Experimental Report

30/10/2013

Proposal: **5-32-773** **Council:** 4/2012

Title: The magnetic interactions between iron and cobalt in the ternary ironarsenide, $(Fe_{1-x}Co_x)_2As$, above the Curie temperature

This proposal is a new proposal

Researh Area: Physics

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Samples: $(Fe_{0.4}Co_{0.6})_2As$
$(Fe_{0.6}Co_{0.4})_2As$

Instrument	Req. Days	All. Days	From	To
D7 He3 Spin Filter	6	6	16/11/2012	22/11/2012

Abstract:

In the Fe-Co-As system the Fe_2P-type structure is stable in the composition range $(Fe_{1-x}Co_x)_2As$, $0.35<x<0.9$, even though the binary phases Fe_2As and Co_2As form other crystal structures. The crystal structure of $(Fe_{1-x}Co_x)_2As$, $0.35<x<0.9$ has a hexagonal unit cell with the metal atoms situated on two different 3-fold positions, one surrounded by four non-metal atoms in a tetrahedral environment and one by five non-metal atoms in a square pyramidal coordination. The cobalt atoms are preferentially occupying the tetrahedral position and the iron atoms the pyramidal site. The two compositions $(Fe_{0.6}Co_{0.4})_2As$ and $(Fe_{0.4}Co_{0.6})_2As$ are ferromagnetic with T_C 320K and 260K respectively. For x=0.4 the magnetic moment are directed along the c-axis, and for x=0.6 in the ab-plane. The saturation magnetization corresponds to 2.4 μ_B/f.u. (x=0.4) and 1.7 μ_B/f.u. (x=0.6). The total magnetic moments on the tetrahedral site are very small (0.1 and 0.0 μ_B/f.u.) and the moments on the cobalt atoms vanish. The aim of the present proposal is to experimentally investigate the dependence of the magnetic short range order correlated to the nuclear short range order.

Introduction

Magnetocaloric compounds have gained an increased interest since the middle 1990s due to environmental and energy benefits from magnetic refrigeration. The large saturation magnetization, first order transition and tunable transition temperature with substitution make the Fe_2P-system a candidate for magnetocaloric applications. Numerous Fe_2P-based compounds have been investigated during the years and many of those have shown improved magnetocaloric properties.

Pure Fe_2P has a first order ferro- to paramagnetic transition at 217K. The Fe_2P-type crystal structure is maintained for extended substitutions both on the iron and phosphorus sites with other transition metals and arsenic or silicon, respectively. The magnetic ordering temperature, the magnetic structure and the magnetization properties are generally drastically changed even for small variations in composition [1, 2]. Another example is the Fe-Co-As system where the Fe_2P-type structure is stable in the composition range $(Fe_{1-x}Co_x)_2As$, 0.35<x<0.9 [3], but the binary phases Fe_2As and Co_2As form other crystal structures.

The crystal structure of $(Fe_{1-x}Co_x)_2As$, 0.35<x<0.9 has a hexagonal unit cell with the metal atoms situated on two different 3-fold positions, one surrounded by four non-metal atoms in a tetrahedral environment and one by five non-metal atoms in a square pyramidal coordination (compare Figure 1). The cobalt atoms are preferentially occupying the tetrahedral position and the iron atoms the pyramidal site. The two compositions $(Fe_{0.6}Co_{0.4})_2As$ and $(Fe_{0.4}Co_{0.6})_2As$ are ferromagnetic with T_C=320K and 260K respectively. The magnetic properties of $(Fe_{1-x}Co_x)_2As$ (x=0.4 and 0.6) are summarized in Figure 2. For x=0.4 the magnetic moment are directed along the c-axis, and for x=0.6 in the ab-plane. The saturation magnetization corresponds to 2.4 μ_B/f.u. (x=0.4) and 1.7 μ_B/f.u. (x=0.6). The total magnetic moment on the tetrahedral site is very small (0.1 and 0.0 μ_B/f.u.) and the moments on the cobalt atoms vanish. For comparison, the magnetic moments 0.9 and 1.7 μ_B, aligned along the c-axis were obtained from polarized neutron diffraction on a single crystal of pure Fe_2P as seen in Figure 1.

