Study of ferroelectric domain switching by domain wall induced light scattering

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The 90°-light scattering on domain walls was probed in various strontium barium niobate (SBN) crystals for studies of the ferroelectric switching under pulsed fields. The validity of this optical method is proved by a good agreement of the switching parameters deduced from optical scattering data with those obtained with electric methods. Scanning of the scattering over the crystal bulk revealed local specialities of the switching, particularly, a marked distribution of the domain wall density D along the polar axis with a maximum close to the negative electrode. In compliance with these *in situ* observations, the electro-optic coefficient r_c reveals a position dependence in all SBN crystals poled in the ferroelectric phase, r_c decreasing from the positive to negative electrode. This regularity is interpreted in terms of the domain density distribution $D(z)$ and accounted for by an asymmetry of the domain nucleation.
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I. INTRODUCTION

Ferroelectric solid solutions like $Sr_xBa_{1-x}Nb_2O_6$ (SBN) are of interest for optical applications due to their extremely high electrooptical and relatively high nonlinear-optical coefficients.[1] These applications are markedly related to the ferroelectric switching and the domain structure. In photorefractive holography the electric fixation of gratings is based on the ferroelectric switching (e.g., Refs. 2–4). Furthermore, the ferroelectric switching forms the basis for the creation of periodically-poled structures aimed for an optical frequency conversion in the quasiphasematched QPM mode of operation. Several attempts of obtaining periodically-poled SBN crystals were reported in literature.[5,6] The known negative features of SBN—an instability of its parameters (the ageing or fatigue effect)—also stem from a specific feature of the polarization or switching processes.[7–11] This specific behavior of SBN is to a great extent a consequence of its relaxor origin.[12] All practical problems as well as fundamentals of relaxor physics require studies of proper ferroelectric properties, particularly in the polarization mechanism. However, for SBN there is minor information reported on these topics. Measurements performed either by electrical[7–9] or optical[13] methods under quasistatic (slowly varying) electric fields revealed specific features caused by "freezing"[8] or, the same, "pinning"[9] effects characteristic for relaxor ferroelectrics. Macroscopically these fundamental effects appear as extremely slow relaxation of parameters, nonreproducibility $P-E$ loops in the course of repeated field cycling and the absence of an unambiguous coercive field. Recently we investigated for the first time the polarization (switching) of SBN crystals under pulse fields[10,11] and found a very unusual behavior, namely, anomalously long times of the polarization reversal even at $E \gg E_c$ and a pronounced difference in switching characteristics of poly- and single-domain crystals. In the present work we continued studies of the pulsed ferroelectric switching in SBN crystals using an optical method by measuring the dynamics of the 90°-light scattering on domain walls, reported by us previously.[14] This method seems to be applicable to other ferroelectric crystals, too. It provides a possibility to scan the switching process over the crystal bulk, which permitted us to reveal local specialities of switching, not detectable by integrating electric methods. These optical data give an insight to some specifics of the optical properties of this material. For example, on the base of the domain density distribution deduced from scattering measurements we propose an explanation for a spatial distribution of electro-optic coefficients presented in the given work.

II. BACKGROUND

In SBN crystals quasielastic 90°-scattering of a laser beam propagating normally to the polar axis was formerly reported.[15,16] It may be undoubtedly attributed to the existence of domains, because the scattering intensity was the largest in a poly-domain state and drastically decreased in a poled state or at temperatures above the phase transition in the paraelectric phase;[16] the scattering intensity could be modulated by applying a low-frequency sinusoidal field.[15] Therefore, a field-induced variation of such light scattering could provide a tool for studies of the ferroelectric switching. The scattering is evidently due to inhomogeneities in refractive indices n_i. As 180°-ferroelectric domains in SBN are

optically equivalent, this scattering is related to domain walls, which are areas of inhomogeneities in the dielectric permittivity. Following Kawai *et al.*[17] we treat domain walls as sandwichlike optical local inhomogeneities in n_i, so a plane wave with a momentum vector k_i propagating normally to the polar axis is partially reflected by a domain wall in the direction $k_r = k_i + 2(k_i q_m) q_m$, where q_m is the unit vector describing the wall geometry. The reflected amplitude due to a differential inhomogeneity dn and wall area dA can be approximated as

