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Domain Switching by Electron Beam Irradiation in SBN61:Ce Single Crystals Covered by Dielectric Layer

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***Abstract*—Formation of domain structure by electron beam irradiation in thermally depolarized Ce-doped strontium barium niobate single crystals with free surface and surface covered by dielectric layer has been studied. The dependences of the domain sizes and domain depth on the irradiated dose have been measured. The circular shape of isolated domains was obtained. Isotropic domain growth was attributed to step generation at the wall as a result of merging with residual nanodomains existed after thermal depolarization. The linear dose dependence of the switched area was attributed to screening of the depolarization field by injected charge. The electrostatic interaction of the approaching charged domain walls was revealed. The better quality of the domain patterns was achieved in the samples with electron localization in the dielectric layer. The obtained results can be applied for creation of the precise domain patterns with arbitrary orientation and shape in order to produce the nonlinear optical devices with improved characteristics.**

***Index Terms*— Čerenkov-type second-harmonic generation microscopy, confocal Raman microscopy, domain structure, electron beam irradiation, local switching, piezoresponse force microscopy, polarization reversal, relaxor ferroelectrics; strontium barium niobate**

I. Introduction

THE kinetics of the ferroelectric domains has been studied intensively due to applications of the ferroelectric crystals with engineered domain structures in optoelectronics and photonics [1,2], mostly by fabrication of the light frequency conversion devices based on periodically poled ferroelectric crystals [3]. The local switching by conductive tip of scanning probe microscope (SPM) and electron beam (e-beam) irradiation are widely used for creation of the precise domain structures in various ferroelectrics, such as lithium niobate [4-11], lithium tantalate [7,11], potassium titanyl phosphate [12,13].

Strontium barium niobate ($Sr_xBa_{1-x}Nb_2O_6$, abbreviated as SBN100x) is uniaxial relaxor ferroelectric, which can be considered as a prospective material for domain engineering. Prominent nonlinear-optical and electro-optical properties of SBN [14] opened up new horizons for commercialization of the domain patterned crystals. The maze-type domain structures with characteristic sizes of about a hundred nanometers was revealed in thermally depolarized crystals at room temperature using piezoresponse force microscopy (PFM) [15-17].

The local polarization reversal on the polar surface of SBN61 crystals in the field of the tip of SPM was demonstrated [18]. The field and exposure characteristics of the SPM domain recording were investigated in pure and Nd, Ce, and Ni doped SBN61 crystals [18-22]. The relaxation of the polarized state created by biased SPM tip was investigated in SBN crystals at various temperatures [23].

The possibility of domain switching of SBN single crystals using e-beam irradiation was demonstrated [22], but detailed studies were not carried out. Recently, technique of e-beam

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domain patterning assisted by surface electron resist layer allowing significant improvement of the domain pattern quality was successfully realized in MgO-doped LN [24,25], and proton exchanged waveguides based on LN single crystals [26]. It was shown that this method allowed reducing drastically the undesirable effect of the electron spreading over the irradiated surface, thus creating the periodical poling in 1-mm-thick MgO-doped LN crystalline plates [25,26].

In this paper, we present the results of investigation of the domain growth during local polarization switching by e-beam irradiation in Ce-doped SBN61 single crystals covered by resist layer.

II. Materials and Methods

The studied samples represented plates of doped by Ce (0.004 wt.% CeO_2) strontium barium niobate single crystals $Sr_{0.61}Ba_{0.39}Nb_2O_6$ (SBN:Ce) grown by modified Stepanov technique [27] in the Institute of General Physics of the Russian Academy of Sciences (Moscow, Russia). The 0.6-mm-thick plates were cut normally to the polar axis and carefully polished with averaged roughness about 1 nm. Before irradiation the samples were thermally depolarized by slow heating (rate about 2°C/min) up to 200°C and zero-field-cooling. Irradiation was carried out at room temperature, which is below freezing temperature for the studied crystal composition [28,29].

