Experimental report

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Proposal: **5-31-2724** **Council:** 10/2019

Title: Study of crystal and magnetic structure changes in La1-xNdxFe12B6 metamagnetic compounds

Research area: Physics

This proposal is a new proposal

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Samples: La1-xNdxFe12B6

Instrument	Requested days	Allocated days	From	To
D1B	3	2	26/08/2020	28/08/2020

Abstract:

La1-xNdxFe12B6 compounds are exhibiting remarkable and unusual itinerant electron metamagnetic behavior. We are particularly interested in the determination of the temperature dependence of the magnetic and crystal structures. We are also interested in the determination of the composition dependence of the magnetic structure. The possible different behavior of the Fe magnetic moment on the inequivalent crystal sites will also be carefully investigated aiming to determine the effect of the local atomic environment on the Fe moment magnitude.

Study of crystal and magnetic structure changes in $La_{1-x}Nd_xFe_{12}B_6$ metamagnetic compounds

Introduction

The rare-earth iron intermetallic compounds are facinating magnetic materials from both fundamental and applied point of views. Indeed they exhibit unique feature resulting from the combination of both localized magnetism of 4*f* electrons of the rare-earth and itinerant magnetism of the 3*d* electrons of an iron group metal. This has led to the discovery of high performance applications such as magnetostrictive effect, the best permanent magnets as well as more recently new materials for giant magnetocaloric applications have all been discovered among the binary and ternary R-Fe system.

Interestingly, unconventional multistep metamagnetic transitions were recently reported in $LaFe_{12}B_6$ compound. These peculiar metamagnetic phase transitions are featured by ultrasharp steps followed by plateaus leading to an unusual and even unique staircase-like magnetization process. The antiferromagnetic itinerant-electron system $LaFe_{12}B_6$ occupies a special place among rare-earth iron-rich intermetallics; it exhibits exotic magnetic and physical properties. The unusual amplitude-modulated spin configuration defined by a propagation vector k = (¼, ¼, ¼), remarkably weak Fe magnetic moment (0.43 μ_B) in the antiferromagnetic ground state, especially low magnetic ordering temperature T_N = 36 K for an Fe-rich phase, a multicritical point in the complex magnetic phase diagram, both normal and inverse magnetocaloric effects, colossal spontaneous magnetization jumps, and huge hydrostatic pressure effects can be highlighted as the most relevant. These singular properties not only stimulate the development of theoretical models and experiments under extreme conditions for a deeper understanding of the striking phenomenology of this magnetic system, but also emphasize the potential interest of $LaFe_{12}B_6$ material in future low-temperature energy technologies. Among the $RT_{12}B_6$ family (where *R* stands for a rare-earth atom and *T* is a 3*d* transition metal element), $LaFe_{12}B_6$ is the sole stable Fe-based phase of the 1:12:6 ternary system. The first Fe-based member of the $RT_{12}B_6$ family to be identified, $NdFe_{12}B_6$ is metastable. On the other hand, the $RCo_{12}B_6$ alloys are stable along the entire rare-earth series [. At room temperature, the intermetallic compounds $RT_{12}B_6$ crystallize in the rhombohedral $SrNi_{12}B_6$-type structure (space group R-3m). Within the unit-cell, there are two crystallographically inequivalent sites for *T* atoms (18*g* and 18*h*). The *R* and B atoms occupy the 3*a* and 18*h* Wyckoff positions, respectively. The $LaFe_{12}B_6$ compound is unique among the ternary system $RT_{12}B_6$ in having an antiferromagnetic ground state. The magnetic transition temperature of $LaFe_{12}B_6$ is much smaller than the Curie point of the Co-based $RCo_{12}B_6$ ferro- (*R* =Y, La-Sm) or ferri- (*R* = Gd-Tm) magnets (T_C =134 - 162 K) and an order of magnitude smaller compared to the magnetic ordering temperature of any rare-earth iron-rich binary intermetallic. Extraordinary magnetotransport effects have been most recently discovered in $RT_{12}B_6$ compounds.

