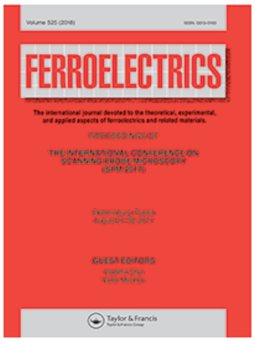

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Local switching in SBN:Ni single crystals with various initial domain states

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ABSTRACT
We present the results of experimental study of the local switching in single crystals of relaxor ferroelectric Ni-doped strontium barium niobate with various initial domain states using conductive tip of scanning probe microscope. We investigated the domain growth in as-grown domain structure and initial domain states created by: (i) scanning by biased tip, (ii) in-field cooling, and (iii) scanning by electron beam. The dependences of effective domain radius on the voltage and pulse duration were derived. The shape factor, domain wall mobility, and start voltage were revealed. The obtained results can be used for the domain engineering in ferroelectrics.

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Relaxor ferroelectrics; piezoresponse force microscopy; strontium barium niobate; polarization reversal; local switching; domain structure

1. Introduction

Local polarization reversal in ferroelectrics using biased tip of scanning probe microscope (SPM) has been studied intensively due to applications of the ferroelectrics with engineered domain structures in optoelectronics and photonics [1–3]. Fabrication of the light frequency conversion devices based on periodically poled ferroelectric crystals [4] and piezoelectric components with improved characteristics [5] are very promising. A number of papers has been devoted to the investigation of the local polarization reversal in various bulk ferroelectrics, such as lithium niobate [6–9], but domain kinetics in relaxor ferroelectrics is still poorly studied.

Strontium barium niobate ($Sr_xBa_{1-x}Nb_2O_6$, abbreviated as SBN100x) is uniaxial relaxor ferroelectric, which is considered to be a prospective material for domain engineering. Prominent nonlinear-optical and electro-optical properties of SBN together with high piezoelectric coefficients [10] opened up new horizons for commercialization of the domain patterned crystals. SBN crystals are very attractive for SPM recording due to low coercive fields [10]. Furthermore, it is considered that the initial nanoscale domain structure appeared due to intrinsic nanoscale compositional inhomogeneity in relaxor ferroelectrics hampers the creation of tailored domain patterns in SBN [11].

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The first high-resolution studies using SPM allowed visualizing initial static domain patterns on the polar surface of SBN crystals [11–13]. In particular, piezoresponse force microscopy (PFM) revealed maze-type domain structures with characteristic sizes of about a hundred nanometers [12] in thermally depolarized crystals at room temperature, as well as their evolution at elevated temperatures [13]. The micro- and nanodomain structures formed during polarization reversal in uniform electric field were also studied using PFM [14].

For the first time, the possibility of the recording of domains on the polar surface of SBN61 crystals in the field of atomic force microscope was demonstrated in Ref. [15]. The field and exposure characteristics of the SPM domain recording were investigated in pure and Nd, Ce, and Ni doped SBN61 crystals [15–18]. The relaxation of the polarized state created by application of a local electric field using a conductive SPM tip was investigated in SBN crystals at various temperatures [19].

In this paper, we present the results of comparative experimental study of the local polarization switching in Ni-doped SBN61 single crystals with as-grown domain structure and having initial domain states created by several methods including: (i) scanning by biased SPM tip, (ii) in-field cooling, and (iii) scanning by electron beam.

2. Experiment

The studied samples represented plates of doped by Ni (0.05 wt.% Ni_2O_3) strontium barium niobate single crystals $Sr_{0.61}Ba_{0.39}Nb_2O_6$ (SBN:Ni) grown by modified Stepanov technique [20] in the Institute of General Physics of the Russian Academy of Sciences (Moscow, Russia). The 0.78-mm-thick plates were cut normally to the polar axis and carefully polished with averaged roughness about 1 nm.

PFM measurements were realized using built-in electronics of NTEGRA Aura (NT-MDT SI, Russia) with silicon NSC-18 tips (MikroMash, Estonia). The tips had platinum conductive coating and typical radius of tip curvature of about 25 nm. The AC modulation voltage with amplitude $U_{mod} = 2{\div}4$ V and frequency $f_{mod} = 440$ kHz (just below the tip resonance) was applied between the conductive tip and bottom electrode to invoke the piezoelectric response [21]. Local switching was performed using external NI-6251 multifunction Data Acquisition board (DAQ, National Instruments, USA) and high-voltage amplifier Trek-677B (TREK Inc., USA). All measurements were performed in dry atmosphere (relative humidity below 5%) provided by constant dry airflow through the microscope chamber to avoid the influence of the thin surface water layer [21].

