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Influence of the domain structure on piezoelectric and dielectric properties of relaxor SBN single crystals

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Abstract. We have studied the influence of initial domain structure on piezoelectric and dielectric properties of $Sr_{0.61}Ba_{0.39}Nb_2O_6$ single crystals slightly doped with Ce and Ni. Initial domain structure was created by zero-field cooling, in-field cooling, and partial switching. The difference in the frequency dependences of macroscopic piezoelectric response and temperature dependences of dielectric permittivity for various initial domain structures was revealed.

1. Introduction

The formation of stable ferroelectric micro- and nanodomain structures with proper parameters for improving piezoelectric, pyroelectric, and dielectric properties is the main direction of the domain engineering. At present, a wide class of coherent light sources based on quasi-phase matching is developed, which allowed creating a qualitatively new direction in the fabrication of electro-optical and nonlinear optical devices [1,2].

It was shown that the formation of domain structure with controlled parameters by domain engineering technique improves significantly the piezoelectric properties [3,4]. In tetragonal barium titanate crystals, the piezoelectric properties were improved due to decreasing domain sizes (increasing domain wall density) [5,6]. The reduction of the domain wall mobility in PZT ceramics was used for creation of the piezoelectric transducers, whereas its increasing was used for manufacturing of precise electromechanical drives [7]. Thus, the domain wall engineering, aimed at creating the maximum possible density of the domain walls of a certain type and orientation, presents significant importance. Based on the observed increase of the dielectric constant in barium titanate ceramics with an average domain size of about 150 nm, it was suggested that at such domain sizes the highest values of piezoelectric coefficients could be obtained [8].

Uniaxial relaxor ferroelectric strontium barium niobate ($Sr_xBa_{1-x}Nb_2O_6$, SBN) is considered as a promising material for domain engineering due to its prominent nonlinear-optical and electro-optical properties along with sufficiently high piezoelectric coefficients [9].

Two alternative theoretical approaches were used for interpretation of the diffused phase transition in relaxor ferroelectrics [10-13]. The relaxor phase is considered: (1) as a dipole-glass state or (2) as a nanoscale mixture of polar nanoregions and nonpolar matrix [14].

In this work, the relaxor phase is considered as a composite consisting of ferroelectric (polar) and paraelectric (nonpolar) nanoregions. Such an approach was supported by high-resolution transmission electron microscopy (HRTEM) [15] and piezoresponse force microscopy (PFM) [16,17]. HRTEM experiments show that the heterophase structure of the relaxor state consists of polar nanoregions with diameters down to 10 nm embedded in a nonpolar matrix [15]. Using PFM, it was observed that in SBN polar nanoregions with opposite polarization directions and nonpolar regions coexisted at the temperature close to the dielectric permittivity maximum [16,17].

It was shown that the nonpolar inclusions influenced integral characteristics of relaxor ferroelectrics. For example, in relaxor PLZT ceramics [18], PZN-PT [19] and SBN [20] single crystals below freezing temperature conventional hysteresis loops were observed, while at higher temperatures double hysteresis loops emerged. It was demonstrated that the dielectric permittivity in PZN-PT single crystals after zero-field and in-field cooling were different [19].

In this paper, we report the experimental findings of investigation of the dielectric properties and macroscopic piezoelectric response in relaxor SBN61 single crystals doped by Ce and Ni with domain structures prepared by different methods.

2. Materials and methods

The studied samples represented plates doped by Ce (0.004 wt.% CeO_2, SBN:Ce) and Ni (0.05 wt% Ni_2O_3, SBN:Ni) $Sr_{0.61}Ba_{0.39}Nb_2O_6$ single crystals grown by modified Stepanov technique [21] in the Institute of General Physics of RAS (Moscow, Russia). The 0.6-mm-thick plates were cut normally to the polar axis and carefully polished. The Cr electrodes were deposited on the sample surfaces.

Three types of the domain states created by various methods have been studied: (1) zero-field cooling (ZFC), (2) in-field cooling (FC), and (3) partial switching (PS). ZFC represent slow cooling from 200°C to the room temperature without application of electric field. The domain structure formed after ZFC consists of nanodomains with random orientation of spontaneous polarization [16,17]. FC procedure was achieved by slow cooling from 200°C to the room temperature under constant external field 600 V/mm [22]. Mostly single-domain state with residual nanodomains was formed after FC. The PS was realized in linearly increasing electric field rising for 300V/(mm·s) until the switching current maximum. The micro-size domain structure with residual nanodomains appeared after PS [23-25].

The temperature control system LINKAM THMSE-600 was used for realization of the proper heating/cooling regime. The heating/cooling rate about 1-5 K/min was used. The dielectric measurements in frequency range from 10 Hz to 10 kHz were carried out in the samples with different domain structures using LCR-meter 2B-1 (Russia) during heating from 25 to 150°C and subsequent cooling. A standard scheme of single-beam Michelson homodyne interferometer was used for measurements of the piezoelectric coefficient at room temperature in frequency range from 70 Hz to 15 kHz [26]. Detailed description of the experimental setup can be found elsewhere [26].

