Highlight #5: QUANTUM MATERIALS

Low dimensionality and frustration promote multiferroicity in $SrMnGe_2O_6$ pyroxene single crystal

Pyroxenes are minerals that possess quasi-one-dimensional magnetic chains, which have been predicted to exhibit multiferroism due to the interplay between low dimensionality and magnetic frustration, leading to complex modulated magnetic phases. In the framework of this research, we have synthesized a single crystal of $SrMnGe_2O_6$ pyroxene based on our recent discovery of multiferroism in this compound. We have thoroughly investigated its magnetic properties and successfully captured its magneto-electric properties using a relatively simple model with three competing isotropic interactions and a weak single-ion anisotropy.

Strong coupling between magnetism and electricity in matter has become a key focus in condensed-matter physics, both for its fundamental significance and technological potential. In spin-driven multiferroics, electric polarization arises from symmetry breaking caused by magnetic ordering. Pyroxenes, historically significant in mineralogy, have garnered interest due to their magnetic arrangement that can be regarded as a spin-frustrated network of quasi-one-dimensional spin chains, thus prone to the formation of incommensurate spin ordering.

The successful growth of a large single crystal of $SrMnGe_2O_6$ pyroxene using the floating zone furnace technique marks a significant advancement in the field. Building on our previous discovery of this phase in polycrystalline form, we have utilized these single crystals to investigate the anisotropic multiferroic properties of $SrMnGe_2O_6$. This exploration employed a variety of experimental techniques, including magnetization, heat capacity, pyroelectric current measurement, and both elastic and inelastic neutron scattering, combined with magnetic phase diagram calculations and spin-wave simulations.

Our findings reveal two incommensurate magnetic orders: a non-polar collinear sinusoidal magnetic structure at 4.36 K and a polar elliptical cycloidal structure below 4.05 K. These observations were confirmed by pyroelectric current measurements demonstrating a spontaneous polarisation concomitant with the second transition. The magnetic phase diagram was calculated taking into account three isotropic exchange couplings, the dominant one (J) between the Mn2+ nearest-neighbor cations along the chains whereas the two others (J1 and J2) bridge the chains. The magnetic excitation spectra of $SrMnGe_2O_6$, measured by inelastic neutron scattering, were successfully modelled using this set of antiferromagnetic exchange interactions and are fully consistent with the magnetic phase diagram. The study demonstrated that the magnetic behaviour of $SrMnGe_2O_6$ is well captured by a model with three competing isotropic interactions and weak single-ion anisotropy, and that the extended Dzyaloshinskii-Moriya mechanism explains the appearance and the direction of spontaneous polarisation.

In conclusion, this study highlights the synergy between the expertise of the MRS and MAGSUP teams and the experimental capabilities of the "Cristaux Massifs" and "Auto-Caractérisation" technological teams, showcasing the strength of Institut Néel collaborative research in advancing our understanding of multiferroic materials.

Reference

Claire V. Colin, Lei Ding, Eric Ressouche, Julien Robert, Noriki Terada, Frederic Gay, Pascal Lejay, Virginie Simonet, Céline Darie, Pierre Bordet & Sylvain Petit, "Incommensurate spin ordering and excitations in multiferroic $SrMnGe_2O_6$", *Physical Review B* **101**, 235109 (2020).

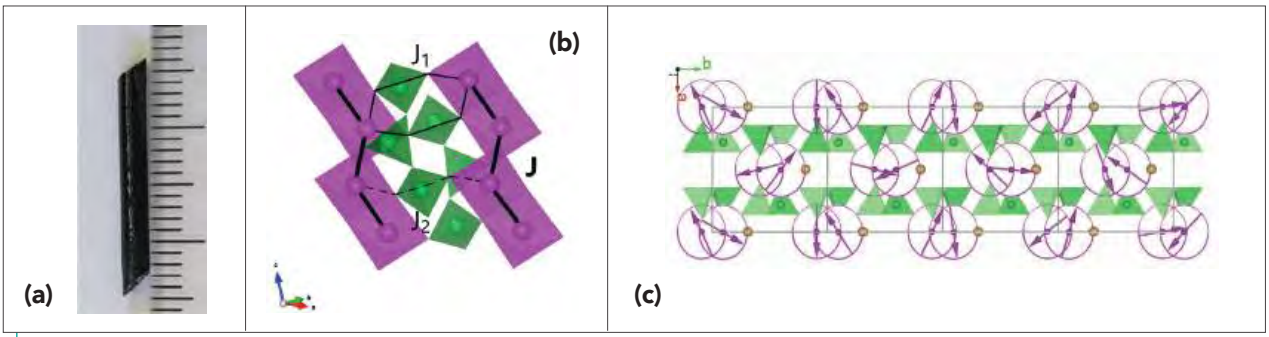

Fig.: *(a) Single crystal of $SrMnGe_2O_6$ grown by floating zone method. (b) The MnO6 zigzag chains connected through GeO4 tetrahedra in $SrMnGe_2O_6$. (c) Polar elliptical cycloidal spin structure.*