

Supporting Information

Tetrahedral Chain Ordering and Low Dimensional Magnetic Lattice in a New Brownmillerite $\text{Sr}_2\text{ScFeO}_5$

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Experimental Section

Synthesis. The brownmillerite $\text{Sr}_2\text{ScFeO}_5$ was synthesized by the standard solid-state reaction route, using in the stoichiometric amounts of SrO (99.9%) Fe_2O_3 (99.99%), and Sc_2O_3 (99.99%). All these oxides were carefully mixed in a glove box under nitrogen atmosphere. The mixture was then pressed in the form of bars, which are subsequently introduced into an alumina finger and sealed in an evacuated quartz tube. The sample was then heated at 1200°C for 24 hours followed by a slow cooling to room temperature in 48 hours.

Raman Spectroscopy. Raman spectra of the powder sample were recorded on a Horiba Labram HR Evolution, using a 532 nm excitation, ca. 5 mW on the sample, in 60 s with 2 accumulations.

Crystal Structure Characterization. X-ray diffraction pattern is collected at room temperature in an X'pert Pro diffractometer working with the CuK_α radiation. The samples for electron microscopy were prepared by dispersing of a small quantity of powder in butanol and depositing obtained suspension on Ni carbon holly grid. Lattice constants and space group were deduced from Electron Diffraction data collected on JEOL 2010. The study High-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) and the atomic level energy-dispersive X-ray spectroscopy (EDS) analyses (mapping) were performed on a JEOL ARM-200F cold FEG probe image aberration corrected microscope operated at 200 kV and equipped with a large solid-angle CENTURIO EDX detector. The oxygen stoichiometry was determined by means of cerimetric titration method [1], the tolerance on the measurement being estimated to 5%.

Magnetic Measurements and Heat Capacity. Direct current (dc) magnetization and ac susceptibility measurements were performed by using a Quantum Design superconducting quantum interference device (MPMS, Quantum Design). Specific-heat measurement was performed by using the two-tau time-relaxation method using the PPMS in zero magnetic field.

Dielectric Measurement. The dielectric measurement were carried out as a function of temperature and magnetic field for various frequencies using an LCR meter (Agilent 4284A) with a home-made sample insert for a Physical Properties Measurement System (PPMS,

Quantum Design). Silver paint was used to make the parallel plate capacitor of a pressed rectangular shaped polycrystalline sample.

Mössbauer spectroscopy. The ^{57}Fe Mössbauer spectra at 4.2 K was recorded in transmission mode, using a conventional spectrometer operating in the constant acceleration mode from -2.5 to 2.5 mm/s. $^{57}\text{Co/Rh}$ was used as the γ -ray source.

[1] T. Motohashi, B. Raveau, M. Hervieu, A. Maignan, V. Pralong, N. Nguyen, V. Caignaert, J. Phys. Cond. Mat. 18 (2006) 2157-2171.

