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Electron beam poling of $[001]_c$-poled PMN-39PT single crystal

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The relaxor-based $Pb(Mn_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMN-PT) ferroelectric single crystal is very attractive material as a potential candidate for optical application due to its high values of electrooptic coefficients (r_{33} is 70 pm/V) [1]. The nonlinear-optical applications require methods of precise control of domain walls positions for precise periodical domain structures [2]. The electron beam (e-beam) poling was elaborated for domain patterning in uniaxial crystals such as lithium niobate (LN) and lithium tantalate with simple 180º-domain structure [3] and applied for multiaxial crystals BT [4], $BaMgF_4$ [5], and ceramics [6]. Recently, the modification of the method using surface covering by buffer artificial dielectric layer allow creation of fine periodically poled structure in bulk MgO-doped LN [7] and LN-based waveguides [8]. The second harmonic generation efficiency comparable with commercial elements was demonstrated.

In this work we have used e-beam patterning for creation of the domain structure in PMN-PT crystal covered by surface dielectric layer. The results explained in terms of kinetic approach [9].

The studied tetragonal PMN-PT single crystals were grown by modified Bridgman technique. The studied samples ($5\times8\times1$ mm^3) were cut normal to [001] direction with sides cuts parallel to (010) and (100) planes. The surface was covered by beam resist AZ nLof 2020 (MicroChemicals GmbH, Germany) deposited by Sawatec SM 180 spin coater. The opposite surface was sputtered by100-nm-thick Cu electrode and grounded during irradiation.

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 irradiated patterns were specified by Raith Nanosuite software. The three exposure modes were used: (1) dot exposure, (2) line exposure by single path line-scan and (3) stripe exposure by meander-scan covering of the rectangular area. The domain patterns after chemical removal of resist layer and electrode were visualized by: optical microscopy, piezoresponse force microscopy (PFM), confocal Raman microscopy (CRM) and scanning electron microscopy (SEM).

We revealed that e-beam irradiation led to switching of c-domains. The design of created domain structures corresponded to irradiated ones. The dose dependence of switched domain area for dot irradiation demonstrates the linear behavior up to 50 pC and saturation with large dispersion of domain sizes at higher doses (Fig. 1a). The saturation has been explained by electrostatic interaction of domain walls and by interaction with a-domains. The domain shape changed from circular at low dose to irregular at high doses (Fig. 1b,c).

The line and stripe exposure mode have been used for creating of 1D pattern (Fig. 1d). The appearance of domain fingers at the walls oriented mainly at the angle close to 45º relative to [100] direction was revealed.

We have demonstrated the possibility to write stripe domains along any direction as well as ring shaped domains (Fig. 1e). Since any area element consist of discrete points the circle domain shape upon dot irradiation at low doses is the key point which allows us to produce domain patterns with arbitrary geometry. The width of stripe domains was independent on direction.

The domain visualization in the crystal bulk using CRM modified for PMN-PT crystals allowed to measure the domain depth down to 200 μm.

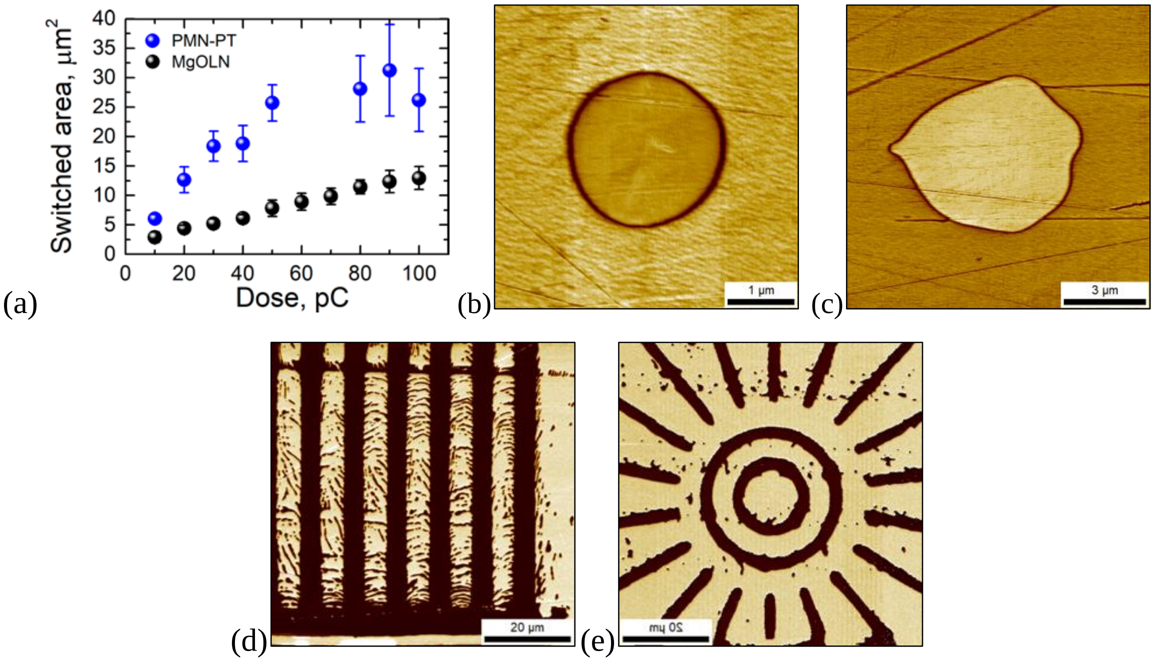

Figure 1. (a) The dose dependence of switched domain area in MgOLN and PMN-PT crystals, (b) – (e) PFM images of c-domains created by e-beam. (b), (c) Dot irradiation with dose: (b) 10 pC, (c) 50 pC. (d), (e) Stripe irradiation along (d) [100] direction, (e) arbitrary direction.

The obtained knowledge can be used for periodical poling in PMN-PT to produce the crystals for light frequency conversion.

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