

Mesoscale Correlative Electro-Chemo-Mechanical Response in Ferroelectric Solid Solutions

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Relaxor-ferroelectric (relaxor-FE) solid solutions on the rhombohedral side of the morphotropic phase boundary (MPB) have substantially higher electromechanical response in single crystalline form – when cut and poled along specific directions – compared to polycrystalline ceramic ferroelectrics. However, the origin of such high response is to date ascribed to a number of different contributors. These include the presence of one or more low-energy, transition phases (e.g., monoclinic or orthorhombic) that contribute to the electric-field-induced phase transition, effects of domain engineering, high electrostriction coefficients correlated to the relaxor nature of the end member, and existence of polar and chemical nano regions. Ultimately, the cause of the large electromechanical response of the relaxor-FE solid solutions should be sought at the micron and submicron scale. Furthermore, due to the compositional dependency of the response, we must determine how the functional properties of these materials evolve as the chemistry progresses through the phase diagram.

This work reports a systematic probing of the local electromechanical response in lead magnesium niobate-lead titanate solid solution, $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3 - x\text{PbTiO}_3$, (1-x)PMN-xPT, single crystals for $x = 0, 10, 28, 40, 60$. All samples were grown with the Bridgeman method and all measurements were performed on the (001) faces. Specifically, piezoresponse force microscopy (PFM) was used in band excitation (BE) mode for local characterization of the electromechanical response and relaxation processes. The response of (1-x)PMN-xPT single crystals to a series of DC pulses at increasing positive and negative integer multiples of the coercive voltage was measured at discrete locations, namely in a 50 by 50 point grid, on regions ranging from $2 \times 2 \mu\text{m}^2$ square to $20 \times 20 \mu\text{m}^2$. The relaxation parameters –saturation value (A_0), total variation over time (R_0), time constant (τ), and stretch exponent (β) – were extracted both from the raw data as well as from the relaxation behavior extracted from independent component analysis (ICA), using in both cases a stretched exponential function (Kohlrausch-Williams-Watts or KWW).

Ferroelectric switching was observed in all samples. The strongest relaxation was observed in the sample near the MPB (72PMN-28PT) with a PFM amplitude signal change substantially larger than that of the tetragonal samples and an order of magnitude larger than that observed in 90PMN-10PT. This behavior is consistent with increasing ferroelectric to relaxor content in the samples, as well as contributions from field-induced phase transitions at the MPB composition. A field-induced phase transition was further corroborated by the changes in the elastic parameters of the 72PMN-28PT, as evidenced by the softening in the contact resonance frequencies. We will further discuss trends in the relaxation parameters as a function of the crystal composition.