$$dE_r = E_i |k_i| H \frac{dn}{2n} dA, \tag{1}$$

where H is the thickness of the wall corresponding to dn (the approximation is valid for $|k_i|H \ll 1$). The total scattered intensity is obtained by integrating dE_r for a typical coherence volume and then—incoherently—by integrating over the illuminated volume. Following Eq. (1) the complete intensity of the 90°-light scattering is proportional to three parameters: (1) the average domain wall thickness, (2) the typical magnitude of the inhomogeneity of n in the vicinity of the wall, and (3) the total wall area (i.e., the number of domain walls, or, the same, the number of domains in the illuminated volume). In its turn, dn varies with applied electric fields due to the linear electro-optic effect. For a field E_3 directed along the polar axis and the wave vector orthogonal to it, for the extraordinary beam the refractive indices in opposite 180°-domains become $n_e^E = \left(n_e \pm \frac{1}{2} n_e^3 r_{33} E_3\right)$, where n_e and n_e^E are the initial and field-induced extraordinary indices, and r_{33} is the associated e.o. coefficient. In a first approximation we attribute the observed variations in the scattered intensity under applied fields to variations in the domain density D. Field-induced changes in H are regarded as a second order effect. A contribution from the electrooptic effect will be commented below. The domain density D controls several parameters of the ferroelectric crystals, particularly the electrooptic properties. As well known, in oxygen-octahedra ferroelectrics the linear electrooptic coefficients can be expressed as follows:[18]

$$r_{33} = 2 g_{33} P_s \epsilon_{33} \epsilon_0, \tag{2}$$

$$r_{13} = 2 g_{13} P_s \epsilon_{33} \epsilon_0, \tag{3}$$

where P_s is the spontaneous polarization, g_{ij} are the coefficients of the quadratic electro-optical effect in the paraelectric centro-symmetrical phase. So, the values of r_{ij} are governed by the domain state of the crystal via P_s. Let an elementary volume of the crystal be v and the volumes of "+" and "−" domains be v^+ and v^-, respectively; then $v^+ + v^- = v$, provided the domain wall thickness is much lesser that the domain size. For example, a positive domain density in a negative matrix is $D^+ = v^+/v$ and the linear electrooptic coefficient is related to the degree of the total volume polarization as $r/r_0 = (v^- - v^+)/v$ (where r_0 corresponds to the single-domain state). One can derive a linear relation

$$\frac{r}{r_0} = 1 - 2D^+, \tag{4}$$

so electro-optic coefficients linearly decrease with D^+. A related parameter, the half-wave voltage

$$V_{\lambda/2} = \frac{\lambda}{r n_e^3} \frac{d}{l}, \tag{5}$$

where d and l are the linear sample dimensions, consequently grows with D. Therefore, all variations of the values of r_{ij} or $V_{\lambda/2}$ in a given crystal may be interpreted in terms of the domain density.

III. EXPERIMENT

Three SBN compositions were under study: SBN-0.75, SBN-0.61, SBN-0.61:0.5 wt. % Nd (further referred to as SBN:Nd). The main reason for this choice is due to the fact that recently we investigated the pulsed switching in the same SBN compositions by means of measuring switching currents.[10,11] The values of the coercive field E_c in these compositions are different due to different phase transition temperatures (52, 82, and 68 °C, respectively[19]). The crystals were grown by the modified Stepanov technique.[20] The samples under study were optically polished cubes of $x*y*z = 5*5*5$ mm^3 in size. In light scattering experiments the beam from a He–Ne laser propagated normally to the polar axis z along the x (or y)-axis and the scattered radiation was registered in the orthogonal direction y (or x). The incident laser beam was linearly polarized along z; the scattered beam was measured through an analyzer with the polarization also parallel to z (i.e., extraordinary polarization for both the incident and scattered waves). On this stage of investigation, only this configuration of observation $X(ZZ)Y$ [or $Y(ZZ)X$] was used, therefore, the degree of depolarization of the scattered beam was not measured yet. The incident beam was focused onto the sample by a lens with a focal length of 60 mm. The intensity of the laser beam was attenuated to approximately 0.05 W/cm^2 in order to avoid any photoconductive effect under applied fields. Only a small part of the beam path cut by a diaphragm disposed directly behind the crystal was imaged to a photomultiplier by another long-focal lens. A low intensity of the propagating laser beam permitted us to neglect the photoconductivity, so a uniform voltage distribution over the crystal bulk was assured. The intensity was measured with an uncertainty of 8%. We studied the behavior of the scattering intensity under external field pulses applied along the z-axis. Field pulses with a rise time less than 1 μs and a duration in the range from 0.4 to 20 ms were used. In this range the results do not depend on the pulse duration.

