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Photorefractive properties of bulk periodically poled $LiNbO_3$:Y:Fe

S. Odoulov [a,*], T. Tarabrova [a], A. Shumelyuk [a], I.I. Naumova [b], T.O. Chaplina [b]

[a] *Institute of Physics, National Academy of Sciences, Kiev 03 650, Ukraine*
[b] *Moscow State University, Moscow, 119899 Russia*

Abstract

We show with specially grown iron-doped lithium niobate crystals that multi-domain periodically poled samples of iron-doped lithium niobate (PPLN) 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 to the single-domain material. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

If the dynamic grating recording is performed in a ferroelectric photorefractive crystal the standard requirement is to use either a perfectly single-domain sample or, at least, the sample with the amount of antiparallel domains as small as possible. This restriction is fully justified for the grating recording in crystals with the diffusion-mediated charge transport or drift in an external electric field: The space charge field that appears in the sample is independent of the polar axis orientation while the effective electrooptic constant does depend on spontaneous polarization direction. This results in π-shift of the recorded grating in every new domain with antiparallel spontaneous polarization and therefore in the reduction of the integral diffraction efficiency of the grating.

We report in this paper on the recording of the efficient volume phase gratings in the bulk iron-doped lithium niobate crystals with the periodic domain structure (periodically poled lithium niobate, PPLN). Such a recording becomes possible in this material because the dominant charge transport process is here the bulk photovoltaic effect [1,2]. It is shown also that the iron-doped PPLN are much more optical-damage-resistant as compared to homogeneously poled crystals with the same impurity content. PPLN attracts attention of the researches mainly as a promising material for such quasi-phase-matched nonlinear interactions as frequency harmonics generation and parametric oscillation [3,4]. Our results suggest that the doped PPLN with bulk photovoltaic effect can be efficiently used also for frequency degenerate four-wave nonlinear interactions.

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[*] Corresponding author. Tel.: +380-44-265-2359; fax: +380-44-265-2359.
E-mail address: odoulov@iop.kiev.ua (S. Odoulov).

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2. Crystal synthesis and characterization

Periodically poled $LiNbO_3$:Y:Fe single crystals are grown along the $\langle 2\bar{1}\bar{1}0\rangle$ crystallographic direction (*X*-axis) by the Czochralski technique from a close congruently melting composition Li/Nb = 0.942. The crystal is pulled at a rate 5 mm/h and a rotation rate 12 rpm, resulting in a 7 m domain structure period. After growth termination the crystal is abruptly separated from the melt and then cooled down at a rate of about 60°C/h. In the Czochralski-grown doped $LiNbO_3$:Y single crystals, a regular domain structure is connected with the rotation-induced growth striations. Periodic temperature oscillations on the crystal–melt interface that arise during the crystal growth lead to the modulations of the instantaneous growth rate [5]. Due to the growth rate dependence of the effective distribution coefficient $k_{\mathrm{eff}} \neq 1$, this results in the modulation of the impurity concentration along the growth direction. The rotational striations are the source of periodically reversed ferroelectric domains with a period (L) that depends on the pulling rate and rotational rate.

Thus, the yttrium impurity leads to the development of the periodic domain structure in Czochralski-grown $LiNbO_3$ and, on the other hand, retains the crystal transparent within the 0.35–4.5 μm wavelength range [6,7]. The iron doping is used to increase the photorefractive sensitivity of the material. The impurity content is measured using wave dispersive spectrum X-ray microanalysis by the scanning electron microscope (JSM-840). Camebax SX-50 is used for impurity concentration measurements, the oxides are used as the standards. The following impurity concentration is measured with the X-ray microanalysis for the studied sample κ–241:Fe–(0.006 ± 0.001) wt%, Y – 0.736 wt%.

It is known that $LiNbO_3$ crystal grown along *X*-axis consists of two basic domains of opposite sign [8]. The boundary between positive and negative basic domains is perpendicular to *Z*-axis. With the technique described above, aiming to get PPLN material both main domains are transformed into a periodic domain structures, still with the single domain area separating the two PPLN area.

The ferroelectric domain structure in the Y:$LiNbO_3$ crystal on the $\{01\bar{1}0\}$ surface was revealed with the selective chemical etching (1:2 v/v HF and HNO_3 mixture in a Pt-crucible during 20 min at boiling temperature). After etching the samples were observed using a metallographic microscope to evaluate the domain spacing and to find the position of the single-domain area. Finally, the sample was cut ($1 \times 4 \times 15$ mm^3 along x, y and z directions, respectively) and optically finished, with the input and output faces parallel to the domain walls and spontaneous polarization. In the central part of the long polished face the sample has a single domain area with a cross-section about 3 mm. This part of the sample was used as a reference to compare the photorefractive properties of PPLN:Fe with those of single domain crystal of the same composition and same impurity content.

3. Experiment

We first compare the divergence of the Ar^+-laser beam transmitted through the periodically-poled and single-domain parts of the sample. The beam with total power 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. The gaussian beam waist at the input face of the sample (full width between 1/e^2 points) is ≈1.5 mm. The far field intensity distribution along the spontaneous polarization direction is shown in Fig. 1 for (a) the incident laser beam (with the crystal removed), for the laser beam transmitted through the single domain part of the sample (b) and for the laser beam transmitted through the PPLN (c). An obvious inhibition of the pronounced optical damage [9] can be seen for the periodically poled part of the sample.