3. Experimental

3.1. Dielectric permittivity

The temperature dependences of the dielectric permittivity at 10 Hz measured during cooling and heating of SBN:Ce and SBN:Ni samples prepared by ZFC, FC, and PS are presented in Figure 1.

It was shown that dielectric permittivity at low temperatures was higher for samples treated by ZFC than by FC and the difference increased with heating (Fig. 2a). This difference is due to a contribution of the charged domain walls to the dielectric response. It was demonstrated earlier that the complex quasi-regular three-dimensional self-similar nanodomain labyrinth with a large number of charged domain walls formed in SBN single crystals treated by ZFC contribute significantly to the dielectric response [16,17]. On the contrary, the small domain wall area in the samples treated by FC decreased the dielectric permittivity. Decrease of depolarization fields and domain wall mobility during heating led to an apparent increase of the dielectric permittivity difference between the samples treated by ZFC and FC (Fig. 2a).

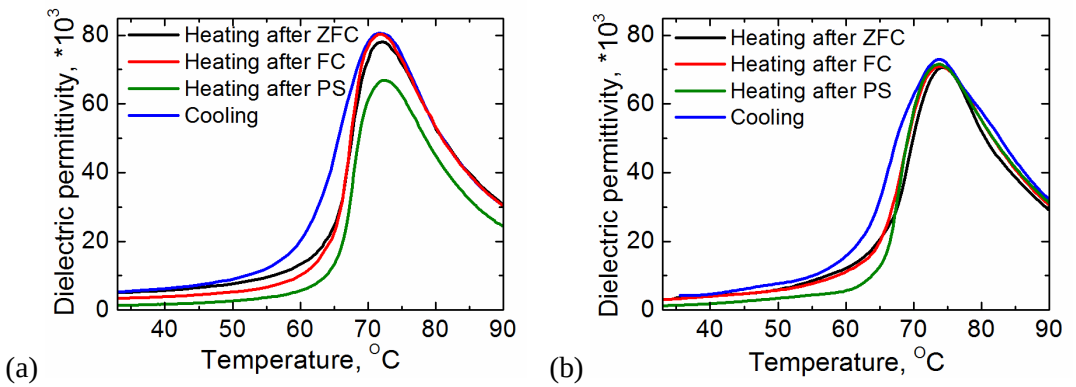

Figure 1. The temperature dependences of dielectric permittivity during cooling and heating (a) SBN:Ce and (b) SBN:Ni with different domain structures. Frequency 10 Hz.

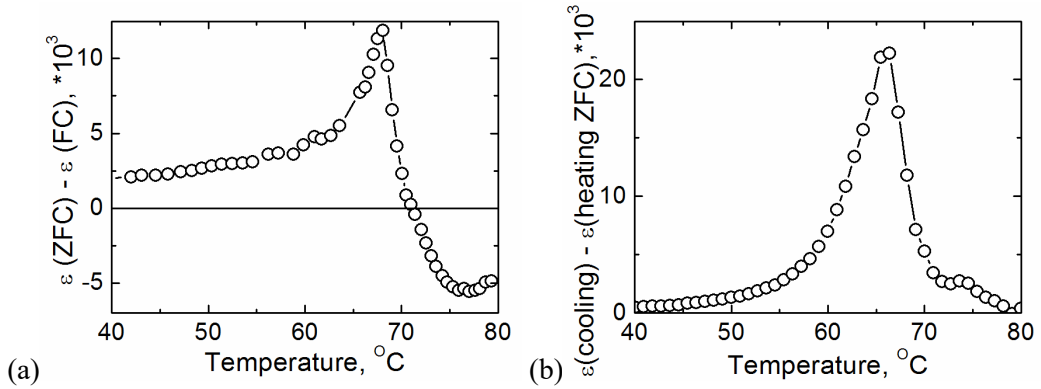

Figure 2. The temperature dependences of difference between dielectric permittivity (a) during heating after ZFC and FC and (b) during cooling and heating after ZFC. SBN:Ce. Frequency 10 Hz.

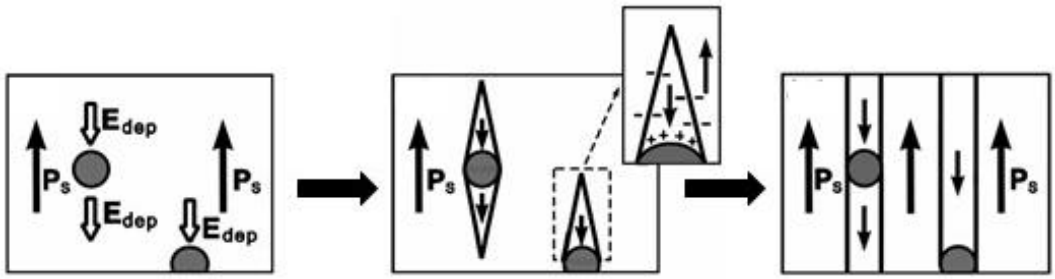

Figure 3. Schematic representation of the evolution of the domain structure samples treated by ZFC during heating. Black arrows - P_s direction, white arrows - E_{dep} direction, dark grey circles - nonpolar inclusions.