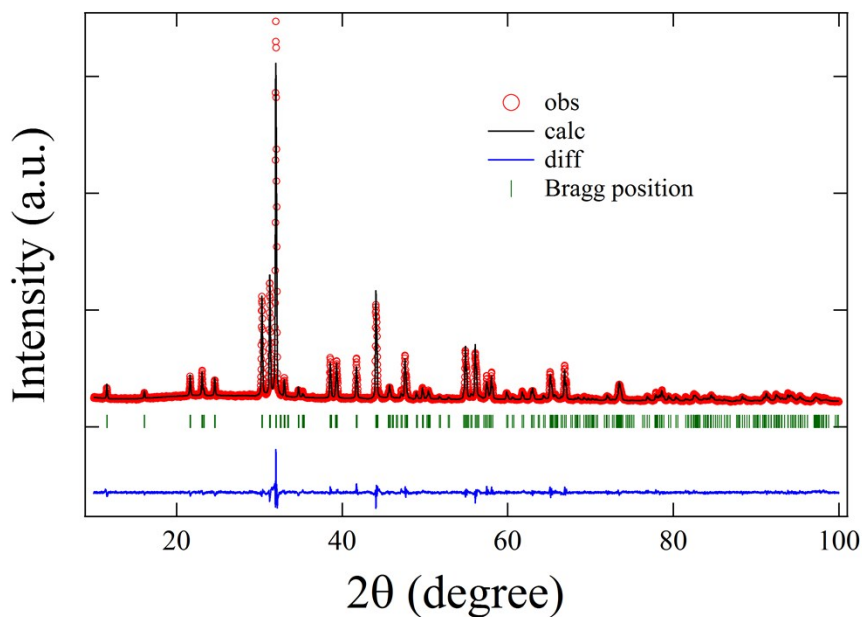


Figure S1. Rietveld refinement fit of the room temperature powder X-ray diffraction pattern for $\text{Sr}_2\text{ScFeO}_5$.

Table 1. Atomic and structural parameters as determined from the Rietveld refined using the room temperature XRD pattern.

	x	y	z	B _{iso}	Occ.
Sr	0.0192(1)	0.1122(1)	0.5157(1)	2.2(1)	1.0
Sc	0	0	0	1.4(1)	1.0
Fe	0.9323(4)	0.25	0.988(1)	1.2(1)	1.0
O1	0.271(2)	0.9864(5)	0.287(2)	0.7	1.0
O2	0.0693(1)	0.1447(4)	0.0352(2)	0.3	1.0
O3	0.823(2)	0.25	0.660(2)	0.5	1.0

$\chi^2 = 4.59$, $R_p = 2.04\%$, $R_{wp} = 2.94\%$, $R_{Bragg} = 5.47$

Space group: *Ibm2*, $a = 5.8965$ (1) Å, $b = 15.2769$ (4) Å and $c = 5.7213$ (1) Å

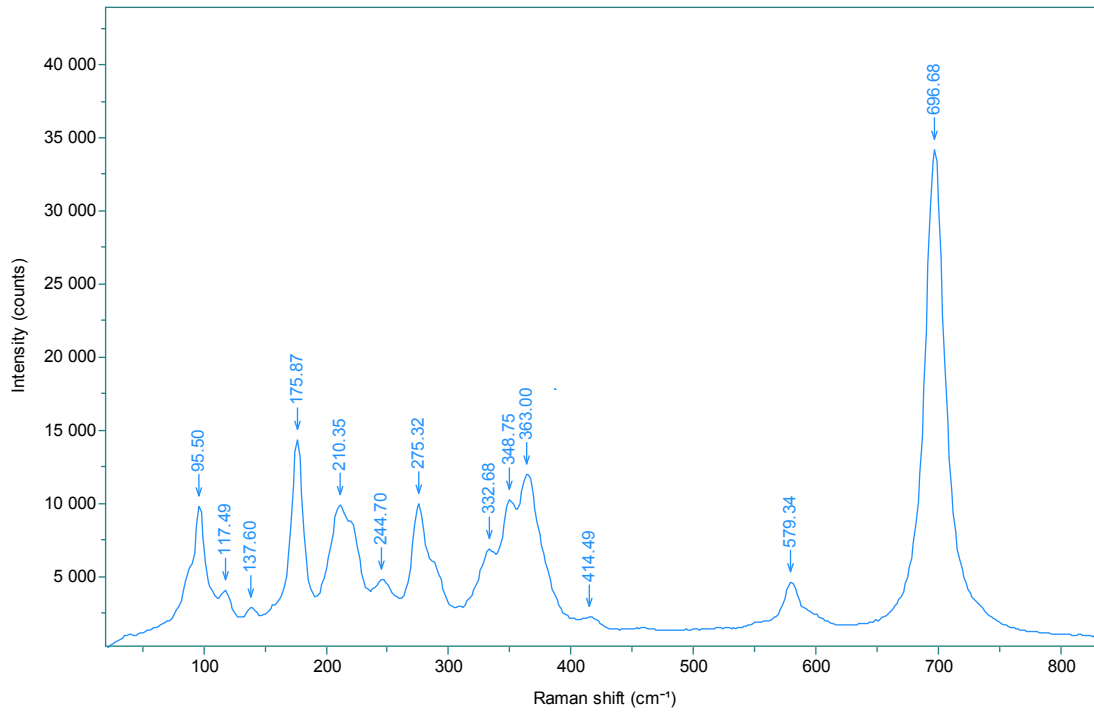


Figure S2. Room temperature Raman spectrum of Sr₂ScFeO₅.