Next, the photorefractive gratings are recorded with two unexpanded extraordinarily polarized coherent light beams that impinge upon the sample at an angle 60° in air forming the fringes with the spacing $\Lambda \approx 0.5$ μm. Two beams have nearly the same intensities to avoid the effect of the fringe bending typical for the crystal with the local

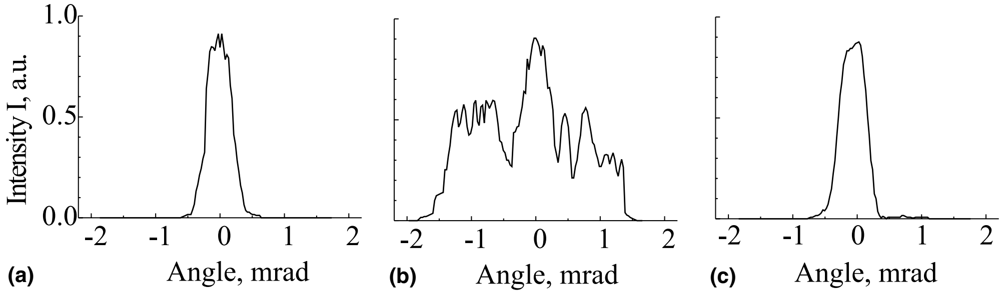

Fig. 1. Far-field intensity distribution of the laser beam incident upon the sample (a), transmitted through the single domain part of the sample (b) and transmitted through the periodically poled part of the sample (c).

nonlinear response [10]. Because of relatively small photoconductivity the recording is slow; it takes more than 10 min to reach the steady-state diffraction efficiency. The ultimate diffraction efficiency (evaluated as the ratio of the intensity of the diffracted beam to the total intensity of two output beams) reaches approximately 80% for the single domain part of a sample and it is about 60–65% for PPLN. Thus we conclude that the photorefractive response of PPLN:Fe at high spatial frequencies is close to that of the sample with uniform poling. This differs strongly from the effect at low spatial frequencies where the photorefractive response is drastically reduced.

4. Discussion

The results of our measurements are in qualitative agreement with the theories [11,12] describing the nonlinear response of the periodically poled ferroelectric photorefractive crystals. The calculated component of the space-charge field is $E_K = -E_{pv}[1(2\Lambda/\pi L)\ \tanh(\pi L/2\Lambda)]$.

For small fringe spacing the space charge field E_K approaches its ultimate value that can be reached in single-domain crystal, equal to the effective photovoltaic field $E_{pv,}$ while for large scale intensity variation $(\Lambda \gg L) E_K$ is decreasing as $E_K \propto (\Lambda/L)^2$.

The domain width in the sample is $L \approx 7$ μm. With the angle between two writing beams $2\theta = 60°$ the fringe spacing is $\Lambda \approx 0.5$ μm so that $(\Lambda/L) \approx 0.07$. The calculated space charge field for this value of (Λ/L) is exactly the same as for grating recorded in a single-domain crystal [13].

The transverse dimension of the laser beams used for grating recording is $d \approx 1.5$ mm, i.e., $(d/L) \approx 200$. For such a value of (d/L) the calculation predicts 5×10^4 times reduction of the space charge field as compared with E_{pv} typical for single domain crystal. With $E_{pv} \approx 2 \times 10^4$ V/cm for $LiNbO_3$:0.005 wt%. Fe [14] we get the large-scale space charge field below 1 V/cm; such a field is too small to produce the detectable effect on the transmitted beam wave front.

The reported inhibition of the optical damage at low spatial frequencies can be of interest for the photorefractive optical memories [15] and for all applications involving volume grating recording in lithium niobate and other crystals with photovoltaic charge separation.

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References

[1] A.M. Glass, D. von der Linde, T.J. Negran, Appl. Phys. Lett. 25 (1974) 233.

[2] B.I. Sturman, V.M. Fridkin, The Photovoltaic and Photorefractive Effects in Noncentrosymmetric Materials, Gordon and Breach Science, Philadelphia, 1992.

[3] A. Armstrong, N. Bloembergen, J. Ducuing, P.S. Pershan, Phys. Rev. 127 (1962) 1918.

[4] G. Rosenman, A. Skilar, A. Arie, Ferroelectrics Rev. 1 (1999) 263.

[5] N.B. Ming, J.F. Hong, D. Feng, J. Mater. Sci. 17 (1982) 1663.

[6] I. Naumova, N. Evlanova, O. Gliko, S. Lavrishchev, J. Cryst. Growth 181 (1997) 160.

[7] I. Naumova, N. Evlanova, S. Blokhin, S. Lavrishchev, J. Cryst. Growth 187 (1997) 102.

[8] N.F. Evlanova, L.N. Rashkovich, Sov. Phys. Solid State 13 (1971) 223.

[9] A. Ashkin, G. Boyd, J. Dzedzich, R. Smith, A. Ballman, H. Levinstein, K. Nassau, Appl. Phys. Lett. 9 (1966) 72.

[10] N. Kukhtarev, V. Markov, S. Odoulov, M. Soskin, V. Vinetskii, Ferroelectrics 22 (1979) 946.

[11] M. Taya, M.C. Bashaw, M.M. Fejer, Opt. Lett. 21 (1996) 857.

[12] B. Sturman, M. Aguilar, F. Agullo-Lopez, V. Pruneri, P. Kazansky, J. Opt. Soc. Am. B 14 (1997) 2641.

[13] S. Odoulov, T. Tarabrova, A. Shumelyuk, I.I. Naumova, T.O. Chaplina, Phys. Rev. Lett. 84 (2000) 3294.

[14] H. Festl, P. Hertel, E. Kraetzig, R. von Balz, Phys. Stat. Solidi B 113 (1982) 157.

[15] L. Hesselink, S. Orlov, A. Liu, A. Akella, D. Lande, R.R. Neurgaonkar, Science 282 (1998) 1089.