The domain patterns were created by local application of the electric field using the SPM tip. The rectangular field pulses were applied in square pattern of 9 dots and period 3.3 μm with pulse duration ranged from 10 ms to 100 s and voltage – from 10 V to 200 V. The domain formation was performed at room temperature, which is below freezing temperature for the studied crystal composition [22].

Four types of the domain states created by various methods have been used for the investigations: (i) as-grown domain structure, (ii) after scanning by biased SPM tip, (iii) after in-field cooling, and (iv) after electron beam scanning. As-grown structure appeared after crystal growth and sample polishing without application of an electric field. Biased SPM tip scanning domain was created by continuous scanning of the area 10×10 μm^2 under external voltage -150 V and recording time 17 min. In-field cooling state was formed by slow (about 5 K/min) in-field cooling of the sample supplied with silver paste electrodes under constant external field 300 V/mm from about 200°C to room temperature [23].

Electron beam scanning domain state was created by continuous scanning of the electron beam with the diameter about 10 nm with accelerating voltage 10 kV, e-beam current 1 nA, and irradiation doses 50 μC/cm^2 [24]. Before irradiation, 2.5-μm-thick layer of negative electron-beam resist AZ nLof 2020 (MicroChemicals GmbH, Germany) was deposited by spin coating method on irradiated sample surface and Cu electrode on the opposite sample surface [25]. The scanning electron microscope (Auriga CrossBeam workstation, Carl Zeiss NTS, Germany) with Schottky field emission gun equipped with the e-beam lithography (EBL) system (Elphy Multibeam, Raith GmbH, Germany) was used for e-beam scanning. The exposure parameters and e-beam positioning were controlled by EBL system. The area mood exposure was done by meander-scan coverage of the desired area.

3. Results and discussion

We have analyzed the maze-type as-grown domains as self-assembled patterns (by fractal dimension) and as quasiperiodic structure (by average correlation radius) (Fig. 1). The fractal dimension $D = 2.8$ was calculated by box counting method [26]. The obtained D value above 2.5 indicates that formation of as-grown domain structure is a correlated process. The average correlation radius about 50 nm has been extracted from the fitting of the autocorrelation function [27, 28].

The domain patterns formed by SPM local switching in the samples: with as-grown domain structure, after scanning by electron beam, and after scanning by biased SPM tip are presented in Figure 2.

The domain area and the domain wall length were measured using the software SPIP 4.8.2 (Image Metrology A/S, Denmark). The shape of the created domains has been characterized by the shape factor $K = L/8R$, where L is the domain wall length, $R = (A/\pi)^{0.5}$ is the effective domain radius, and A is the domain area. For circular domain $K = 1$.

It is clear that the minimal value of K corresponds to switching from the single domain state. We propose to use the shape factor of domains formed by local switching for

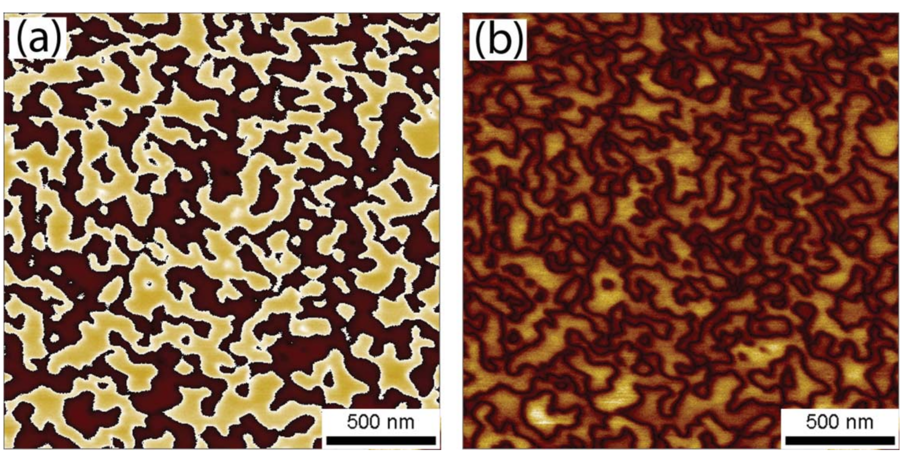

Figure 1. Typical PFM images of as-grown domain structure: (a) phase, (b) amplitude.

