

Dynamical studies of the polarized nano-regions in relaxor ferroelectrics using the soft polar mode as a probe.

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The lead-oxide class of relaxor ferroelectrics $(1-x)\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3 + x\text{PbTiO}_3$ (PZN- x PT) and PMN- x PT ($M=\text{Mg}$) possess exceptional piezoelectric properties far exceeding those of conventional $\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$ (PZT) ceramics used in current device applications. A correspondingly intense scientific effort has been devoted to studying the structural phase diagram of this system as a function of the PbTiO_3 concentration x . By contrast, far less research has been done on the lattice dynamics of these novel systems. Neutron inelastic scattering measurements performed at the NIST Center for Neutron Research on single crystal of PZN-8%PT in its cubic phase reveal an anomalous ridge of scattering intensity centered $\sim 0.2 \text{ \AA}$ from the Brillouin zone center.¹ When plotted as a standard dispersion diagram, this ridge of scattering resembles a waterfall, and extends vertically from the TO phonon branch near 9 meV down to the TA phonon branch near 4 meV. This anomalous feature has subsequently been observed other relaxor systems including single crystals PZN and PMN ($M=\text{Mg}$). We have been able to model the presence of the waterfall in PZN at 500 K using a simple coupled-mode model that assumes a q -dependent TO phonon linewidth $\Gamma_{\text{TO}}(q)$ that increases sharply near $q = 0.2 \text{ \AA}$ as one approaches the zone center. The dramatic increase in $\Gamma_{\text{TO}}(q)$ is believed to occur when the wavelength of the TO mode becomes comparable to the size of the polarized nano-regions that first condense around $T_d \sim 750 \text{ K}$, far above the ferroelectric transition temperature $T_c = 410 \text{ K}$.² Recent high temperature measurements on PMN above T_d confirm the recovery of a normal propagating zone-center TO mode.

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