Photorefractive Response of Bulk Periodically Poled $LiNbO_3$:Y:Fe at High and Low Spatial Frequencies

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Polydomain periodically poled samples of iron-doped lithium niobate allow for phase grating recording with the diffraction efficiency close to that ensured by a single-domain sample with identical thickness. At the same time the optical damage induced by a finite transverse size light beam is considerably reduced in the periodically poled sample as compared with single-domain material.

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For the study and various practical applications of ferroelectric nonlinear optical crystals either the single-domain samples or the samples with specially designed perfectly periodic domain structure are used (see, e.g., [1,2]). We show in this Letter that the phase grating recording in multidomain ferroelectric photorefractive crystals with photovoltaic charge transport may be as effective as in the single-domain material. At the same time the multidomain crystals ensure the inhibited nonlinear response at low spatial frequencies, i.e., suppress the undesirable optical damage.

The reversible light-induced deterioration of optical quality known as optical damage was first discovered in bulk crystals of lithium niobate [3]. When the sample was illuminated with a green or blue laser beam the strong anisotropic nonlinear lens developed gradually, resulting in dramatic wave front distortion of the transmitted beam. This obvious drawback of $LiNbO_3$ for possible applications such as frequency conversion, electro-optic modulation, and parametric oscillation appeared to be an advantage for holographic recording [4]. Even a relatively small refractive index change of the order of $\Delta n \simeq 5 \times 10^{-5}$ allowed for the recording of a volume phase holographic grating with nearly 100% diffraction efficiency in a 5-mm-thick sample.

The first photorefractive optical memory was created soon thereafter [5]. The great expectations about the possibility to build up high-density holographic optical memory with photorefractive crystals are still alive [6,7]. Unfortunately, optical damage leads to degradation of the diffracted beam quality. Any spatial variation of the recording wave intensity, for example, Gaussian transverse profile of the reference wave, results in the large-scale variations of the refractive index, leading to the distortion of the wave fronts. One task of the research developing optical memories is to find a compromise between the diffraction efficiency of the recorded hologram and acceptable quality of the reconstructed wave front. This paper describes, to our knowledge, the first experimental evidence of high-efficient holographic recording in multidomain $LiNbO_3$:Y:Fe with periodically reversed spontaneous polarization and demonstrates a considerable reduction of the large-scale space-charge field in these samples.

The samples with the periodic domain structure (PDS) are grown by the Czochralski technique with close congruently melting composition $Li/Nb = 0.942$ in usual atmosphere (i.e., with oxygen) and no after-growth treatment. The structure of inverted ferroelectric domains is formed by modulation of the yttrium concentation during the crystal growth [8,9]. For the first time the PDS samples are deliberately doped with Fe (about 0.006 wt %) to increase the photorefractive sensitivity. The domain walls are nearly parallel to the spontaneous polarization P_s and the PDS period L is about 7 μm (Fig. 1). As distinct from the peripheral area where the domain structure is well developed the central part of the boule, near the growth axis, is a perfect single domain. This gave us a reliable reference in the same sample: the lateral displacement of a few mm is sufficient to move from the PDS region to the single-domain part of the sample. The thickness of the test sample with

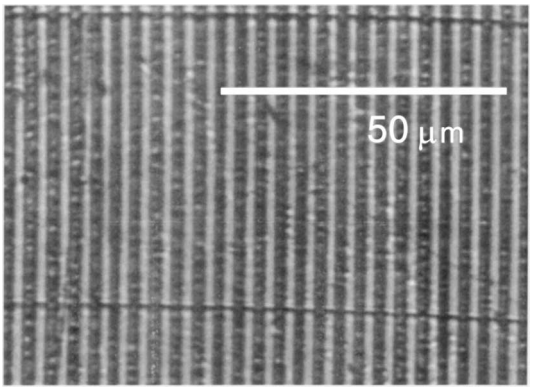

FIG. 1. Domain pattern in periodically poled $LiNbO_3$ as seen with the polarization microscope. Spontaneous polarization is vertical. Horizontal black straight lines are scratches from polishing.

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the faces parallel to the spontaneous polarization axis (and domain walls) is 1 mm.

We first compare the divergence of the Ar^+-laser beam transmitted through the multidomain and single-domain parts of the sample. The unexpanded beam with total power of about 100 mW is directed nearly along the normal to the input face; the light polarization is chosen to excite the extraordinary wave of the crystal.

