Investigation of the density of phonon states of the Mn5-xFexSi3 Magnetocaloric Materials

M. Herlitschke[1,2], M. Gottschlich[1], B. Klobes[1], J. Voigt[1], J. Perßon[1], Th. Brückel[1] and R. P. Hermann[1,2]

[1]*Jülich Centre for Neutron Science JCNS and Peter Grünberg Institut PGI, JARA-FIT Forschungszentrum Jülich, Jülich, Germany*
[2]*Faculté des Sciences, Université de Liège, Liège, Belgium*

1. Aims of the experiment and scientific background

The magneto-caloric effect (MCE) based on entropy changes of magnetic materials in an applied magnetic field, holds the potential for applications such as refrigeration without moving mechanical parts. Therefore it has recently attracted the attention of many scientific groups [1]. The magnitude of the MCE depends on magnetic and structural changes influencing the entropy of the system. This effect is largest at (or near) phase transitions. Many materials have been investigated to explore their usefulness for applications and of course to understand the effect itself [2,3].

For the MCE to occur, an energy transfer has to take place between two reservoirs in the material: the lattice and the spin entropy hence related to the phonon and magnon thermal bath. This transfer is driven by an applied magnetic field and varies in temperature [4,5]. In the context of the MCE inelastic neutron scattering experiments have already been performed on several compounds to investigate magnetic dynamic properties [6,7].

In order to understand the origin of the MCE in those materials, nuclear inelastic x-ray experiments are crucial to determine the density of the phonon states. Once those states are known complete information on several important thermodynamical properties such as the internal energy per atom, vibrational entropy is available [8]. Furthermore, by calculating the lattice entropy and comparing it to MCE measurements, for example, detailed knowledge of the origin of the MCE will be obtained as it has been done in [7]. One hypothesis that we are hoping to confirm with the proposed experiment is the close relationship between MCE and magnon-phonon coupling. Polycrystalline samples enriched with 57Fe for the compositions x=1,2,3,4 were prepared for the experiment reported herein.

2. Measured data - analysis

During the beamtime the nuclear inelastic scattering data for x=1,2,3 and 4 was obtained for different temperatures. Fig. 1 shows the results for x=3 and 4. A phonon softening for x=4 near room temperature seems to be found.

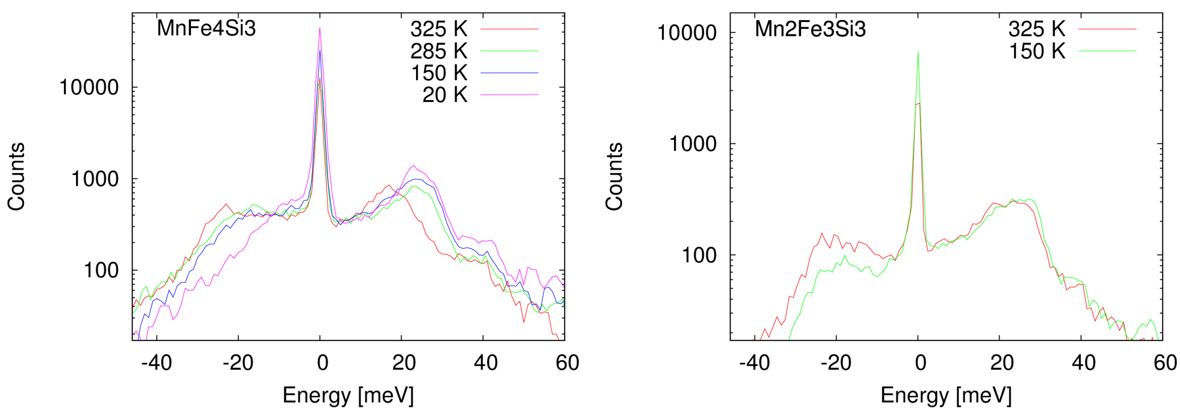

Figure 1: Nuclear inelastic Scattering data from x=3 and 4 for different temperatures. A phonon softening seems to be occur in$x$=4 near room temperature.

But a detailed analysis of the raw data reveals strong discrepancies in all measured spectra. A comparison of the left and the right side of the inelastic spectra normalised by the Bose-Einstein statistic shows anomaly in the behaviour of the phonon states (see Fig 2a).

