Periodic Nano-domain Patterns in Relaxor Single Crystals

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$(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3$-$xPbTiO_3$ (PMN-PT) ferroelectric relaxors, especially with PT ($PbTiO_3$) concentration near its morphotropic phase boundary (MPB), have been employed for a broad range of applications such as ultrasound transducers, sensors, and actuators. Two engineering techniques, dopants and micro-patterned electrodes, are usually used to manipulate piezoelectric properties of relaxor ferroelectrics. Piezoelectric properties of ferroelectric crystals can also be enhanced using other domain engineering techniques, which may involve crystal cut, electrical field, stress field, or thermal field to obtain designed domain configurations. Wada *et al.* have recently reported a domain engineering technique that used micro-patterned electrodes to effectively manipulate the domain size of $BaTiO_3$ for enhanced piezoelectric properties. Yamashita *et al*. reported PMN-PT with double-side composite electrodes (randomly distributed Mn oxide and Au) showing 40% d_{33} improvement. In our previous work, the 200 nm grating pattern electrode (Mn oxide and Au composite) was prepared on one surface of PMN-PT crystal, exhibiting 25% d_{33} enhancement. In this study, we studied the size effect of nano-patterned composite electrode on the dielectric and piezoelectric properties of PMN-PT single crystals.

Lloyd's-mirror interference lithography, metal deposition, and an annealing process were used to fabricate the nano-patterns of electrode on PMN-PT single crystals. The dielectric and piezoelectric properties were investigated for the composite electrode on PMN-PT with different pitch sizes of Mn oxide patterns (200 nm, 500 nm, 800 nm and 1 μm) and a uniform gold layer (100 nm). The domain distribution was observed using a piezoresponse force microscopy (PFM) for samples with different thicknesses. Mn diffusion was examined using a time-of-flight secondary ion mass spectrometry (ToF-SIMS). Initial results on PMN-PT crystals with one side 800 nm nanocomposite electrode showed a higher piezoelectric constant (d_{33} = 2250 pm/V) than the ones with conventional plain electrodes (d_{33} = 1600 pm/V).

In summary, periodic nanocomposite electrodes with different pitch sizes were fabricated on PMN-PT single crystals as a domain engineering technique to enhance the piezoelectric properties of relaxor PT single crystals. Initial results showed that PMN-PT samples with one side nanocomposite electrode exhibited 35% d_{33} enhancement. Further experiments will be focused on the double side nano-pattern fabrication and mechanism study of nanocomposite electrode associated phase transformation, so that the domain distribution (domain wall density) and depth of domain structure and the associated effect on the crystal's properties can be studied, and hence the electrode-domain structure-property relationships can be established.