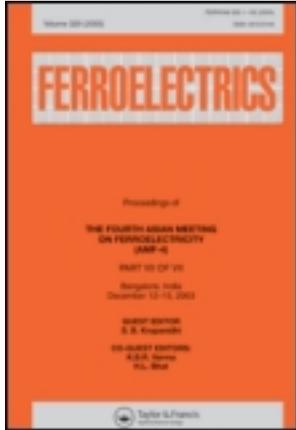


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K. Matyjasek^a, S. M. Kaczmarek^a & L. I. Ivleva^{a b}

^a Institute of Physics, Faculty of Mechanical Engineering and Mechatronics, West Pomeranian University of Technology, Al. Piastów 48, 70-310, Szczecin, Poland

^b General Physics Institute of the Russian Academy of Sciences, Vavilov Str. 38, 119991, Moscow, Russia

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Temperature Dependence of Domain Switching in Cr Doped $\text{Sr}_{0.61}\text{Ba}_{0.39}\text{Nb}_2\text{O}_6$ Single Crystals

K. MATYJASEK,* S. M. KACZMAREK, AND L. I. IVLEVA**

Institute of Physics, Faculty of Mechanical Engineering and Mechatronics, West Pomeranian University of Technology, Al. Piastów 48, 70-310 Szczecin, Poland

Domain nucleation and growth during the switching process in ferroelectric relaxor $\text{Sr}_{0.61}\text{Ba}_{0.39}\text{Nb}_2\text{O}_6$ doped with Cr ions have been observed by means of a nematic liquid crystal (NLC) method. Micro-scale studies of the switching process in conjunction with electrical measurements allowed us to establish a relationship between local properties of domain dynamics and macroscopic response i.e., polarization hysteresis loop and dielectric permittivity measurements.

Keywords Polarization reversal; relaxor; strontium barium niobate

PACS: 77.80.Dj; 77.80.Fm, 77.84.Dy

1. Introduction

Strontium barium niobates, SBN ($\text{Sr}_x\text{Ba}_{1-x}\text{Nb}_2\text{O}_6$, $0.25 \leq x \leq 0.75$), are ferroelectric crystals that have received a great interest in their applications in optoelectronics, due to their large electro-optical and relatively high non-linear optical coefficients [1]. Compared to conventional ferroelectrics, a unique property of relaxors is the appearance of a very broad and frequency—dependent dielectric anomaly with high dielectric permittivity values [2]. These unusual dielectric properties of relaxors have been explained by proposing a model, in which relaxor behavior can be attributed to the development of a quenched random field, associated with the compositional/structural disorder [3]. Despite numerous experimental investigations of dielectric relaxation, the microscopic origin of unusual physical properties of relaxors still remains a subject of discussion [4]. The fundamental point is determination of a relationship between the ferroelectric polar structures and their respective dynamical response. The open tungsten bronze structure of SBN allows various possibilities to accommodate different doping ions, taking into account that only five out of six available positions of Sr^{2+} and Ba^{2+} host cations are occupied [5]. It was reported that doping improves the properties via a significant increase of the electro-optical and piezoelectric coefficients [6, 7]. There are very limited works devoted to study the influence of doping on ferroelectric properties, especially polarization switching mechanism. Most domain structure studies were limited to piezoresponse force microscopy (PFM) techniques,

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**General Physics Institute of the Russian Academy of Sciences, Vavilov Str. 38, 119991 Moscow, Russia

*Corresponding author. E-mail: km@zut.edu.pl

providing the visualization of the domain structure at the nano-scale [8–11]. We have recently examined the effect of Ni doping on ferroelectric and dielectric properties of $\text{Sr}_{0.61}\text{Ba}_{0.39}\text{Nb}_2\text{O}_6$ single crystals [12]. In presented report we investigate the influence of temperature on the micro- scale domain structure and switching properties in Cr doped $\text{Sr}_{0.61}\text{Ba}_{0.39}\text{Nb}_2\text{O}_6$ (SBN61: Cr) relaxor crystal. The nematic liquid crystal (NLC) method was applied to visualize domain structure dynamics. This method do not require complex instrumentation and enables non-invasive investigation of domain structure dynamics with a micrometer resolution.

