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Tip-induced domain growth in the non-polar cuts of SBN:Ce single crystals

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Abstract. The local switching by conductive tip of scanning probe microscope was studied in the non-polar cuts of Ce-doped $Sr_{0.61}Ba_{0.39}Nb_2O_6$ single crystals after creation of the single-domain state. The switched domains possessed the egg-shaped heads and wedge-like tails. The dependences of lengths of the domain head and tail and width of the domain head on the voltage and pulse duration were derived. The start voltage for growth of the domain tail was revealed. The fast relaxation of the domain head and slow relaxation of the domain tail were observed. The model of the forward domain growth by step generation and kink motion was used for explanation of the experimental results. The obtained knowledge can be used for the domain engineering in ferroelectrics.

1. Introduction

The domain growth in ferroelectrics under the action of the local electric field produced by conductive tip of the scanning probe microscope (SPM) has been studied intensively due to applications of the ferroelectrics with engineered domain structures in optoelectronics and photonics [1-4]. Fabrication of the light frequency conversion devices based on periodically poled ferroelectric crystals [3] and piezoelectric components with improved characteristics [5] is very promising. The local switching by conductive SPM tip has been used for creation of the domain patterns in lithium niobate [6-9]. However, most papers to the date explore the lateral domain growth on the polar surfaces and have a lack of information about forward domain growth along polar direction due to some experimental difficulties.

Strontium barium niobate ($Sr_xBa_{1-x}Nb_2O_6$, SBN100x) is a uniaxial relaxor ferroelectric, which is considered to be a prospective material for domain engineering. Prominent nonlinear-optical and electro-optical properties of SBN [10] opened up new horizons for commercialization of the domain patterned crystals. Moreover, SBN crystals are very attractive for SPM recording due to their low coercive fields [10]. The initial domain state at room temperature in thermally depolarized SBN crystals represents maze-type domain structures with characteristic sizes of about a hundred nanometres, which was revealed using piezoresponse force microscopy (PFM) [11-14].

For the first time, the possibility of recording domains on the polar surface of SBN61 single crystals using conductive SPM tip was demonstrated in Ref. [15]. Afterward, the field and exposure characteristics of the SPM domain recording on the polar surface were investigated in pure and Nd, Ce, and Ni doped SBN61 single crystals [16-19]. The influence of the initial domain state on the sizes of the domains created by the biased conductive SPM tip was studied in Ni doped SBN single crystals [20]. The relaxation of the polarized state created by conductive SPM tip was investigated in Z-cut SBN crystals at various temperatures [21].

The formation of the microdomain patterns on the nonpolar surface using conductive SPM tip was investigated in SBN61 single crystals with initial multi-domain and single-domain states [18,22].

In this paper, we present the results of experimental study of tip-induced domain growth and its relaxation in the non-polar cuts of Ce-doped SBN61 single crystals after creation of single-domain state.

2. Experimental

The studied samples represented plates of $Sr_{0.61}Ba_{0.39}Nb_2O_6$ single crystals doped by Ce (0.004 wt.% CeO_2) (SBN:Ce) grown by modified Stepanov technique [23] in the Institute of General Physics of the Russian Academy of Sciences (Moscow, Russia). The 2-mm-thick plates were cut normally to the nonpolar axis (X-cut) and carefully polished.

The single domain state was created in the studied samples before measurements using the cyclic switching at elevated temperature [24]. The sinusoidal electric field with amplitude 300 V/mm and frequency 1 Hz was applied along the polar axis during 100 s at 50°C. The switching was performed using NI-6251 multifunctional Data Acquisition board (DAQ, National Instruments, USA) and high-voltage amplifier Trek-677B (TREK Inc., USA). The silver paste electrodes were deposited for electric field application.

The PFM measurements were realized using scanning probe microscope Asylum MFP-3D (Oxford Instruments, UK) with silicon NSC-18 tips (MikroMasch, Estonia). The domain structure was created by application of dc voltage pulses perpendicular to nonpolar axis with amplitudes from 10 to 30 V and duration from 0.5 to 16 s via conductive SPM tip. The tip had a platinum conductive coating and a typical curvature radius of about 25 nm. The domains were formed at room temperature, which was below freezing temperature for the studied crystal composition [25,26].

3. Results and discussion

The domains formed by local switching (amplitude 20 V, duration from 0.5 to 16 s) on the nonpolar surface of the SBN are presented in Figure 1a. All created domains possessed the egg-shaped head and wedge-like tail.

The ZX distribution of the electric field, produced by SPM tip in contact with the nonpolar surface of SBN:Ce single crystals was calculated for the our experimental conditions (Fig. 2a) [27-29]. It is seen that the shape of the created domain head correlates with electric field distribution (Fig. 2b).