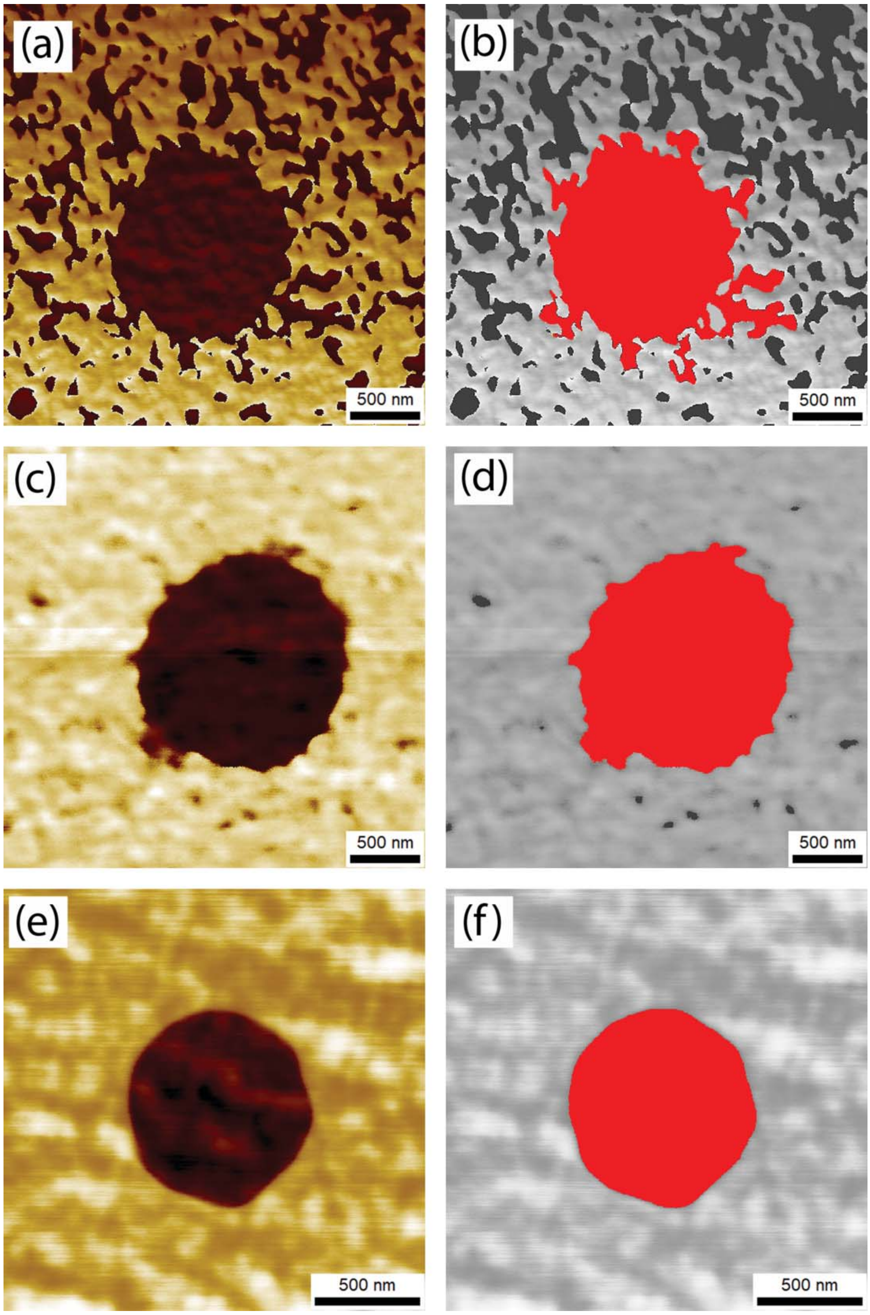

Figure 2. The PFM images of domains formed by local switching in the samples with: (a) as-grown domain structure, (c) after scanning by electron beam, (e) after scanning by biased SPM tip; (b), (d), (f) marked domains. $U_{sw} = 150$ V. $t_{sw} = 1$ s. Phase signal.

quantitative characterization of the domain states created by various methods. The averaged K values for the samples with various initial domain states are presented in Table 1. According to the obtained results, the in-field cooling is the most effective method for creating single domain state in SBN:Ni.

Table 1. The shape factor (*K*); coefficient proportional to the domain wall mobility (a); start voltage (U_{st}); deviation from the value of the start voltage (ΔU_{st}) in the samples with different initial states.

Initial state	*K*	*a*, nm/(s*V)	U_{st}, V	ΔU_{st}, V
As-grown	2.2 ± 0.3	0.082	−24	0
SPM tip scanning	1.6 ± 0.2	0.070	−17	7
In-field cooling	1.4 ± 0.2	0.063	−99	−75
Electron beam	1.5 ± 0.2	0.135	5	29

The dependences of the effective domain radius on the pulse duration with voltage 150 V and duration ranging from 10 ms to 100 s in the samples with different initial states are shown in Figure 3. The largest value of the effective domain radius obtained after electron beam scanning can be attributed to the presence of the bias produced by e-beam irradiation, which facilitated the polarization reversal. The least effective domain radius obtained after scanning by biased SPM tip is likely to originate from the injection of a charge of the opposite sign, which hampered the domain growth.