Electro-optic properties of SBN crystals were measured with the aid of a dynamical method with a phase sensitivity of $2\pi 10^{-6}$. The signal was excited by an ac-measuring field with an amplitude of 6 V/cm and a frequency of 1000 Hz (case of an unclamped crystal). For the used transverse geometry ($E \| z, k \perp z$) the measured electro-optic coefficient was

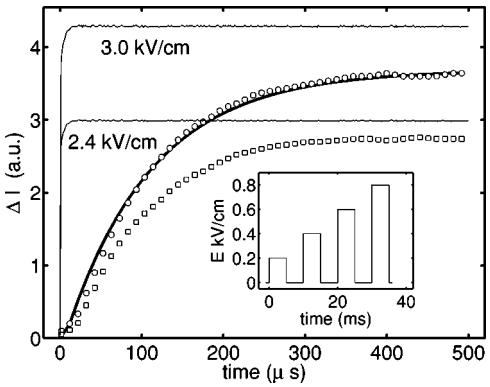

FIG. 1. The thin steplike curves show the risetime of the pulses in μs. The time evolution of the corresponding scattering intensities (scattering kinetics) is given by the markers. The solid line close to the upper curve represents a fit of the experimental data with an exponential function according to Eq. (7). The inset shows the used field pulse train in a ms time scale.

$$r_c = r_{33} - \left(\frac{n_o}{n_e}\right)^3 r_{13}. \tag{6}$$

All measurements have been performed at room temperature with an accuracy of 10% for r_c. To obtain spatial distributions of the light-scattering or electro-optic coefficients we probed the crystal bulk with the focused laser beam of 0.1 mm in diameter.

IV. 90°-LIGHT SCATTERING AS A TOOL FOR STUDIES OF FERROELECTRIC SWITCHING

A. Scattering kinetics under field pulses

Figure 1 shows examples of two typical scattering flashes of electric pulses on a SBN-0.75 sample. It should be emphasized that for $E=0$ in both polydomain and poled ("single-domain") SBN crystals a nonzero scattering intensity (a "background" I_b) exists. It may be due to a variety of origins, for example, to growth layers,[21] and only partially to domains. Below in terms of domain evolutions, we discuss a change of the scattered intensity $\Delta I = I_s - I_b$ (a "flash") under an applied field pulse. A flash of the beam trace within the crystal bulk under a high field may be so luminous that it is detected by naked eyes. Below we denote by $+I$ or $-I$, respectively, an increase or decrease of the scattering intensity with respect to an initial I_b. All experiments were performed by means of applying in turn pulse trains of opposite polarities with increasing amplitudes (see the inset in Fig. 1).

The temporal dependence $\Delta I(t)$ (the curves with markers in Fig. 1) is essentially retarded with respect to the front of the field pulse, which means that a contribution from the electrooptic effect $\left(\delta n = -\frac{1}{2} n_e^3 r_{33} E_3\right)$ is negligible, otherwise the scattering response would raise synchronically with the field. Therefore, the kinetics of $\Delta I(t)$ are related to domain evolutions. In ferroelectrics the kinetics of pulsed switching are characterized by a switching time, which corresponds to a drop down the switching current by 90% of its peak value.[22,23] For our study we defined it as a characteristic time for $\Delta I(t)$ reaching a saturation under a given pulse amplitude (Fig. 1). Fitting the measured $\Delta I(t)$ by a function

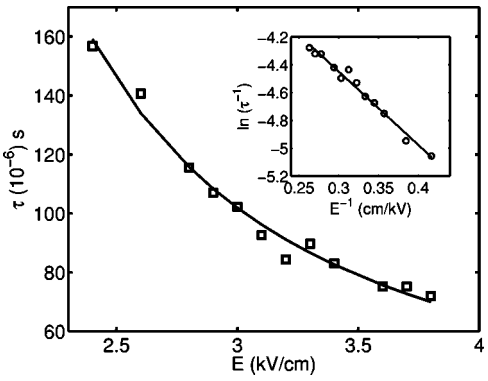

FIG. 2. Switching times τ and switching velocities τ^{-1} as a function of the field amplitude in a polydomain SBN-0.75. The line in the inset represents a fit to $\tau_s^{-1} = A \exp(-(E_a/E))$ with $A = (5.4 \pm 0.4) 10^4\ \mathrm{s}^{-1}$ and $E_a = (5.1 \pm 0.5)$ kV/cm.