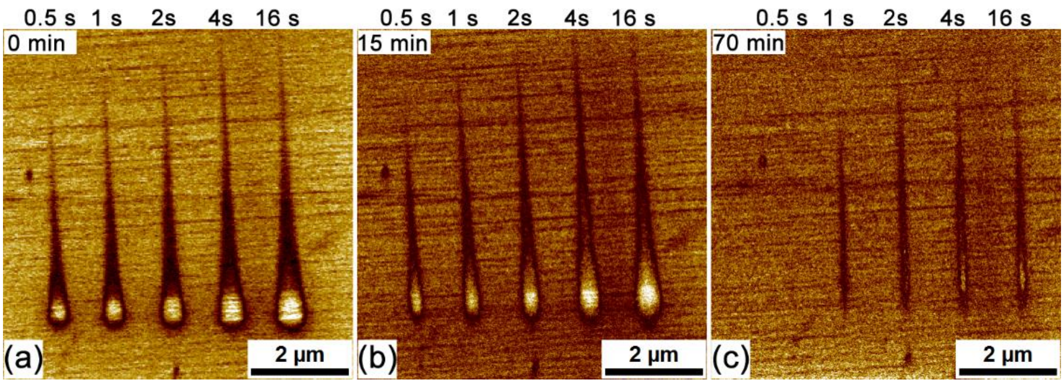

Figure 1. PFM images of domains formed on SBN nonpolar surface: (a) immediately after SPM local switching, (b) 15 min after switching, (c) 70 min after switching. U_{sw} = 20 V. t_{sw} = 0.5-16 s. Amplitude signal.

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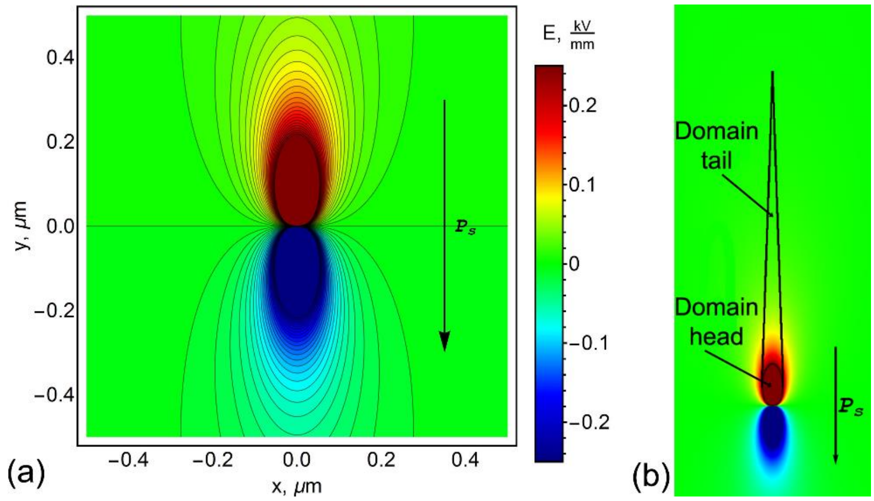

Figure 2. (a) Contour maps of calculated electric field E_z across the ZX plane (y = 0 nm), produced by SPM tip in contact with the nonpolar surface of SBN:Ce (ε_c = 2800, ε_a = 450, r_{tip} = 25 nm, U_{sw} = 30 V). (b) Domain growth scheme.

The maximum width of the created domains corresponded to the width of the domain head (Fig. 3a,d). The length of the domain head was extracted from the measured head width according to the calculated spatial distribution of the polar component of the electric field (Fig. 3b,e). The domain lengths were divided in two parts: 1) length of the domain head and 2) length of the domain tail. So, the length of the domain tail was equal to the difference between the domain length and the domain head length (Fig. 3c,f). The growth of the domain tail in the area with negligible value of the applied field was attributed to kink motion under the field produced by neighbouring kinks [30]. Thus, we observed the forward domain growth by step generation [31] and kink motion.

All the domain sizes (lengths of the domain head and tail and width of the domain head) increased with pulse duration and voltage. The observed linear dependence of the domain sizes on time logarithm (Fig. 3a-c) and linear voltage dependence (Fig. 3d-f) are typical for ferroelectrics [29].

