Experimental Report

Proposal: 5-53-245 **Council:** 4/2014

Title: Probing magnetic short-range orderin the paramagnetic regime of (Mn,Fe)2(P,Si) magnetocaloric compounds

This proposal is a new proposal

Researh Area: Materials

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Samples: (Mn,Fe)2(P,Si)

Instrument	Req. Days	All. Days	From	To
D7	7	5	04/12/2014	09/12/2014

Abstract:

(Mn,Fe)2(P,Si) compounds show a giant magnetocaloric effect (GMCE) at the first-order paramagnetic-ferromagnetic phase transition, making them one of the most promising candidates for room-temperature magnetic refrigeration application. Previous magnetic susceptibility measurements led to the identification of magnetic short-range order in the paramagnetic state. Understanding the nature of short-range order is essential for clarifying the magnetic phase transition and the GMCE in (Mn,Fe)2(P,Si) compounds. 'xyz' neutron polarization analysis is well suited to study the nature of the magnetic short-range order as well as their spin dynamics in (Mn,Fe)2(P,Si) magnetocaloric compounds.

Probing magnetic short-range order in the paramagnetic regime of $(Mn,Fe)_2(P,Si)$ magnetocaloric compounds

Introduction

The giant magnetocaloric effect (GMCE), associated with a first-order magneto-elastic transition, makes near room-temperature magnetic refrigeration attractive as a highly efficient and environment-benign cooling technology. We discovered the GMCE in hexagonal $(Mn,Fe)_2(P,Si)$ compounds.[1-3]. This high-performance and low-cost system is a promising candidate for magnetic cooling and power conversion applications.[4]

$(Mn,Fe)_2(P,Si)$ compounds crystallize in the so-called Fe_2P-type structure where there are two metal (3*f* and 3*g*) and two non-metal (2*c* and 1*b*) sites. Our first-principle electronic-structure calculations indicate a mixed magnetism in this system.[2] The layers occupied by Mn are strongly magnetic, implying that the magnetic long-range order is only lost at the Curie temperature T_C. The size of the Mn moment (3*g* site) is reduced from 2.8 μ_B in the ferromagnetic (FM) phase to 2.6 μ_B in the paramagnetic (PM) phase. By contrast, the Fe-layers show weak itinerant magnetism: the Fe moment (3*f* site) of 1.54 μ_B in the FM phase almost vanishes in the PM phase. The first-order PM-FM phase transition in $(Mn,Fe)_2(P,Si)$ compounds is related to the redistribution of electron density around magnetic atoms.

Magnetization measurements in the paramagnetic phase show that the inverse magnetic susceptibility deviates from the Curie-Weiss law up to about 2.2 T_C.[5] Also, an anomaly in the thermal evolution of lattice parameters above T_C has been observed from X-ray diffraction experiments. These results imply the existence of magnetic short-range order above T_C. The aim of this study is to understand the nature of short-range order as well as their spin dynamics in $(Mn,Fe)_2(P,Si)$-type magnetocaloric compounds.

Experimental details

xyz neutron polarization analysis is able to unambiguously separate magnetic scattering cross section from nuclear coherent and spin-incoherent contributions, which has been widely used for studying magnetic short-range ordered materials.[6, 7] Also, a huge contrast in the scattering length of Mn and Fe may allow us to specify the magnetic atoms or crystallographic sites involved in the short-range magnetic correlations.

We performed 6-point *xyz* neutron polarization experiments on the D7 spectrometer for 2 compositions: $Mn_{1.0}Fe_{0.95}P_{0.67}Si_{0.33}$ (T_C = 131 K) and $Mn_{1.7}Fe_{0.25}P_{0.50}Si_{0.50}$ (T_C = 167 K), which exhibits a 1st-order and 2nd-order para-ferromagnetic transition, respectively. The powder sample was put between the inner wall of a cylindrical aluminium can and an insert. The measurements were taken in a temperature range of $1.3T_C \leq T \leq 3\ T_C$ for each sample. An incident neutron wavelength of 3.12 Å was chosen in order to get good energy integration for the high temperature measurements. Standard background determination and polarization correction was performed

using cadmium and quartz, respectively. The flipping ratio was also measured for the whole temperature range, which allows us to easily determine the magnetic state of the samples.

