

# Supplemental Material: Helical bunching and symmetry lowering inducing multiferroicity in Fe langasites

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## DEVIATION FROM A 120° MAGNETIC ARRANGEMENT

The magnetic structure of the Fe langasite is described by three magnetic helices propagating perpendicular to the  $ab$ -plane with a propagation vector  $(0, 0, \tau)$ , based on magnetic moments at 120° from each other in the  $ab$ -plane. The magnetic structure factor of this structure, at the reciprocal space position  $H = \pm\tau$  with  $H$  a reciprocal lattice vector, writes :

$$\vec{F}_M(\vec{Q} = \vec{H} \pm \vec{\tau}) = p \sum_{\nu=1,3} f(|\vec{Q}|) \vec{m}_{\pm\vec{\tau},\nu} e^{i\vec{Q} \cdot \vec{r}_\nu}$$

with  $p=0.2696 \cdot 10^{-12}$  cm,  $f$  the magnetic form factor of the  $\text{Fe}^{3+}$  atoms,  $\vec{r}_\nu$  the position of the atom  $\nu$  in the cell:  $\vec{r}_1 = (x, 0, \frac{1}{2})$ ,  $\vec{r}_2 = (0, x, \frac{1}{2})$  and  $\vec{r}_3 = (-x, -x, \frac{1}{2})$  with  $x=0.2496$ . The Fourier components of the magnetization distribution for the atom  $\nu$  is  $\vec{m}_{\pm\vec{\tau},\nu} = [\frac{\mu\hat{u} \pm i\mu\hat{v}}{2}] e^{\mp i\phi_\nu}$  with  $\mu$  the magnetic moment amplitude,  $(\hat{u}, \hat{v})$  an orthonormal basis in the  $ab$ -plane, and  $\phi_\nu$  a phase which should account for the relative 120° dephasing between the three Bravais lattices.

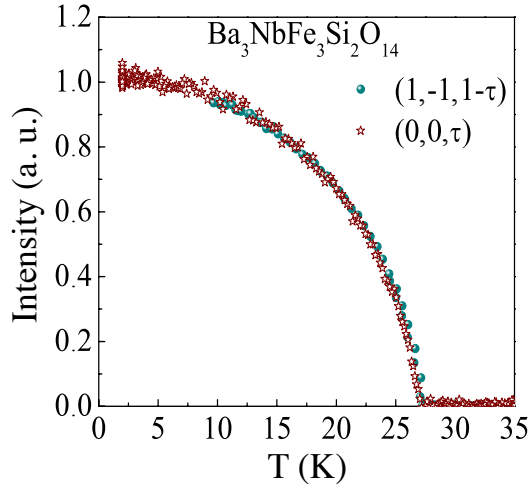


FIG. 1: Temperature dependence of the magnetic contributions of the  $(0, -1, 1-\tau)$  and  $(0, 0, \tau)$  satellites measured by neutron scattering and rescaled for comparison.

For  $H = (0, 0, \ell)$ , the magnetic structure factor becomes zero:

$$\vec{F}_M(0, 0, \ell \pm \tau) = pf(|\vec{Q}|) \sum_{\nu=1,3} \left[ \frac{\mu\hat{u} \pm i\mu\hat{v}}{2} \right] e^{\mp i\Phi_\nu} e^{i2\pi \frac{(\ell \pm \tau)}{2}} = pf(|\vec{Q}|) \left[ \frac{\mu\hat{u} \pm i\mu\hat{v}}{2} \right] e^{i2\pi \frac{(\ell \pm \tau)}{2}} (1 + e^{\mp i\frac{2\pi}{3}} + e^{\mp i\frac{4\pi}{3}}) = 0$$

For in-plane magnetic moments, the only way to observed a magnetic signal on the  $(0, 0, \ell \pm \tau)$  satellites implies a deviation from the  $120^\circ$  arrangement of the magnetic moments. Scagnoli *et al.* have also observed, by magnetic resonant X-ray diffraction, the presence of a  $(0, 0, \tau)$  magnetic satellite and attributed it to an out-of-plane (butterfly) component of the magnetization [2]. This component presumably originates from the Dzyaloshinskii-Moryia interaction. In addition to the  $D$  vector along the  $c$  axis, a  $D$  vector component along the side of each triangle is also allowed by symmetry, responsible for this small out-of-plane butterfly component of the magnetic arrangement [2, 3]. However, neutron scattering is only sensitive to the magnetization component perpendicular to the scattering vector  $\vec{Q}$ . Hence, this out-of-plane component cannot contribute to the  $(0, 0, \ell \pm \tau)$  satellites measured by neutron diffraction.