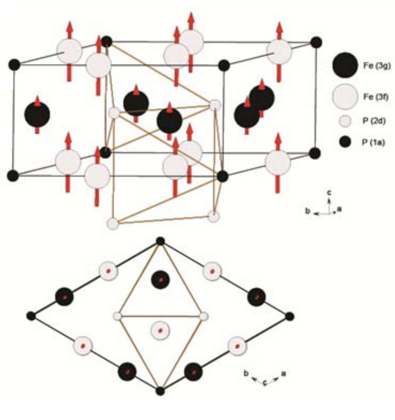

Figure 1.: The magnetic structure of Fe_2P. In $(Fe_{1-x}Co_x)_2As$, Co substitutes preferentially on the 3g-site (black).

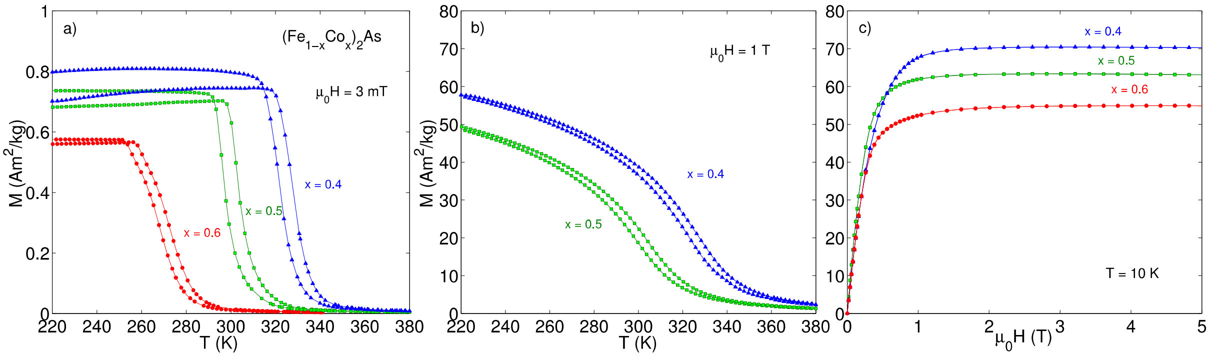

Figure 2.: Magnetic properties of $(Fe_{1-x}Co_x)_2As$ (x=0.4, 0.5 and 0.6) as a function of temperature a) + b) and magnetic field at 10K c).

Experimental details and analysis

Powder samples of $(Fe_{0.6}Co_{0.4})_2As$ and $(Fe_{0.4}Co_{0.6})_2As$ were fabricated using the drop synthesis method and pre-characterized for structural properties using a Bruker D8 as well as magnetic properties using a Quantum Design MPMS-XL 5 T. Neutron experiments were performed on the diffuse scattering beamline D7. The D7 beamline measures information on local atomic and magnetic arrangements such as clustering or short-range ordering [4]. The most basic form of diffuse scattering is incoherent scattering which can be of structural (nuclear incoherent scattering) or magnetic (spin incoherent) origin. The D7 beamline was chosen to obtained complete separation of magnetic and structural diffuse scattering. For the measurements $(Fe_{0.6}Co_{0.4})_2As$ and $(Fe_{0.4}Co_{0.6})_2As$ powder samples were mounted on the inner wall of a cylindrical aluminum can and a wavelength of 3.1171 Å was used. When analyzing the diffuse part of a neutron scattering experiment we study the information not contained in the nuclear and magnetic Bragg peaks careful calibration and background handling is hence essential. Our experimental neutron results are in absolute units of the magnetic scattering cross-section (barns/st/f.u.), which is proportional to the square of the magnetic moments.

