

# Experimental report

03/04/2024

**Proposal:** 5-31-2928

**Council:** 10/2022

**Title:** Investigation of the crystal and magnetic structures of ScFe<sub>2</sub>D<sub>x</sub> type compounds

**Research area:** Physics

**This proposal is a new proposal**

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**Samples:** ScFe<sub>2</sub>D<sub>x</sub> and (Sc,Nb)Fe<sub>2</sub>D<sub>x</sub>

Instrument	Requested days	Allocated days	From	To
D1B	4	3	14/04/2023	17/04/2023

## Abstract:

From macroscopic magnetic measurements we have observed both a large change of the magnetization and huge unit cell expansion upon insertion of hydrogen/deuterium in ScFe<sub>2</sub>. Here we intend to investigate the ScFe<sub>2</sub> type hydrides, determine the hydrogen location within the crystal and its effects on the crystal lattice distortion. We are also interested in the determination of the magnetic structure in order to know the magnetic moments magnitudes on the two inequivalent Fe atomic positions. A further aim is to investigate the effect of the local atomic environment on the magnitude of the magnetic moments of the Fe atoms. The study starting with ScFe<sub>2</sub>D<sub>x</sub> will be extended to the Sc rich side of the (Sc,Nb)Fe<sub>2</sub>D<sub>x</sub> compounds. The possible occurrence of a magnetic moment on the Sc position will also be investigated. We request for neutron diffraction experiments at 1.28Å in order to explore a wider range of the reciprocal space and determine the crystal structure and experiments at 2.52Å at low temperature to investigate the magnetic structure in a series of selected (Sc,Nb)Fe<sub>2</sub>D<sub>x</sub> compounds for a total of 4 days on D1B instrument.

## Investigation of the crystal and magnetic structures of (Sc,Nb)Fe<sub>2</sub>D<sub>x</sub> type compounds

### Experiments

Neutron powder diffraction (NPD) experiments were performed on the high-intensity two-axis powder diffractometer D1B with a detector angular range coverage  $5^\circ \leq 2\theta \leq 128^\circ$  which is especially suited for magnetic structure determination. About 2 g of fine powder were introduced into a cylindrical vanadium container ( $D = 6$  mm,  $H = 5$  cm) and mounted on the stick of a He cryostat, whose contribution to the diffraction patterns was eliminated using a radial oscillating collimator. Several diffractograms were collected at selected temperatures ranging between 1.5 and 300 K. The data were collected using a <sup>3</sup>He multicounter containing 1280 detection cells with a step of  $0.1^\circ$  between neighbouring cells. A neutron incident wavelength of 2.52 Å was selected by a (002) Bragg reflection of a pyrolytic graphite monochromator, the take-off angle being  $44.2^\circ$  in  $2\theta$ . All measurements were performed upon heating after a stabilization time of 3 minutes with typical acquisition times of 5 minutes per isotherm. Due to the high flux available on the instrument, a second set of diffraction patterns was recorded *in situ* every 5 K while ramping the temperature from 1.5 K to 300 K in order to follow the thermal evolution of the lattice parameters and the possible presence of magnetoelastic phenomena across the magnetic transition.

### Preliminary results

NPD measurements were carried out to determine the crystalline and magnetic structure of (Sc,Nb)Fe<sub>2</sub>D<sub>x</sub> compounds, as well as to localize hydrogen in the crystal structure of these compounds. NPD patterns were collected at room temperature with incident wavelength of 1.28 Å. Figure 1 presents an example of Rietveld refinement for ScFe<sub>2</sub>D<sub>3.14</sub>. The presence of iron impurity in the ScFe<sub>2</sub>D<sub>3.14</sub> sample was observed in the initial ScFe<sub>2</sub> intermetallic with a very small proportion (less than 2%). Achieving correct refinement for ScFe<sub>2</sub>D<sub>3.14</sub> required the inclusion of an impurity phase (Sc with a hexagonal structure). Analysis of the refinement results revealed that the hexagonal MgZn<sub>2</sub>-type structure ( $P6_3/mmc$  space group) remains preserved upon hydrogen (H) or deuterium (D) insertion for all (Sc,Nb)Fe<sub>2</sub> compounds studied. In this hexagonal structure, deuterium localization was tested at the four sites richest in neighboring Sc atoms, namely the four Wyckoff positions  $24l$ ,  $12k$ ,  $6h$  and  $6h$ . Deuterium partially occupies the three sites D1( $24l$ ),