Before irradiation, 2.5-µm-thick layer of negative e-beam resist AZ nLOF 2020 (MicroChemicals GmbH, Germany) was deposited by spin coating method on irradiated surface and Au-Pd electrode by magnetron sputtering on the opposite polar surface [25]. The scanning electron microscope (Auriga CrossBeam workstation, Carl Zeiss NTS) with Schottky field emission gun equipped with the e-beam lithography (EBL) system (Elphy Multibeam, Raith GmbH) was used for e-beam domain patterning. The exposure parameters and e-beam positioning were controlled by EBL system. The irradiation pattern was specified by the Raith Nanosuite software. Two types of exposure regimes were used: (1) dot exposure - discrete irradiation by isolated dots with the distances between the points 7 and 100 µm; (2) stripe exposure - continuous irradiation by stripes with period 7 µm. The irradiation was performed at accelerating voltage of 10 kV and electron beam current (I) 1.5 nA. The irradiation doses (charge) for dot exposure (dot dose) were defined as $D_d = I*t$ and for stripe exposure (area dose) — as $D_A = (I*t)/A$, where t is exposure (d_{well}) time and A is area step size (defined as 40 nm^2).

The created domain patterns were imaged at the surface by PFM and in the bulk by confocal Raman microscopy (CRM) [30-33] and by Čerenkov-type second-harmonic generation microscopy (ČSHGM) [34,35] after removing of the resist layer and electrode.

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} = 2-4 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.

CRM imaging was done using Alpha 300AR (WITec GmbH, Germany). The Raman spectra were recorded in backscattering configuration at the room temperature using He-Ne laser (633 nm, 30 mW) as a pumping source. It was shown that integrated intensity of Raman lines around 593 cm^{-1}, corresponding to A_{1g} vibrational mode noticeably changes their frequency in the vicinity of the domain walls [30]. The estimated CRM lateral resolution was about 300 nm and resolution in Z direction – about 500 nm.

The images of the domain structures in the crystal bulk were obtained by ČSHGM [34,35] using the modified experimental setup based on NTEGRA Spectra (NT-MDT, Russia). The strong Čerenkov-type second-harmonic generation was obtained only when the focal spot is crossing a domain wall, which allows precise domain wall imaging [34,35]. The ČSHGM images of XY section were recorded with depth increments of 2.4 µm.

III. Results and Discussion

A. Dot Exposure

The dot exposure performed with dose ranged from 1 to 50 pC. The ensemble of isolated submicron-size domains appeared in the samples with free surface (Fig. 1 a), whereas the isolated circular domains were revealed in the samples covered by resist (Fig. 1 b). This fact can be attributed to the charge localization in the resist [25]. The circular shape of isolated domains (isotropic domain growth) is caused by step generation at the wall by merging with isolated residual nanodomains existed in the initial state after thermal depolarization.

The domain area was measured by analysis of PFM images

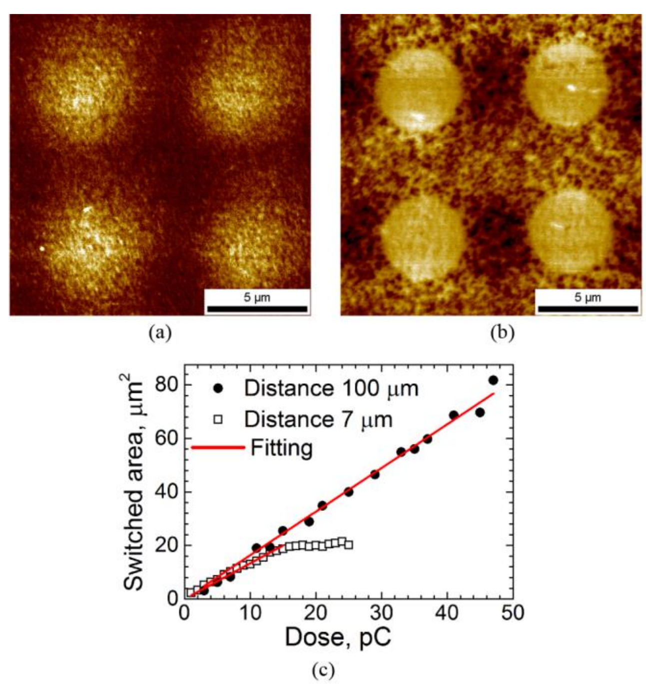

Fig. 1. PFM image of domain structure after dot exposure in the samples (a) with free surface and (b) with surface covered by resist. (c) Dose dependence of the switched domain area in the samples with resist layer. (a, b) Distance between dots 7 µm, dose 21 pC. Out-of-plane response. Phase signal.

