Experimental report

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Proposal: **4-01-1618** **Council:** 4/2019

Title: Polarized neutron investigation ofthe magnetic excitation spectrum of the inverse magnetocaloric compound Mn5Si3

Research area: Physics

This proposal is a continuation of 4-01-1580

Main proposer: **Stephane RAYMOND**

Experimental team: Stephane RAYMOND
Karin SCHMALZL
Nikolaos BINISKOS

Local contacts: Paul STEFFENS

Samples: Mn5Si3

Instrument	Requested days	Allocated days	From	To
THALES	7	7	26/09/2019	03/10/2019

Abstract:

Mn5Si3 has attracted interest for exhibiting an inverse magnetocaloric effect that corresponds to the cooling by adiabatic magnetization. This effect is opposite to the more common direct magnetocaloric effect, the cooling by adiabatic demagnetization.

Using inelastic neutron scattering (INS), we have shown that, among the two stable antiferromagnetic phases of this compound, the high temperature one (AF2) is characterized by an unusual magnetic excitation spectrum where propagative spin waves and diffuse spin fluctuations coexist. Moreover, it was evidenced that the inverse magnetocaloric effect of Mn5Si3 is associated with field induced spin fluctuations.

Polarized neutron scattering experiments provided an essential input to reach these conclusions. In order to further separate spin-waves from spin fluctuations in the full energy range of the dynamical magnetic response, it is proposed to carry out a polarized INS investigation of the AF2 state in the energy range 0.2-3 meV at zero field. Beyond providing a complete microscopic landscape of this peculiar spin dynamics, this will allow to consistently analyze the unpolarized INS data obtained as a function of magnetic field.

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}≈100K towards a collinear antiferromagnetic state (AF2). A second transition occurs at T≈66K 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

In previous inelastic neutron scattering experiments we have shown that amongst the two stable antiferromagnetic phases the lower temperature AF1 phase shows propagative spin waves whereas the high temperature AF2 phase has an unusual magnetic excitation spectrum. We could show with polarized neutron scattering experiments that the spectra of the AF2 phase differs in the M_y and M_z channels from those of the paramagnetic state and the spectra of the AF2 phase were interpreted as coexistence of spin waves and spin fluctuations.
The aim of the experiment was to add to the understanding of the magnetism and the excitation spectrum of the AF2 phase with polarized neutrons.
To this aim energy and also Q scans have been performed around the magnetic Bragg position in the AF2 phase at 80K and in the paramagnetic state.

Experimental set-up

The Thales spectrometer was set-up in W-configuration (-1,1,-1) in a polarized set-up and Cryopad has been used for spherical polarization analysis. . Data have been taken with a final wavevector of $k_f=1.3Å^{-1}$ as well as with $k_f=1.1Å^{-1}$. 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 at the magnetic Bragg position (1,2,0) for the diagonal channels of the polarization matrix at 80K in the AF2 phase and for 120K in the paramagnetic state. At 80K at the elastic position the magnetization M_y was found to be greater than M_z. Away from the elastic

position M_z was found greater than M_y and further for inelastic energies M_y approximates M_z in agreement with previous experiments.
In the paramagnetic state M_y and M_z appeared to be isotropic.

Fig.1 shows the resulting magnetization in z-direction of an exemplary energy scans at 80K at a Q-positions, Q=(1.06,2,0).

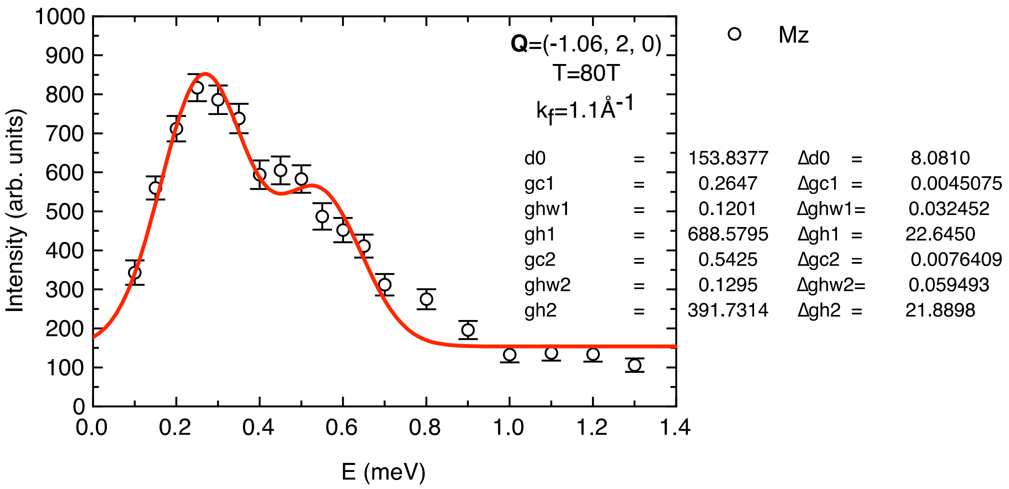

Fig.1: Energy scan at Q=(1.06,2,0) at 80K. Shown is the magnetization in z-directions. The red curve represents the fit of the data with two Gaussians. The symbols are the raw data.

Unlike from previous experiments where the data suggested that the inelastic spectra in the AF2 phase consist of a combination of propagative spin waves and diffuse fluctuations, the present data suggest that in the AF2 phase broad spin waves dominate the inelastic picture.