

Experimental report

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Proposal: 5-41-1192

Council: 10/2022

Title: Magnetic structure of the emergingspin density wave phase in Nb_{1-y}Fe_{2+y}

Research area: Physics

This proposal is a new proposal

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Samples: NbFe₂

Instrument	Requested days	Allocated days	From	To
ORIENTEXPRESS	1	1	21/06/2023	22/06/2023
D10	7	7	21/06/2023	28/06/2023

Abstract:

Our previous neutron scattering of ferromagnetic (FM) Nb_{1-y}Fe_{2+y} revealed that chemical composition (y) tuning leads to a FM quantum-critical point masked by an emerging spin-density wave (SDW). The latter could be connected to many exciting phenomena, including field-induced quantum-critical lines, quantum tri-critical points, and quantum multi-critical points. Our previous neutron scattering experiments revealed the SDW ordering wave vector, but here we propose a dedicated magnetic neutron diffraction experiment to reveal the detailed magnetic structure, which might resemble a modulation of a ferrimagnetic building block. The results would form an important test for, e.g., order-by-disorder models that predict the emergence of modulated order at the border of ferromagnetism. Here, we apply for 7 days of beamtime at D10.

Magnetic structure of the emerging spin density wave phase in $\text{Nb}_{1-y}\text{Fe}_{2+y}$

Experimental report for Proposal 5-41-1192

1. Scientific background

The transition metal Laves phase compound $\text{Nb}_{1-y}\text{Fe}_{2+y}$ presents a rare opportunity to study the border of itinerant ferromagnetism (FM) at low temperature at ambient pressure (Fig. 1a). Slight iron excess of the order of 1% induces FM, whereas the stoichiometric compound exhibits spin density wave order (SDW), and slightly Nb-rich $\text{Nb}_{1-y}\text{Fe}_{2+y}$ remains a paramagnet (PM) down to low temperatures and displays signatures of Fermi-liquid breakdown [1]. Empirical observation in other materials as well as theoretical arguments [2, 3] suggest that the masking of itinerant FM quantum critical points (QCPs) by SDW order may be intrinsic in many clean systems. The SDW in NbFe_2 could be connected to many exciting phenomena including field-induced quantum-critical lines, quantum tri-critical points [4] and quantum multi-critical points. Hence, it is important to obtain detailed insight into $\text{Nb}_{1-y}\text{Fe}_{2+y}$ including its emerging SDW.

