Experimental Report

Proposal: **5-31-2257** **Council:** 10/2012

Title: The Effect of Deuterium Absorptionon Magnetic Ordering, Crystal Structure and Phase Transformations in Mn3Ga Materials.

This proposal is a new proposal

Researh Area: Materials

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Samples:
Mn3Ga D022
Mn3Ga D019
MnAlC
Mn3Ga
Mn65Ga35

Instrument	Req. Days	All. Days	From	To
D1B	3	3	26/07/2013	29/07/2013

Abstract:

Mn3Ga with the D022 structure has ferrimagnetic ordering whereas in Mn3Ga D019, triangular antiferromagnetic ordering is observed. Both of these structures have a low magnetic moment which makes them unsuitable as a permanent magnet material. Expansion of the crystal lattice by absorbing deuterium onto the interstitial sites strongly affects the magnetic coupling between the Mn atoms and may lead to a much higher magnetic moment. Neutron powder diffraction experiments are required to characterise the effect of deuterium absorption on the local magnetic moments in D019 and D022 Mn3Ga. Neutron thermodiffractometry will also be used to study the as yet unknown nature of the high temperature phase transformation in the system. The results will not only be critical to the developement of Mn3Ga-based materials as permanent magnets but will also contribute more detailed knowledge of the fundamental physical properties of the system.

ILL report for experiment 57513

Experiment Date: 26.07.2013 – 29.07.2013
Proposer: Thomas G. Woodcock
Local contact: Vivian Nassif

Changes to the proposed experiment

The proposal was originally to investigate the effect of deuterium absorption on the properties of the samples. Although initial experiments showed some possible hydrogen absorption, more detailed work before the start of the experiment showed that hydrogen absorption was minimal and that no effect on the crystal lattice or magnetic properties could be seen. In light of this, the focus of the experiments at ILL was shifted to studying phase transitions and magnetic structures in the samples, which are the topic of current discussions in the literature and are still not fully understood.

Measured samples:

Five different kind of sample were provided for the experiment. For each sample a sufficient amount of powder to fill the 5 mm in diameter sample holder was available. The different kinds of sample were the following:

- Cubic Mn_3Ga powder
- Tetragonal $D0_{22}$ Mn_3Ga powder
- Tetragonal $L1_0$ $Mn_{65}Ga_{35}$ powder
- MnAlC powder in tau phase
- MnAlC powder in epsilon phase

Measurement:

The mounting of the sample into the sample holder was carried out under argon atmosphere in a glove box to avoid the contact with oxygen.

The neutron experiments took place at the D1B detector. To calibrate the detector measurements with a NAC and Si sample were done. To subtract the background the empty sample holder was measured for 1 hour.

After a long time measurement of 1 hour the Mn_3Ga samples were heated up with a heating rate of 1 K/min. During the heating process every 5 min a diffraction pattern was captured. Additional long term measurements were carried out at 200°C, 300°C, 400°C, 600°C and 800°C. Thereby the phase transition from the tetragonal $D0_{22}$ to the hexagonal $D0_{19}$ structure could be observed. Unfortunately a second phase transition of the $D0_{19}$ structure could not be seen. At 800°C the measurement signal became very noisy and no peaks could be identified. This was not due to melting of the sample. On cooling through the phase

transition temperature, the peaks of the low temperature phase reappeared and the measurement continued as planned.

For the MnAlC tau phase powder long exposure spectra were captured at room temperature, 400°C, 700°C, 850°C and 250°C. During the heating and cooling period every 5 min one spectrum was measured. As for the Mn-Ga samples, when passing through the high temperature phase transition, the measurement became very noisy and no peaks were visible. On cooling through the phase transition, the peaks of the low temperature phase reappeared. No explanation was found for this behaviour. The MnAlC sample with the epsilon phase showed no peaks at all when measured at room temperature although x-ray diffraction patterns carried out in our own lab showed clear peaks. This sample was not measured further.

Problems:

- The detector was offline for a short while due to the hot weather and overheating of the detector.
- No peaks could be observed in the epsilon structure of the MnAlC sample at room temperature. The reason was unknown.
- After heating the Mn_3Ga and MnAlC samples above 800°C the diffraction pattern vanished. Until now it is unknown if a phase transition occurs at this temperature or if there different reasons.

Results/Plans:

The crystal structures of the Mn_3Ga $D0_{22}$, $D0_{19}$ and MnAlC $L1_0$ structure could be fitted to the obtained patterns. The Curie temperature of the MnAlC material was < 300°C and long exposure patterns recorded above and below the Curie temperature revealed very clearly which peaks have a magnetic contribution. The Curie temperature of the Mn3Ga sample is above the structural phase transition and therefore it was not possible to obtain a neutron diffraction pattern without magnetic contributions. This would have been very useful in order to determine the magnetic structure of the compound, which is currently under discussion in the literature. With the patterns gathered during the heating and cooling some of the structural phase transformations are clearly visible. First attempts to refine the magnetic structure are promising, but further work is needed to obtain a fully refined crystal and magnetic structure of the Mn3Ga sample. If this can be achieved, the results will be published.