$$\Delta I(t) = \delta I_{\mathrm{sat}} \left[1 - \exp\left(\frac{-t}{\tau_s}\right) \right] \tag{7}$$

we can derive the parameters τ_s and their dependence on E. An example of approximating $\Delta I(t)$ by this exponential function is shown by the solid line in Fig. 1. Figure 2 presents the field dependence $\tau_s(E)$ in an initially polydomain sample SBN-0.75 obtained from a set of curves $\Delta I(t)$ under applying a pulse train. The values of τ_s (tens of microseconds) are in a satisfactory agreement with switching times observed in the same crystal by the switching current method.[10,11]

The inverse parameter $v_s(E) \propto \tau_s^{-1}(E)$ shown in the inset in Fig. 2 has the meaning of a switching velocity. Its field dependence at moderate fields often obeys an exponential law $\tau_s^{-1} = A \exp(-(E_a/E))$ reported earlier.[22,23] A fit to the measured values of τ_s^{-1}—shown as a line in the inset in Fig. 2—yields $A = (5.4 \pm 0.4) 10^4\ \mathrm{s}^{-1}$ and $E_a = (5.1 \pm 0.5)$ kV/cm. The latter value characterizing an "activation field" for switching is reasonable and close to the values of E_a found in other ferroelectrics, such as $BaTiO_3$ and TGS.[23] Here we should dwell on a fundamental divergence of the switching process in SBN crystals from the model scenario.[22,23] In model ferroelectrics under a field pulse $E > E_c$ a switched charge approaches the spontaneous polarization, $Q_s \to P_s$, so during the switching time a crystal becomes practically single-domain. In contrast, in SBN crystals Q_s practically never achieves P_s under a short field pulse,[10,11] because the total polarization reversal requires times being larger by orders of magnitude. Thus, under our experimental conditions after applying a pulse $E > E_{\max}$ the selected crystal volume is poled to a small extent only ($Q_s \ll P_s$), and τ_s describes a partial switching. That is why in the case of pulse switching of a polydomain crystal characterized by Fig. 1 and Fig. 2 and below by Fig. 3, the background intensity I_b is practically unchanged after applying a pulse train.

B. Field dependencies of light scattering in poly- and single domain crystals

We continue with a discussion of the field dependence of scattering intensities, i.e. of the domain density. First, we present and discuss the scattering data in poly-domain SBN crystals. Figures 3(a)–3(c) present the field dependencies of

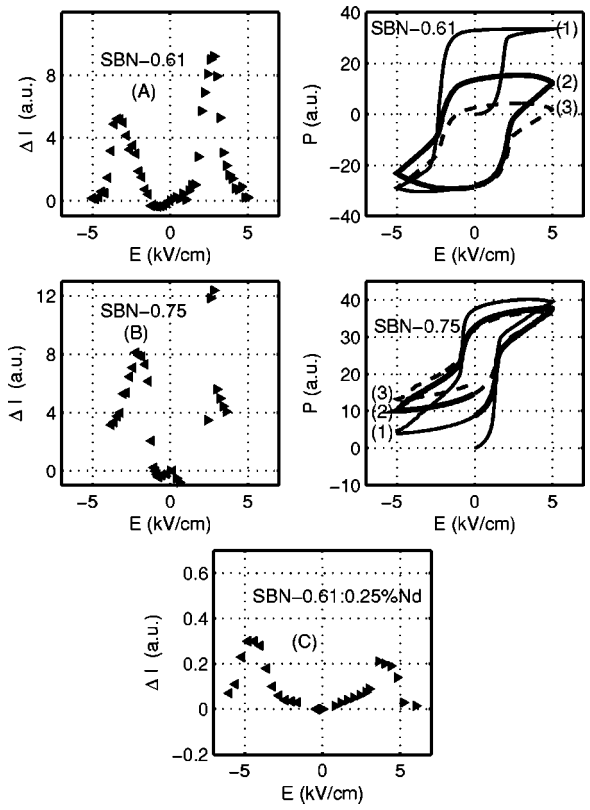

FIG. 3. Light scattering intensities as a function of the field amplitude in polydomain crystals SBN-0.61 (a), SBN-0.75 (b), and SBN-0.61:Nd (c). The sign of the field is settled in the laboratory coordinate system. The insets show $P-E$ loops obtained under quasistatic fields in the same crystals, labels 1, 2, 3 denote successive field cycling.

the scattering intensities $\Delta I(E)$ in SBN-0.61, SBN-0.75, and SBN-Nd crystals observed under field pulses of two opposite polarities. The right and left branches in all curves $\Delta I(E)$ shown below correspond to the field signs "+" and "−," respectively, uniquely settled in the laboratory coordinates. Prior to apply a field pulse train of any polarity, the crystal was annealed at $T>T_c$ and cooled slowly to room temperature. This means that $+E$ and $-E$ branches of $\Delta I(E)$ curves were measured under identical conditions starting from a polydomain state.