At a certain temperature, the domain contribution reached its maximum and, upon further heating, began to decrease sharply (Fig. 2a). This behaviour can be attributed to the appearance of insulated nanometre nonpolar inclusions in ferroelectric multidomain matrix [16,17]. The bound charges arising at the phase boundaries created the depolarization fields (Fig. 3) in the samples treated by FC, which could be compensated only by slow bulk screening process [27]. These fields led to a partial switching and formation of nanodomain structure with charged domain walls in the vicinity of nonpolar inclusions. Charged domain walls may contribute to the dielectric constant [27,28] due to an abnormally large concentration of steps that were mobile even in weak electric fields used for dielectric measurements.

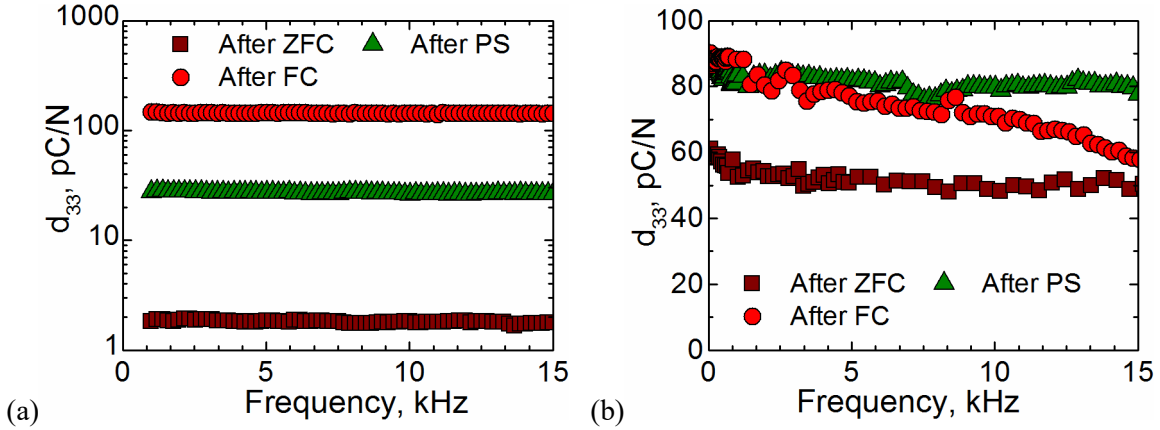

Figure 4. The frequency dependences of integrated piezoelectric response of (a) SBN:Ce and (b) SBN:Ni with different domain structures.

At low temperatures, dielectric permittivity values measured during cooling and heating for samples treated by ZFC were the same due to a small mobility of the domain walls at these temperatures. During heating, the difference between dielectric permittivity values observed (Fig. 2b). The dielectric permittivity is larger under cooling than under heating due to a higher mobility of the "fresh" domain walls formed during cooling as compared with the screened walls formed as a result of ZFC.

The dielectric permittivity for the sample prepared by PS at low temperatures is less than for those processed by other methods. It can be attributed to the lower concentration of the charged domain walls.

3.2. Piezoelectric coefficient

The frequency dependences of the integrated piezoelectric response of SBN single crystals with different domain structure measured at room temperature (below freezing temperature for the studied samples) are presented in Figure 4. Slight decrease of the macroscopic piezoelectric response with increasing frequency was seen for crystals prepared by all methods. The most pronounced decrease was obtained in SBN:Ni single crystal treated by FC.

The largest piezoelectric response was obtained in the samples treated by FC (146 pm/V (SBN:Ce) and 89 pm/V (SBN:Ni) at 1 kHz). The smallest piezoelectric response was measured in the samples treated by ZFC (1.8 pC/N (SBN:Ce) and 55 pC/N (SBN:Ni) at frequency 1 kHz). The piezoelectric response in the samples treated by PS was 28 pm/V for SBN:Ce and 80 pm/V for SBN:Ni (at frequency 1 kHz). This behaviour can be attributed to the contribution of the crystal polarization P to the piezoelectric response of the system. The polarization of the crystals treated by ZFC is rather small, while in the crystals treated by FC its value is much larger.

4. Conclusion

The influence of the initial domain structure on piezoelectric response and dielectric permittivity has been studied in SBN single crystals slightly doped by Ce and Ni. The initial domain structure was formed by three methods: (1) zero-field cooling, (2) in-field cooling, and (3) partial switching. The difference in the temperature dependences of dielectric permittivity for various initial domain structures has been attributed to the existence of charged domain walls in the samples with multi-domain structure and appearance of the non-polar inclusions during heating above the freezing temperature. The difference in the macroscopic piezoelectric response for various initial domain structures can be attributed to the contribution of the crystal polarization to the piezoelectric response.

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