2. Experimental Results

SBN crystal exhibits only 180° domains because the paraelectric phase has a tetragonal symmetry (4/mmm), and the transition to ferroelectric phase (4 mm) occurs along [001] direction [13]. Single crystals SBN61:Cr were grown by the modified Stepanov method [14]. Doping was performed by adding Cr_2O_3 (0.01 wt.%) in the melt. Plateled-shaped samples were cut perpendicular to the [001] direction and polished to optical quality. The samples were annealed at high temperature in order to relax possible residual strain that could be introduced during the crystal growth. The results presented refer to a single sample of SBN61:Cr with surface area of 0.16 cm^2 and thickness 0.1 cm (in the polar direction). However, these results are typical to a fairly large number of specimens.

In order to observe optically indistinguishable 180° domain walls the NLC mixture of p-methoxybenzylidene-p-n-butylaniline (MBBA) and pethoxybenzylidene-p-n-butylaniline (EBBA) was used. For dynamic observation in an electric field, a crystal plate with a thin NLC layer, on its upper and lower surface, was sandwiched by two glass plates with a tin oxide coating as transparent electrodes. The observation of the domain structure was carried out using a polarizing microscope. The method of visualizing the domain structure is based on the contrast in NLC layer in the region above moving domain walls. In the region where the repolarization occurs, switching current flows followed by a decrease in the impedance of the ferroelectric; hence the major part of the applied voltage falls on the liquid crystals. Thus the reversed regions, where reorientation of domains still occurs, look somewhat darker, as in these regions a certain electrohydrodynamic instability, particularly dynamic scattering, takes place [15]. Some limitations of this method is connected with a slow response time (of the order of several ms) of NLC layer. Moreover, the voltage is applied to the sandwich structure consisting of ferroelectric sample and NLC layers (the thickness of NLC layer of about several μm). Thus it is difficult to estimate the real magnitude of the voltage applied to the ferroelectric sample, especially during the switching process. Therefore, the visualization of the domain structure by NLC method can only be used in checking the domain distribution and growth during the switching process, which obviously reflects internal disorder of the crystal.

Hysteresis loops (D-E dependence) were recorded using a modified Sawyer-Tower circuit by using an ac field of 50 Hz and digital oscilloscope. Dielectric permittivity dependence on temperature and frequency was measured by HP 4284A LCR meter. The electrical measurements were carried with air-drying silver paste as electrodes.

3. Results and Discussion

Ferroelectric polarization reversal, which proceeds by nucleation and subsequent domain wall motion, is significantly more complex in SBN relaxors in comparison with convectional ferroelectric crystals. In conventional ferroelectrics the growth of existing domains is more