D2(12*k*) and D4(6*h*), but site D3(6*h*) appears to be less favorable and is almost unoccupied in the ScFe<sub>2</sub>D<sub>3.14</sub> compound.

Figure 2 represents the Rietveld refinement of the NPD pattern for ScFe<sub>2</sub>D<sub>3.14</sub> recorded at a low temperature of 3.5 K with a wavelength of 2.52 Å. All peaks are indexed in the hexagonal structure, *P6<sub>3</sub>/mmc* space group. The coincidence of the nuclear and magnetic unit cells indicates the ferromagnetic behavior of this compound. Various ferromagnetic configurations were tested to refine the magnetic structure, including one where Fe magnetic moments align along the *c*-axis, as observed in ScFe<sub>2</sub> at low temperatures, and another where the magnetic moments align in the basal plane of the hexagonal structure. The Fe magnetic moments have been refined independently for both Fe(2*a*) and Fe(6*h*) sites. For ScFe<sub>2</sub>D<sub>3.14</sub> at 3.5 K, the deduced magnetic moments of Fe at crystallographic positions 2*a* and 6*h* are 1.94 and 2.36 μ<sub>B</sub>, respectively. These values are twice larger than the iron magnetic moment observed for the ScFe<sub>2</sub> intermetallic at 1.5 K (0.96 and 1.04 μ<sub>B</sub> for Fe(2*a*) and Fe(6*h*), respectively). The insertion of deuterium induces remarkable changes in magnetic properties, such as a large increase in iron magnetic moment and a change of the easy magnetization direction.

**These results are part of the PhD thesis work of Soufiane El Bidaoui (Université Grenoble Alpes).** They have been presented as an oral contribution in the *XV* International Conference on Crystal Chemistry of Intermetallic Compounds [1] and in the multidisciplinary scientific congress “Journées de la Diffusion Neutronique 2023” [2].

#### **References:**

[1] S. El Bidaoui, L.V.B. Diop and O. Isnard, “Structural and magnetic properties of (Sc,Nb)Fe<sub>2</sub> intermetallics and their deuterides”, the *XV* International Conference on Crystal Chemistry of Intermetallic Compounds (IMC-*XV*2023), (25-27 September 2023), Ukraine – online.

[2] S. El Bidaoui, L. V. B. Diop and O. Isnard, “Structural and magnetic properties of (Sc,Nb)Fe<sub>2</sub> intermetallics and their deuterides”, Journées de la Diffusion Neutronique (JDN2023), (09-12 October 2023), France.

