Hybrid Goldstone modes in multiferroics

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(Dated: November 16, 2008)

By using polarized inelastic neutron scattering measurements, we show that the spin-lattice quantum entanglement in multiferroics results in hybrid elementary excitations, involving spin and lattice degrees of freedom. These excitations can be considered as multiferroic Goldstone modes. We argue that the Dzyaloshinskii-Moriya interaction could be at the origin of this hybridization.

Spontaneous symmetry breaking is a powerful concept at the basis of many developments in physics, especially in condensed matter and high energy physics. The low symmetry phase is described by an order parameter associated with low energy and long wavelength excitations, restoring the original high temperature phase symmetry. These Goldstone modes are nothing but longitudinal and transverse phonons in solids, or spin waves in magnets. In case of multiferroic materials, two order parameters, namely the ferroelectric polarization and the magnetization, coexist and are strongly coupled by a spin-lattice interaction. As a result of this entanglement, the multiferroic Goldstone modes are expected to be new spin and lattice hybrid excitations called electromagnons. While their existence has been theoretically predicted for a long time, their dual nature as both spin and lattice excitations makes them challenging to observe and study experimentally. Due to their dipole electric activity, they first could be detected by optical measurements: evidence for their existence has been recently reported in different orthorhombic multiferroics, namely GdMnO₃, TbMnO₃, Eu₀₉₄₅Y₀₂₅₃MnO₃, DyMnO₃, Y₃Mn₂O₅ and TbMn₂O₅. However, their magnetic counterpart is still not clearly evidenced. Moreover, as optical techniques probe the zone center, the shape of their dispersion and thus the underlying mechanism responsible for the hybridization, is still unknown. In this letter, we report polarized inelastic neutron scattering experiments performed on the particular case of hexagonal Y₃MnO₅ to unravel the spin-lattice dual nature of these hybrid modes. Moreover, as neutron scattering allow a global survey of the reciprocal space, we report the dispersion of these hybrid modes throughout the Brillouin zone. This result is discussed in the framework of the dynamical magnetoelastic coupling theory, where the Dzyaloshinskii-Moriya interaction plays a central role.

Y₃MnO₅ becomes ferroelectric below 900K, with an electric polarization along the c-axis, due to alternatively long and short yttrium-oxygen bonds (parallel to the c-axis). Despite a strong geometric frustration, the Mn spin order below the Neél temperature T_N = 75K in a classical triangular arrangement (Curie-Weiss temperature θ = 500K). The magnetic and electric order parameters strongly interact, as recently shown in reference, claiming the existence of a giant magnetoelastic coupling.

Through the comprehensive investigation of hybrid spin and lattice excitations, inelastic neutron scattering on a triple axis spectrometer combined with longitudinal polarization analysis (LPA) offer an efficient way to determine hybrid modes. Indeed, this technique allows to measure separately the spin-spin and nuclear-nuclear correlation functions in different channels (called...
spin flip SF and non-spin flip NSF respectively. It is worth noting that the magnetic cross section probes spin fluctuations perpendicular to the wave vector Q only. On the contrary, the nuclear cross section probes the atomic displacements parallel to Q. Measurements were carried out on the IN22 triple axis spectrometer at the Institute Laue Langevin (ILL, Grenoble, France). The sample was aligned in the scattering plane (100),(001) such that momentum transfer of the form Q=(H,0,L) in units of reciprocal lattice wave vectors were accessible and mounted into an ILL-type orange cryostat (1.5K-300K). All data were obtained with a fixed final wave vector of 2.662Å⁻¹ providing an energy resolution less than 1meV. Heussler crystals were used as analyzer and monochromator, together with a flipper of Mezei to reverse the spin of the scattered neutrons. The elastic and inelastic measurement of the polarisation efficiency (flipping ratio), as determined from different magnetic Bragg peaks (100),(105),(003) and from the magnon signal at Q=−(0.4 0 6) and (1 0 1), was of about 16 (elastic) and 14 (inelastic signal). The amplitude of the expected peak from the SF channel to the NSF channel is therefore less than 7% of the SF intensity. Using an unpolarized beam (PG monochromator), we measured a polarization parallel and perpendicular in- and out-of-plane to the wave vector less than 10⁻² excluding the presence of chiral terms and of the nuclear-magnetic interference terms [20,21].