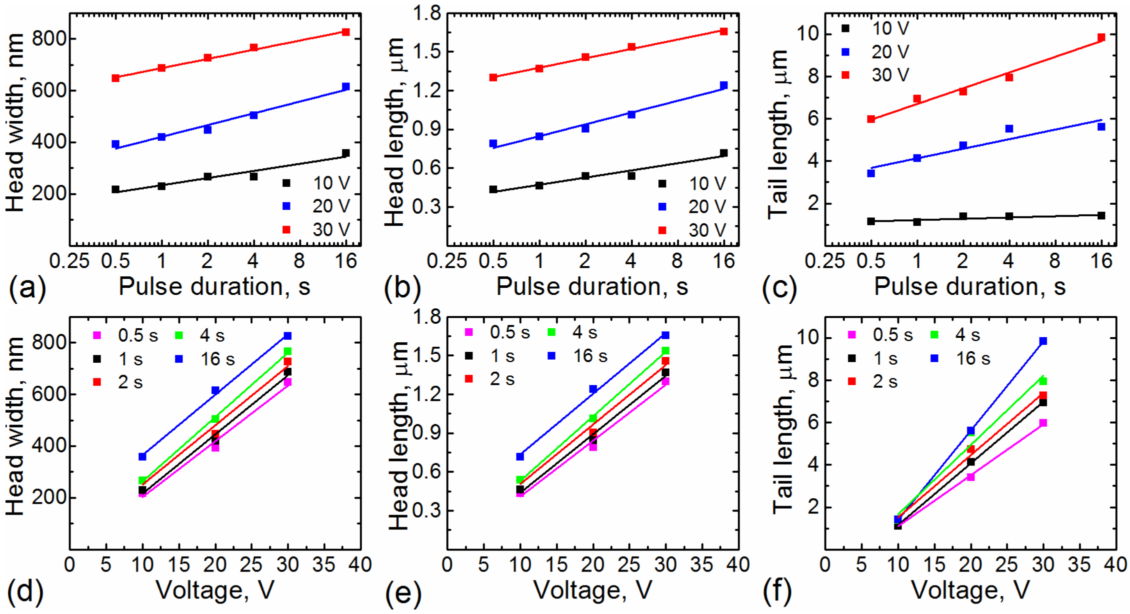

Figure 3. The dependences of domain head width (a, d), domain head length (b, e), and domain tail length (c, f) on pulse duration (a, b, c) and voltage (d, e, f).

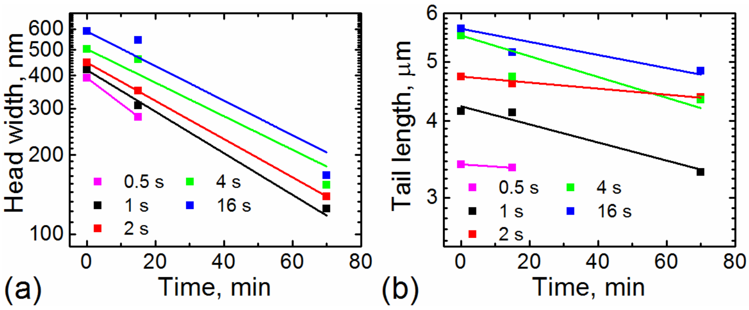

Figure 4. The relaxation of (a) domain head width and (b) domain tail length with time. Logarithmic scale on the Y axis. U_{sw} = 20 V. t_{sw} = 0.5-16 s.

The obtained voltage dependences of the domain tail length were fitted by linear function [32,33] $L_t(U) = a\ (U\text{-}U_{st})$, where a is proportional to the domain wall mobility, U_{st} is the start voltage. The obtained start voltage for the tail length ranged from 5 to 6.5 V.

The changes in created domains were revealed over time. The domain images created by application of the pulse with amplitude 20 V were obtained just after local switching and 15 and 70 minutes later (Fig. 1). The domain created with pulse duration 0.5 s disappeared on the third scan. The length of the other domains decreased slightly, while their width decreased significantly.

The time relaxation of the domain sizes was investigated for created domains. The time dependences of the domain head width (Fig. 4a) and domain tail length (Fig. 4b) were fitted by exponential function. The fast time relaxation of the domain head width and slow relaxation for the domain tail length were observed. The relaxation time for domain head width was in the range from 50 to 70 minutes; for domain tail length, it was in the range from 300 to 800 minutes. The fast relaxation of the domain head can be attributed to the high concentration of the kinks near domain head. The slow decrease of the length of domain tails can be attributed to electrostatic interaction of the approaching walls [3,34].

4. Conclusion

We studied the local switching by conductive tip of scanning probe microscope on the non-polar surface of Ce-doped $Sr_{0.61}Ba_{0.39}Nb_2O_6$ single crystals after creation of the single-domain state. It was found that created domains consisted of egg-shaped heads and wedge-like tails. The shape of the domain head correlated with spatial distribution of the polar component of electric field, produced by SPM tip in contact with the non-polar surface of SBN:Ce single crystals. The dependences of domain sizes (lengths of the domain head and tail and width of the domain head) on the voltage and pulse duration were derived. The linear dependence of domain sizes on time logarithm and linear voltage dependence were observed. The start voltage was revealed for growth of the domain tail. The fast relaxation of the domain head and slow relaxation of the domain tail were revealed. The obtained knowledge can be applied for improvement of the domain engineering methods in SBN crystals.

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