The dependencies $\Delta I(E)$ in all compositions are qualitatively very similar. However, in SBN:Nd the scattering intensity is essentially lower than in undoped samples. Partly it may be assigned to different optical absorptions (for $\lambda=633$ nm, $\alpha=0.03$ and 0.3 cm^{-1} in SBN and SBN:Nd, respectively), however, more likely it is related to a difference either of the domain density, or the domain wall thickness. The following common features may be recognized in the curves $\Delta I(E)$ in the crystals under study: (1) in the range of relatively low fields of both signs, ΔI only very slightly varies with E; (2) at certain E values depending on the composition, ΔI increases rather sharply with E, passes over a maximum at $E_{\max}$ and then comes to a low saturation value; (3) the curves are asymmetrical, namely, in a given crystal the values of $E_{\max}$ and peaks ΔI are different for the two field polarities. In the language of ferroelectricity the curves in Fig. 3 may be interpreted as a weak switching (or polarization process) under fields approximately within 1–2 kV/cm,

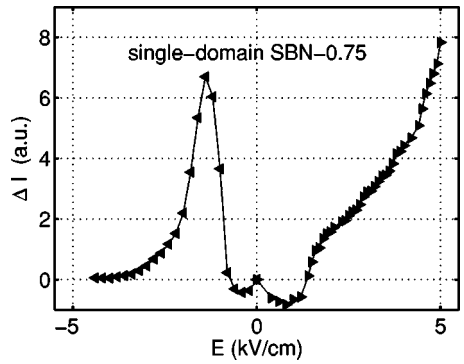

FIG. 4. Light scattering intensity as a function of the field amplitude in a single-domain SBN-0.75 crystal. After poling the crystal with a negative $E_p=-1.2$ kV/cm, a positive (repolarizing) pulse train was applied yielding the right branch. Afterwards a negative pulse train was applied resulting in the left branch.

an acceleration of switching in the ranges close to $E_{\max}$ and then a saturation of a remnant polarization at $E>E_{\max}$. Thus, the values of $E_{\max}$ should correlate to the coercive field E_c. This is corroborated by a higher $|E_{\max}|$ in SBN-0.61 than in SBN-0.75. A negative slope of $\Delta I(E)$ at $E>E_{\max}$ reflects the switched charge saturation Q_s, thus a decrease of the domain density. Different absolute values of $E^+_{\max}$ and $E^-_{\max}$ and different peak intensities of ΔI for $+E$ and $-E$ branches in a given crystal characterize a unipolarity, i.e., a preferential direction of P_s in a crystal on a whole or at a given crystal position.

To justify this interpretation, in Figs. 3(a) and 3(b) we present $P-E$ hysteresis loops obtained in the same samples in a quasi-static regime with a cycling loop of about 2 h. The observed specific features of the $P-E$ hysteresis, namely, open loops and a non-coincidence of their trajectories on several first cycles is characteristic for all SBN crystals. It was detected by various methods[7–9,13] and accounted for by freezing or pinning effects. Averaged values of E_c estimated from several $P-E$ cycles after the start for SBN-0.75 and SBN-0.61 are (1.5 ± 0.3) and (2.5 ± 0.5) kV/cm, respectively, being in reasonable agreement with averaged values of $|E_{\max}|$ (2 and 3 kV/cm, respectively). The degree of the unipolarity determined from the $P-E$ hysteresis loops is characterized by a bias field $E_b=(|E_1-E_2|)/2$ (where E_1 and E_2 are coercive fields corresponding to the left and right halves of the loop). Averaged values of E_b are 0.15 and 0.3 kV/cm in SBN-0.75 and SBN-0.61, respectively. For the optical curves $\Delta I(E)$ we define a bias field in a similar way $E_b=(|E^+_{\max}-E^-_{\max}|)/2$, which gives $E_b=0.3$ kV/cm and 0.4 kV/cm for SBN-0.75 and SBN-0.61, respectively. These values are in reasonable agreement with dielectric results. The values of $E_{1,2}$ in $P-E$ loops slightly differ from $E^{\pm}_{\max}$ in $\Delta I(E)$ curves because of different conditions of measurements and evaluation. The existence of an unipolarity in a polydomain (thermally de-poled) SBN crystal is supported by electrical measurements.[8]

