Spin and lattice dynamics in multiferroic $BiFeO_3$

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$BiFeO_3$ is a room temperature multiferroic material with great application potentials. The G-type antiferromagnetic (AFM) order in this material is weakly coupled to the ferroelectric polar order, resulting in a cycloid spin structure with long wavelength modulations (~ 62 nm). There has also been growing interest on couplings between the dynamical properties as well. For example, a "magneto-optical" resonance has been proposed theoretically, and extra modes below the lowest lying optic phonon energies have been observed in Raman measurements. A full understanding of both the spin and lattice dynamics in $BiFeO_3$ is highly desired.

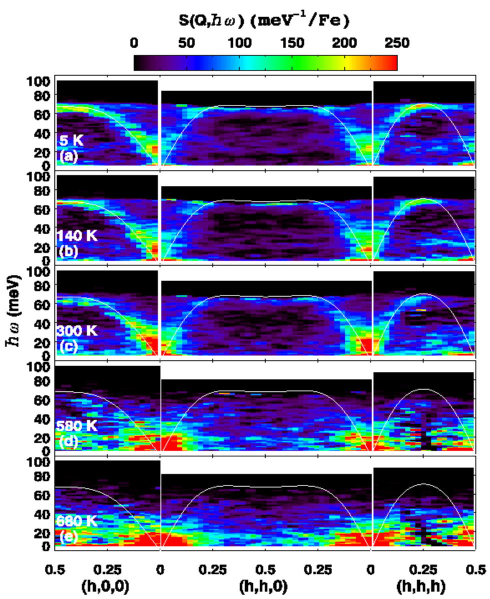

Fig.1 Magnetic excitations measured at 5, 140, 300, 580, and 680 K. The horizontal axis denotes $\mathbf{Q} - \mathbf{Q}_{AF}$. The data shown here are measured with Ei = 120 meV.

With inelastic neutron scattering, we have been able to systematically map out the complete three-dimensional spin-waves in this system. In addition, low energy (~<20 meV) phonons have also been studied. Here we would like to present our work on both the magnetic and lattice dynamics in this system for a large temperature range between 10K and 700K.

The sample being used is a cylindrical single crystal sample which weighs about 3.5 g. The experiments have been performed using ARCS and HYSPEC time-of-flight spectrometers at SNS. The magnetic excitations are measured on ARCS with incident energies of 40 and 120 meV to cover the full dispersion. The low energy phonons are measured on HYSPEC with incident energy of 20 meV.

In Fig.1, magnetic excitations measured along different crystallographic directions are shown. At low temperatures, the spin-waves are well-defined coherent modes. We can use a simple Heisenberg spin Hamiltonian, taking into account the first, second and third nearest neighbor exchange interactions (J_1, J_2, and J_3) to model the spin-waves. The exchange parameters can be obtained from the modeling. Warming up the sample resulting in a reduction of the effective exchange parameter and the life-time of the spinwave excitations. When the system is heated above the Neel temperature (T_N=640 K), the spin-waves become heavily damped and broad in space as well. In addition, our analysis on the the magnetic form factor suggests that a strong hybridization between the Fe and O orbitals needs to be taken into account when the magnetic response is considered. Even as a good insulator, the electrons responsible for the magnetic moments are not restricted at the Fe sites, and the local magnetic moment could extend well beyond the typical ionic radius of Fe^{3+} into the adjacent O^{2-} sites.

We were also able to measure low energy phonons in $BiFeO_3$. The phonon excitations in different zones are summarized in Fig.2. The three acoustic phonon modes (TA1, propagating along [010], polarized along [100]; TA2, propagating along [1,-1,0],polarized along [110]; and LA, propagating along [100], polarized along [100]), and the lowest lying optic modes are mapped around a number of Bragg peaks. The measured phonon dispersions and the intensities after removing the Bose-factor do not show any significant variation with temperature within measurement errors. A clear broadening of the transverse acoustic mode is observed near the zone-boundary when heating toward T_N, suggesting a possible coupling between the lattice dynamics and the antiferromagnetic order in the system.

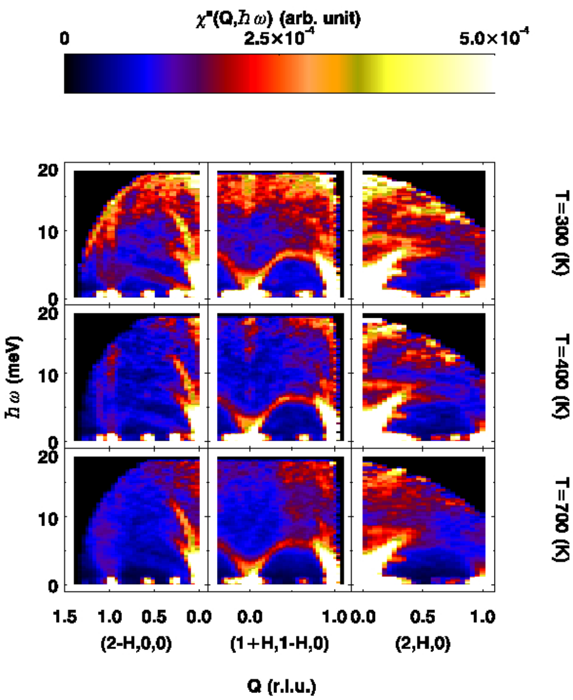

Fig.2 The dynamic response measured from the single crystal BiFeO3 sample, at T=300 K (top row), 500 K (central row), and 700 K (bottom row). The intensities are shown in the (HK0) plane, for q along [100] (left column), [1,-1,0] (central column), and [010] (right column) directions, respectively. The intensities shown near the top of the measurement energy range are spurious intensities when ~! becomes close to the incident energy Ei. These intensities are temperature independent. They only appear to be stronger at 300 K (top frames) in the plot of ′′(Q, ~!) because the data here are divided by the Bose factor.

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