Experimental report

25/06/2020

Proposal: 5-31-2645 **Council:** 10/2018

Title: Study of room-temperature spin reorientation and low-temperature magnetic structure in Sm0.7Tm0.3FeO3 perovskite

Research area: Materials

This proposal is a new proposal

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Samples: 152-Sm0.70Tm0.30FeO3
La-Mn-Nb-O

Instrument	Requested days	Allocated days	From	To
D1B	2	1	26/06/2019	28/06/2019

Abstract:

Spin reorientation (SR) transitions which consist in rotation of the easy axis of magnetic moments are well known for numerous metal transition oxides. SmFeO3 is one example of perovskite displaying SR above room temperature (RT) that can be tuned to room temperature by mixing samarium with other lanthanides. In the Sm1-xTmxFeO3 system, magnetization measurements of Sm0.70Tm0.30FeO3 suggest SR between 205 K and 353 K. We have acquired 152Sm2O3 to prepare a sample without high neutron absorption as natural samarium and we are asking for neutron diffraction experiments using D1B, to collect thermodiffraction data versus temperature together with measurements at fixed temperatures for Rietveld analysis. We wish 48 hours on D1B equipped with a

Introduction

Multiferroic and magnetoelectric materials have recently gained notoriety due to their possible technological applications. Coupling electric and magnetic properties is an important feature in a material, and knowing the order parameters of their properties is of great interest **[1]**. The novel field of spintronics is also concerned with the behaviour of the electrons spins and also with the anisotropy of promising materials **[2]**. Accurate control over the magnetic structure of a material can thus provide both valuable knowledge on materials design and possible applications in emerging fields. The rare-earth orthoferrite perovskites are interesting candidates for these uses, since they display a magnetic transition called spin reorientation (SR). This phenomenon consists in a rotation of the easy axis of the magnetic coupling and is closely related to the magnetic anisotropy of the material. We synthesized a perovskite with formula $Sm_{0.7}Tm_{0.3}FeO_3$ with the goal of obtaining this SR transition at room temperature, which is valuable for technological uses. Determining the magnetic structure at each temperature would provide insight into the magnetic anisotropy of the material, and also into possible magnetic couplings that may lead or suggest other order parameters.

Experimental

We synthesized a perovskite with formula $^{152}Sm_{0.70}Tm_{0.30}FeO_3$. Natural samarium has a very high absorption cross-section for thermal neutrons, while ^{152}Sm is noticeably less absorbent **[3]**. The synthesis was performed using isotopically pure $^{152}Sm_2O_3$ to ensure the lowest possible absorption while retaining the crystal structure and the magnetic properties. Preliminary structural characterization was performed with XRPD at room temperature using Rietveld refinement. Unit cell is orthorhombic and the crystal structure belongs to the space group *Pbnm*, as expected. The experiments performed under proposal 5-31-2645 were aimed at the proper determination of magnetic structure in a wide temperature range with the use of instrument **D1B**. The temperature-dependent data of **D1B** can provide detailed information about the particular SR transition present in our sample. Patterns were collected both while cooling and warming the samples in order to maximize data output. To further characterize the magnetic structures, we also collected four diffraction patterns at constant temperatures far from observed magnetic transitions. All patterns were collected with $\lambda = 2.528$ Å.

Results and discussion

As mentioned before, all NPD data confirmed that the space group *Pbnm* is suitable for the description of the structure. Within this space group, the interpretation of magnetic structures with propagation vector $\boldsymbol{k} = (0, 0, 0)$ can be performed using the formalism enunciated by Bertaut **[4]**. Essentially, all the magnetic structures observed can be correctly described by using G-type arrangements for the Fe^{3+} magnetic moments orderings. **Figures 1a** and **1b** show NPD patterns from **D1B** for $^{152}Sm_{0.70}Tm_{0.30}FeO_3$ at two different temperatures, above (450 K) and below (1.5 K) the room-temperature SR transition, together with their corresponding Rietveld analysis results. Since the Néel temperature of orthoferrites is generally well above 450 K **[5]**, both diffraction patterns have contributions from both crystal and magnetic structures. The main change between these two patterns is the relation of intensities between the first two peaks. For the determination for magnetic structures, Rietveld analysis was carried out using the FULLPROF suite software, with the assistance of the BasIreps for generation of suitable candidates for magnetic structure refinement. After careful analysis, we arrived to the conclusion that the observed magnetic structures can be described as G_x and G_z AFM orderings of the Fe^{3+} substructure above and below the SR transition, respectively. No additional contributions from the lanthanide magnetic moments were observed, discarding possible long-range orderings that had been suggested in previous reports **[6]**.

D1B data allowed us to follow the SR closely in the 200 K-350 K range. The change in the intensities of the first two peaks can be seen in the temperature-dependent diffraction data shown in **Figure 2**. By performing Rietveld refinements over different integrations of this data, we were able to follow the rotation of the Fe^{3+} magnetic moments. **Figure 3** shows the refined variation of the *x* and *z* components of the magnetic moment of one Fe^{3+} cation representative for the rotation of all cations in the substructure. The Fe^{3+} substructure displays a mixed G_x-G_z ordering during the transition. The exchange of the module from one direction to the other signals the SR transition. As it can be seen, there is perfect correspondence with a drop in magnetization observed in a ZFC magnetization curve, included in **Figure 3a**.