We now shortly present results in a single-domain crystal. Figure 4 shows $\Delta I(E)$ curves obtained in a sample SBN-0.75 poled by cooling from $T>T_c$ to room temperature under a dc-field $E_p=-1.2$ kV/cm. First, the positive (repolarizing) pulse train was applied to the crystal [the right

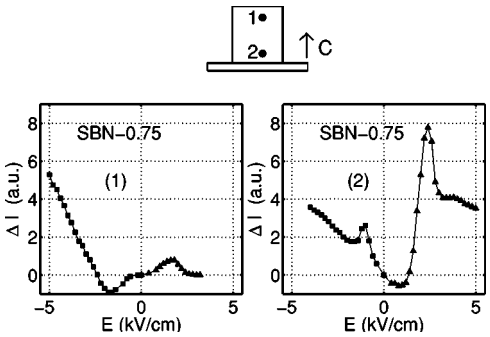

FIG. 5. Light scattering intensities as a function of the field amplitude in the vicinity of electrodes (1, the upper; 2, the lower) in a polydomain SBN-0.75 crystal. The upper inset represents schematically the layout of the sample.

branch of the $\Delta I(E)$ curve]. As seen, $\Delta I(+E)$ progressively grows with no saturation or maxima. This indicates that under repolarizing fields the crystal becomes depoled, the degree of depoling increasing with the pulse amplitude. No total reversal of P_s occurs, otherwise $\Delta I(+E)$ would pass over a maximum or tend to saturate. This is consistent with the data on switching currents,[10,11] which evidenced a minor switching of single-domain SBN crystals under repolarizing short pulses. After applying a positive field train, a negative train coincident with E_p was applied (the left branch of the curve in Fig. 4). One may see a maximum in the curve $\Delta I(-E)$ with a following decrease of I_b to zero at $|E|>|E_{\max}|$. This means that the crystal, partially depoled by $+E$ pulses, is now restored to its single-domain state. We mention that in a single-domain state the background scattering I_b is strongly decreased compared to the poly-domain one after applying field pulses.

C. Spatial variations in the ferroelectric switching

Now we show an example of a local speciality of ferroelectric switching, which was observed by scanning the bulk, and hence not found by integrating electrical methods. The field dependencies $\Delta I(E)$ presented in Fig. 3 and Fig. 4 (in poly and single domain state, respectively) with a small scatter in the values of $E^{\pm}_{\max}$ are qualitatively similar in any region of a given crystal apart from the electrodes. However, when approaching them, the curves $\Delta I(E)$ become qualitatively different. Figure 5 (1) and (2) present $\Delta I(E)$ obtained in a polydomain SBN-0.75 sample [the same as in Fig. 3(b)] in spots adjacent to electrodes, approximately 0.1 mm apart from the upper and lower ones, respectively (the upper inset in Fig. 5). Both branches of these curves were obtained again after a preliminary annealing of the crystal in the paraelectric phase. These edge-related dependencies $\Delta I(E)$ show a pronounced asymmetry of the branches $\Delta I(-E)$ and $\Delta I(+E)$. When a positive potential is applied to a given electrode (right branches of the curves in Fig. 5 1,2), the dependence $\Delta I(+E)$ is similar to $\Delta I(\pm E)$ apart from electrodes, it shows a maximum at $E^{+}_{\max}$ coinciding with the average $E^{\pm}_{\max}$ in the bulk and comes to a low saturation level at $E>E^{+}_{\max}$. In contrast, if a negative potential is applied to the same electrode (left branches of the curves), then ΔI after passing over a slight maximum, gradually increases with E without tending to saturate. It should be emphasized that these dependencies unambiguously correlate with the polarity of the electrode: a gradual increase of $\Delta I(E)$ in a polydomain crystal is always observed close to the negative electrode. This means that with increasing field amplitude, the domain density is growing at the negative electrode only and becomes nonuniformly distributed along the polar axis. The possible explanation of this nonuniformity is a preferential domain nucleation at the negative electrode and subsequent evolution of domains in the region adjacent to it, whereas the positive electrode is inactive. Such a nonequiprobability of the domain nucleation is not unique, for example, a preferential domain nucleation was also observed at the negative electrode in $LiNbO_3$.[24] In SBN it might be related to the above mentioned unipolarity, inherent to a polydomain relaxor ferroelectric.