Experiments

Neutron powder diffraction (NPD) experiments were performed on the high-intensity two-axis powder diffractometer D1B with a detector angular range coverage $5° \leq 2\theta \leq 128°$ 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

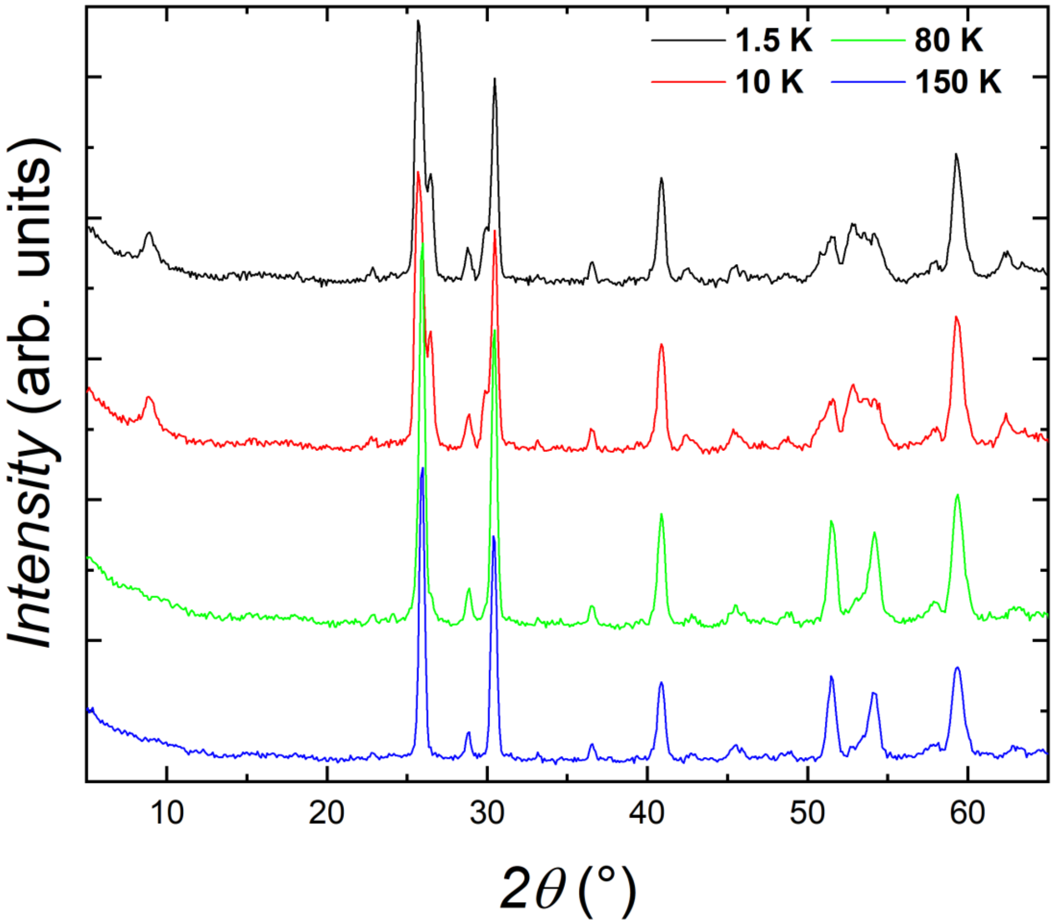

Figure 1: Neutron diffraction patterns of $La_{0.85}Nd_{0.15}Fe_{12}B_6$ recorded at 1.5 K, 10 K, 80 K and 300 K.

The observation of anomalous features and multiple magnetic transitions by magnetization measurements urged us to carry out NPD investigations to establish the crystallographic and magnetic structures of $La_{0.85}Nd_{0.15}Fe_{12}B_6$ as a function of temperature. Diffraction patterns recorded at some selected representative temperatures are displayed in Figure 1. For the sake of clarity, only the region from 5° to 65° 2θ is depicted. From the visual inspection of the spectra, it is immediately apparent that there are big differences between them. The diffraction profile is considerably altered upon cooling and new reflections appear. NPD data reveal a temperature-induced structural phase transition associated with the AFM-FM and PM-FM transformations. The lattice distortion is driven by magneto-elastic coupling and converts the crystal structure from rhombohedral to monoclinic. The AFM and PM states are related to the rhombohedral structure, whereas the FM order develops in the monoclinic symmetry. The Rietveld refinement of the obtained diffraction patterns are in progress.