Figure 2 shows the far-field patterns recorded with a CCD camera and the intensity distribution along the P_s direction. The initial divergence of the laser beam (without the $LiNbO_3$ sample) has to be compared with the divergence of the beam transmitted through the PDS part and through the single-domain part of the sample. An obvious inhibition of the optical damage can be seen for the PDS part of the sample. This proves that the photorefractive response at low spatial frequencies is strongly suppressed in PDS samples.

There have been several reports that periodically poled lithium niobate is qualitatively more resistant to optical damage than homogeneously poled crystals are [10–12]. As distinct from these studies where pure $LiNbO_3$ crystals and crystals with Mg doping were used to reduce optical damage we report on the reduction of much stronger optical damage for deliberately Fe-doped material.

In the second experiment we compare the dynamics of the diffraction efficiency of the 3D photorefractive grating for these two regions of the sample. Two unexpanded coherent light beams of nearly equal intensities ($\simeq 2$ W/cm^2) impinge upon the sample in the plane parallel to the axis of spontaneous polarization. Both beams are polarized identically with the electric field vector in the plane of incidence. The full angle in air 2θ between the recording waves is 60°, i.e., the spatial frequency K of the fringe pattern is $K = 4\pi \sin\theta/\lambda \simeq 12\,500$ mm^{-1}. Every 20 s one recording beam is stopped for a fraction of a second and the intensity of the diffracted beam is measured. The diffraction efficiency η is evaluated as the ratio of the intensity of diffracted from the grating beam to the total intensity of the transmitted readout beam and diffracted beam.

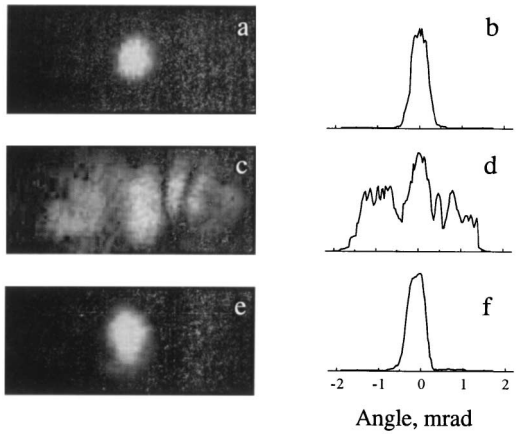

FIG. 2. Steady-state far-field intensity distribution for single light beam incident to the sample (a),(b) transmitted through the single-domain sample (c),(d) and transmitted through the sample with periodic domain structure of the same thickness (e),(f).

The temporal variation of the diffraction efficiency is plotted in Fig. 3 for the PDS part (squares) and the single-domain part (circles) of the sample. One can see that the dynamics of the recording are quite similar and the difference in absolute steady-state values of the diffraction efficiency is less than 20%. This proves that photorefractive response at high spatial frequencies remains nearly the same in the PDS sample as in the single-domain sample.

These experiments confirm the possibility of optical damage reduction in multidomain photorefractive crystals with photovoltaic charge transport, predicted by Taya *et al.* [12] and efficient grating recording predicted by Sturman *et al.* [13]. We recall the main line of explanation of this unusual effect before the description of the additional experiments.

The optical nonlinearity of photorefractive crystals is due to the spatial redistribution of the photoexcited charge carriers, formation of the space-charge field E_{sc}, and modification of the high-frequency dielectric constant ϵ via the linear electro-optic effect [14]. The light-induced refractive index change is given by

$$\Delta n = -(1/2)n^3 r_{\text{eff}} E_{sc}\,, \tag{1}$$

where n is the refractive index and r_{eff} is the effective electro-optic constant. The space-charge field E_{sc} may be a consequence of charge redistribution in external electric field or may appear because of photoexcited carrier diffusion. The ultimate value of the space-charge field E_{sc} is limited either by the external electric field E_0 or by the diffusion field $E_D = (2\pi/\Lambda)(k_B T/e)$ [where k_B is the Boltzmann constant, T is the temperature, e is the electron charge, and Λ is the characteristic scale of the spatial intensity distribution, e.g., $\Lambda = \lambda/(2\sin\theta)$ is the fringe spacing for grating recording].

