Experimental report

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Proposal: **5-31-2719**

Council: 10/2019

Title: Neutron diffraction study of the itinerant electron system Sc1-xNbxFe2

Research area: Physics

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Samples: (Sc,Nb)Fe2

Instrument	Requested days	Allocated days	From	To
D1B	4	2	21/08/2020	23/08/2020

Abstract:

We propose to determine the magnetic structure of the Sc1-xNbxFe2 compounds which are exhibiting interesting and unusual itinerant electron magnetic behavior. We are also interested in the determination of the composition dependence of the magnetic structure. The possible different behavior of the Fe atoms on the inequivalent crystal sites will also be investigated. A further aim is to determine the effect of the local atomic environment on the magnitude of the magnetic moments of the Fe atoms.

Neutron diffraction study of the itinerant electron system $Sc_{1-x}Nb_xFe_2$

Introduction

The Laves phase compounds MFe_2 where M can be a 3*d*, 4*d*, 5*d* elements are showing various kind of crystal structure and magnetism depending upon the species of M. Furthermore deviation from stoechiometry can also significantly modify the magnetic properties of the compounds. The MFe_2 alloys have attracted much interest due to the complexity of their magnetic phase diagram resulting from the itinerant character of the 3*d* electron magnetism. Indeed, they have been found to present both antiferromagnetic and ferromagnetic ordering and large metamagnetic transition have been reported for some composition. This is promising since such metamagnetic behavior can lead to significant change of magnetic entropy. The magnetocaloric effect around room temperature is of outmost interest in the nowadays context of intense researches worldwide for new high performance magnetocaloric materials for room temperature magnetic refrigeration. In addition to the fundamental physics of itinerant electron magnetism, this potential for magnetocaloric application is a further interest for these materials.

$ScFe_2$ is a ferromagnet with a Curie temperature of about 535 K whereas $NbFe_2$ is an itinerant system close to quantum criticality. It is established that as function of the Nb/Fe stoichiometry, that is as function of the control parameter *y* in $Nb_{1-y}Fe_{2+y}$, three distinct ground state phases are traversed across a narrow compositional range. Nb-rich material displays a ferromagnetic-like ground state, material close to stoichiometry orders with a low-moment spin density wave, while Fe-rich material is ferromagnetically ordered. In between Nb-rich and stoichiometric material there is a quantum critical point/regime (QCP). Recently, the bulk nature of these distinct magnetic phases has been established by means of μSR and ESR experiments. The identification of QCP in $NbFe_2$ opens up the new phenomenon of quantum tricriticality for experimental studies in a whole class of systems with buried or avoided ferromagnetic QCP. This provides a fresh perspective on other materials with the same universality, including prototypical heavy-fermion materials, in which multiple and competing low-energy scales have in the past prevented the detection of a QCP and obscured the investigation of its consequences.

Experiments

Neutron powder diffraction (NPD) experiments were performed on the high-intensity two-axis powder diffractometer D1B with a detector angular range coverage $5° \leq 2\theta \leq 128°$ which is especially suited for magnetic structure determination. About 3 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 3He multicounter containing 1280 detection cells with a step of 0.1° 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° in 2θ. All measurements were performed upon heating after a stabilization time of 3 minutes with typical acquisition times of 20 minutes per isotherm. Due to the high flux available on the instrument, a second set of diffraction patterns was recorded *in situ* every 3.5 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

Rietveld analysis of the diffraction data was done using the FullProf suite software package, which allows the simultaneous refinement of structural and magnetic profiles.

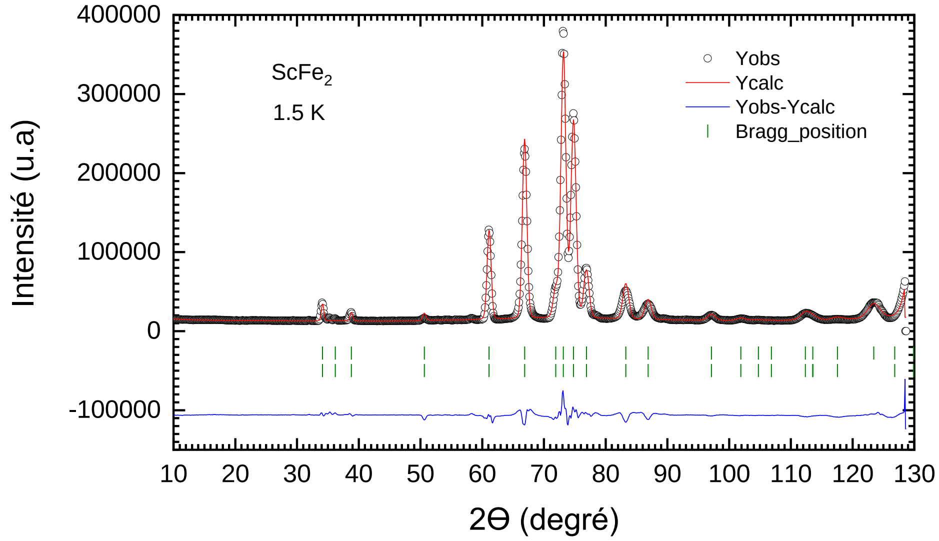

Figure 1: Refinement of the neutron diffraction pattern recorded at 1.5 K for $ScFe_2$. The top and second rows of Bragg markers are referring to the nuclear and magnetic contributions of the $ScFe_2$ phase.