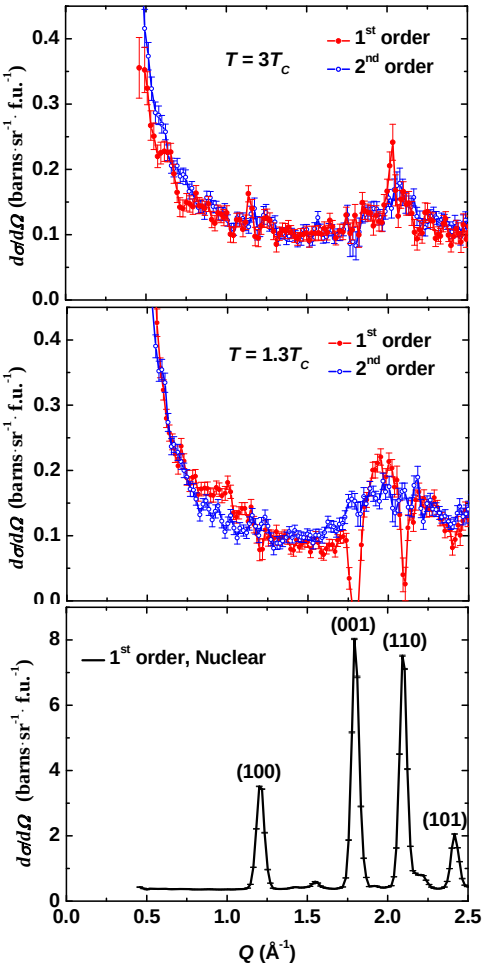

Figure 1. Magnetic and nuclear-coherent scattering cross section of $Mn_{1.0}Fe_{0.95}P_{0.67}Si_{0.33}$ (1^{st}-order phase transition) and $Mn_{1.7}Fe_{0.25}P_{0.50}Si_{0.50}$ (2^{nd}-order phase transition).

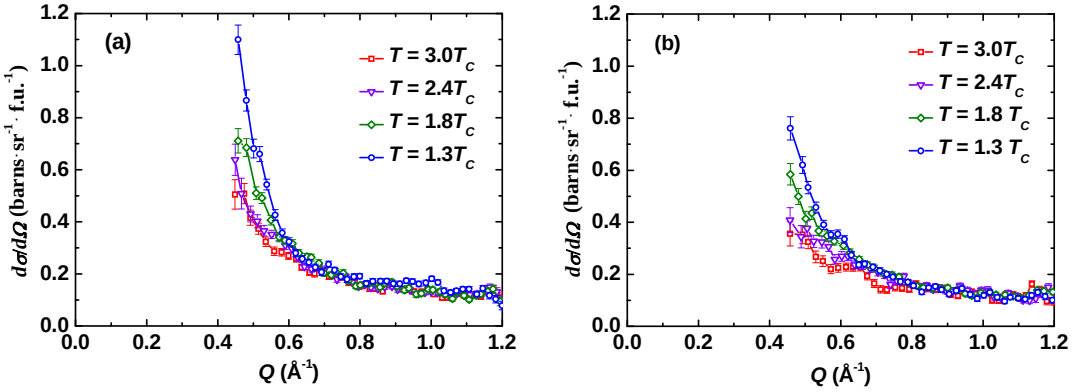

Figure 2. Temperature-dependent magnetic scattering cross section of (a) $Mn_{1.0}Fe_{0.95}P_{0.67}Si_{0.33}$ and (b) $Mn_{1.7}Fe_{0.25}P_{0.50}Si_{0.50}$.

Preliminary results

Magnetic, nuclear and spin-incoherent scattering cross sections are extracted from the 6 separate polarization-dependent neutron cross sections. A magnetic diffuse scattering was observed at $T = 3T_C$ in both 1st-order and 2nd-order phase transition samples, as shown in figure 1. The magnetic diffuse scattering is centred around Q = 2.1 Å, which coincides with the (110) nuclear Bragg peak. This suggests the presence of short-range ferromagnetic correlations within basal plane in both samples far above the Curie temperatures. However, at low temperatures (*e.g.* $T = 1.3T_C$) we got some negative magnetic scattering cross sections at the nuclear Bragg peak positions for the 1st order sample (see figure 1), but the sample was still in paramagnetic state since we did not see a significant drop of the flipping ratio. The unexpected negative magnetic scattering cross sections is due to the improper polarization corrections. We did the polarization corrections based on an amorphous quartz sample, which gives feature-less diffuse scattering which is very different from the well-defined scattering at Bragg peak positions in our samples. As a result, the polarization corrections do not work properly at the Bragg peak positions in our samples. The improvement of polarization corrections is in progress.

Figure 2 shows the temperature dependence of the magnetic scattering cross section in the small Q range for the 1st-order and 2nd-order phase transition samples. The increase of magnetic scattering cross-section with the decrease of temperature suggests the development of magnetic correlations, which is more pronounced for the 1st-order phase transition sample. From the forward scattering centred at $Q = 0$, the magnetic correlation length may be determined. The thermal evolution of the magnetic correlation length will provide insights into the origin of the phase-transition process of $(Mn,Fe)_2(P,Si)$-type compounds.

References

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