Raw data for our measurements on $(Fe_{0.6}Co_{0.4})_2As$ and $(Fe_{0.4}Co_{0.6})_2As$ are shown in Figure 3 a) and b), respectively. Co has a comparable high neutron absorption cross section (37.18 barn) which gave rise to increased data noise when comparing x=0.4 and 0.6. Additional measurements were performed on an antiferromagnetic $(Fe_{0.4}Mn_{0.6})_2P$ sample at 10K, 355K, 425K and 525K (not shown here).

Preliminary results

The two datasets of the average diffuse magnetic neutron scattering (ds/dw) for $(Fe_{0.4}Co_{0.6})_2As$ and $(Fe_{0.6}Co_{0.4})_2As$, depicted in Figure 3 a) + b), exhibit a qualitative similar behavior. Due to the larger momentum transfer (up to 3.8 $Å^{-1}$) a broad distribution (FWHM ~1.6 $Å^{-1}$) for the $(Fe_{1-x}Co_x)_2As$ diffuse magnetic neutron scattering, centered at about 2.3 $Å^{-1}$, is observed. This suggests short-range ordering on length scales of ~4 Å, which correspond roughly to the 2nd to 3rd NN interaction of Fe2-Fe2 (pyramidal sites) in the Fe_2P structure. Due to a smaller momentum transfer in our earlier measurements for Fe_2P (up to 2.5 $Å^{-1}$) this could not be observed [5].

A qualitative comparison of the diffuse magnetic neutron scattering data (up to 2.5 $Å^{-1}$) slightly above the Curie temperature, ~T_C, and at 1.5x T_C for $(Fe_{1-x}Co_x)_2As$ (x=0.4 and 0.6) and Fe_2P is shown in Figure 3 c) + d), respectively.

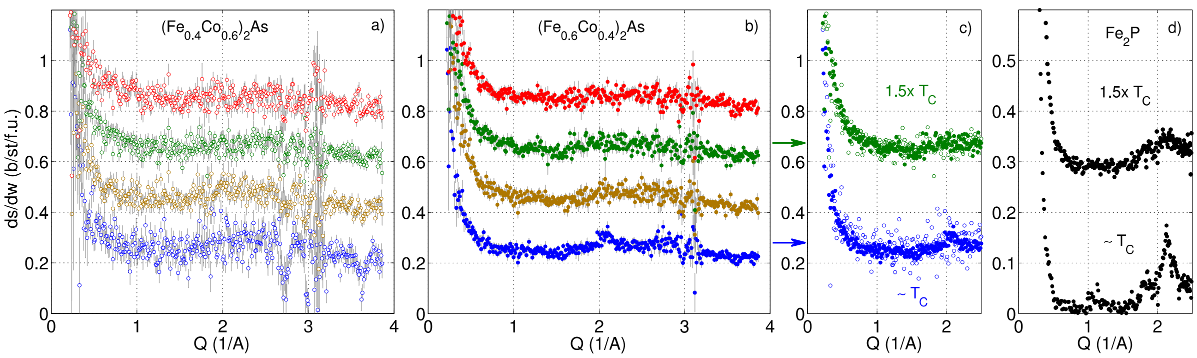

Figure 3.: Magnetic scattering cross section (ds/dw) as a function of wave vector transfer Q for a) $(Fe_{0.4}Co_{0.6})_2As$ measured at 270, 275, 390, and 525 K (bottom up) and b) $(Fe_{0.6}Co_{0.4})_2As$ at 325, 425, 480, and 525 K (bottom up). C + d) shows comparison of the raw data for $(Fe_{1-x}Co_x)_2As$ and Fe_2P at ~T_C and 1.5x T_C.

Our measurements of the $(Fe_{1-x}Co_x)_2As$ system will clearly help to understand the interesting magnetic behavior of Fe_2P. A joint data analysis of both $(Fe_{1-x}Co_x)_2As$ and Fe_2P measurements concerning modeling of the average diffuse magnetic neutron scattering and a subsequent determination of the magnetic moments of Fe_2P are in progress.

Acknowledgements

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References

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