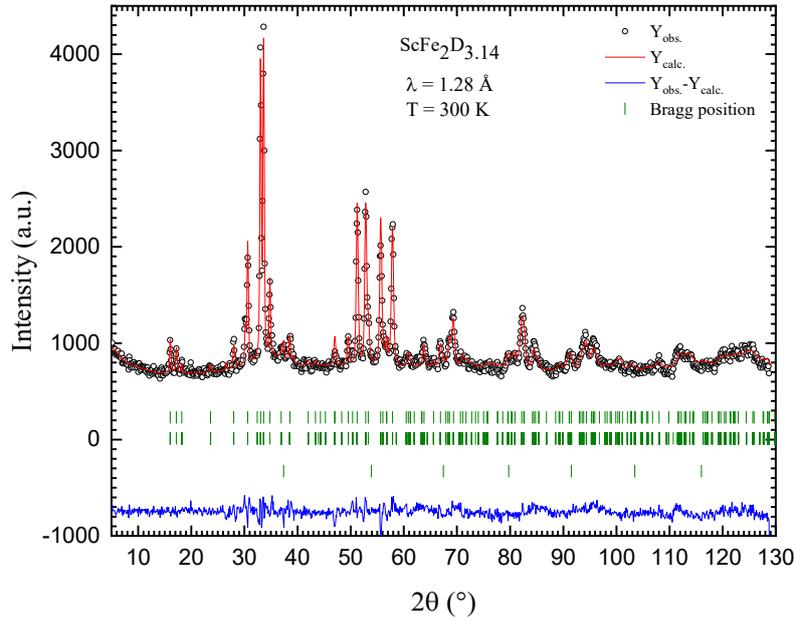


Figure 1. Rietveld refinement of the neutron diffraction pattern of  $\text{ScFe}_2\text{D}_{3.14}$  at 300K with wavelength  $\lambda=1.28\text{\AA}$  ( $R_p = 3.93\%$ ,  $R_{wp} = 5.01\%$ ,  $\chi^2 = 2.21$ ,  $R_{\text{Bragg}} = 11$ ,  $R_{\text{Mag}} = 17$ ). Black dots represent the experimental diffractogram. The red line represents the calculated diffractogram. The first two series of vertical lines represent the positions of the Bragg peaks of the nuclear and magnetic phase of  $\text{ScFe}_2\text{D}_{3.14}$  respectively. The last series corresponds to the  $\alpha\text{-Fe}$  impurity with a very low fraction, less than 2%. The difference between the experimental and calculated profile is shown at the bottom of the figure in blue.

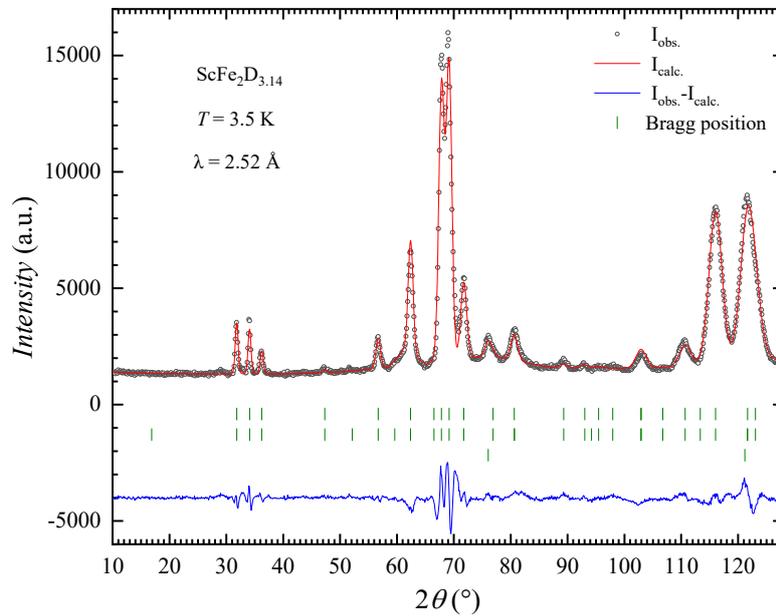


Figure 2. Rietveld refinement of the neutron diffraction pattern of  $\text{ScFe}_2\text{D}_{3.14}$  at 3.5K with wavelength  $\lambda=2.52\text{\AA}$  ( $R_p = 14.2\%$ ,  $R_{wp} = 13.6\%$ ,  $\chi^2 = 3.12$ ,  $R_{\text{Bragg}} = 4.59$ ,  $R_{\text{Mag}} = 9.64$ ). The two series of vertical lines represent the positions of the nuclear and magnetic phase Bragg peaks of  $\text{ScFe}_2\text{D}_{3.14}$  respectively. The third series corresponds to the  $\alpha\text{-Fe}$  impurity.