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using the SPIP 4.8.2 software (Image Metrology A/S, Denmark). It was found that the domains area linearly increases with irradiation dose for distance between dots 100 µm, while the area of domains recorded with 7-µm-distance linearly increases with saturation at dose above 15 pC (Fig. 1 c).

The obtained dose dependence of the switched area can be attributed to screening of the depolarization field by injected electrons. The electron beam is an analog of external current for traditional switching by application of the field by means of electrodes. The observed domain growth retardation and saturation of the switched domain area for short period is due to the electrostatic interaction of the approaching charged domain walls.

The ČSHGM [34,35] was used for bulk imaging of the domains created by dot irradiation with distance between dots 7 µm (Fig. 2 a, b). The length of needle-like domain for dose 25 pC achieved 75 µm with aspect ratio about 30 (Fig. 2 c).

It is seen that at the images obtained close to the crystal polar surface the continuous optical contrast inside the created domains in addition to conventional contrast of the boundary of the switched area (domain wall) (Fig. 2 a). It has been found that this contrast decreased and completely disappeared with depth (Fig. 2 b). Since this imaging method is sensitive to the domain walls only [34,35], the observed results can be attributed to formation of needle-like nanodomains close to the polar surface (Fig. 2 a). The existence of nanodomains inside the micro-scale domains is confirmed by imaging of the surface relief of sample after etching in hydrofluoric acid using atomic force microscopy (AFM) (Fig. 2 d). The appearance of such surface domain structure has been previously theoretically predicted by Marchenko [36]. Moreover, the existence of a high density of nanodomains inside the micro-scale domains was earlier obtained in SBN single crystals using confocal Raman microscopy (CRM) [30, 37].

It should be noted that domain structure was very stable at room temperature. The area and PFM contrast of the created domains did not change noticeably at least for several months. The PFM contrast of created domains decreased with heating and disappeared completely at 120°C.

B. *Stripe Exposure*

The stripe exposure was performed with dose ranged from 100 to 500 µC/cm^2. An ensemble of isolated submicron-size domains appeared along e-beam irradiation in the samples with free surface and stripe periodical domain structure - in the samples covered by resist (Fig. 3 a,b).

The domain width was the same for stripe recording along the X and Y directions due to the C_{4v} symmetry of the SBN single crystals and equality Y and X directions. The dose dependence of the averaged width of stripe domains in SBN covered by resist after stripe irradiation along Y(X) and 45 degrees to Y(X) is shown on Fig. 3 c. It was shown that the width of the obtained stripe domains linearly increased with irradiation dose and saturated at high doses, which was attributed to the electrostatic interaction of the approaching

