Experimental report

09/03/2017

Proposal:	**CRG-2380**	**Council:**	10/2016
Title:	Spin and lattice dynamics of magnetocaloric Mn5Si3 compound		
Research area:	Physics		

This proposal is a new proposal

Main proposer:	**Nikolaos BINISKOS**
Experimental team:	Nikolaos BINISKOS
Local contacts:	Stephane RAYMOND

Samples: Mn5Si3

Instrument	Requested days	Allocated days	From	To
IN22	6	6	21/02/2017	27/02/2017

Abstract:

We propose to study the spin and lattice dynamics of the magnetocaloric compound Mn5Si3. Two ingredients could play an important role in the magnetocaloric effect of this compound: i) the phonon-magnon coupling and ii) coexistence of well-defined spin-waves and strong magnetic fluctuations within a magnetically ordered phase, that has been found in a previous experiment with focus on a,b plane. Here we propose to continue our study along c direction for different energy transfers in the different magnetic phases as well as in the paramagnetic phase. For this microscopic investigation using a TAS instrument (IN22 or IN12) seems the most efficient method.

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 cooling process is based on the magnetocaloric effect (MCE). MCE is the reversible temperature change of a magnetic material upon the application or removal of a magnetic field. The MCE can be characterized as direct or inverse if a magnetocaloric compound heats up or cools down by applying an external magnetic field adiabatically. An entropy transfer between crystal lattice and the magnetic spin system has to take place. Among different compounds under investigation, the system $Mn_{5-x}Fe_xSi_3$ shows a modestly large MCE close to room temperature at low magnetic fields, which is promising for magnetic refrigeration applications. The parent compound Mn_5Si_3 on cooling undergoes a first phase transition at $T_{N2} \approx 100$ K toward a collinear antiferromagnetic ground state (AF2) and a second transition to a non-collinear antiferromagnetic phase (AF1) that occurs at $T_{N1} \approx 66$ K. Its specificity is to exhibit positive and negative magnetic entropy change in relation with two distinct magnetic phase transitions at $T_{N1}{\approx}66$ K and $T_{N2}{\approx}100$ K, respectively.

Aim of the experiment:

The aim of this experiment was to investigate the spin dynamics along c direction at 10K (AF1 phase), 80K (AF2 phase) and at 120K (PM state) and to follow the magnon dispersion in plane at higher energy transfers $3<E<25$meV (the orthorhombic cell is derived from the ortho-hexagonal cell of the PM space group $P6_3/mcm$, therefore the a, b plane will be referred to as the ‘‘plane’’).

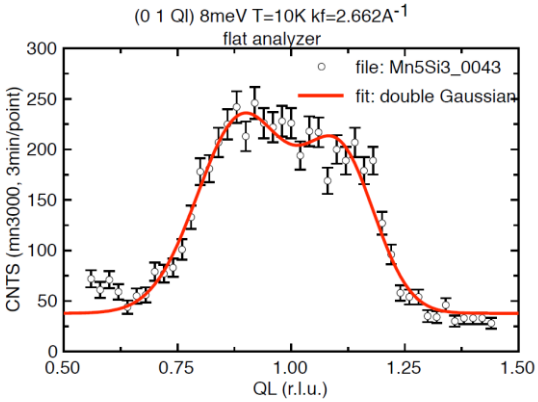
Experimental setup:

The IN22 spectrometer was set up in W configuration. We used a PG monochromator (double focusing mode), a monitor, slits before and after the sample, a PG filter after the sample and a PG analyser. All data have been collected with a fixed $k_f=2.662Å^{-1}$. In order to investigate the spin dynamics along c direction the single crystal (with a mass of about 6g) was mounted with the [010] – [001] directions in the scattering plane inside an orange cryostat.

Results:

Experiments were performed on a single crystal of about 1.5 cm^3 on the IN22 spectrometer. In order to investigate the spin dynamics along [001] direction spectra were collected in three temperature regions: in the PM state ($100<T<120$K), in the collinear antiferromagnetic AF2 phase ($66<T\leq100$K) and in the non collinear antiferromagnetic AF1 phase ($T\leq66$K).