In doped crystals with low conductivity the bulk photovoltaic effect [15,16] may also become an efficient source

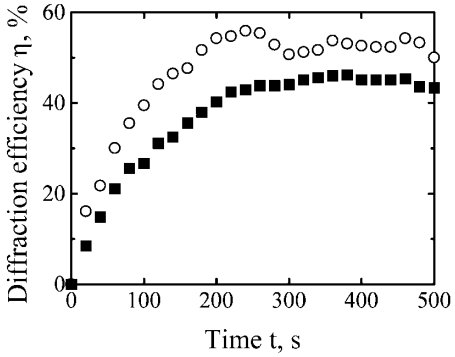

FIG. 3. Temporal variation of the diffraction efficiency for single-domain sample (circles) and multidomain sample (squares).

of the charge separation and development of large space-charge fields up to 10^4–10^5 V/cm. The short-circuited illuminated sample generates the steady-state electric current proportional in the simplest scalar case to the light intensity I, $j = \beta_{\text{eff}} I$, where β_{eff} is the effective photovoltaic constant. The amplitude of the space-charge field which is due to photovoltaic charge separation is equal to the effective photovoltaic field $E_{\text{pv}} = j/\sigma$, where σ is the crystal conductivity [16]. The refractive index change for photovoltaic photorefractive material is

$$\Delta n = -(1/2) n^3 r_{\text{eff}} \beta_{\text{eff}} I/\sigma . \quad (2)$$

Let us now analyze the effect of the domain inversion on grating recording. If we consider the diffusion-mediated or drift-mediated charge separation the sign of the space-charge field is independent of the orientation of the spontaneous polarization axis P_s and remains the same in adjacent domains. At the same time the effective electro-optic constant in Eq. (1) does depend on the sign of P_s. This means that the contrast of the phase grating recorded in the sample is reversed in every new domain with an inverted P_s direction. In other words there is a π shift in phase of the recorded phase grating every time when two light waves enter a new domain. The total recorded structure looks like a sequence of thin stripes (domains) with complimentary phase gratings: maxima changed to minima and vice versa in adjacent domains. It is quite clear that the partial contributions to the diffracted wave from the gratings in two adjacent domains will be also π-out-of-phase and integration over total sample thickness will give a vanishing to zero intensity for the diffracted wave.

The result is quite different in the case of photovoltaic charge separation. Here the direction of the photovoltaic current depends on the orientation of the ferroelectric axis. If the axis of spontaneous polarization is inverted both the effective electro-optic constant and the effective photovoltaic constant change their signs so that the sign of Δn remains unchanged [see Eq. (2)]. This means that there is no phase shift between the photorefractive gratings recorded in domains with the opposite direction of ferroelectric axis. A question remains: Is it possible to find the conditions where the amplitude of photorefractive grating is also not too strongly affected by the domain inversion?

The theory gives the analytical solution for the space-charge field in photovoltaic crystals with PDS [13]. For light fringes with spacing Λ much smaller than the domain period L the amplitude of the space-charge field in the PDS sample approaches that in a perfect single-domain sample. If, on the contrary, the spatial variation of the light intensity becomes much larger than the typical domain period the corresponding space-charge field vanishes.

The explanation of this behavior is quite simple: If the period of the domain structure is much larger than the fringe spacing Λ the space-charge field inside each domain approaches the field that would be induced in a single-domain crystal with the same orientation. The influence of the charges in other domains on the space-charge field in consideration is relatively small. In adjacent antiparallel domains, the space-charge fields have the same strength but opposite direction. A strong variation of the space-charge field occurs near the domain wall where the field passes a zero value.

When the fringe spacing Λ becomes larger the effect of the transitional layers becomes important and the space-charge field starts to decrease. For $\Lambda \gg L$ (low spatial frequency K) the space-charge field becomes strongly reduced because of a quick alternation of positive and negative photoinduced charges along each fringe. The calculated space-charge field component along the grating vector $\mathbf{K}$ is [13]

$$E_K = -E_{\text{pv}}[1 - (2G/\pi K)\tanh(\pi K/2G)], \quad (3)$$

where $G = 2\pi/L$ is the spatial frequency of the domain grating. The normalized space-charge field E_K/E_{pv} is plotted in Fig. 4 as a function of the normalized spatial frequency (K/G). For small fringe spacing (large K/G) the space-charge field E_K approaches its ultimate value E_{pv} that can be reached in a single-domain crystal while for large-scale intensity variation ($K \ll G$) E_K is decreasing as $E_K \propto (K/G)^2$ [12,13].