For SBN crystals needlelike domains (fourfold pyramides with an angle by vertices of about 0.5°–1°) are characteristic with lengths up to 0.5–1 mm and several microns for the cross section.[25–27] Under external fields the stage of the frontal growth of these needles is very pronounced,[27] whereas a coalescence via the side domain wall movement typical for model ferroelectrics[22,23] is weakened. One may imagine that in SBN crystal close to the negative electrode a pattern of these small needles is accumulated, which cannot grow through the crystal bulk during short field pulses.

D. Discussion

The switching parameters (coercive and bias fields, switching times and velocities) estimated from the data on the light scattering are in a good agreement with the results of conventional electrical methods. Therefore, 90°-light scattering is an appropriate tool for studies in the ferroelectric switching. The details of switching, particularly a different switching in a poly and single-domain state detected from the optical data, are consistent with the specific switching scenario in SBN proposed on the base of electrical studies.[7–11] The scattering method offers advantages over electrical measurements, first because it provides a possibility of spatial scanning over the crystal bulk. Due to this we revealed, for example, a speciality in a coordinate dependence of the switching, namely an enhanced domain density near the negative electrode. The second advantage is a good time resolution of the scattering signal, which exceeds the resolution of the switching current method, limited by a relatively high capacity of SBN crystals ($\epsilon \sim 10^3$–10^4). The method is nondestructive, because it requires the use of very weak light intensities which cannot affect the switching properties, hence the method provides certain advantages over related optical methods, such as field-induced switching of the two-beam coupling gain Γ (Refs. 13 and 28) or of the "fanning."[29] The latter methods can be realized in photorefractive crystals only and their resolution is limited by rather long Maxwell relaxation times. Note that the fanning effect in photorefractive crystals (a photoinduced light scattering caused by a nonlinear coupling of the transmitted light-wave with the scattered ones) requires a seed or primary light scat-

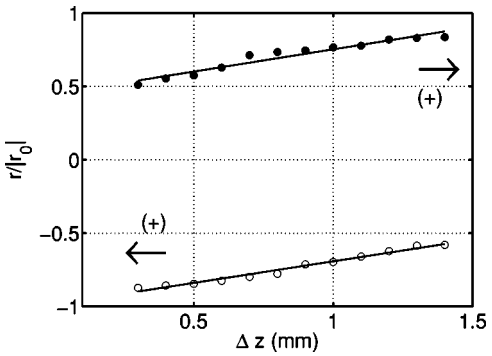

FIG. 6. The distributions of the electro-optic coefficient r_c along the polar axis in a SBN-0.61 sample poled by two fields of the opposite polarities at room temperature. The negative (upper plot) and a positive (lower plot) polarities are fixed in the laboratory coordinates.

tering (e.g., Ref. 30), whose most probable origin in SBN crystals could be the light scattering on domain walls discussed in the present work.

The domain wall induced light scattering is obviously common for a wide group of ferroelectric crystals. For example, a stationary light scattering at domains was also observed in Gd_2MoO_4 (Ref. 31) and $Sn_2P_2S_6$.[32] In SBN crystals this effect is very pronounced, perhaps because of a relatively large thickness of a domain wall, or, to be precise, of a perturbed layer adjacent to it. A possible reason may be either an elastic strain or a space-charge field in the vicinity of the domain wall. These would lead to a high inhomogeneity in the refractive indices or even to an appearance of a birefringence via the elasto- or electro-optic effects, respectively. For example, elastic strains of the order of 10^{-4} were detected in the vicinity of domain walls in switched $LiNbO_3$ crystals.[33,34] A large elastic strain at a moving domain wall may be specific for ferroelectric crystals with a high piezoelectric effect; in SBN the latter is even higher than in $LiNbO_3$ [e.g., $d_{33}=140\cdot10^{-12}$ C/N (Ref. 35) and $\sim20\cdot10^{-12}$ C/N, respectively]. From this point of view the presented technique of measuring the domain wall induced light scattering, could be suitable for studies of the switching in $LiNbO_3$, too. A space-charge field arises, if the domain wall is inclined with regard to P_s, which is realized in SBN.[25–27]