Conclusions

The room temperature SR transition in $Sm_{0.70}Tm_{0.30}FeO_3$ was characterized by NPD in the D1B instrument. Thanks to the large output and high quality of the data, a very detailed picture of the magnetic structure at every temperature was obtained. The reorientation consists in a gradual rotation from a G_x to a G_z ordering in the Fe^{3+}

substructure. There are no indications of long range ordering in the Sm^{3+}/Tm^{3+} substructure. A manuscript covering these and other relevant findings in the system is in preparation.

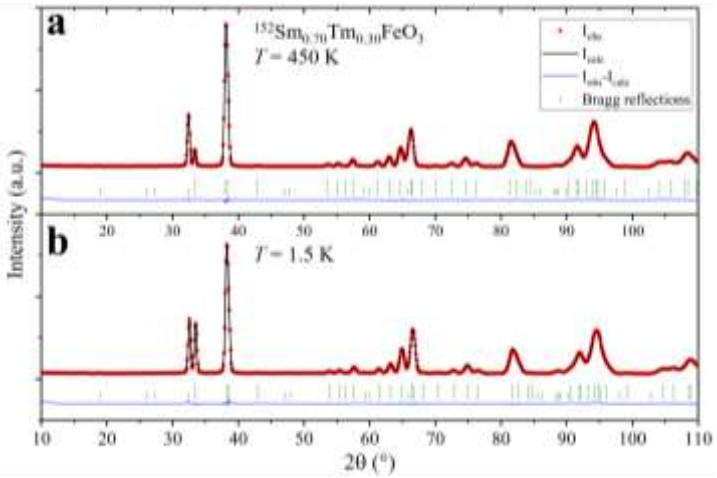

Figure 1) D1B NPD patterns for $^{152}Sm_{0.70}Tm_{0.30}FeO_3$ at 450 K **(a)** and 1.5 K **(b)**.

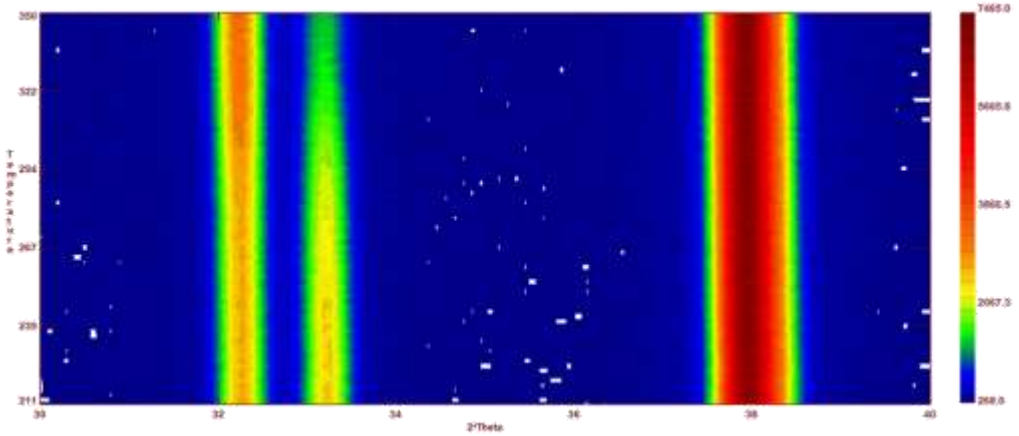

Figure 2) D1B temperature-dependent NPD for $^{152}Sm_{0.70}Tm_{0.30}FeO_3$. Change of intensities in the first two peaks signals the SR transition.

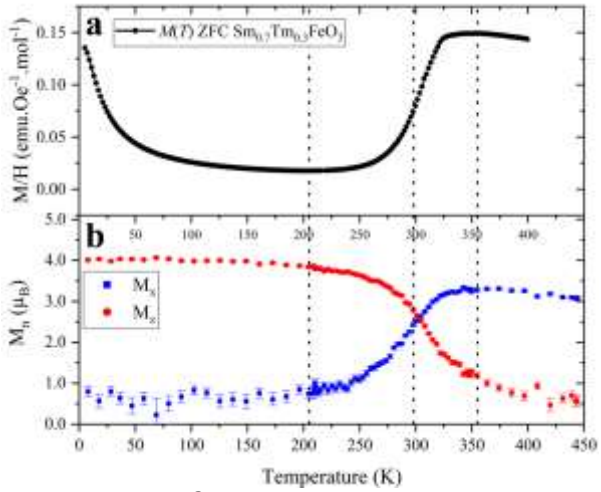

Figure 3) Refined magnetic moments for the Fe^{3+} sublattice during the SR transition together with ZFC magnetization data.

[1] Eerenstein, W. et al. (2006). Multiferroic and magnetoelectric materials. *Nature* **442**, 759-765.

[2] Jungwirth, T. el al. (2016). Antiferromagnetic spintronics. *Nat. Nanotechnol.*, **11**(3), 231-241.

[3] Sears, V. F. (1992). Neutron scattering lengths and cross sections. *Neutron News*, **3**(3), 26-37.

[4] Bertaut, E. F. (1968). Representation analysis of magnetic structures. *Acta Crystallogr. A*, **24**(1), 217-231.

[5] Kuo, C. Y. et al. (2014). *k*= 0 magnetic structure and absence of ferroelectricity in $SmFeO_3$. *Phys. Rev. Lett.*, **113**(21), 217203.

[6] Khan, A. A. et al. (2019). Tuning of spin reorientation temperature of $SmFeO_3$ by doping of Tm^{3+} ion: Role of exchange interaction between 4*f* & 3*d* electrons. *J. Alloy. Compd.*, **808**, 151603.