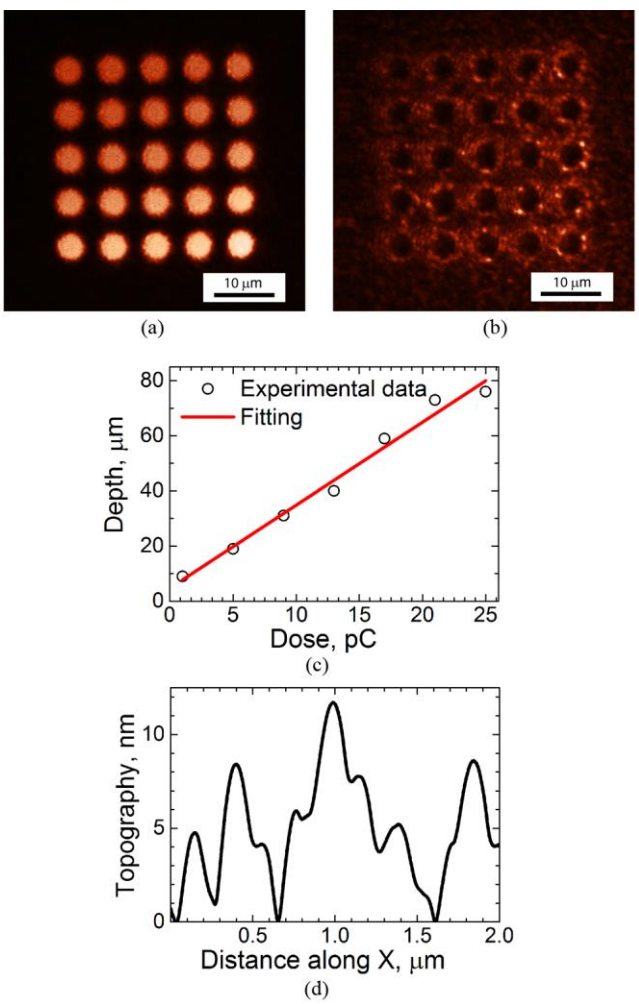

Fig. 2. (a-b) The ČSHGM images of domain structure created by dot irradiation, (c) the dose dependence of the domain depth in SBN single crystal with resist layer, (d) AFM profile of topography after etching. Depth (a) 5 µm, (b) 14 µm. (a, b, d) Dose 13 pC.

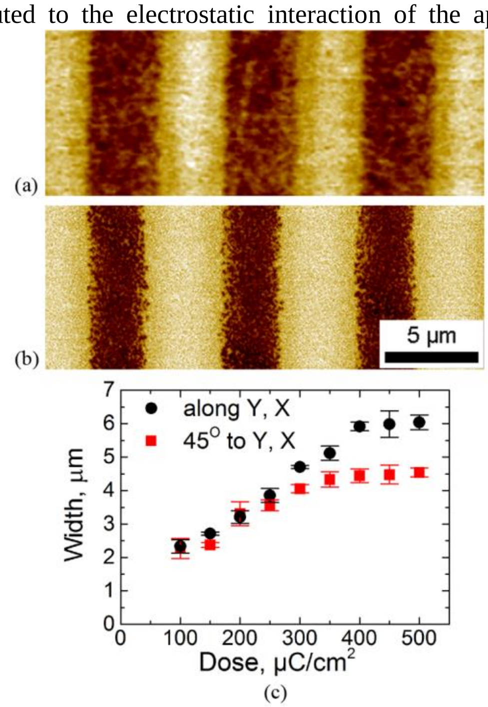

Fig. 3. PFM image of stripe domains obtained at the surface in crystal with resist after stripe irradiation (a) along Y, (b) 45 degrees to Y, and (c) dose dependence of the domain width. (a,b) Dose 350 µC/cm^2. Out-of-plane response. Phase signal.

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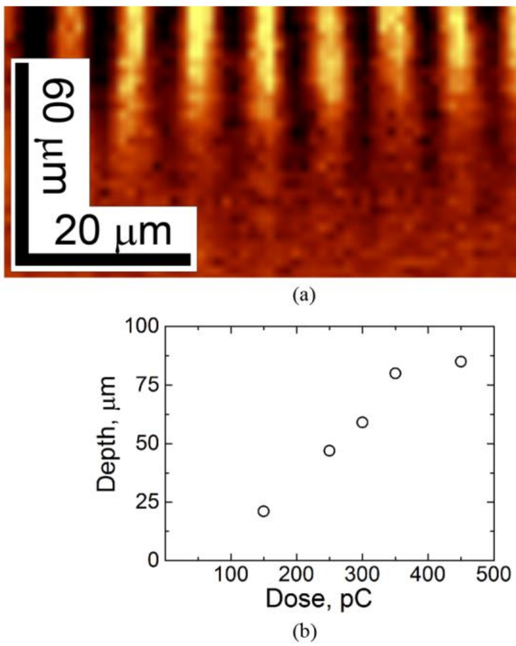

