Experimental report

29/10/2018

Proposal: **CRG-2444** **Council:** 4/2017

Title: Magnon-phonon coupling in a magnetocaloric effect compound

Research area:

This proposal is a new proposal

Main proposer: **Nikolaos BINISKOS**

Experimental team: Nikolaos BINISKOS

Local contacts: Stephane RAYMOND

Samples: $MnFe_4Si_3$

Instrument	Requested days	Allocated days	From	To
IN12	5	5	19/03/2018	26/03/2018

Abstract:

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) refers to a change of temperature/entropy of a magnetic material exposed to a change of magnetic field. The MCE requires the exchange of magnetic entropy, lattice entropy and/or electronic entropy during an adiabatic (de-)magnetization process. A large MCE at room temperature and low magnetic field for a material with abundant and environmentally friendly elements opens the way for magnetic cooling devices [1-3]. The ferromagnetic compound $MnFe_4Si_3$ is a promising candidate material for such devices as it has a magnetic phase transition of about T_c =300 K and shows a moderate MCE of 2.9 J/(kg·K) at a reasonable magnetic field change from 0T to 2T. So far studies of the $MnFe_4Si_3$ compound have concentrated mainly on the crystallographic and magnetic structure in powder and single crystal samples [4]. To add to the understanding of the fundamental driving forces of the MCE on the magnetocaloric compound $MnFe_4Si_3$ a study of spin and lattice dynamics and their interactions is necessary, a topic never covered for magnetocaloric compounds so far.

Aim of the experiment:

The aim of the experiment was to investigate further: (i) the phonon-magnon coupling in complement to measurements already performed [5-7] and (ii) the difference of intensity between the two non-spin flip (NSF) channels that is related to the nuclear magnetic interference term (NMI) close to the Brillouin zone boundary [8].

Experimental setup:

The IN12 spectrometer was set up in -1, 1, -1 configuration. For the experiment we used: a velocity selector, polarizer, guiding field, double focusing PG (002) monochromator, two flippers, monitor and a Heussler analyzer. Also slits were put before and after the sample. The single crystal (with a mass of about 7g) was mounted with the [100] – [001] directions in the scattering plane inside a 2.5T magnet. Vertical field of H=1T was applied parallel to the b axis of the hexagonal system of the sample, which is corresponding to the easy axis. After tuning the flipper using a graphite sample (flipping ratio: 16) the $MnFe_4Si_3$ single crystal was cooled down from 316K to 1.5K under magnetic field of 1T in order to have a single domain sample. Data have been collected with a fixed k_f=2.0Å^{-1}.

Results:

In order to further investigate a possible phonon-magnon interaction, polarized INS experiment was performed using spin-flippers before and after the sample to access the four possible longitudinal polarized INS cross-sections. The main result obtained from our pervious studies is the observation a large difference of intensities in the two non-spin-flip (NSF) channels (flippers ‘off-off’, ‘on-on’) for the TA phonon propagating along [1 0 0] and polarized in [0 0 1] for low energy transfers far below the crossing point of phonons and magnons, e.g. at 4meV [8].

At low temperatures (T=1.5K), it is also found that the full TA branch shows a complete cancelation of intensities in the two NSF channels (see Fig.1 for data at E= 6 meV), except in the vicinity of the zone Brillouin boundary (E>9 meV). In the range of wave-vectors where the phonon is measured in our experiment, there are no spin-waves and no diffuse spin fluctuations (E≤9 meV), therefore the only possible mixed nuclear-magnetic correlation function involves the uniform static magnetization of the system. This suggests that, in this case, the origin for the NMI is the magnetovibrational scattering, a scattering elastic in the magnetic system and inelastic with respect to atomic vibrations. The nuclear part is almost constant consistently with the weak variation of the Debye-Waller factor in a small temperature range with respects to our limited statistics [8]. The magnetic part rises as ferromagnetic ordering is growing up. The phonon intensity cancellation at low temperature is due to an accidental coincidence of the nuclear and magnetic amplitudes. In the vicinity of the zone Brillouin boundary the acoustic approximation [9-10] is not valid significant intensity is observed in the two NSF channels.

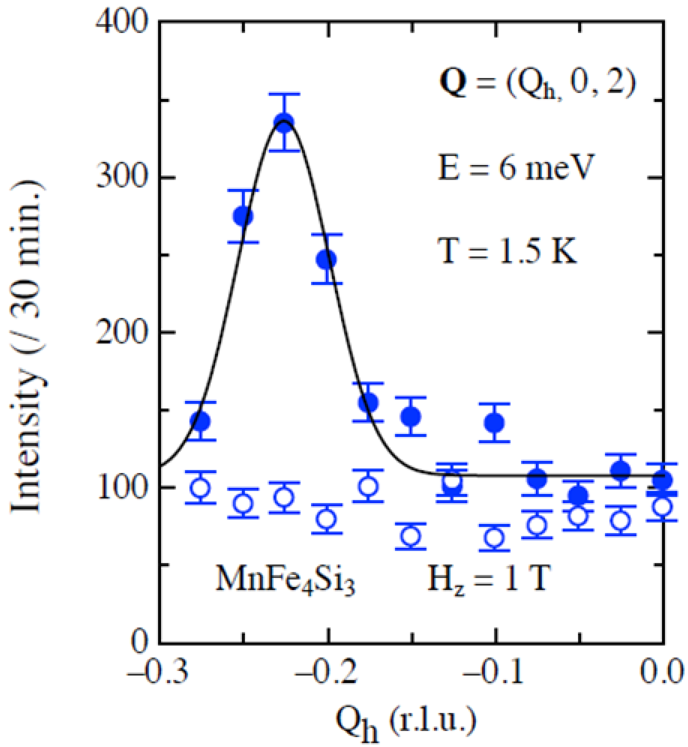

Fig. 1: Polarized INS spectra (raw data: **Q**= (Q_h, 0, 2) at const. E=6meV) obtained at 1.5K under constant vertical magnetic field H_Z=1T. Full symbols: NSF measurement with two flippers on, open symbols: NSF measurement with two flippers off.

[1] O. Tegus et al., Nature 415 (2002) 150. [2] N.H. Dung et al., Adv. Energy Mater. 1 (2011) 1215. [3] X. Moya et al., Nature materials, 13 (2014) 439, [4] P. Hering et al., Chem. Mater. (2015), 27 (20), 7128–7136, [5] CRG-2263 Experimental Report, [6] CRG-2172 Experimental Report, [7] CRG-2292 Experimental Report, [8] CRG-2331 Experimental Report, [9] Squires G. L. Introduction to the Theory of Thermal Neutron Scattering (1978) (Cambridge : Cambridge University Press). [10] Xu G. et al. Rev. Sci. Instrum. 84 083906 (2013).