The Raman spectrum shows several peaks of comparable intensities below 400 cm^{-1} with only two peaks above 420 cm^{-1} , a main intense band at 697 cm^{-1} and a small band at 579 cm^{-1} .

Although the Raman spectrum of this compound has not been reported before, it can be compared to that of the related Brownmillerite $\text{SrFeO}_{2.5}$ [1]. The spectrum of $\text{SrFeO}_{2.5}$ shows several peaks between 400 and 600 cm^{-1} which are nearly absent in the spectrum of $\text{Sr}_2\text{ScFeO}_5$. Following the studies of several iron oxide compounds, bands in this region were assigned to the presence of FeO_6 octahedra while FeO_4 tetrahedra are expected between 660 and 720 cm^{-1} [2]. The absence of noticeable bands between 400 and 570 cm^{-1} likely indicates the absence of iron in octahedral environment. From the computed values of the A_g modes for $\text{CaFeO}_{2.5}$ [1], the bands at 579 and 697 cm^{-1} can be attributed to the vibrations of iron in tetrahedral environment. The strong band at 697 cm^{-1} and its large relative intensity compared to bands around 400 cm^{-1} is consistent with the prominent presence of iron in tetrahedral environment.

Regarding the presence of scandium in the structure, Sc_2O_3 was reported to yield bands around 420 cm^{-1} [3]. Similarly, scandium in octahedral environment is expected to yield bands in the same region [4]. Some of the bands between 320 and 420 cm^{-1} must thus be indicative of ScO_6 groups.

References

- [1] Andrea Piovano, Monica Ceretti, Mark R Johnson, Giovanni Agostini, Werner Paulus and Carlo Lamberti, *J. Phys.: Condens. Matter* 2015, 27, 225403.
- [2] J. Kreisel, G. Lucazeau and H. Vincent, *Journal of Solid State Chemistry*, 1998, 137, 127–137.
- [3] J A. Ubaldini and M. M. Carnasciali, *Journal of Alloys and Compounds*, 2008, 454, 374–378.
- [4] W. W. Rudolph and C. C. Pye, *J. Phys. Chem. A*, 2000, 104, 1627–1639.

