Experimental report

23/03/2026

Proposal: 5-55-47 **Council:** 4/2025

Title: Characterizing the magnetic structures in Fe2TiO4

Research area: Physics

This proposal is a new proposal

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Samples: Fe2TiO4

Instrument	Requested days	Allocated days	From	To
D3	3	3	27/09/2025	30/09/2025
D9	3	3	02/10/2025	05/10/2025
ORIENTEXPRESS	1	1	24/09/2025	25/09/2025

Abstract:

Fe2TiO4 is highly magnetostrictive below the ordering temperature of 120 K. While the structure is ferrimagnetic, there is a temperature region near 75 K where the moments are compensated. We propose to apply polarized neutrons at this compensation temperature to measure the magnetic structure. The goal is twofold - first to measure any moment canting that will be used to model the magnetic excitations, and second to establish the magnetic moment values to obtain information on orbital degrees of freedom in this material.

Experimental Report: D3 & D9
Characterizing the Magnetic and Nuclear Structures in Fe_2TiO_4

March 23, 2026

Sample and Alignment

A large high-quality single crystals of Fe_2TiO_4 (total mass: 5g, volume: 3000 mm^3) was used. The crystal was grown at Oxford University by the floating zone method, with quality confirmed by Laue X-ray diffraction and preliminary neutron diffraction on ALF. The sample was aligned and glued with the [110] direction vertically. Susceptibility measurements confirmed the magnetic/structural transition near 120 K and identified the compensation temperature at $T_{\text{comp}} \approx 75$ K where magnetization is minimized.

D9: Nuclear Structure Refinement

Large high-quality data sets were collected on D9 ($\lambda = 0.84$ Å) at 150 K (cubic phase, $Fd\bar{3}m$) and 2 K (tetragonal phase), with roughly 900 unique reflections and 100 symmetry-equivalent ones. Refinements confirm that the tetrahedral 8a site is almost pure Fe (98.7% Fe, 1.3% Ti), while the octahedral 16d site shows approximately 50:50 mixing (50.6% Fe, 49.4% Ti). This mixed occupancy places Jahn-Teller active Fe^{2+} on the octahedral site, providing a mechanism for magnetoelastic coupling.

D3: Spherical Neutron Polarimetry

The SNP experiment on D3 used $\lambda = 0.84$ Å with initial polarization $\approx 93\%$. Scattered neutrons were analyzed by a ^{3}He spin filter, regularly monitored on nuclear peaks, e.g. (888) at high momentum transfer ($\sin\theta/\lambda = 0.81$ with negligible magnetic contribution. Zero-field cooling resulted in significant depolarization throughout the whole temperature range of the magnetically ordered state. Surprisingly, field cooling with $H \parallel [110]$ only revealed weak depolarization effects allowing for a reasonable SNP data collection at different temperatures (10, 90 and 130 K) for the magnetic structure refinement.

Magnetic Structure Refinement

Full polarization matrices were refined against models with moments along [100], [111], and [110], the true orientation being unknown due to the cubic symmetry of the parent space group. Our correlated refinement between D3 and D9 data suggests $\mu \parallel [110]$ to be the best match. The final refinement at 10 K converged with $\chi^2_r = 73.5$ and RF(Int) = 5.88 (Fig. 1), while the other two models revealed χ^2_r of roughly 80 ($\mu \parallel [100]$) and 90 ($\mu \parallel [111]$), respectively.

The refined magnetic moments (spherical coordinates) are:

$$\mathbf{M}_{\text{Fe(tet)}} = [3.720\ 45.000^\circ\ 90.000^\circ]$$
$$\mathbf{M}_{\text{Fe(oct)}} = [3.852\ 135.000^\circ\ -90.000^\circ]$$