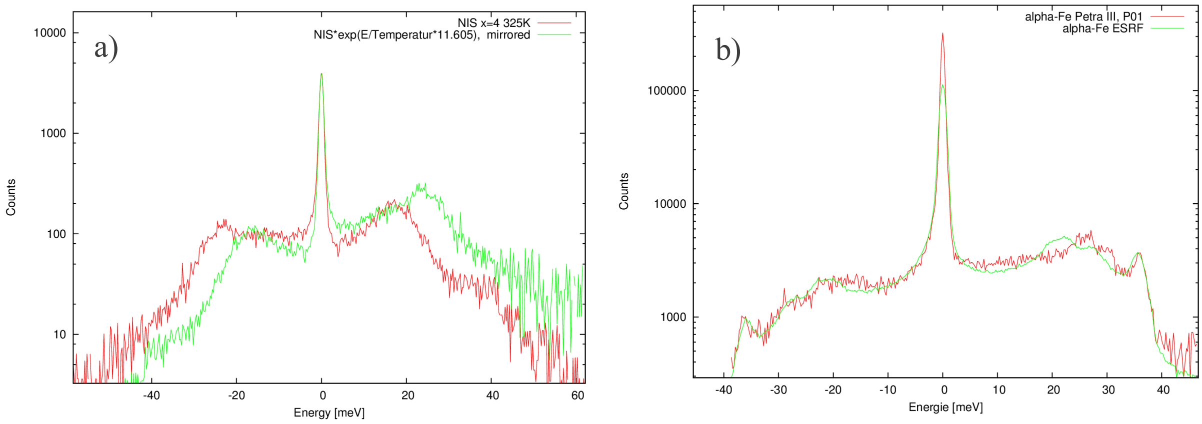

Figure 2: (a) Inelastic scattering from x=4 at 325 K. The discrepancies in the behaviour of the phonon states are probably caused by non-linearities in the high resolution monochromator. The x-axis was obtained by normalizing the 1064-Theta-Angle of the monochromotaor. (b) nelastic scattering from alpha-Fe measured at ESRf and Petra III. The comparison of both spectra shows strong discrepancies in the phonon states between 10 to 30 meV.

This discrepancies, which can not explained by physically effects, occurred due to problems with drive of the high resolution monochromator. A comparison with α-Fe data measured during this beamtime and α-Fe measured at ESRF reveals this directly (see Fig 2b).

This behaviour was observed for nearly every spectrum and also for two spectra with same conditions. This means that a shift of phonons observed in two different spectra could be explained either with a physical effect or a with incorrect positioning of the high resolution monochromator.

An additional problem could be a non constant background. A higher background on the negative side of the spectrum compared to the positive side was detected in nearly all spectra.

3. Results

As consequence of this observations every observed shift can not be trusted and the data is not applicable for calculation of lattice entropy. All of this data that was measured is not usable and would have to be remeasured.

References

[1] A. M. Tishin and Y I Spichkin, “The magnetocaloric Effect and its applications”, Institute of physics, Series in condensed matter (2003).

[2] K. A. Gschneider Jr, V K Pecharsky and A O Tsokol, “Recent developments in magnetocaloric materials”, Rep. Prog. Phys. 68, 1479-1539 (2005).

[3] Lui Min and Yu Bing-feng, “Development of magnetocaloric materials in room temperature magnetic refrigeration application in recent six years”, J. Cent. South Univ. Technol. , 0001-0012 (2009).

[4] R. Szymczak, R. Kolano, A. Kolano-Burian, V.P. Dyakonov and H. Szymczak, “Giant Magnetocaloric Effect in Manganites”, Acta. Phys. Pol. A 117, 203-206 (2010).

[5] Th. Svobodny and Z. Kalva, “Magnetocalorice Effect in Ferromagnetic and Superconducting Materials”, Phys. Stat. Sol. (b) 208, 187-196 (1998).

[6] C. Vecchini et al., “Dynamic susceptibility of Gd5Si2Ge2 and Gd4YSi1.9Ge2.1”, J. App. Phys. 95, 11, 7207-7209 (2004).