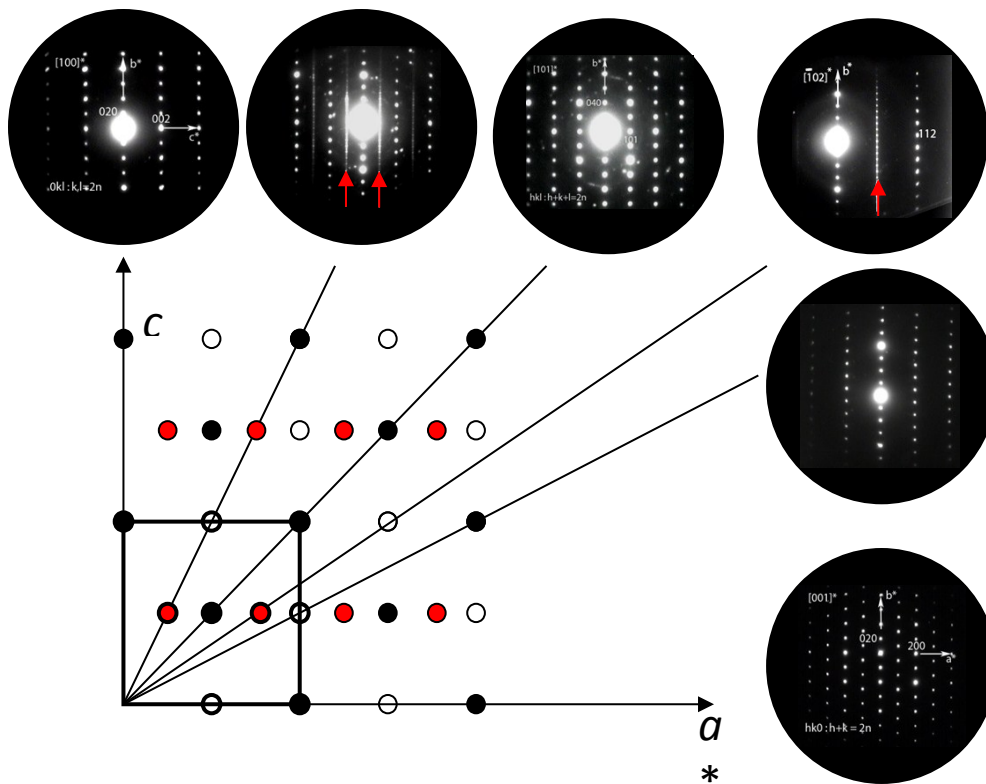


Figure S3a. Room temperature electron diffraction patterns along different zone axis and the reciprocal space reconstruction for $\text{Sr}_2\text{ScFeO}_5$ [$\text{Ibm}2(\alpha 00)0s0$ superspace group with the modulation vector $q = \alpha a^* + \beta b^*$].

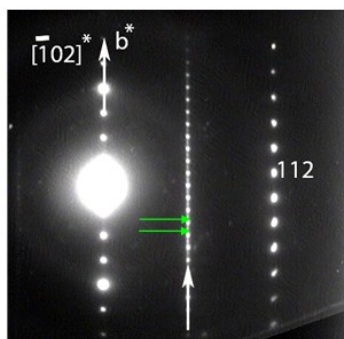


Figure S3b. Doubling of the cell parameters along the stacking b^* axis is highlighted for brownmillerite $\text{Sr}_2\text{ScFeO}_5$.

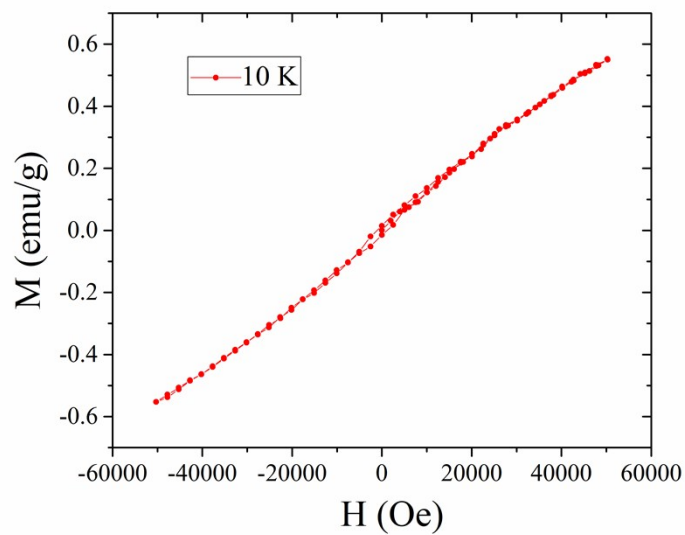


Figure S4. Magnetic field variation of the magnetization for the brownmillerite $\text{Sr}_2\text{ScFeO}_5$.