The voltage dependences of the effective domain radius in the samples with as-grown domain structure, after scanning by biased SPM tip, after in-field cooling, and after electron beam scanning are displayed in Figure 4.

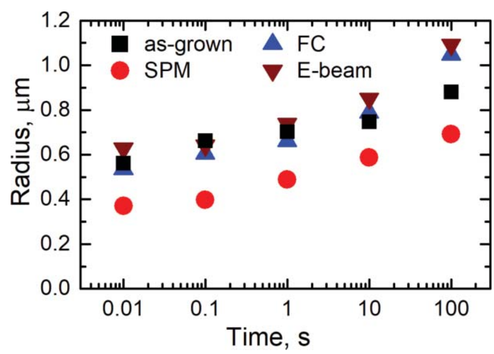

Figure 3. The dependences of the effective domain radius on pulse duration in the samples with as-grown domain structure (■), after biased SPM tip scanning (●), after in-field cooling (▲), and after electron beam scanning (▼). U_{sw} = 150 V.

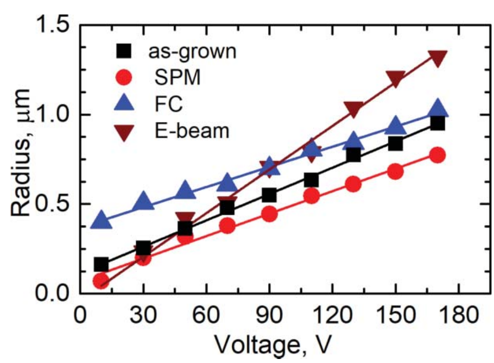

Figure 4. The dependences of the effective domain radius on voltage in samples with as-grown domain structure (■), after biased SPM tip scanning (●), after in-field cooling (▲), and after electron beam scanning (▼). t_{sw} = 60 s.

The obtained voltage dependences of the effective domain radius have been fitted by Equation 1 [21, 29]. The obtained parameters are given in Table 1.

$$R(U) = v(U)\ t_p = a\ (U - U_{st})\ t_p, \tag{1}$$

where v is the domain wall velocity, t_p is the pulse duration, a is proportional to the domain wall mobility, U_{st} is the start voltage. $U_{st} = U_{th} + U_b$, where U_{th} is the threshold voltage, U_b is the internal bias.

The values of the domain wall mobility after biased SPM tip scanning and after in-field cooling are approximately the same and are slightly less than those in as-grown domain structure. This fact can be attributed to internal bias appeared as a result of both treatments. The highest mobility observed after electron beam scanning can testify that this treatment decreased the pinning on the domain walls. The mechanism of this decrease is unclear at this moment and will be studied in the future work.

The start voltage includes threshold voltage and internal bias. Thus, we calculated the deviation from the start voltage after various treatments of the as-grown domain structure ΔU_{st}. After scanning by biased tip, ΔU_{st} is the least. However, after in-field cooling, ΔU_{st} is the highest and, surprisingly, has the opposite sign. This difference can be attributed to the appearance of the internal bias field after used sample treatments.

4. Conclusion

The influence of the initial domain state on the parameters of isolated domains created by local switching using biased conductive tip of scanning probe microscope was studied in Ni-doped SBN single crystals with as-grown domain structure and domain states created by scanning with biased SPM tip and electron beam, and by in-field cooling. The dependences of the effective domain radius on voltage and pulse duration were analyzed. The geometry of the as-grown domain structure was characterized by fractal dimension ($D = 2.8$) and average correlation radius of about 50 nm. We proposed to use the shape factor of domains formed by local switching for quantitative characterization of the domain states created by various methods. According to the obtained results, the in-field cooling was the most effective method for the creation of a single domain state. The largest value of the effective domain radius obtained after electron beam scanning was attributed to the presence of the internal bias field produced by e-beam irradiation, which facilitated the polarization reversal. The least effective domain radius obtained after scanning by biased SPM tip was attributed to injection of a charge of the opposite sign which hampered the domain growth. The values of the domain wall mobility after biased SPM tip scanning and after in-field cooling were approximately the same being slightly less than those for as-grown domain structure. This fact was attributed to the internal bias field appeared as a result of both treatments. The highest mobility observed after electron beam scanning could be a result of the reduced pinning of the domain walls after this treatment. The deviation from the start voltage after studied treatment from as-grown domain structure could be attributed to the appearance of internal biases after all the studied treatments. The obtained results can be used for domain and domain wall engineering in ferroelectrics.

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