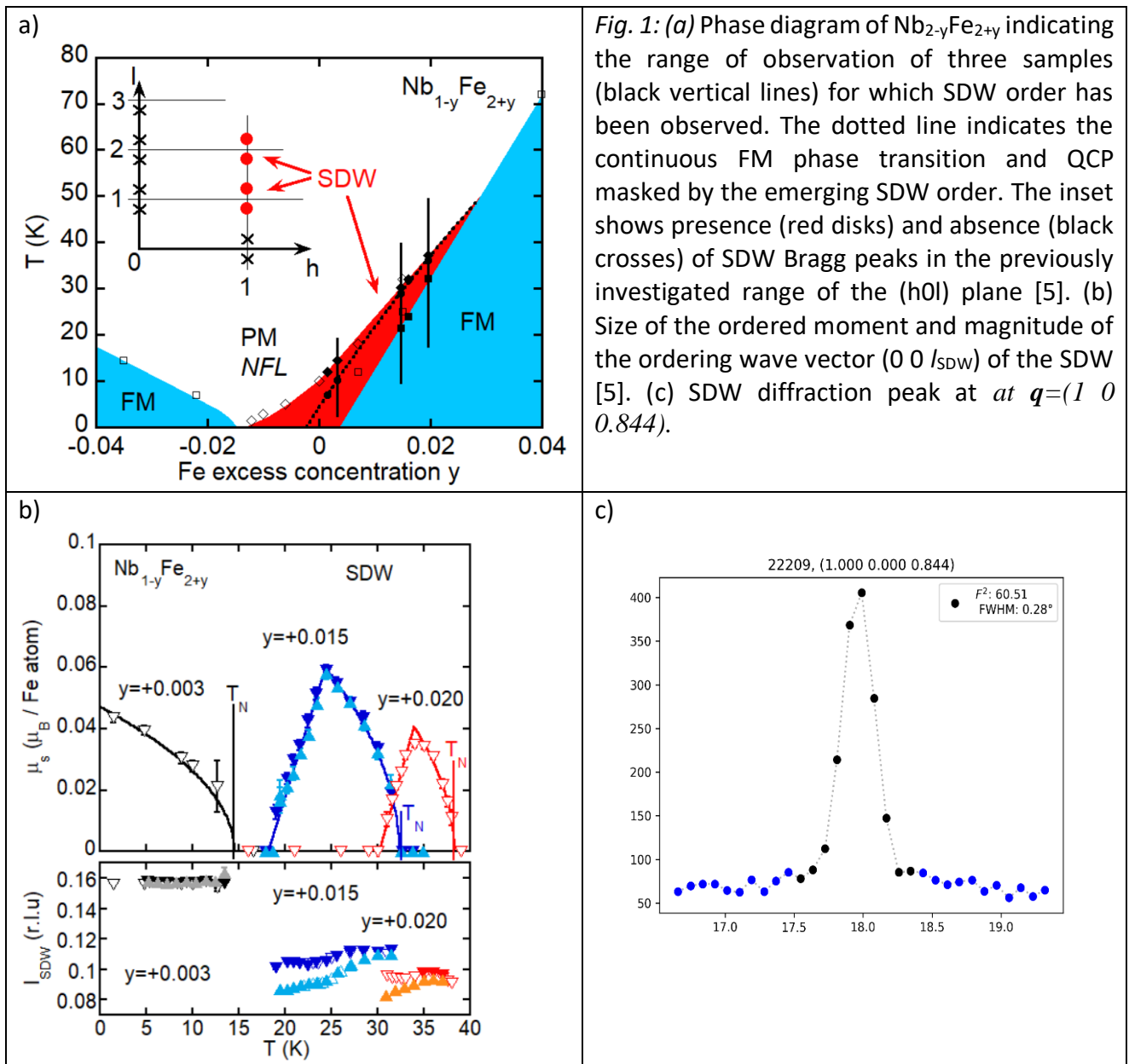


Fig. 1: (a) Phase diagram of $\text{Nb}_{1-y}\text{Fe}_{2+y}$ indicating the range of observation of three samples (black vertical lines) for which SDW order has been observed. The dotted line indicates the continuous FM phase transition and QCP masked by the emerging SDW order. The inset shows presence (red disks) and absence (black crosses) of SDW Bragg peaks in the previously investigated range of the $(h0l)$ plane [5]. (b) Size of the ordered moment and magnitude of the ordering wave vector $(0\ 0\ l_{\text{SDW}})$ of the SDW [5]. (c) SDW diffraction peak at $q=(1\ 0\ 0.844)$.

Previous neutron scattering of $\text{Nb}_{1-y}\text{Fe}_{2+y}$ had revealed the SDW ordering wavevector \mathbf{q}_{SDW} and its y and T dependence (Fig. 1b) [5]. However, further details of the SDW structure were still unknown.

2. Aim of the experiment

We proposed neutron diffraction to explore the detailed magnetic structure of the SDW phase in the nearly stoichiometric sample $\text{Nb}_{0.997}\text{Fe}_{2.003}$ at low temperatures and zero magnetic field.

3. Experimental setup

After optimization of the sample orientation with OrientExpress we used D10+ during the obtained seven days of beamtime to investigate a $\text{Nb}_{1-y}\text{Fe}_{2+y}$ sample with $y=+0.003$ and wave vector $(0\ 0\ l_{\text{SDW}}=0.155\ \text{r.l.u.})$ for which the presence of the SDW phase (Fig. 1b) had previously been shown [5]. The samples was glued to an Al pin holder that was attached to a He-flow cryostat. Measurements were done between 20K and 2K using neutrons with $k=2.66\ \text{\AA}^{-1}$. For the structure measurements at 20K an area detector was used. For measurements of small magnetic peaks it became clear that to sufficiently reduce the background, an analyzer needed to be used, which was combined with a single He detector.

