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Intrinsic localized modes observed as a precursor to polar nanoregion formation in a relaxor ferroelectric

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Abstract– Intrinsic localized modes (ILMs) have been proposed as an explanation for the formation of polar nanoregions (PNRs) in technologically important relaxor ferroelectric materials [1-3]. While displacive ferroelectric transitions have been understood in terms of soft mode theory for over 50 years an equivalent explanation for relaxors has remained elusive. The ILM theory provides a possible explanation for a number of properties including: (1) the arrested softening of the ferroelectric soft mode by ILM renormalization; (2) dynamic polar nanoregions as slowing ILMs; (3) static PNRs as a freezing in of ILMs; (4) crossover from relaxor to displacive behavior with doping; and (5) the high frequency dielectric response [1-3]. The ILM model has yet to be fully accepted, however, since the supporting evidence has been indirect or, in the case of inelastic neutron scattering from single crystals, limited by narrow views of momentum-energy space and/or temperature. Here, a comprehensive neutron scattering study will be presented that provides direct evidence of ILMs playing a role in relaxor behavior in the classic PMN-PT system. First, an extra mode spontaneously localizes at the Burns temperature [4] (the characteristic temperature where relaxor behavior begins). Second, the localized mode softens with decreasing temperature, indicating a slowing down of the local dynamics in the relaxor region. Third, the local mode sharpens (decouples) as it localizes, a property of ILMs in thermal equilibrium. Finally, the local mode delocalizes (becomes dispersive) and broadens when the PNRs delocalize below the Curie temperature. Apparently, ILMs play an important role in a material property that is central to several multi-billion dollar industries.

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