TeraHertz properties of multiferroic compounds

Electromagnetic waves outside the spectrum of visible light have been used for over a century to carry information (radio waves) or probe matter (X-Rays). However the spectral range known as the "TeraHertz gap" (Fig. 1) has hardly been explored because of a lack of good sources and detectors. Broadband spectroscopy at TeraHertz frequencies has now become available at synchrotron radiation sources. This spectral range is of considerable interest in Condensed Matter Physics, especially in magnetic or dielectric compounds, since both magnetic excitations (called magnons) and their electric analogue (optical phonons) have energies in this range. We have explored the TeraHertz response of compounds where new phenomena are expected, the so called "electro-magnons".

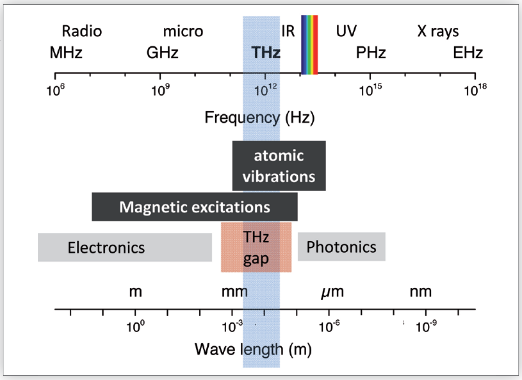

***Fig. 1:** Electromagnetic spectrum from radio-waves to X-Rays. In the THz range, probed at SOLEIL and ILL, both atomic vibrations (phonons) and magnetic excitations (magnons and crystal-field excitations) can be studied.*

The manganite compounds of formula $RMnO_3$, where R is the transition element Y (Yttrium) or a rare earth element such as Dysprosium, Holmium, Erbium, are ferroelectric at room temperature: They show a spontaneous electric-charge ordering, giving an electric polarization directed along the principal axis (the c-axis) of the crystal's hexagonal lattice structure. These materials also present an antiferromagnetic order, typically below 80 K, associated with the Mn^{3+} ions (see Fig. 2). Compounds of this type, where both an electric and a magnetic ordering prevail simultaneously, are called "multiferroics". They are attracting considerable attention since they have potential applications for recording and manipulating information using both electric and magnetic fields.

We have focused on two members of this family, namely $YMnO_3$ and $ErMnO_3$. We have fully characterized their energy spectra in the TeraHertz (THz) range by combining two techniques available at large-scale facilities: THz spectroscopy at the synchrotron SOLEIL near Paris and inelastic neutron scattering at the Institut Laue-Langevin (ILL) in Grenoble. Both techniques allow measurements of the THz spectra, but bring different information.

Since the THz electromagnetic wave has perpendicular electric and magnetic field components, one can differentiate electric and magnetic excitations by their polarization dependence. That is, one can distinguish magnons, which are associated with the magnetic order and excited by the THz magnetic field, from optical phonons, associated with the ordered lattice of positive and negative ions and excited by the THz electric field. In $YMnO_3$, where the Y^{3+} ion is non magnetic, a sharp excitation is observed at 1.2 THz whenever the THz magnetic field is polarized perpendicular to the c-axis, for all orientations of the perpendicular electric field. It has all the characteristics of a magnon corresponding to the precession of the Mn magnetic moment around the c-axis. In $ErMnO_3$, where the rare earth ion Er^{3+} is magnetic, an excitation at the same energy is observed but polarization measurements show that it is excited only by the electric field, along the c-axis, not at all by the magnetic field!

One can also study THz excitations with neutron scattering. Since neutrons carry a magnetic moment, they can distinguish magnetic excitations from atomic excitations. Our neutron scattering measurements have confirmed the magnetic nature of the 1.2 THz (5.2 meV) excitation. These combined results demonstrate that a Manganese magnon has become electro-active in the Erbium compound. This is a new kind of excitation called an "electro-magnon". We attribute its electro-activity to the presence of the second magnetic ion in the material, the rare-earth Erbium, as follows.

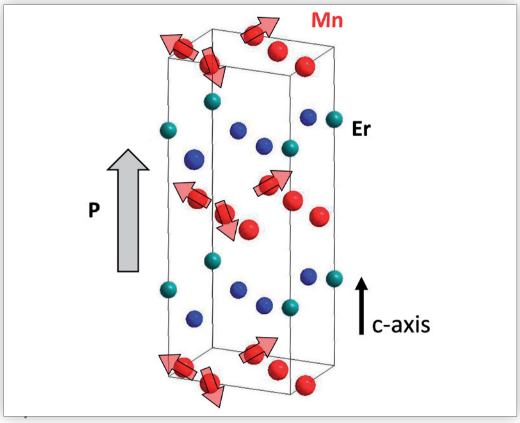

***Fig. 2:** $ErMnO_3$ magnetic and electric orders. The electric polarisation is along the c-axis. The Mn magnetic moments are oriented at 120° from each other.*

It is known that the electronic levels in rare earth ions are shifted and split by the electrostatic field of the surrounding ions, the so-called "crystal field". Transitions between the crystal field-split levels may be allowed depending on the local symmetry. Thanks to our complementary probes, THz waves and neutrons, we have identified six such transitions for $ErMnO_3$. One of these transitions, at 2.1 THz, has the correct symmetry to be electro-active and to couple strongly to the magnon at 1.2 THz. The coupling interaction is then responsible for the complete loss of the magnetic character of the magnon: It has been transmuted into a purely electro-active excitation. This new mechanism for electro-magnons may be general to other rare-earth-based multiferroics and suggests new possibilities for manipulating these excitations through the action of magnetic and electric fields.

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FURTHER READING

"Magneto- to electroactive transmutation of spin waves in $ErMnO_3$"
L. Chaix, S. de Brion, S. Petit, R. Ballou, L.-P. Regnault, J. Ollivier, J.-B. Brubach, P. Roy, J. Debray, P. Lejay, A. Cano, E. Ressouche and V. Simonet
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