Energy scans at the wave vector Q=(0,0,6) in both channels measured above and below TN are depicted on the panel B of the Figure 1. On the one hand, the strong magnetic quasi-elastic signal observed at 150K in the SF channel confirms the presence of strong spin-spin correlations in the paramagnetic phase arising from the geometrically frustrated Mn moments [16,17]. Below TN, 3 almost doubly degenerate spin wave modes are known to rise up [16,22,23]. At the zone centre, they are characterized by a spin gap ΔSG, typical of a magnetic anisotropy. Upon cooling below TN, the magnetic long range order develops and ΔSG gradually shifts to higher values (Figure 2c), reaching its maximum of about 5.3 meV around 40K. On the other hand, the NSF intensity shows at 150 K a quasi-elastic signal as well. At 1.5K, the NSF data demonstrate the emergence of an additional inelastic nuclear mode. The energy of this mode coincides with the spin gap ΔSG, pointing out its close connection with the spin subsystem. To understand the temperature dependence of both quasi-elastic and inelastic modes, NSF energy scans have been performed at different temperatures around TN (Figure 2a). From these NSF data, we extract the lattice susceptibilities, presented on Figure 2b. This analysis shows that the additional mode rises upon cooling on the top of the quasi-elastic signal as a supplementary intensity. Below 40K, the energy of this nuclear mode is found to follow the spin gap ΔSG (Figure 2). Above 40K, its presence becomes however hard to distinguish. To overcome this difficulty, we measured the NSF intensity at Q=(0,0,6) for sampling temperatures at 2.2 meV, well below the low temperature energy of the nuclear mode (Figure 2a). According to the above analysis, the quasi-elastic signal is expected to give a contribution proportional to the standard detailed balance factor (grey line). Figure 2 shows however subtle deviations from it in the intermediate region surrounding TN (blue line), that we attribute to the raising of the nuclear mode starting from the lowest energies. These measurements allow estimating the energy of the nuclear mode for temperatures higher than 40K. Finally, we conclude that both spin gap and nuclear mode energies follow the same temperature dependence from 1.5 K till TN. Since the nuclear cross section probes fluctuations along Q=(0,0,6), which is parallel to the c-axis, we attribute the nuclear quasi-elastic signal to relaxation vibrations along this particular direction. The nuclear mode corresponds to collective vibrations along the same c-axis. Moreover, since it is found at a zone center, it corresponds to vibrations within the decoration of the unit cell. Now the question is: how do such internal motions propagate through the crystal?

To address this question, it is essential to determine the dispersion of the additional nuclear mode, by repeating the same measurements for different Q=(H,0,6) with varying H (Figure 3). We first determine the dispersion of the modes which are trivially expected, namely the spin wave modes (red points in SF channel on Figure 3b) and the transverse acoustic phonon22 (denoted by P on Figure 3). Amazingly, the NSF data demonstrate unambiguously the existence of additional nuclear modes all along the low energy spin wave dispersions from Q=(0,0,6) to Q=(0.25,0,6) (denoted by HM on Figure 3). Their intensity is found to decrease as the wave vector increases. For instance, at Q=(0.325,0,6), the nuclear intensity measured in the NSF channel becomes comparable to the expected leak of the intensity from the SF channel. Figure 3 evidences the fact that the dispersions of these nuclear modes match the spin wave dispersions for a wide range of wave vectors. It therefore appears obvious that they must be attributed to collective vibrations within the decoration that propagate through the crystal in the Néel state by hybridizing with the spin waves.

