Magneto-electric chiral excitations in the Fe langasite

After having established the remarkable structural and magnetic chiral properties of the multiferroic Fe langasite compound, we have been exploring their consequences on its dynamical properties by neutron scattering and by THz spectroscopy. We have observed the first evidence of the intrinsic chirality of the spin waves and have measured a new type of magnetoelastic excitation interpreted as atomic rotations excited by the magnetic component of the THz wave.

A novel chiral magnetic order was discovered at the Institut NEEL in the structurally chiral langasite Ba₃NbFe₃Si₂O₁₄. Chiral compounds in general are synthesized in a mix of the two chiral phases, mirror-image of each other, but in the case of Ba₃NbFe₃Si₂O₁₄ the grown single crystals are enantiopure that is single domain in chirality. The magnetic moments are distributed in a 120° arrangement over small triangle units whose centers form a triangular lattice. This order is helically modulated in the direction perpendicular to the triangles (see Figure a). A single triangular magnetic chirality together with a single helical magnetic chirality (unique sense of rotation of the magnetic moments around the triangles and along the helix) is stabilized in an enantiopure Ba₃NbFe₃Si₂O₁₄ single crystal. This totally chiral magnetic state generates new magnetoelastic and multiferroic behaviors.

We used inelastic neutron scattering without and with polarized neutrons and polarization analysis at the Institut Laue Langevin, Grenoble, in order to study the influence of this remarkable magnetic order on the corresponding elementary spin waves excitations. In Ba₃NbFe₃Si₂O₁₄, the spin waves are visualized in the reciprocal space by two branches rising at finite energy from the elastic Bragg positions characteristic of the magnetic order. They could be accurately modeled by 5 superexchange interactions and a Dzyaloshinski-Moryia term. Most remarkably, the chiral scattering measured using polarized neutrons revealed that the spin wave branch that corresponds to the Fourier Transform of planar spin-spin correlations is found to be totally chiral, again corresponding to a unique sense of rotation of the magnetic moments dynamical modulation (see Figure b). This is the first observation of chiral dynamics over the whole energy range of the spin-wave excitations in the absence of an external magnetic field. This is a consequence of the absence of opposite chirality domains whose equipopulation usually averages the chiral scattering to zero. In this special case, the single chiral domain allowed evidencing this generic chiral property of the spin waves. It also highlighted how the static chiral properties are extended to the dynamics.

Normally, phonons and magnons couple to the electric and magnetic components of the electromagnetic field respectively. Recently, a novel type of excitation was discovered, called the “electromagnon”, which has attracted much attention because it is a magnetic excitation that can couple to the oscillatory electric field. In this perspective, we pursued our study of the langasite dynamics by combining our inelastic neutron scattering results with THz spectroscopy on the synchrotron source SOLEIL (Saclay) in the energy range where magnons and low energy phonons are present. The highly sensitive polarized THz spectroscopy allows identifying the way the observed excitations are activated by the oscillating electric and/or magnetic field of the incoming radiation. We clearly identified the expected magnons in the antiferromagnetic phase below the NEEL temperature. At somewhat higher temperature we observed a dual non-magnetic mode. We determined that this phononic mode is excited by the electric field component of the electromagnetic radiation and also, more surprisingly, by its magnetic field component (see Figure c). From symmetry arguments, we could interpret our results as rotational atomic vibrations that can be excited by a THz electric field, as usual optical phonons, but also by a THz magnetic field (via the Lorentz force). These findings demonstrate that not only magnons but also phonons can acquire a combined magnetoelastic character.