Fig. 4. (a) CRM image of YZ-section of domain structure in crystal with resist layer after stripe irradiation by e-beam along 45 degrees to Y and (b) dose dependence of the domain depth. (a) Dose 300 μC/cm^2.

charged domain walls. The width of stripe domains created by scanning along Y(X) and 45 degrees to Y(X) was almost equal for doses less than 250 μC/cm^2. The value of saturation the width of stripe domains oriented along 45 degrees to Y(X) was less than ones oriented along Y(X), which may be associated with more favorable growth domains along the crystallographic directions.

The CRM was used for imaging of the YZ-section of the domain structure created by stripe irradiation along 45 degrees to Y in crystal with resist layer (Fig. 4 a). The stripe domain structure depth increased with irradiated dose up to 85 μm in used dose range (Fig. 4 b). The obtained depth of the stripe domains was also confirmed by ČSHGM images of domain structure.

It is important to note that the isotropic domain growth allowed to create: (1) the stripe domains oriented along any direction and (2) the arbitrary domain shapes. This fact is very promising for domain engineering and allowed creation of the domain structures of complex geometry.

IV. Conclusion

In summary, the domain patterning by electron beam irradiation was studied in thermally depolarized Ce-doped strontium barium niobate single crystals with free surface and surface covered by resist layer. The created domain patterns were visualized by: piezoresponse force microscopy at the surface, confocal Raman microscopy and Čerenkov-type second-harmonic generation in the bulk. The dependences of the domain sizes and domain depth on the irradiated dose have been revealed. The circular shape of isolated domains was attributed to isotropic domain growth with step generation at the wall by merging with isolated residual nanodomains existed in the initial state. The linear dose dependence of the switched area can be attributed to screening of the depolarization field by injected charge, and its saturation for domains in the matrix is due to the electrostatic interaction of the approaching charged domain walls. The better quality of the domain patterns in samples covered by resist was attributed to the charge localization. The obtained results can be applied for creation of the precise domain patterns with arbitrary orientation and shape in SBN single crystals in order to produce the nonlinear optical devices with improved characteristics.

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References

[1] N. Balke, I. K. Bdikin, S. V. Kalinin, and A. L. Kholkin, “Electromechanical imaging and spectroscopy of ferroelectric and piezoelectric materials: state-of-the-art and prospects for the future,” *J. Amer. Ceram. Soc.*, vol. 92, no. 8, pp. 1629-1647, Aug. 2009, 10.1111/j.1551-2916.2009.03240.x.

[2] V. Ya. Shur, E. L. Rumyantsev, E. V. Nikolaeva, E. I. Shishkin, R. G. Batchko, G.D. Miller, M. M. Fejer, and R. L. Byer, “Regular ferroelectric domain array in lithium niobate crystals for nonlinear optic applications,” *Ferroelectrics*, vol. 236, pp. 129-144, 2000, 10.1080/00150190008016047.

[3] V. Ya. Shur, A. R. Akhmatkhanov, and I. S. Baturin, “Micro- and nano-domain engineering in lithium niobate,” *Appl. Phys. Rev.*, vol. 2, no. 4, Oct. 2015, Art. no. 040604, 10.1063/1.4928591.

[4] B. J. Rodriguez, R. J. Nemanich, A. Kingon, A. Gruverman, S. V. Kalinin, K. Terabe, X. Y. Liu, and K. Kitamura, “Domain growth kinetics in lithium niobate single crystals studied by piezoresponse force microscopy,” *Appl. Phys. Lett.*, vol. 86, no. 1, 2005, Art. no. 012906, 10.1063/1.1845594.

[5] G. Rosenman, P. Urenski, A. Agronin, Y. Rosenwaks, and M. Molotskii, “Submicron ferroelectric domain structures tailored by high-voltage scanning probe microscopy,” *Appl. Phys. Lett.*, vol. 82, no. 1, pp. 103-105, Jan. 2003, 10.1063/1.1534410.