The discovery of a strong mixing down to the zone centre H=0 shows that the long range properties of the system are affected. In explaining these findings, we thus propose to consider a coupling of the spin subsystem with atomic displacements within the unit cell [19]. This in turn implies a hybridization mechanism with an optical phonon, as in the dynamical magnetoelastic coupling theory developed for orthorhombic RMnO3 [4,5,24]. In this scenario, the spin current Jij=Sᵢ×Sⱼ (defined for neighbouring manganese spins Sᵢ and Sⱼ sitting at distance rij) plays a crucial role. Owing to the
spiral magnetic ordering typical of these materials, $J_{ij}$ acts, via the inverse Dzyaloshinskii-Moryia coupling, as a force pushing the oxygen atom located between adjacent manganese off the Mn-Mn bond. This mechanism gives rise at $T_N$ to an electric polarization $P = r_{ij} \times J_{ij}$ lying within the spiral plane and perpendicular to $r_{ij}$. The multiferroic Goldstone mode is predicted to be a hybrid mode, rising at $\Delta_{SG}$, and made of a mixing between the optical phonon associated with the oxygen displacement, and the spin wave mode involving spin fluctuations out of the spiral plane. At first glance, this mechanism would not hold in the hexagonal case, since due to the triangular symmetry, the resulting magnetic force experienced by oxygen atoms is zero: indeed, each oxygen ion is located at the centre of a triangle formed by 3 neighbouring Mn ions. The system can however benefit from the Dzyaloshinskii-Moryia interaction by spontaneously moving the oxygen atoms along the c-axis. This distortion is expected to create an electric polarization parallel to $J_{ij}$ and is accompanied by a slight rotation of the spins towards the same direction. In that case, the coupled spin and atomic motions look like those of the ribs of an umbrella that would be put up or down (Figure 4). In close analogy with the orthonhornbic case, hybridized spin-lattice Goldstone modes are expected at $\Delta_{SG}$, in agreement with the present results.

To test the validity of this scenario, several predictions have to be further examined. First, the magnetic structure should be characterized by a tiny ferromagnetic moment superimposed on the AF structure, as sketched in the right panel of Figure 4. In that case, specific Bragg peaks should exhibit a specific contribution at $T_N$. Calculations of the structure factor show that this effect is best observed for Bragg peaks with a forbidden AF magnetic intensity and a very weak nuclear one. The (2-11) Bragg peak fulfils these conditions. As shown on Figure 4, its intensity increases below $T_N$, and this is a good indication for the validity of this scenario. Next, as the high temperature ferroelectric distortion is mainly due to atomic displacements parallel to the c-axis, the oxygen displacements proposed in this umbrella scenario should result at $T_N$ in a slight change of the ferroelectric moment. Evidence for such an evolution has been recently reported by Lee et al [19], thanks to high resolution X rays and neutrons diffractions measurements. This result is another argument supporting our interpretation.

In conclusion, polarized inelastic neutron scattering ex-
FIG. 4: (Color online) Umbrella scenario. Left figure (a) displays the temperature dependence of the (100) antiferromagnetic Bragg peak and of the (2-11) Bragg peak (unpolarized neutron). The umbrella mechanism discussed in the text is sketched on the right panel (b). It evidences the existence of a small ferromagnetic component perpendicular to the Manganese (Mn)-Oxygen (O) layers when the oxygens are moving out of the plane.

Experiments demonstrate the existence of hybrid spin and lattice low energy modes in $YMnO_3$, that can be considered as Goldstone modes of the multiferroic phase. The neutron polarization analysis directly shows their hybrid nature, revealing both spin and structural counterparts. The mechanism responsible for this hybridization could be the Dzyaloshinskii-interaction, in close analogy with the model recently proposed for orthorhombic multiferroic materials.

We would like to thank Yvan Sidis, Martine Hennion and Fernande Moussa for fruitful discussions.

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