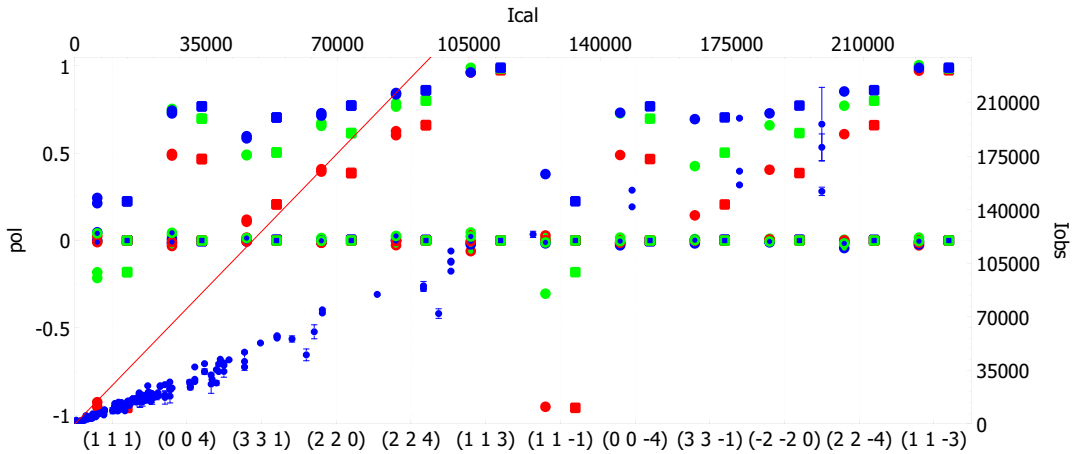


Figure 1: Final results of the [110] model corefinement against D3 and D9 data at 10 K. Calculated vs. observed integrated intensities are shown as blue dots which lie very close to the "perfect" red line. Observed polarization matrix elements are shown in colored dots (reg/green/blue marker face for P_i along $x/y/z$ and red/green/blue marker edge for P_f along $x/y/z$), while calculated ones are shown in squares using the same color code. The respective Bragg reflections are labeled along the horizontal axis.

12 magnetic domains were included in the [110] model: 6 for the different ¡110¿ directions and their corresponding 180° domain. With respect to the direction of applied magnetic field during cooling, 8 domains have the same inclined orientation between the ordered ferrimagnetic moment and $H \parallel z$, 2 are (anti)parallel to z and 2 are perpendicular to z. The first as well as the latter were contrained to have the same population, respectively, leading to 3 refinable parameters, since the sum of the domain population is constrained to 1.

Discussion of Depolarization

Strong depolarization below 150 K, even at T_{comp}, indicates the simple two-sublattice ferrimagnetic model with equal and opposite moments is not fully realized. However, in-field cooling with $H \parallel$[110] suppresses beam depolarization throughout the measured temperature range, which can be explained by an induced compensation of magnetic sublattices or the creation of sufficiently small magnetic domains that may avoid depolarization. The investigation of related compounds may shed light onto this interesting observation.

Group Theory Analysis and Coupling to Strain

To understand the magnetoelastic coupling in Fe_2TiO_4, a group theory analysis was performed using the ISOTROPY software suite. The analysis aimed to identify invariant terms in the free energy coupling the magnetic order parameter to the strain modes associated with the Jahn-Teller distortion.

- The magnetic structure refines with symmetry **mGM4+**, a 3D irreducible representation with order parameters η_1, η_2, η_3. The experimentally observed moment direction [110] corresponds to the order parameter direction $(\eta_1, \eta_2, \eta_3) = (a, a, 0)$.
- The tetragonal shear strain (E_g symmetry, corresponding to the $c_{11} - c_{22}$ elastic constant) transforms as the 2D irreducible representation **GM3+** with order parameters ϵ_4, ϵ_5.

- The orthorhombic shear strain (T_{2g} symmetry, corresponding to the c_{44} elastic constant) transforms as the 3D irreducible representation **GM5+** with order parameters $\epsilon_4, \epsilon_5, \epsilon_6$.

The linear-quadratic coupling terms (linear in strain, quadratic in magnetization) in the free energy were derived:

- For coupling between **mGM4+** ($\boldsymbol{\eta}$) and **GM3+** ($\boldsymbol{\epsilon}$):

$$\mathcal{F}_{\text{coup}}^{(3+)} \propto (0.500)\eta_1^2\epsilon_4 + (0.866)\eta_1^2\epsilon_5 + (0.500)\eta_2^2\epsilon_4 - (0.866)\eta_2^2\epsilon_5 - (1.000)\eta_3^2\epsilon_4 \quad (1)$$

- For coupling between **mGM4+** ($\boldsymbol{\eta}$) and **GM5+** ($\boldsymbol{\epsilon}$):

$$\mathcal{F}_{\text{coup}}^{(5+)} \propto \eta_1\eta_2\epsilon_4 + \eta_1\eta_3\epsilon_6 + \eta_2\eta_3\epsilon_5 \quad (2)$$

Key result: Inserting the experimentally determined order parameter direction $(\eta_1, \eta_2, \eta_3) = (a, a, 0)$ into these invariants yields:

- For **GM3+** coupling: the expression simplifies to $a^2\epsilon_4$, predicting a non-zero macroscopic strain ϵ_4 — precisely the tetragonal distortion observed in our D9 data.
- For **GM5+** coupling: the expression vanishes identically for this order parameter direction.

This analysis confirms that the magnetic order parameter (mGM4+) couples selectively to the tetragonal shear mode (GM3+), and that the observed [110] moment direction naturally selects this coupling channel. This provides a quantitative explanation for the spontaneous strain observed in D9 and confirms that the Jahn-Teller active Fe^{2+} on the octahedral site drives the structural distortion via its coupling to the magnetic order.

Acknowledgements

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