[6] A. V. Ievlev, S. Jesse, A. N. Morozovska, E. Strelcov, E. A. Eliseev, Y. V. Pershin, A. Kumar, V. Ya. Shur, and S. V. Kalinin, “Intermittency, quasiperiodicity, and chaos during scanning probe microscopy tip-induced ferroelectric domain switching,” *Nature Physics*, vol. 10, pp. 59-66, Nov. 2014, 10.1038/nphys2796.

[7] V. Ya. Shur, “Kinetics of ferroelectric domains: Application of general approach to $LiNbO_3$ and $LiTaO_3$,” *J. Mater. Sci.*, vol. 41, no. 1, pp. 199-210, Jan. 2006, 10.1007/s10853-005-6065-7.

[8] H. Ito, C. Takyu, and H. Inaba, “Fabrication of periodic domain grating in $LiNbO_3$ by electron beam writing for application of nonlinear optical process,” *Electron. Lett.*, vol. 27, no. 14, pp. 1221-1222, Aug. 1991, 10.1049/el:19910766.

[9] L. S. Kokhanchik, M. V. Borodin, N. I. Burimov, S. M. Shandarov, V. V. Shcherbina, and T. R. Volk, “Surface periodic domain structures for waveguide applications,” *IEEE Trans. Ultrason. Ferroelectr. Freq. Control*, vol. 59, no. 6, pp. 1076-1084, Jun. 2012, 10.1109/TUFFC.2012.2298

[10] T. R. Volk, L. S. Kokhanchik, R. V. Gainutdinov, Y. V. Bodnarchuk, and S. D. Lavrov, “Domain formation on the nonpolar lithium niobate surfaces under electron-beam irradiation: A review,” *J. Adv. Dielectr.*, vol. 8, no. 2, Apr. 2018, Art. no. 1830001, 10.1142/S2010135X18300013.

[11] X. Li, K. Terabe, H. Hatano, and K. Kitamura, “Domain patterning in $LiNbO_3$ and $LiTaO_3$ by focused electron beam,” *J. Cryst. Growth*, vol. 292, no. 2, pp. 324-327, Jul. 2006, 10.1016/j.jcrysgro.2006.04.028.

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[12] M. C. Gupta, W. P. Risk, A. C. G. Nutt, and S. D. Lau, "Domain inversion in $KTiOPO_4$ using electron beam scanning," *Appl. Phys. Lett.*, vol. 63, no. 9, pp. 1167-1169, Jun. 1993, 10.1063/1.109811.

[13] J. L. Hart, S. Liu, A. C. Lang, A. Hubert, A. Zukauskas, C. Canalias, R. Beanland, A. M. Rappe, M. Arredondo, and M. L. Taheri, "Electron-beam-induced ferroelectric domain behavior in the transmission electron microscope: Toward deterministic domain patterning," *Phys. Rev. B*, vol. 94, no. 17, Nov. 2016, Art. no. 174104, 10.1103/PhysRevB.94.174104.

[14] A. M. Prokhorov, Yu. S. Kuz'minov, *Ferroelectric crystals for laser radiation control*. Bristol, England: Adam Hilger, 1990, ch. 4, pp. 101-175.

[15] P. Lehnen, W. Kleemann, Th. Woike, and R. Pankrath, "Ferroelectric nanodomains in the uniaxial relaxor system $Sr_{0.61-x}Ba_{0.39}Nb_2O_6$:$Ce^{3+}{}_x$," *Phys. Rev. B*, vol. 64, no. 22, Nov. 2001, Art. no. 224109, 10.1103/PhysRevB.64.224109.

[16] K. Terabe, S. Takekawa, M. Nakamura, K. Kitamura, S. Higuchi, Y. Gotoh, and A. Gruverman, "Imaging and engineering the nanoscale-domain structure of a $Sr_{0.61}Ba_{0.39}Nb_2O_6$ crystal using a scanning force microscope," *Appl. Phys. Lett.*, vol. 81, no. 11, pp. 2044-2046, Sept. 2002, 10.1063/1.1506945.

