Study of slow lattice dynamics in relaxor PMN-30%PT by neutron spin echo technique

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Relaxor ferroelectrics are characterized by a giant piezoelectricity associated with significant frequency dispersion. The variation of the diffuse maximum of the dielectric constant in wide frequency range gives evidence for an important role of slow dynamics. A key-concept to understand the relaxor behavior is slowly relaxing polar nanoregion (PNR), a local nanometer-sized region with ferroelectric polarization and atomic shift. Such an atomic shift in PNR was observed for a typical relaxor such as PMN and PZN as characteristic diffuse scattering. However, there is no decisive understanding of the microscopic mechanism of the PNR formation at present. In addition, recent neutron spin-echo measurement concluded that the diffuse scattering is purely elastic and is not related to polarization fluctuations[1]. The purpose of the present work is to reinvestigate the intrinsic energy width of the diffuse scattering and clarify its mechanism. Neutron spin-echo experiments were performed on the *i*NSE installed at the JRR-3 Guide-hall of the JAEA. The sample was a high-quality PMN-30%PT single crystal with a volume of 1cc grown at JFE Mineral. The data were taken with incident neutron energies of 3.38 meV which enables access to diffuse scattering near Q =(100) at $2\theta = 74.5$ degree.

Figure 1 shows the normalized intermediate scattering functions of the diffuse scattering at Q =(0.98, 0.02, 0) measured at various temperatures. Below $T = 400$ K, the normalized intermediate scattering functions $I(Q,t)/I(Q,0)$ do not decay, which is associated with the long-range ordered ferroelectric phase for $T < T_C = 400$ K. Note that the averaged $I(Q,t)/I(Q,0)$ are less than 1, indicating fast relaxations even in the ordered phase. At $T = 450$ K, $I(Q,t)/I(Q,0)$ is flat as $T \leq 400$ K, but it drops from 0.9 to 0.6, suggesting enhancement of fast relaxations above T_C. This is consistent with enhancement of meV-range broad quasi-elastic scattering at high temperatures[2]. Above $T = 500$ K, the $I(Q,t)/I(Q,0)$ has maximum at 0.5 ns. Assuming this peak is the first peak of oscillation, it corresponds to excitation of 4 μeV. This ultra-low energy mode may be related to the ferroelectric fluctuations in PNR. Further study is highly required to clarify the nature of this mode.

References

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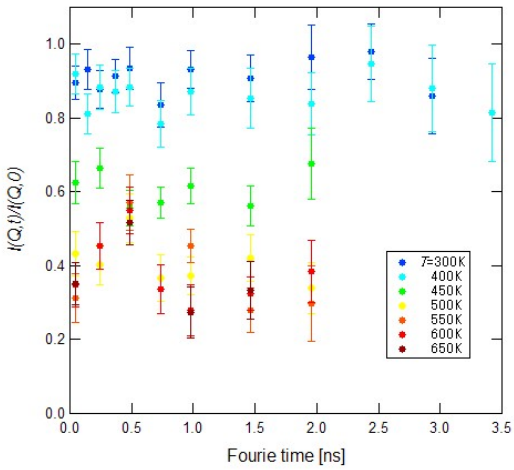

Fig. 1. Normalized intermediate scattering functions I(Q,t)/I(Q,0) of the diffuse scattering at (0.98, 0.02, 0).