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Charged domain walls in lithium tantalate with compositional gradients produced by VTE process

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We investigated the shape of charged domain walls (CDWs) in $LiTaO_3$ (LT) single crystals with inhomogeneous spatial distribution of Li concentration along polar axis. Vapor Transport Equilibration process (VTE) was used for changing the Li concentration from congruent (48.8 mol.%) to stoichiometric (50 mol.%) composition. Spatially inhomogeneous distributions were obtained for various durations of VTE at 1100 °C (far above Curie temperature) [1, 2].

The spatial distribution of Li concentration was measured by confocal Raman spectroscopy (Alpha AR300, WITec). CDWs were visualized at the surface using optical (BX61, Olympus, Japan) and scanning electron (Merlin, Carl Zeiss) microscopes after selective chemical etching and in the bulk using Cherenkov-type Second Harmonic Generation microscopy (Ntegra Spectra, NT-MDT) and confocal Raman microscopy.

The geometry of the initial domain structure formed during cooling below Curie temperature after VTE depends on the spatial distribution of LT composition. The wide polydomain layer appeared in the volume with congruent composition and single domain layers appeared near polar surfaces. The polydomain layer represent 3D maze domain structure with head-to-head and tail-to-tail CDWs. The increase of VTE duration resulted in Li concentration increase and thinning of polydomain layer. The narrow CDW was formed for long enough VTE due to presence of built-in fields produced by gradient of Li concentration [1-3].

The subsequent cooling to room temperature resulted in change of CDWs morphology and formation of the domain bumps under the action of the pyroelectric field. Moreover, the isolated non-through domains with diameter below 5 μm appeared in the volume between polar surfaces and CDW. The shape of domain cross-section changed with depth from hexagonal close to the surface layer with stoichiometric composition to trigonal for congruent composition in the bulk.

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