[17] V. V. Shvartsman, W. Kleemann, T. Łukasiewicz, and J. Dec, "Nanopolar structure in $Sr_xBa_{1-x}Nb_2O_6$ single crystals tuned by Sr/Ba ratio and investigated by piezoelectric force microscopy," *Phys. Rev. B*, vol. 77, no. 5, Feb. 2008, Art. no. 054105, 10.1103/PhysRevB.77.054105.

[18] K. Terabe, S. Higuchi, S. Takekawa, M. Nakamura, Y. Gotoh, and K. Kitamura, "Nanoscale domain engineering of a $Sr_{0.61}Ba_{0.39}Nb_2O_6$ single crystal using a scanning force microscope," *Ferroelectrics*, vol. 292, pp. 83-89, Jun. 2003, 10.1080/00150190390222862.

[19] R. V. Gainutdinov, T. R. Volk, O. A. Lysova, I. I. Razgonov, A. L. Tolstikhina, and L. I. Ivleva, "Recording of domains and regular domain patterns in strontium-barium niobate crystals in the field of atomic force microscope," *Appl. Phys. B*, vol. 95, no. 3, pp. 505-512, Jun. 2009, 10.1007/s00340-009-3507-y.

[20] T. R. Volk, L. V. Simagina, R. V. Gainutdinov, A. L. Tolstikhina, and L. I. Ivleva, "Ferroelectric microdomains and microdomain arrays recorded in strontium-barium niobate crystals in the field of atomic force microscope," *J. Appl. Phys.*, vol. 108, no. 4, Aug. 2010, Art. no. 042010, 10.1063/1.3474965.

[21] V. A. Shikhova, V. V. Fedorovyh, A. P. Turygin, L. V. Gimadeeva, D. S. Chezganov, E. O. Vlasov, D. O. Alikin, L. I. Ivleva, A. L. Kholkin, and V. Ya. Shur, "Local switching in SBN:Ni single crystals with various initial domain states," *Ferroelectrics*, vol. 525, pp. 100-107, Mar. 2018, 10.1080/00150193.2018.1432745.

[22] T. R. Volk, R. V. Gainutdinov, Ya. V. Bodnarchuk, L. V. Simagina, E. D. Mishina, N. A. Ilyin, V. V. Artemov, and L. I. Ivleva, "Microdomain arrays fabricated in strontium-barium niobate crystals by microscopic methods," *Ferroelectrics*, vol. 442, pp. 63–73, May 2013, 10.1080/00150193.2013.776360.

[23] V. Ya. Shur, V. A. Shikhova, D. O. Alikin, V. A. Lebedev, L. I. Ivleva, J. Dec, D. C. Lupascu, and V. V. Shvartsman, "Temperature effect on the stability of the polarized state created by local electric fields in strontium barium niobate single crystals," *Scientific Reports*, vol. 7, Mar. 2017, Art. no. 125, 10.1038/s41598-017-00172-1.

[24] V. Ya. Shur, D. S. Chezganov, M. M. Smirnov, D. O. Alikin, M. M. Neradovskiy, and D. K. Kuznetsov, "Domain switching by electron beam irradiation of Z^+ polar surface in Mg-doped lithium niobate," *Appl. Phys. Lett.*, vol. 105, no. 5, Aug. 2014, Art. no. 052908, 10.1063/1.4891842.

[25] V. Ya. Shur, D. S. Chezganov, A. R. Akhmatkhanov, and D. K. Kuznetsov, "Domain patterning by electron beam of MgO doped lithium niobate covered by resist," *Appl. Phys. Lett.*, vol. 106, no 23, Jun. 2015, Art. no. 232902, 10.1063/1.4922372.

