Experimental report

| Proposal: | 4-01-177 | /3 | Council: 10/2022 | | | | | |
|---------------------------------|----------|---|-------------------------|----------------|------------|------------|--|--|
| Title: | Paramagr | Paramagnetic scattering of the magnetocaloric compound MnFe4Si3 | | | | | | |
| Research area: Materials | | | | | | | | |
| This proposal is a new proposal | | | | | | | | |
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| Samples: MnFe | e4Si3 | | | | | | | |
| Instrument | | | Requested days | Allocated days | From | То | | |
| IN22 | | | 7 | 6 | 07/04/2023 | 18/04/2023 | | |

Abstract:

We propose to study the paramagnetic scattering of the magnetocaloric compound MnFe4Si3 in order to obtain its characteristic length and energy scales. We will investigate possible deviations of the temperature dependence of the inverse spin correlation length from know scaling laws and we will address the question of the nature of magnetism in this system. To this aim a mapping in reciprocal space of the paramagnetic scattering for several wave vectors and temperatures using the thermal TAS IN22 with polarization analysis seems the most efficient method. We hope that this microscopic investigation will shed light on the fundamental ingredients playing a pivotal role in the magnetocaloric effect of this compound. Such a study has never been performed before in other promising magnetocaloric compound.

Scientific background:

One way for saving energy in daily life is using the magnetocaloric effect (MCE) and this has led to a growing interest for the research field of magnetocaloric materials. The MCE corresponds to the change of magnetic entropy and adiabatic temperature following a change in an applied magnetic field around the magnetic transition temperature. A large MCE at room temperature and low magnetic field for a material with abundant and environment friendly elements opens the way for magnetic cooling devices. The

ferromagnetic compound MnFe₄Si₃ (S.G.: $P\overline{6}$) is a promising candidate material for such devices as it has a magnetic phase transition in the range of 300 K and shows a moderate MCE of 2.9 J/(kg·K) at a reasonable magnetic field change from 0T to 2T.

In order to understand the fundamental driving force of the MCE on the magnetocaloric compound Mn₄FeSi₃, a study of magnetism, lattice dynamics, spin dynamics and their interaction is necessary with inelastic neutron scattering experiments.

Aim of the experiment:

The aim of the experiment was to investigate the paramagnetic scattering in the magnetocaloric compound $MnFe_4Si_3$ in several temperatures above T_c .

Experimental setup:

The instrument was set up in -1, 1, -1 configuration. For the experiment, a Heusler monochromator, a monitor, a PG filter and a Heusler analyzer were used. All along the neutron path, guide fields were installed to maintain the polarization of the beam. In order to access different polarization channels Helmholtz coils were employed. The single crystal (with a mass of about 7g) was mounted with the [100] – [001] directions in the scattering plane inside a cryofurnace. All data have been taken with fixed $k_f=2.662$ Å⁻¹. The offset of the analyzer was measured with a vanadium sample.

Results:

Inelastic neutron scattering measurements at IN22 spectrometer were performed on a MnFe4Si₃ single crystal at several temperatures above the Curie temperature (Tc=305 K). The flipping ratio was about 12. Data have been collected along the hexagonal reciprocal directions (*h00*) and (*001*) around the zone centers Q=(2,0,0) and Q=(0,0,2) for energy transfers between $-4\leq E\leq 4$ meV in two non-spin flip channels (NSF_{xx} and NSF_{zz}). The measuring time per point per channel was about 10 minutes. Using canonical subtractions of intensities measured in the two different polarization channels we managed to remove the complex background which is q, E and T dependent (see Fig.1). Further analysis allowed us to extract the magnetic relaxation rates $\Gamma(q)$ and the q-dependent susceptibility $\chi(q)$ along (*h00*) and (*001*) at different temperatures.



Fig. 1: Energy dependence of magnetic fluctuations at Q=(0, 0, 1.925) at T=367 K and Q=(1.8, 0, 0) at T=457 K. The lines represent a fit corresponding to a relaxation function. Monitor corrections were applied to the data and for all fittings the center was fixed. The center was determined from a measurement of the incoherent scattering of the sample.