A coupling between an acoustic phonon and the diffuse scattering in the relaxor ferroelectric \((1-x)\text{Pb(Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\text{PbTiO}_3\) near morphorobic phase boundary

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Relaxor ferroelectrics are a class of disordered crystals showing extremely high, broad and frequency dependent peak in dielectric susceptibility. It is widely believed that the polar nanoregions (PNR) play an important role in relaxor behavior occurring at temperatures much above \(T_c\), the so-called Burns temperature \(T_d\) [1]. Neutron scattering measurements by Naberezhnov et al. on the typical relaxor \(\text{Pb(Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\) (PMN) revealed the onset of strong diffuse scattering at or very near \(T_d\) [2], indicating a close connection between the PNR and the diffuse scattering. A solid solution of PMN and ferroelectric \(\text{PbTiO}_3\) (PT) exhibits a maximum dielectric susceptibility near PMN-30\%PT, the boundary between the rhombohedral phase and the tetragonal one, which is the so-called morphorobic phase boundary (MPB). Last year, we investigated PMN-34\%PT which locates on the tetragonal side of the MPB and reported that the \(Q\)-pattern of the diffuse scattering changes from ellipsoid elongating along \((1 \bar{1} 0)\) direction in the cubic phase to cross elongating along \((100)\) direction in the tetragonal phase. This year, we report an overdamped acoustic phonon coupled with the diffuse scattering.

Figures 1 show constant \(Q\) scan spectra at \((0.944 \ 1.056 \ 0)\) and \((0.92 \ 1 \ 0)\) taken at \(T = 440\) and \(410\) K. At \(T = 440\) K (cubic), the diffuse scattering extends along the \([1\bar{1}0]\) direction; the diffuse scattering is strong at \((0.944 \ 1.056 \ 0)\) but weak at \((1.021 \ 1 \ 0)\). We found the acoustic phonon at \((0.944 \ 1.056 \ 0)\) is overdamped while that at \((1.021 \ 1 \ 0)\) is underdamped, as shown in Fig.1(a) and (b). At \(T = 410\) K (tetragonal), the overdamped phonon becomes underdamped concomitantly with the disappearance of the diffuse scattering at \((0.944 \ 1.056 \ 0)\). On the other hand, at \((1.021 \ 1 \ 0)\), the well-defined phonon peak observed in the cubic phase disappears presumably due to over-damping. These results indicate a coupling between the acoustic phonon and the diffuse scattering. Similar damped acoustic modes were observed in PMN by Stock \textit{et al.} [3] and the authors successfully simulated the phonon spectra by Michel-Naudts model which takes into account a coupling between a phonon mode and a relaxation mode [4]. From their model, an acoustic mode is overdamped and peak at \(E = 0\) appears when a characteristic relaxational frequency is less than or nearly equal to phonon frequency. The current results indicate slow relaxation mode may exist along the particular direction at which the diffuse scattering is strong.

References

Fig. 1. Constant Q scans at (a),(c) (0.944 1.056 0) and (b),(d) (0.92 1 0) taken at $T = 440$ and 410 K. Solid lines show fits to the sum of a sharp Gaussian peak at $E = 0$ for the central peak and a Lorentzian peak for the acoustic phonon.