Experimental Report

17/03/2014

Proposal: **4-01-1318** **Council:** 10/2012

Title: Lattice and magnetic dynamics in Magneto-Caloric MnFe4Si3

This proposal is a new proposal

Researh Area: Physics

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Samples: MnFe4Si3

Instrument	Req. Days	All. Days	From	To
IN4	5	3	25/02/2013	28/02/2013

Abstract:

We propose to investigate the lattice and magnetic dynamics in Magneto-Caloric MnFe4Si3. A large MCE in often observed when the magnetic phase transition is combined with changes of the underlying lattice, so that not only the magnetic but also the lattice entropy is changed. Using time-of-flight spectroscopy we aim for the extraction of the magnetic and the lattice entropy in MnFe4Si3 by combination with data from inelastic nuclear scattering and a non-magnetic reference sample. We intend to measure the temperature dependence in the magnetically ordered state and in the paramagnetic state. These results will provide a better understanding of the MCE in MnFe4Si3, which is a potential candidate for an abundant and non-toxic magnetic refrigerant.

Novel technologies for refrigeration have an enormous potential to increase the energy efficiency. The MCE base refrigerators are in use today mainly for low temperature applications, but their potential for domestic applications is well acknowledged. Best materials contain often toxic (As, P) elements or expensive Rare Earth elements. The observation of a modestly large MCE in $MnFe_4Si_3$ [2] has stimulated a project to investigate the dynamical properties with a focus on the lattice excitations. We therefore synthesized single crystals of this compound by the Czochralski method. The quality and phase purity of the sample was checked by magnetization measurements and by powder diffraction measurements on a crushed fragment of the large single crystal specimen. The results correspond well to the ones reported in [1][2]. 10.6 g of the crushed material have been put into the cryofurnace on IN4 and neutron spectra have been recorded at several temperatures on the magnetically ordered and in the paramagnetic state. For faster temperature changes the sample environment was changed to the cryoloop, to control the sample temperature between 200 K and 400 K more promptly. For this configuration a new background measurement was performed, while the same vanadium measurement has been used to calibrate all measurements. To account for the detector efficiency we measured the scattering from a standard vanadium sample. Furthermore we determined an instrumental background by empty can measurements for all instrument configurations that we used. The data has then been reduced to $S(Q,\omega)$ using the 'Lamp' software.

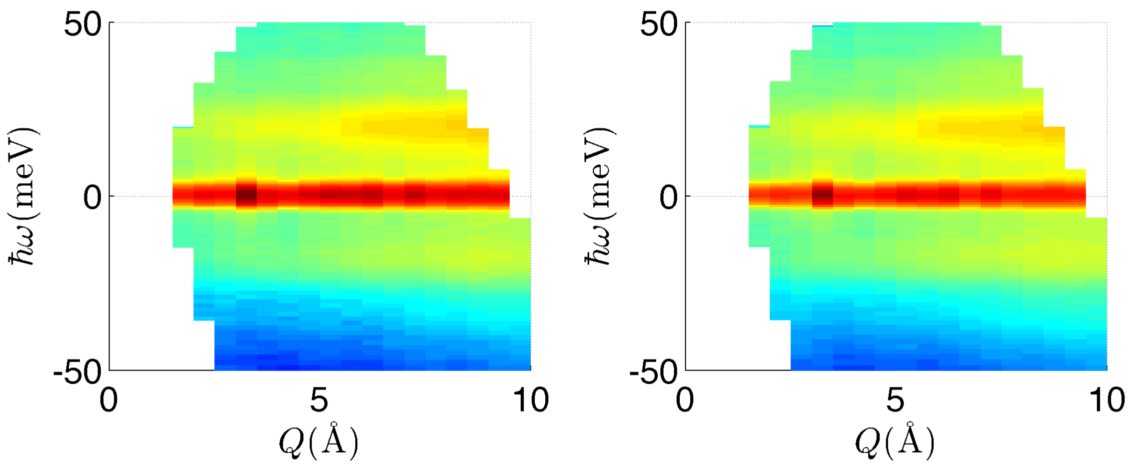

Figure 1: Q,ω maps at 200 K (left) and 400 K (right) measured with an initial neutron energy $E = 65$ meV. The scattered intensity is given on a logarithmic color scale.

