

Diffuse Scattering from Critical Ferroelectric Fluctuations

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Introduction: KTaO_3 (KTO) belongs to the class of dielectric materials called quantum paraelectrics, such as SrTiO_3 (STO), in which the ferroelectric phase transition is suppressed by quantum fluctuations. Unlike STO, KTO does not undergo any structural phase transition. When KTO is doped with a small amount of more polarizable elements such as Li and Nb these impurity atoms induce strong polarization cloud around them, resulting in relaxor ferroelectric behavior with record-high values of permittivity. In $\text{K}_{1-x}\text{Li}_x\text{TaO}_3$ (KLT) Li^+ ions substitute for K^{2+} in the KTO lattice, and form a system of randomly distributed reorientable off-centered dipoles in a highly polarizable matrix of KTO. As a result various multi-scale dipole ordered states appear in the KLT at low temperatures, depending on the Li concentration. The most essential characteristics of KLT are large ($\sim 1\text{\AA}$) dipolar $\langle 100 \rangle$ displacements and the slow relaxation of Li^+ ions, dipole cluster formation, and softening of the TO_1 mode. In $\text{KTa}_{1-y}\text{Nb}_y\text{O}_3$ (KTN) slightly off-centered Nb^{5+} ions show very small displacements in the $\langle 111 \rangle$ direction. Fast relaxation of related dipoles cause the TO_1 mode to soften and enhance the polarization. At small concentrations of Li ($x \leq 0.01$) or Nb ($y \leq 0.007$) the properties of the compounds are close to the nominally pure KTO, remaining cubic (O_h^1) in symmetry at the lowest temperature where the quantum effects prevent the ferroelectric ordering (quantum paraelectricity).

Results: We have measured diffuse and Bragg intensities for the sample with x and $y < 0.01$ i.e. in the “quantum paraelectric” regime during our recent beam time allocation (September 2001). Our preliminary results show several interesting points regarding mesoscopic structural transitions. In agreement with neutron scattering data we do not observe any deviations from the overall cubic symmetry. However, at low temperature we observe significant broadening of the Bragg peaks. This is seen in the Figure 1. This result clearly indicates that at low temperatures, $T < 50$ K, there is formation of static (or dynamic but beyond our time scale resolution) polarized domains induced most likely by freezing into dipole-glass-like state. In addition we noticed that we could quench structure from 80 K to 10 K with only small increase in the peak width. After about 20 minutes the structure at 10 K recovers original width. It suggests slow relaxation of electro-elastic strain after freezing. The preliminary analysis of the diffuse scattering studies revealed strong anisotropy in the principal directions. The intensity along H and K was twice as high as along L. This result is best explained by the scattering geometry and confirms strong transverse $\langle 100 \rangle$ type displacements. On the other hand longitudinal displacements observed during L scans are much smaller. In addition there is significant difference in power law ($I \sim 1/q^\alpha$) behavior. Again power appears to be anisotropic. We did not see significant temperature dependence for the scans along L, however there is difference for the transverse scans below the “freezing temperature”. We speculate that this result suggests appearance of nano-domain structure with well defined system of domain walls.