[26] D. S. Chezganov, E. O. Vlasov, M. M. Neradovskiy, L. V. Gimadeeva, E. A. Neradovskaya, M. A. Chuvakova, H. Tronche, F. Doutre, P. Baldi, M. P. De Micheli, and V. Ya. Shur, "Periodic domain patterning by electron beam of in proton exchanged waveguides in lithium niobate," *Appl. Phys. Lett.*, vol. 108, no. 19, May 2016, Art. no. 192903, 10.1063/1.4949360.

[27] L. I. Ivleva, "Physicochemical and technological peculiarities of multicomponent oxide crystal growth from melt by modified Stepanov technique," *Bulletin of the Russian Academy of Sciences: Physics*, vol. 73, no. 10, pp. 1338-1340, Oct. 2009, 10.3103/S1062873809100074.

[28] S. M. Kaczmarek, M. Orowski, T. Skibiñski, A. Jasik, and L. I. Ivleva, "Ferroelectric properties of relaxor type SBN single crystals pure and doped with Cr, Ni, and Ce," *Rev. Adv.* Mater. Sci., vol. 23, pp. 80-87, Mar. 2010.

[29] V. A. Shikhova, A. D. Ushakov, V. V. Fedorovyh, V. A. Anikin, A. A. Esin, V. Ya. Shur, A. L. Kholkin, and L. I. Ivleva, "Influence of the domain structure on piezoelectric and dielectric properties of relaxor SBN single crystals", IOP Conference Series: Materials Science and Engineering, vol. 443, no. 1, Nov. 2018, Art. no. 012031, 10.1088/1757-899X/443/1/012031.

[30] P. S. Zelenovskiy, V. A. Shikhova, A. V. Ievlev, M. M. Neradovskiy, and V. Ya. Shur, "Micro-Raman visualization of domain structure in strontium barium niobate single crystals," *Ferroelectrics*, vol. 439, pp. 33-39, Dec. 2012, 10.1080/00150193.2012.746890.

[31] R. Hammoum, M. D. Fontana, P. Bourson, and V. Ya. Shur, "Characterization of PPLN-microstructures by means of Raman spectroscopy," *Appl. Phys. A*, vol. 91, no. 1, pp. 65-67, Apr. 2008, 10.1007/s00339-007-4356-3.

[32] P. S. Zelenovskiy, M. D. Fontana, V. Ya. Shur, P. Bourson, and D. K. Kuznetsov, "Raman visualization of micro- and nanoscale domain structures in lithium niobate," *Appl. Phys. A*, vol. 99, no. 4, pp. 741-744, Jun. 2010, 10.1007/s00339-010-5621-4.

[33] V. Ya. Shur and P. S. Zelenovskiy, "Micro- and nanodomain imaging in uniaxial ferroelectrics: joint application of optical, confocal Raman and piezoelectric force microscopy," *J. Appl. Phys.*, vol. 116, no. 6, Art. no. 066802, Aug. 2014, 10.1063/1.4891397.

[34] Y. Sheng, A. Best, H.-J. Butt, W. Krolikowski, A. Arie, and K. Koynov, "Three-dimensional ferroelectric domain visualization by Čerenkov-type second harmonic generation," *Opt. Express*, vol. 18, no. 16, pp. 16539-16545, Aug. 2010, 10.1364/OE.18.016539.

[35] M. Ayoub, P. Roedig, K. Koynov, J̈. Imbrock, and C. Denz, "Čerenkov-type second-harmonic spectroscopy in random nonlinear photonic structures," *Opt. Express*, vol. 21, no. 7, pp. 8220-8230, Apr. 2013, 10.1364/OE.21.008220.

[36] V. I. Marchenko, "Domain structure of ferroelectric materials," *Sov. Phys. JETP*, vol. 50, no 6, pp. 1168-1169, Dec. 1979.

[37] V. Shur, V. Shikhova, A. Ievlev, P. Zelenovskiy, M. Neradovskiy, D. Pelegov, and L. Ivleva, "Nanodomain structures formation during polarization reversal in uniform electric field in strontium barium niobate single crystals," *J. Appl. Phys.*, vol. 112, no. 6, Sept. 2012, Art. no. 064117, 10.1063/1.4754511.