Two typical $S(Q,\omega)$ maps are shown in fig. 1, measured at 200 K and at 400 K, respectively. The lower temperature data is in the magnetically ordered state, the high temperature data was measured well above the Curie temperature $T_C \approx 305$ K. The most prominent features are observed in the energy range from 18 to 60 meV. From the Q-dependence it is clear that they originate from optical phonon branches. The individual branches become clearer when the data is angle integrated. We show here a generalized density of phonon states by integrating $S(Q,\omega)$ in Q. Besides a slight increase in intensity as expected for phonon contributions we observe no significant change of the phonon spectrum across the phase transition.

To measure the energy gain response with better energy resolution we have performed additional measurements with a longer wavelength $\lambda = 2.4$Å corresponding to a energy $E_i \approx 14$ meV. As an example we show the $S(Q, \omega)$ measured at 400 K. We clearly resolve here some acoustic mode with better resolution. Beside this additional information we will use the spectra measured with different wavelength to get a reliable estimate of the multiple scattering contributions. The data will also be compared to nuclear resonance inelastic scattering that has been measured to establish the partial density of phonon states for the Fe in the sample.

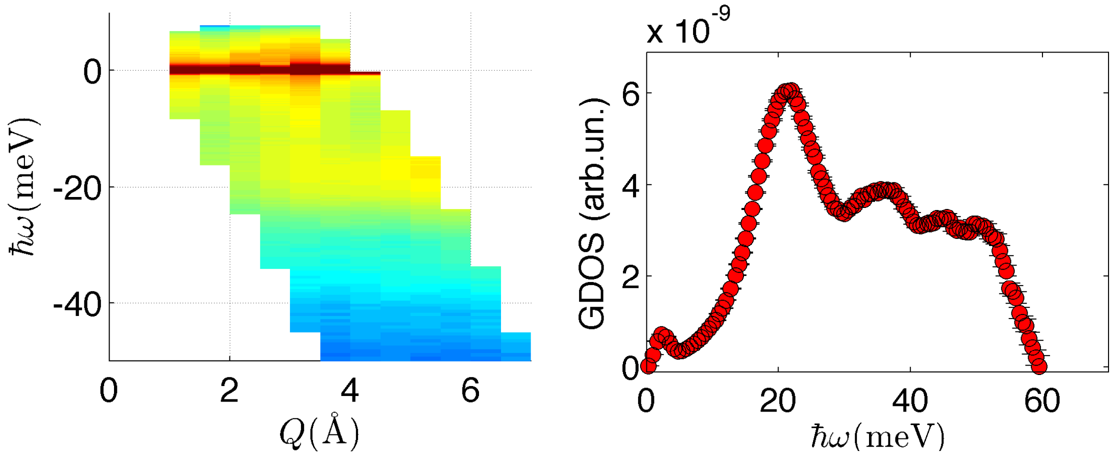

Figure 2: Left: $S(Q, \omega)$ measured at 400 K with an initial neutron energy $E = 14$ meV. Right: Generalized density of phonon states measured at 400 K with an initial neutron energy $E = 65$ meV.

References

[1] A. Candini, O. Moze, W. Kockelmann, J. M. Cadogan, E. Brück, and O. Tegus. Revised magnetic phase diagram for FexMn5xSi3 intermetallics. *Journal of Applied Physics*, 95(11):6819–6821, June 2004.

[2] Songlin, Dagula, O. Tegus, E. Brück, J. Klaasse, F. de Boer, and K. Buschow. Magnetic phase transition and magnetocaloric effect in Mn5xFexSi3. *Journal of Alloys and Compounds*, 334(1–2):249–252, Feb. 2002.