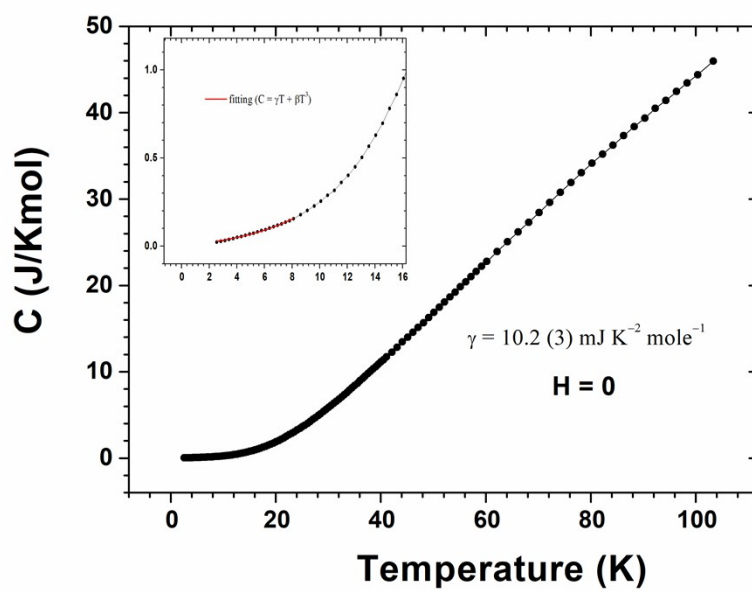


Figure S5. The temperature dependence of the specific heat data for the brownmillerite $\text{Sr}_2\text{ScFeO}_5$.

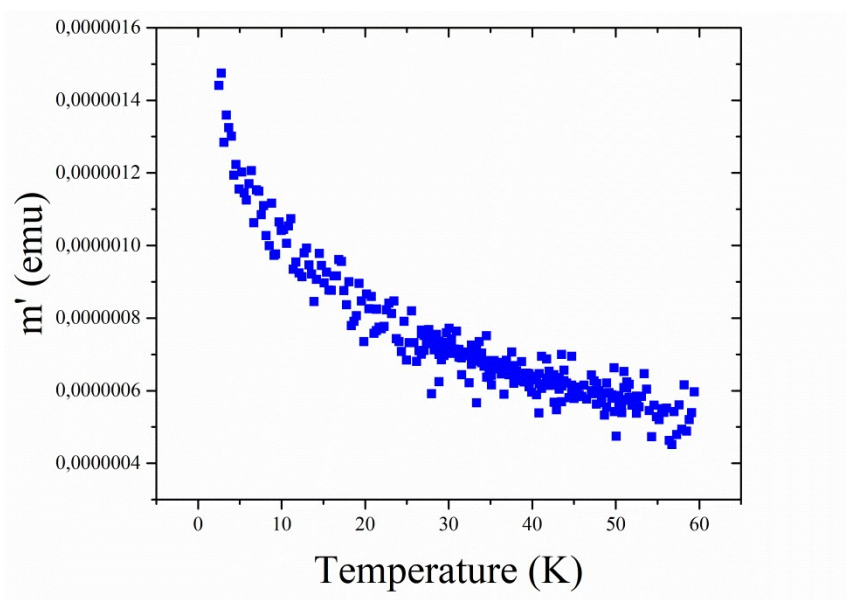


Figure S6. Temperature variation of the ac susceptibility ($f = 999$ Hz, ac excitation field = 1 Oe) for the brownmillerite $\text{Sr}_2\text{ScFeO}_5$.

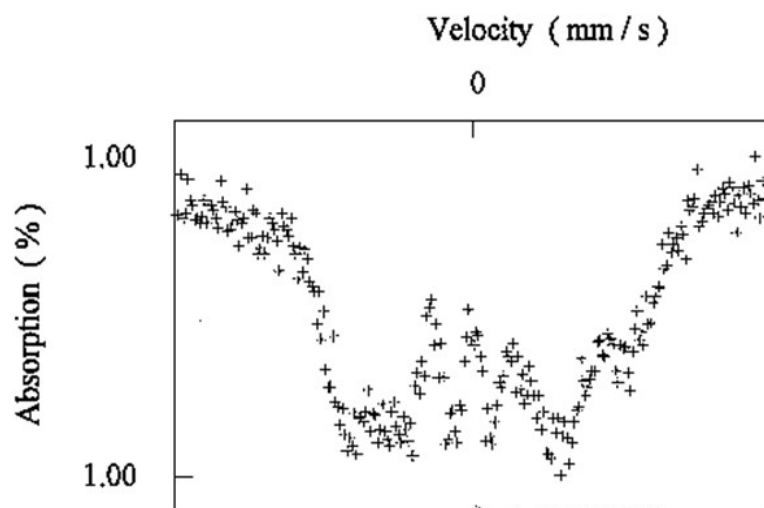


Figure S7. The Mössbauer spectrum at 4.2 K for the brownmillerite $\text{Sr}_2\text{ScFeO}_5$, shows the absence of magnetic order (absence of a well-defined sextuplet) at low temperature.