Periodic Domain Patterning by Electron Beam in Lithium Niobate Single Crystals Modified by Proton Exchange

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The formation of ferroelectric domains by electron (e-beam) irradiation of polar surfaces in single crystals of 5 mol% MgO-doped lithium niobate (MgOLN) and congruent lithium niobate (CLN) with surface layer modified by soft proton exchange (SPE) has been studied experimentally [1,2]. The periodical domain patterns in bulk MgOLN and SPE channel waveguides have been produced and effective second harmonic generation (SHG) has been demonstrated.

The irradiation of polar surfaces was performed by scanning electron microscope (Auriga Crossbeam, Carl Zeiss) attached with electron-beam lithography system (Elphy Multibeam, Raith) with different doses and periods using two exposure modes: arrays of dots and stripes. The irradiated surface was covered by 2.5-μm-thick resist layer, whereas the opposite surface with solid electrodes was grounded. The studied samples were prepared on a 0.5-mm-thick Z-cut congruent lithium niobate (LN) and a 1-mm-thick Z-cut MgO doped LN wafers. The soft proton exchange (SPE) procedure was carried out by dipping the crystal for 72 hours into a benzoic acid bath diluted by 3.1 weight % of lithium benzoate (BL) and melted at 300°C. The domain patterns were visualized by optical microscopy and piezoresponse force microscopy. The dependences of the domain sizes on dose and period of irradiated areas have been measured. The irradiation parameters for uniform patterning were obtained experimentally. It has been shown that the resist layer is required for successful domain poling in MgOLN and mandatory to periodical poling in LN crystal with PE layers. The formation of isolated submicron domains has been attributed to the discrete switching caused by ineffective screening of the depolarization field provided by the modified surface layer.
The space charge formation and distribution of electric field were studied by computer simulation. It was shown that the localized in resist space charge slightly touching the crystal surface produced the maximum value of electric field at the resist/LN interface thus resulting in the best pattern quality. The mechanism of domain nucleation and growth under the field produced by space charge was proposed and explained in terms of the kinetic approach [3].

The developed technique has been used for creation of periodically poled LN. SHG of green light was demonstrated in MgOLN with 6.89 μm period. The high pattern homogeneity was confirmed by high SHG efficiency. The e-beam poling with surface resist layer was done after channel waveguide fabrication. SHG with normalized nonlinear conversion efficiency up to 48%/(W·cm^2) has been achieved [4].
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