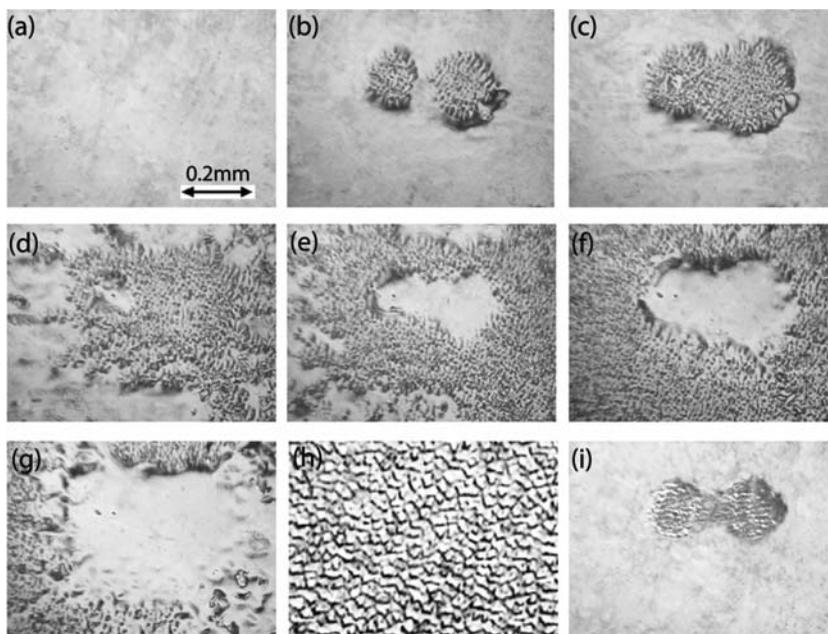


Figure 1. Domain pattern evolution observed after application of the electric field $E = 2.4 \text{ kVcm}^{-1}$: (a) initial single domain state. Time from the moment of applying E in [s]; (b) 2; (c) 4; (d) 8; (e) 10; (f) 12; (g) 14; (h) shows the zoom image of the small fragment of the image (f); (i) domain pattern obtained after applying the negative electric field $E = -2.4 \text{ kVcm}^{-1}$.

favorable than the creation of new ones. So that the nucleated domains expand with little or no resistance under an electric field and start to coalesce into larger ones, and consequently this process is accompanied by a decrease of the domain density. Figure 1 shows the series of video frames illustrated the domain pattern evolution, starting from a single—domain state, in the electric field of 2.4 kVcm^{-1} . This field is high enough to complete the micro scale domain switching in the entire volume of the sample, because the further increase of the electric field do not induce any domain switching. The dark areas shown in Fig. 1 correspond to the areas that actually reversed their polarization direction and clear areas correspond to the regions where the switching process has not started yet (Fig. 1a) or has already been completed (central part of images 1(e)-1(g)). The nucleated domains do not grow continuously, as local structure irregularities provide strong pinning sites at which the domain walls remain fixed, forming a maze type domain pattern [see Fig. 1(h)]. The observed complexity of the domain structure of relaxor-type crystals could be understood in terms of the slow inhomogeneous domain growth in the presence of random pinning fields [3]. In such crystals fast growing domains coexist with slower ones, corresponding to different local barrier heights, giving rise to suppression of the switching process. The doping additionally deteriorates the crystal homogeneity. The inhomogeneous distribution of the domain nuclei can reflect the non uniform defect structure distribution within the crystal sample. Similar picture of the nucleated domains has been observed for the positive [Fig. 1(b)] and negative [Fig. 1(i)] electric field. It demonstrates the presence of a frozen polarization state, due to locally accumulated defects as observed in some region of the crystal sample. We use the term “nucleation” to describe emergence of new antiparallel domains within the original domains as they appear in the video images. However it is an

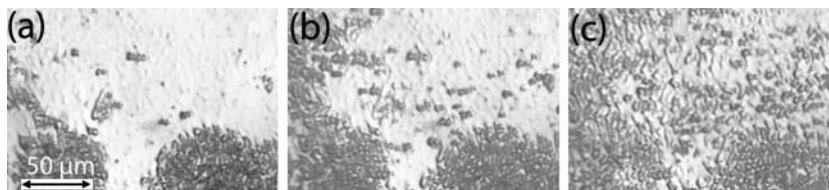


Figure 2. Domain pattern obtained after electric field application of 2.4 kVcm^{-1} at various temperatures: (a) 320 K; (b) 330 K; (c) 340 K.

open question whether the initial domain state [Fig. 1(a)] is single-domain or contains the nanoscale residual domains, which are not resolved by NLC method.