The measured temperature dependence of the  $(0, 0, \tau)$  extra-satellites, produced by the deformation of the  $120^\circ$  magnetic arrangement is shown in Fig. 1. It follows the temperature dependence of strong first order magnetic satellites.

### NOTE ON THE CHIRALITY

The excellent agreement between the calculated and observed spectral weight extinction in the spin waves when including a single-ion anisotropy in the Hamiltonian is only observed for one triangular chirality (anticlockwise  $120^\circ$  rotation of the spins on each triangle), in agreement with previous determination of the magnetic chirality [4]. Both Ta and Nb single-crystals thus have the same triangular chirality. Their structural chirality is opposite, as determined from anomalous X-ray scattering. This structural chirality, in turn, imposes the helical chirality to be opposite in both compounds.

### NEUTRON POLARIMETRY

The simplest neutron polarimetry technique to probe the chiral scattering was first introduced by Moon, Riste and Koehler [1], and is called longitudinal polarization analysis (LPA). In this case the final and initial polarizations are parallel, which can be achieved typically on a triple-axis spectrometer with polarizing monochromator/analyser (*e.g.* Heusler crystals). In addition, two flippers select the polarization states  $+$  and  $-$  (parallel or antiparallel to the polarization axis). The spin-flip terms (scattering processes changing the sign of the polarization),

$$\left( \frac{d^2\sigma}{d\Omega dE_f} \right)^{+-} = \sigma^{+-} \quad \text{and} \quad \left( \frac{d^2\sigma}{d\Omega dE_f} \right)^{-+} = \sigma^{-+}$$

and the non-spin-flip terms (scattering processes leaving the sign of the polarization unchanged),

$$\left( \frac{d^2\sigma}{d\Omega dE_f} \right)^{++} = \sigma^{++} \quad \text{and} \quad \left( \frac{d^2\sigma}{d\Omega dE_f} \right)^{--} = \sigma^{--}$$

of the partial differential cross-section can be measured independently.

In neutron polarimetry, a right-handed coordinated system is usually chosen with the  $x$ -axis along the scattering vector  $\vec{Q}$ , the  $y$ -axis in the scattering plane and the  $z$ -axis perpendicular to the scattering plane, so that the magnetic interaction vector has zero  $x$  component. We shall use in the following the simplified notations: the nuclear cross-section  $\sigma_N$  is proportional to  $|N|^2$  with  $N$  the nuclear structure factor. The magnetic cross-section  $\sigma_M$  is proportional to  $|M_\perp|^2$  with  $M_\perp$  the magnetic interaction vector, which is the projection of the magnetic structure factor  $F_M$  onto the plane perpendicular to the scattering vector  $Q$ . The chiral cross-section  $\sigma_{ch}$  is proportional to  $M_{ch}$  defined as:

$$M_{ch} = i(M_\perp^Z M_\perp^Y - M_\perp^Y M_\perp^Z)$$

These contributions can then be easily determined from linear combinations of the spin-flip and non-spin-flip cross-sections, knowing the initial polarisation  $P_i$ :

$$|N|^2 \propto \sigma_x^{++} = \sigma_x^{--}$$

$$|M_{\perp}|^2 \propto \frac{\sigma_x^{+-} + \sigma_x^{-+}}{2}$$

$$M_{ch} \propto \frac{\sigma_x^{+-} - \sigma_x^{-+}}{2P_i}$$

Alternatively, one can use an initial unpolarized beam (produced by a graphite monochromator for instance) and perform polarization analysis. In this case, the chiral scattering can be derived from the difference:

$$\frac{\sigma_x^{0-} - \sigma_x^{0+}}{2} = M_{ch}$$

where the symbol 0 is meant for zero beam polarization of the incoming neutrons.

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