V. COORDINATE DEPENDENCE OF THE ELECTRO-OPTIC COEFFICIENTS IN SBN CRYSTALS AND ITS RELATION TO A DISTINCT BEHAVIOR OF FERROELECTRIC SWITCHING

A spatial nonuniformity of the domain density under applied fields discussed above gives an insight into a specific of the electrooptic effect in SBN crystals described below. Figure 6 exemplifies the coordinate dependencies of r_c measured along the polar axis in a poled SBN-0.61 sample. The experimental values of r_c are normalized to the maximum value r_0 [see Eq. [4]] achieved in the given crystal (thus approaching one in the single-domain state). The two presented $r_c(z)$ plots were obtained after poling the crystal at room temperature by applying fields of opposite polarities and equal amplitudes. In the given case the poling fields were $|E_p|=5.6>E_c=(2-2.6)$ kV/cm. Before poling, the crystal was every time brought to the polydomain state by annealing it in the paraelectric phase. Solid lines in Fig. 6 present a fit of the experimental data to linear functions. One may see that the slope $r_c(z)$ in the laboratory coordinates is reversed after a change of the poling field sign. Therefore, this slope is controlled by the sign of the poling field rather than by possible growth reasons, for example, a compositional variation along the growth axis. The slope of $r_c(z)$ is always unambiguously settled by the polarity of the electrode, namely, r_c shows a minimum close to the negative electrode and smoothly increases when scanning towards the positive one. In the crystals, where $V_{\lambda/2}(z)$ instead of $r_c(z)$ was scanned, the data are qualitatively the same, namely after poling at room temperature $V_{\lambda/2}$ has its maximum close to the negative electrode. These regularities persist even after a very long poling, they were found in a variety of SBN crystals [e.g., SBN-0.75, SBN-0.61, SBN-0.61:Cr, SBN-0.61:Ce, SBN -0.61:(Ce+La)]. Coordinate dependencies of $r_c(z)$ are reproducible provided that prior to applying a poling field, the sample was brought to an initial poly-domain state. In more detail it is described in Ref. 36. Note that a nonuniformity of the electro-optic coefficient within the crystal bulk may be responsible for certain scatter of r_{ij} values in the literature. An almost linear plot of $r_c(z)$ (Fig. 6) may account for an effect of scanning of a laser beam under external fields in SBN crystals reported recently.[37]

We now discuss the electro-optic data in terms of the domain density. According to Eq. (4) a spatial distribution of $r_c(z)$ means that after poling in the ferroelectric phase, a stable smooth distribution of a residual domain density along the polar axis is formed in the crystal. The maximum of $D(z)$ is always adjacent to the negative electrode, decreasing smoothly towards the positive one. This type of distribution correlates qualitatively to $D(z)$ observed *in situ* under applied fields (see Sec. IV C), the coordinate dependencies both of them revealing an increased domain density near the negative electrode. If extending this assumption of a polarity of the preferential domain nucleation and taking into account a weakened stage of their coalescence, one may conclude that even after a very long field application in the ferroelectric phase certain domain density still persists close to the negative electrode. In other words, poling this crystal in the ferroelectric phase does not produce a uniform single-domain state and the distribution of the electro-optic coefficient cannot be completely smoothed.

VI. SUMMARY

In SBN crystals an essential 90°-scattering of light propagating normally to the polar axis is observed. The scattering centers are domain walls. In a first approximation, the scattering intensity under applying fields is controlled by the domain wall density. Studies in this scattering under applied fields provide a convenient tool for studies in the pulsed ferroelectric switching. This was concluded from a good agreement of the switching parameters estimated from field and temporal dependencies of the scattering intensity with the data of the electrical measurements. The scattering data fit well in the scenario of the pulsed switching of SBN pro-

posed recently on the base of the switching currents analyzed. The method permits uses to particularly scan the ferroelectric switching within the crystal bulk. In poly-domain SBN crystals, a nonuniform distribution of the domain density D under applying fields was found indicating a preferred domain nucleation at the negative electrode. The domain density distribution correlates to a spatial distribution of the electro-optic coefficient $r_{ij}(z)$ observed in SBN crystals after poling in the ferroelectric phase. In agreement with the distribution $D(z)$ deduced from the scattering experiments, r_{ij} smoothly decreases from the positive to negative electrode. The domain wall induced light scattering seems to be appropriate for studies in switching in other ferroelectric crystals.

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