It is well known that the activation field for nucleation and the domain growth decrease with increasing the temperature [16]. Figure 2 shows the domain pattern observed just after application of an electric field of 2.4 kVcm^{-1} for three different temperatures. With the increase of the temperature the number of nucleated domains remains nearly unchanged until a temperature of 320 K is reached. The observed increase in the polarization in this temperature range is mostly due to the decrease in the threshold nucleation field and thus coercive field and the increase in the domain walls mobility. The pronounced increase in the density of domain nuclei was observed in the vicinity of 340 K. At this temperature the domains start to emerge nearly on the whole surface area, after application of the electric field, [see Fig. 2(c)]. A close inspection revealed that beside the sites where intensive nucleation takes place [seen as dark regions in Fig. 2(c)] the macroscopically visible domain walls arise with lower wall velocity. This is reflected in a pour contrast of NLC above the slowly moving domain walls. The specific mechanism driving the nucleation in the high temperatures results from the local structure irregularities, and could be interpreted in term of a wide distribution of activation energies for domain nucleation and growth. This is closely reflected to distribution of random electric fields, inherent to relaxor-type crystals. It will result in the distribution of the waiting time for local switching over different places of the crystal sample.

Figure 3(a) presents the shape of hysteresis loops (H-L) obtained at various amplitudes of an ac-field at frequency 50 Hz. The macroscopic in nature, polarization versus electric field, data obtained in high electric fields support the optically observed nonuniform switching process. At low electric field H-L has a near square shape relative to P-E axis, indicating an abrupt change of the polarization orientation. At higher E, the H-L becomes

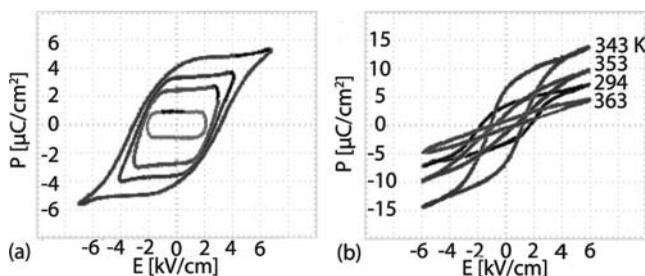


Figure 3. Hysteresis loops of SBN61:Cr crystal; under different ac-electric field at room temperature (a); at various temperatures (b).

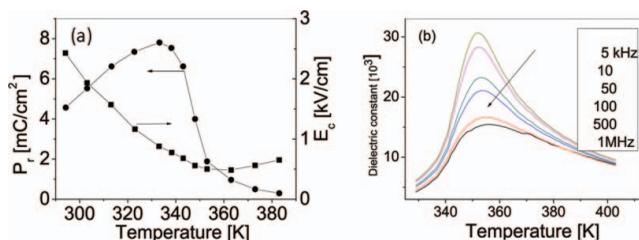


Figure 4. Temperature dependences of: remnant polarization P_r and coercive field E_c (a); dielectric constant (b).

more slanted. It means that the slowly switching regions, initially frozen, become activated and start to participate in the polarization reversal process. The SBN61: Cr crystal sample has no precisely defined coercive field, E_c , which increases with the increase of electric field amplitude. Figure 3(b) illustrates the H-Ls taking at various temperatures. As temperature increases the H-L becomes more saturated because of the significant decrease of the coercive field. Typical ferroelectric H-Ls are observed up to 353 K when large electric fields up to 6 kVcm^{-1} were applied. At 363 K and higher, typical linear dielectric responses are observed, suggesting that the investigated SBN61: Cr crystal becomes paraelectric at macroscopic scale. The variation in E_c and remnant polarization P_r with respect to the temperature is presented in Fig. 4(a). It is seen that E_c decreases initially, with the increase of temperature, reaching a minimum at 353 K and then gradually increases. It means that at high temperature region the large E applied tends to realign more freezing domains. Figure 4(b) presents the dielectric characteristics of the examined crystal sample as a function of temperature at various frequencies. The SBN61: Cr crystal exhibits very broad and frequency-dependent dielectric anomaly.

