Experimental report

11/02/2020

Proposal: CRG-2620 **Council:** 4/2019

Title: Evolution of the spin fluctuationsand spin dynamics in the inverse magnetocaloric effect compound Mn5Si3

Research area: Physics

This proposal is a new proposal

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Samples: Mn5Si3

Instrument	Requested days	Allocated days	From	To
IN12	7	4	20/09/2019	24/09/2019

Abstract:

Mn5Si3 shows the inverse magnetocaloric effect (MCE), the cooling by adiabatic magnetization. This effect is opposite to the more common direct MCE, the cooling by adiabatic demagnetization.
In previous inelastic neutron scattering experiments we could show that among the two stable antiferromagnetic phases the high temperature one (AF2) shows an unusual magnetic excitation spectrum with coexisting propagative spin waves and diffuse spin fluctuations. Moreover, we could associate the inverse magnetocaloric effect with field induced spin fluctuations.

In a recent test experiment we explored the low energy inelastic spectra in the AF2 phase and in the paramagnetic (PM) state under the influence of a magnetic field. The aim of this proposal is to further investigate the temperature and magnetic field dependence of spin fluctuations in the PM state as well as the spin dynamics in the low temperature magnetic phase AF1.
The field dependence of the PM state will help us to understand the behaviour of spin fluctuations under field in the AF2 phase. The evolution of the spin dynamics in the AF1 phase is strongly related to transition to the AF2 phase under field and the inverse MCE.

Background

The search for more efficient use of energy has been leading to a growing interest for the research field of magnetocaloric materials. The magnetocaloric effect (MCE) is the reversible temperature change of a magnetic material upon application or removal of an external magnetic field. The MCE can be characterized as direct or inverse depending on whether the material heats up or cools down when applying an external magnetic field adiabatically.
The system $Mn_{5-x}Fe_xSi_3$ shows a reasonable large MCE close to room temperature at low magnetic fields being promising for magnetic refrigeration applications.
The parent compound Mn_5Si_3 undergoes with cooling a first order phase transition at $T_{N2}\approx100K$ towards a collinear antiferromagentic state (AF2). A second transition occurs at $T\approx66K$ towards a non-collinear antiferromagnetic phase (AF1). The inverse MCE is related to the AF1-AF2 phase transition.
In previous experiments we could show that the high temperature AF2 phase shows an unusual magnetic excitation spectrum with coexisting propagative spin waves and diffuse spin fluctuations where well-defined spin waves are characteristic for the low temperature AF1 phase.

Aim of the experiment

The aim of the experiment was to investigate further the evolution of the inelastic spectra in temperature and magnetic field dependence.
Scans have been taken between 10K deep in the AF1-phase and 150K far in the paramagnetic regime with different magnetic fields and temperatures.
An emphasis lied on the magnetic field dependence of energy scans at 50K (AF1) and 80K (AF2) for different Q close to Q=(1,2,0).

Experimental set-up

The IN12 spectrometer was set-up in W-configuration (-1,1,-1) and a vertical 10T-magnet has been installed. To improve the background 80' collimation was in place after the monochromator. Data have been taken with a final wavevector of $k_f=1.05Å^{-1}$ as well as with $k_f=1.5Å^{-1}$. For optimum resolution a 40' collimator has been installed after the monochromator in combination with the lowest k_f. The single crystal (with a mass of about 6g) has been mounted with the [100]-[010] directions in the scattering plane.

Results

Energy scans have been performed in the AF2 phase at 80K at several Q points close to the magnetic Bragg position (1,2,0) in different vertical magnetic fields ranging from 0T to 10T.
The results hint to a double gap structure evolving in dependence of magnetic field, for (1,2,0) from 6T onwards clear

(From previous experiments with polarized neutron set-up and without an external magnetic field a hint to a double peak structure has been already visible away from the Bragg position (1.06,2,0,0) with an indicated gap of approximately 0.25meV), see left side of Fig.1.
Energy scans in the AF1 phase in a field of 10T at Q=(1,2,0) showed a slight broadening of the spin waves compared to scans without external magnetic field.

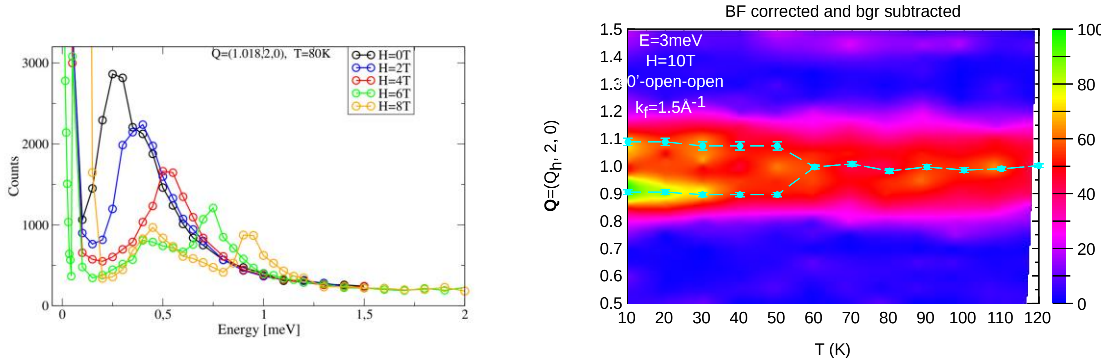

Fig.1: Right: Energy scans at Q=(1.018,2,0) at 80K for different vertical magnetic fields. Left: Q_h-scans at Q=(Q_h,2,0,3) in an external vertical magnetic field of 10T for temperatures 10K-120K.

The right hand side of Fig.1 shows Q_h scans at the magnetic Bragg position (1,2,0) with an energy transfer of 3 meV and a magnetic field of 10T in the temperature range of 10K (AF1 phase) up to 120K (paramagnetic state). Field induced spin fluctuations persist deep into the AF1 phase.
The excitations measured in an external magnetic field of 10T starting at 10K (AF1 phase) up to 120K (paramagnetic state) showed an increase of excitations mainly in the temperature range of approximately 50K to 100K, corresponding to the AF2 phase.