Experimental report

15/06/2018

Proposal: **CRG-2331** **Council:** 4/2016

Title: Anomalous phonon in MnFe4Si3

Research area:

This proposal is a new proposal

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Samples: MnFe4Si3

Instrument	Requested days	Allocated days	From	To
IN12	7	13	30/08/2016	06/09/2016
			10/02/2017	16/02/2017

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 the phonon-magnon coupling in complement to measurements already performed [5-7].

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=2.0$Å^{-1}$.

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 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. As can be seen in Fig. 1 this phonon mode has significant intensity in only one NSF channel ('on-on'), while in the other NSF channel ('off-off') the intensity equals the background level. This difference of intensity between the two NSF channels is related to the nuclear magnetic interference term (NMI): $I_{NSF}^{on-on} - I_{NSF}^{off-off} = 2|I_{NMI}|$. To further characterize the evidenced NMI term in $MnFe_4Si_3$, its temperature dependence was studied (Fig. 3; the corresponding spectra at 316 K are shown in Fig.2). The temperature dependence of the NMI term mimics the one of the magnetic order parameter (with finite value above T_c due to the finite magnetic field). This might suggest that the observed NMI term is elastic in the magnetic subsystem.

Establishing the origin of the observed NMI in this system could pave the way for first microscopic explanations of the magnetocaloric effect in these materials. Changes in temperature and magnetic field are major parameters for the magnetocaloric effect and their impact will further elucidate the underlying mechanism.

Fig. 1

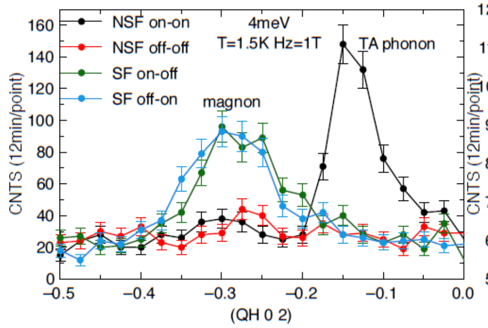

Fig. 2

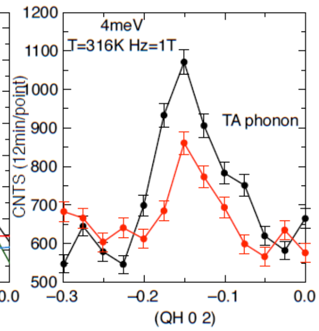

Fig. 3

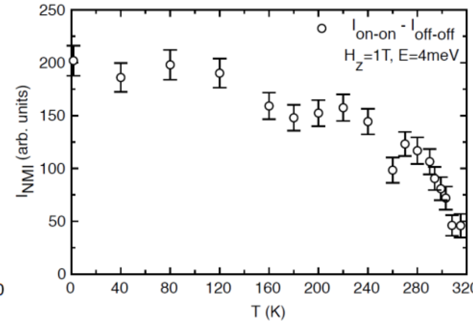

Fig. 1 - 2: Polarized INS spectra (raw data: (Q_h, 0, 2) scans at const. E=4meV) obtained at 1.5K (Fig.2) and 316K (Fig.3) under constant vertical magnetic field H_Z=1T. Black: NSF measurement with two flippers on, red: NSF measurement with two flippers off, green: SF measurement with 1st flipper on and 2nd off, blue: SF measurement with 1st flipper off and 2nd on. Lines are used as guides for the eyes. **Fig. 3:** Temperature dependence of $\left|I_{NMI}\right| = I_{NSF}^{on-on} - I_{NSF}^{off-off}$. Points correspond to the difference of phonon intensities in the peak Q=(-0.15, 0, 2) at 4meV. For I_{NSF}^{on-on} and $I_{NSF}^{off-off}$ background and Bose factor corrections were applied. All data have been collected under vertical magnetic field H_z=1T. During the temperature change lattice parameters were corrected when necessary.

[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