The domain walls in ferroelectric crystals have considerable influence on the value of their dielectric permittivity. It is reported that the domain walls contribute to the dielectric permittivity proportionally to their total surface [17]. One can presume that a weak measuring ac-field in dielectric measurements did not affect the domain configuration but creates nuclei on the domain walls. The high field characteristics correlates well with low field response of SBN61:Cr sample under much smaller electric fields (0.05 kVcm^{-1}). The maximum value of P_r was observed in the vicinity of 340 K. This appears to correspond to the sharp increase of dielectric permittivity near the phase transition. Associated with the dielectric peak, we also observed a minimum in the E_c at ~ 353 K. We speculate that the compositional disorder in SBN61:Cr crystal sample could play an important role on the phase transition broadening, assuming the formation of polar regions with locally different Curie temperature. It was reported that the Curie temperature for a ferroelectric transition depends on the material composition [18]. Note that, nanopolar structure and local ferroelectricity has been revealed by PFM technique well beyond the phase transition temperature in SBN:61 crystals [10].

4. Conclusions

Visualization of the domain structure using NLC method can be used to check the quality of the sample, for selecting homogeneous crystalline samples with an uniform distribution of the domain nuclei. We have shown that Cr doping deteriorates SBN crystals homogeneity, as evidenced by spatially inhomogeneous nucleation process. The observed broad dielectric

anomalies have a strong link to configuration of the ferroelectric microdomains and may reflect an internal disorder of SBN61:Cr crystal.

References

1. R. R. Neurgaonkar, J. R. Oliver, W. K. Cory, L. E. Cross, and D. Viehlan, *Ferroelectrics* **160**, 265 (1994).
2. J. Dec, W. Kleemann, Th. Woike, and R. Pankrath, *Eur. Phys. J.* **B14**, 627 (2000).
3. W. Kleemann, *J. Mater. Sci.* **41**, 129 (2006).
4. M. Venet, J. S. Guerra, I. A. Santos, J. A. Eiras, and D. Garcia, *J. Phys. Condens. Matter* **19**, 026207 (2007).
5. A. M. Glass, *J. Appl. Phys.* **40**, 4699 (1969).
6. T. Woike, U. Dörfler, L. Tsankov, G. Weckwerth, D. Wolf, M. Wöhlecke, T. Granzow, R. Pankrath, M. Imlau, and W. Kleemann, *Appl. Phys.* **B72**, 661 (2001).
7. T. Volk, L. Ivleva, P. Lykov, N. Polozkov, V. Salobutin, R. Pankrath R, and M. Woehlecke, *Optical Materials* **81**, 179 (2001).
8. P. Lehnen, W. Kleemann, Th. Woike, and R. Pankrath, *Phys. Rev.* **B64**, 224109 (2001).
9. V. V. Shvartsman, W. Kleemann, T. Łukasiewicz, and J. Dec, *Phys. Rev. B* **77**, 054105 (2008).
10. X. Y. Liu, Y. M. Liu, S. Takekawa, K. Kitamura, F. S. Ohuchi, and J. Y. Li, *J. Appl. Phys.* **106**, 124106 (2009).
11. R. F. Gainutdinov, T. R. Volk, O. A. Lysova, I. I. Razgonov, A. L. Tolstikhina, and L. I. Ivleva, *Appl. Phys.* **B95**, 505 (2009).
12. K. Matyjasek, K. Wolska, S. M. Kaczmarek, J. Subocz, and L. Ivleva, *Appl. Phys. B*, **106**, 143 (2012).
13. P. B. Jamieson, S. C. Abrahams, and J. L. Bernstein, *J. Chem. Phys.* **48**, 5048 (1968).
14. L. I. Ivleva, N. V. Bogodaev, N. M. Polozkov, and V. V. Osiko, *Opt. Mater.* **4**, 168 (1995).
15. N. A. Tikhomirova, L. J. Dontsova, S. A. Pikin, and L. A. Shuvalov, *JETP Lett.* **29**, 34 (1979).
16. M. E. Lines and A. M. Glass, *Principles and Applications of Ferroelectrics and Related Materials*. Oxford University Press: Oxford (2001).
17. J. Fousek and V. Janoušek, *Phys. Stat. Sol.* **13**, 195 (1966).
18. C. Perrin, N. Menguy, O. Bidault, C. Y. Zahra, A.-M. Zahr, C. Caranoni, B. Hilczer, and A. Stepanov, *J. Phys.: Condens. Matter* **13**, 10231 (2001).