4. Results

Measurements of 254 nuclear peaks were done at 20 Kelvin (paramagnetic phase) and 149 at 4K (in the SDW phase). Obtaining this comprehensive set took three days, as approximately two days were required to overcome technical issues concerning the cooling apparatus and mechanical issues that also led to the disconnection of the sample and need to remount.

After that three days were spent to measure 104 magnetic peaks at 2K to collect comprehensive data for magnetic Rietveld refinement. The magnetic intensities were found at the anticipated $\mathbf{q}_{\text{SDW}} = (0\ 0\ l_{\text{SDW}})$ with $l_{\text{SDW}} = 0.156$ positions (see example in Fig. 1c)).

During the final day, we investigated nuclear peak positions for any difference between the intensities in the SDW phase at 2K and in the PM phase at 20K. Those measurements revealed small differences that could be indicators of a $k=0$ antiferromagnetic contribution to the modulated order. However, the robustness of these results will need to be investigated further.

5. Conclusions and outlook

Nuclear refinement with FullProf has already produced a robust refinement of the previously known crystal structure of the $\text{Nb}_{1-y}\text{Fe}_{2+y}$ system.

Integrated magnetic Bragg intensities have also been fed into FullProf. A preliminary wide search in parameter space seems to confirm the suspected confinement of magnetic moments to the easy c -axis. Refinement carried out so far also favours antiparallel alignment of 2a and 6h Fe sites and resulting ferrimagnetic building block that are modulated with \mathbf{q}_{SDW} . (Fig. 2)

Understanding the emerging SDW order in detail would create an important test for, e.g., order-by-disorder models [6] that predict the emergence of modulated order at the border of ferromagnetism.

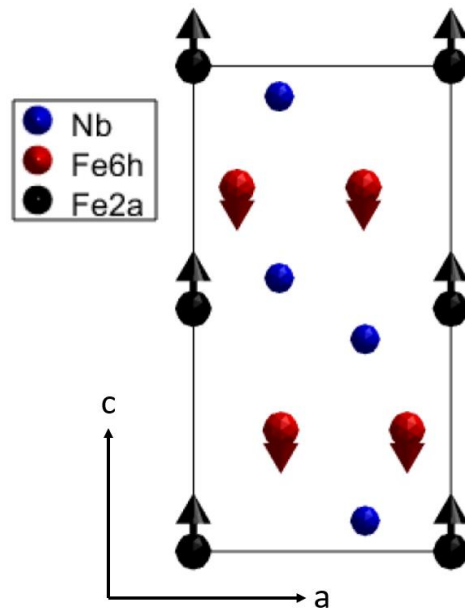


Figure 2. Projection of the unit cell of $\text{Nb}_{1-y}\text{Fe}_{2+y}$ with ferrimagnetic moment arrangement on to the a - c plane. An incommensurate modulation of this ferrimagnetic arrangement may be a candidate for the SDW structure.

- [1] M. Brando et al., Phys. Rev. Lett. **101**, 026401 (2008), D. Rauch et al., Phys. Rev. B **91**, (2015).
 [2] G. Abdul-Jabbar et al, Nat Phys **11**, 321 (2015). [3] see, e.g. M. Brando, D. Belitz, F. M. Grosche, T. R. Kirkpatrick, Rev. Mod. Phys **88**, 025006 (2016), T. Vojta et al., Ann. Phys. (Leipzig) **8**, 593 (1999). [4] S. Friedemann et al, Nat Phys **14**, 62 (2018). [5] P. G. Niklowitz et al., Phys. Rev. Lett. **123**, 247203 (2019). [6] U. Karahasanovic, F. Krüger, and A. G. Green, Phys. Rev. B **85**, 165111 (2012).