generation process initiated by the wave 1 and corresponding to the phase-matching condition with $|s|=1$ is not possible because one of the oscillation waves would coincide with the main pump wave p. Therefore, generation should occur for $|s|=2$. Here one of the generated waves just corresponds to the first diffraction order, $\theta^{(1)}$. Its conjugate generated wave, propagating inside of the crystal at the angle $\theta_1+\theta_2$, cann leave the crystal due to essentially total internal reflectic from the rear face.

In conclusion, the domain structure of PPLN results in modification of the spatial frequency spectrum of the photc refractive grating and, as a consequence, in new schemes fo four-wave mixing. The modified phase-matching conditior correctly predicts the directions of unusual diffractior maxima discovered in PPLN. The main features of the self-organized light-induced scattering described in Ref. 1 are explained on the basis of the concept of optical oscillation, using the published data on photovoltaic transport in $LiNbO_3$ crystals.

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