For obtaining the magnon dispersion along c at T=10K (AF1 phase) q-scans at constant energies below 25meV were carried out around magnetic Bragg peaks (0 3 1), (0 1 1) and (0 5 0). Because of the observed high steepness of the magnon dispersion the analyser was set up in flat mode for all scans in order to improve the q resolution. Such scans were mostly conducted between intervals of +q and -q. Therefore every peak (if an excitation could be detected) was measured in focusing and defocusing mode. The exact position of a magnon was evaluated by fitting the excitation spectrum with a double Gaussian with same center at the focusing and defocusing side, but with different widths and amplitudes. In Fig. 1 such typical scan is presented. Before fitting, every spectrum was analyzed carefully looking for spurions, in particular Al contamination, and the corresponding regions were cut out. The preliminary obtained magnon dispersion curves along [010] and [001] directions of the orthorhombic system at 10K (AF1 phase) are shown in Fig. 2 and 3.



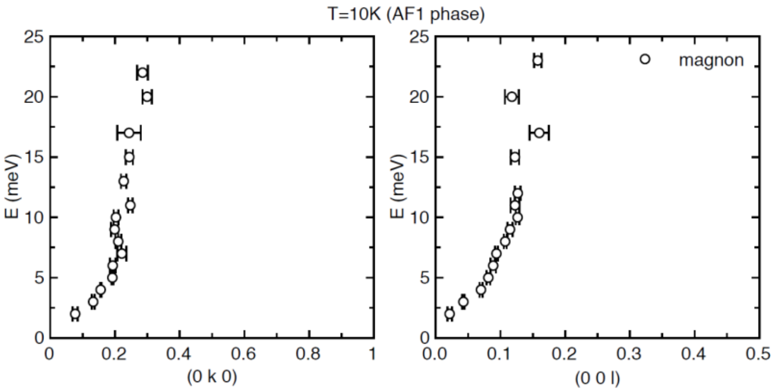


Figure 1 (left): Q-scan (0, 1, Ql) at 8meV on IN22 spectrometer. Red line corresponds to a double Gaussian fit. **Figures 2 and 3 (middle and right):** Preliminary magnon dispersion of Mn$_5$Si$_3$ along b and c direction at T=10K.

Spectra obtained in the collinear antiferromagnetic AF2 phase at 80K indicate that the scattering is characterized by a diffuse signal that resembles the one of the paramagnetic (PM) phase, a result which is consistent with our previous experiments performed in the a-b plane. The spin excitation spectrum is markedly different in the two magnetically ordered phases and this is illustrated by a color-coded intensity plot of the INS data collected at 6 meV as a function of Q$_l$ (c-axis) and temperature (Fig.4-bottom).

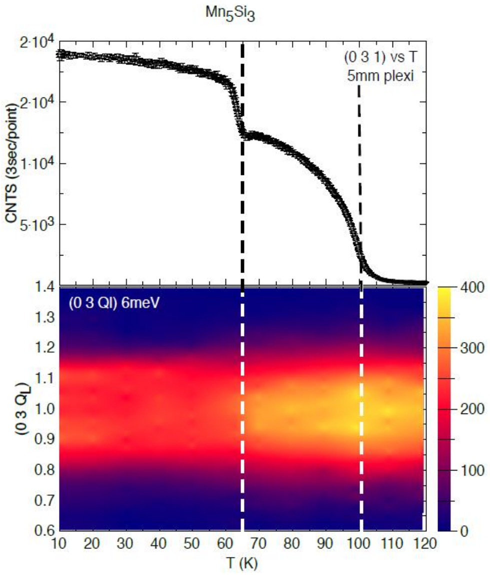


Figure 4: Top: Temperature dependence of magnetic Bragg peak (0 3 1) while cooling from PM state (120K) to base temperature (10K). Bottom: Ql-scans at constant energy 6meV. Intensity corrected for background and Bose factor. Vertical dashed lines indicate the onset of AF2 and AF1 phases, respectively.