Terahertz magneto-electric excitations in a chiral material

The term "multiferroics" designates a remarkable class of solid materials where both ferromagnetism (i.e. an alignment of magnetic dipoles) and ferroelectricity (an alignment of electric dipoles) can coexist. Multiferroics are receiving much attention as a promising way to achieve electric-field control of magnetic dipoles and, conversely, magnetic-field control of electric dipoles, this for application in a number of future hybrid technologies, for instance novel electronics based on both spins and charges. These electric/magnetic cross-manipulation effects can have signatures in the THz frequency range, in the form of resonances (phonons and magnons) that are characteristic of the multiferroic order in the material.

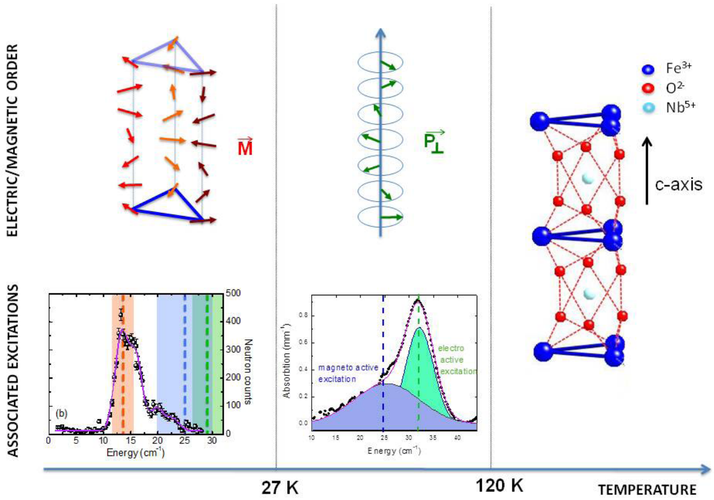

Fig. 1: At right: a simplified drawing of the chiral, tubular lattice structure of the langasite compound $Ba_3NbFe_3Si_2O_{14}$, characterized by its triangles of Fe^{3+} ions along the vertical c-axis.
At centre: below 120 K, a helical electric polarization exists (green arrows) which supports dual magneto-electric excitations. They can be induced by either the magnetic or the electric field components of incident electromagnetic radiation, with maximum efficiency at 25 cm^{-1} (blue fitted peak) and 32 cm^{-1} (green peak) respectively, as probed by polarized THz spectroscopy.
At left: Below 27 K, additionally, a helical ordering of the Fe^{3+} spins (horizontal arrows) exists along the c-axis. In this "multiferroic" phase, we also observe magnon excitations (spin waves) as shown by the peak at 13 cm^{-1} in the inelastic neutron scattering measurements.

Phonons and magnons are quantized states of two kinds of wave propagation in a solid. They correspond to coherent deviations of the atoms' positions or spins away from their equilibrium in an atomically or magnetically ordered phase. Normally, phonons and magnons couple to the electric and magnetic components of the electromagnetic field respectively. In 2006, a novel type of excitation was discovered, called the "electro-magnon", which is a magnetic excitation that can couple to the oscillatory electric field. Now, by combining several sophisticated techniques we have obtained evidence for a new kind of magneto-electric excitation, a phonon that is magnetically active.

We used terahertz spectroscopy on the synchrotron source SOLEIL (Paris), and inelastic neutron scattering at the Institut Laue Langevin, Grenoble, working in the energy range where magnons and low energy phonons might be present. Radiation of frequency 1 THz has a wavelength of 0.3 mm (energy of 5 meV). The highly sensitive polarized THz spectroscopy allows us to identify the way the observed excitations are activated by the oscillating electric and/or magnetic field of the incoming radiation. The neutron scattering measurements clearly identify magnetic excitations, which interact with the neutron's magnetic moment.

We applied these two tools to study the energy spectrum of an iron langasite ($Ba_3NbFe_3Si_2O_{14}$), a complex magnetic material first synthesized at Institut NEEL and studied for its remarkable magnetic and structural properties. It has static magneto-electric properties. Moreover, it is totally "chiral" : both its atomic structure and its magnetic order cannot be superimposed on their images in a mirror. Thus this material is a good candidate for multi-ferroic behaviour. In this langasite, we see clearly the expected ordinary magnons in the antiferromagnetic phase below the Néel temperature of 27 K (see at left in Fig.1). At somewhat higher temperature we identified a dual non-magnetic mode, a phonon mode, at frequencies around 0.8 THz (= 28 cm-1). This mode is excited by the electric field component of the electromagnetic radiation and also, more surprisingly, by its magnetic field component (see in centre of Fig. 1).

These findings demonstrate that not only magnons but also phonons, i.e. atomic vibrations, can acquire a combined magneto-electric character. From symmetry arguments, we interpret our results as rotational atomic vibrations that can be excited by an oscillating magnetic field (via the Lorentz force) if a spontaneous helical electric polarisation is present. We deduce that a complex ferroelectric phase exists below a temperature (120 K) four times higher than the temperature (27 K) where the magnetic order sets in, and that below the Néel temperature the compound is multiferroic. The microscopic polarization is formed by a helical arrangement of electric dipoles (see Fig.1 at centre) which, due to the chirality of the compound, has a definite sense of rotation.

Finally, our findings widen the type of magneto-electric excitations that could be used in "magnonics", a new information science that would use magnetic excitations to carry and process information.

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FURTHER READING

THZ MAGNETOELECTRIC ATOMIC ROTATIONS IN THE CHIRAL COMPOUND $Ba_3NbFe_3Si_2O_{14}$
L. Chaix, S. de Brion, F. Lévy-Bertrand, V. Simonet, R. Ballou, B. Canals, P. Lejay
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