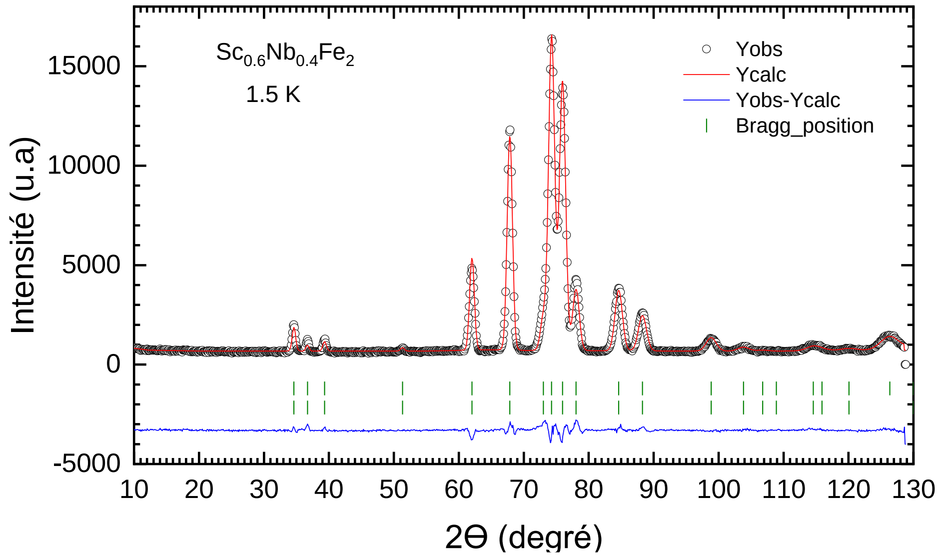

Figure 2: Refinement of the neutron diffraction pattern recorded at 1.5 K for $Sc_{0.6}Nb_{0.4}Fe_2$. The top and second rows of Bragg markers are referring to the nuclear and magnetic contributions of the $Sc_{0.6}Nb_{0.4}Fe_2$ phase.

Representative patterns of the Rietveld refinements are shown in Figures 1-2. The analysis of the NPD patterns obtained at 1.5 K shows that the crystallographic structure is retained at low temperature. The 1.5 K diffractograms present the same Bragg peaks as the spectra collected in the paramagnetic regime. No additional magnetic reflections are detected, which implies that the magnetic unit cell coincides with the crystallographic one. $ScFe_2$ and $Sc_{0.6}Nb_{0.4}Fe_2$ exhibit a ferromagnetic (FM) ordering of Fe moments. These FM structures only modify intensities of the nuclear peaks and magnetic scattering was found on several lines. The Fe magnetic moments have been refined independently for the two sites and align along the six-fold symmetry axis *c*. For $ScFe_2$ and at 1.5 K, the deduced magnetic moments of Fe at the 2*a* and 6*h* crystallographic positions are about 0.96 and 1.04 μ_B respectively. For $Sc_{0.6}Nb_{0.4}Fe_2$, the refined moments for Fe(2*a*) and Fe(6*h*) atoms are 0.21 and 0.26 μ_B, respectively. Nb for Sc substitution induces dramatic changes on the magnetic properties such as a strong decrease of the ordering temperature and a significant reduction of the Fe magnetic moment.

Not knowing a priori the orientation of the magnetic moments in the FM ground state, Rietveld refinements of the 1.5 K diffraction patterns were also conducted considering an alignment of the moments in the basal-plane of the hexagonal lattice. The two principal basal-plane directions, [100] and [120], were tested. The magnetic reliability factors (Rmag) obtained for the two directions are very close: 23.9% and 21.8% for [100] and [120] orientation, respectively. The orientation of the magnetic moments in the basal-plane leads to much worse fits compared to that along the *c*-axis [001] which leads to much smaller reliability factor 6.07%. Consequently, at 1.5 K, Rietveld refinement gives a significantly better fit when the magnetic moments are along the [001] direction giving confidence to such orientation of the magnetic moments.

In $Sc_{1-x}Nb_xFe_2$ series of compounds, the obtained Fe magnetic moments are remarkably decreased in comparison with the pure Fe moment of 2.22 μ_B. This experimental result is consistent with the presence of non-magnetic Sc and Nb atoms as near neighbours.