The vertical dashed lines in Fig. 4 show two particular (K/G) values for our experiment. For the recording of a grating with $\Lambda = 0.5$ μm in the PDS part we have $(K/G) = (L/\Lambda) = 14$ and E_{sc} is only 2% smaller than E_{pv}. With transverse dimensions of the laser beam $d \simeq 1.5$ mm $(K/G) \simeq 0.01$), calculation predicts $\simeq 10^4$ times reduction of the space-charge field as compared with E_{pv}, typical for a single-domain crystal. By taking $E_{\text{pv}} \simeq 2 \times 10^4$ V/cm for $LiNbO_3$:0.006 wt % Fe [17] we get the large-scale space-charge field of the order of 1 V/cm; such a field cannot provide any detectable effect on the transmitted beam wave front.

In the experiment we have not seen the difference in the divergence of the light beam transmitted through the multi-domain part of the sample and divergence of the beam incident to the sample, within our experimental accuracy (see

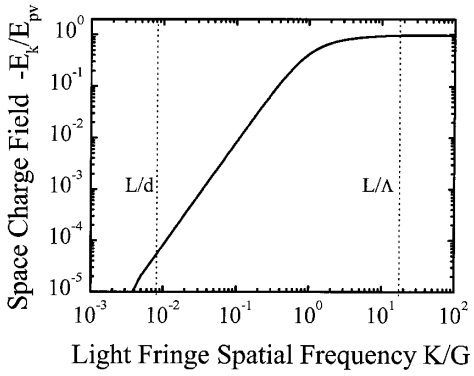

FIG. 4. Spatial frequency dependence of the space-charge field normalized to the ultimate value in the single-domain sample.

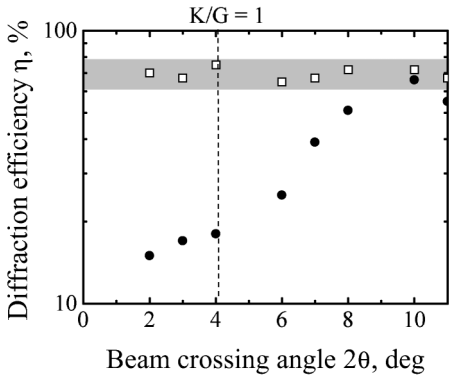

FIG. 5. Angular dependence of the steady-state diffraction efficiency for single-domain sample (squares) and multidomain sample (circles).

Fig. 2). At the same time a small but well detectable reduction of the diffraction efficiency ($\Delta\eta \simeq 10\%$) for the PDS sample as compared to the single-domain sample is clearly seen from Fig. 3. This difference in η can be attributed in part to the additional contribution of the diffusion-mediated recording [18] in a single-domain crystal (suppressed in the multidomain sample). With fringe spacing $\Lambda \simeq 0.5\ \mu$m the diffusion field $E_D \simeq 3200$ V/cm is not negligible when compared to photovoltaic field $E_{\text{pv}} \simeq 2 \times 10^4$ V/cm.

Finally we compare the angular dependences of the diffraction efficiency for single-domain and multidomain samples for small crossing angles θ (Fig. 5). For single-domain crystal the efficiency is nearly constant within the error range shown by the horizontal stripe while for the multidomain part it becomes smaller with the decreasing angle. Qualitatively it fits our expectations: For $2\theta = 4°$ the light fringes spatial frequency K is equal to the spatial frequency of the domain structure G. According to Eq. (3) the space-charge field for $K = G$ becomes $E_{\text{sc}} \simeq 0.4E_{\text{pv}}$ and the estimated diffraction efficiency should be $\eta \simeq (15\%–18\%)$. The spread of experimental data for η is too high to check the theoretical dependence given by Eq. (3), but qualitatively the tendency to smaller η for the decreasing beam crossing angle is quite obvious.

The inhibition of optical damage demonstrated in this Letter is also of importance for all applications of periodically poled $LiNbO_3$:Y:Fe for nonlinear wave mixing including phase conjugation, amplification of image bearing beams, and coherent self-oscillation.

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