Experimental report

20/02/2020

Proposal:	**5-32-886**		**Council:**	4/2019
Title:	Antiferromagnetically coupled anti-phase domains under external field			
Research area:	Materials			

This proposal is a new proposal

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Samples: Ni2MnAl
Ni2MnAl0.5Ga0.5

Instrument	Requested days	Allocated days	From	To
D33	5	3	27/09/2019	30/09/2019

Abstract:

We propose to measure the magnetic small-angle neutron scattering on powders of Ni2MnAl and Ni2MnAl0.5Ga0.5 in various annealing conditions under various external fields at cryogenic temperatures at D33. Our aim is to clarify whether the ferromagnetic domains, which correspond to the structural anti-phase domains as verified in previous experiments, are inherently antiferromagnetically coupled over the anti-phase domain boundaries.

Antiferromagnetically coupled anti-phase domains under external field

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Ni_2MnZ based Heusler compounds have attracted a considerable amount of attention due to their various appealing properties such as the ferromagnetic shape-memory effect or the magnetocaloric effect. Since the magnetic properties are very sensitive to the degree of structural order in these systems [1], understanding the correlation of these two ordering parameters is crucial. The magnetic moments are mainly carried by the Mn atoms, which interact ferromagnetically in the $L2_1$-ordered state. However, it has been found that at structural antiphase domain boundaries (APBs) the magnetization tends to reverse, leading to atomically sharp ferromagnetic domain walls [2]. In a previous small angle neutron scattering experiment at SANS-1 at the FRM II, Garching, under zero magnetic field we investigated the relation of structural and magnetic microstructure in powder samples of Ni_2MnAl and $Ni_2MnAl_{0.5}Ga_{0.5}$ in different annealing states and demonstrated that their characteristic length scales are of the same size.
To clarify the reason of this relation we studied further the influence of coupling across APBs by applying small-angle neutron scattering under external magnetic fields. Since the scattering signal is sensitive to the component of the magnetization perpendicular to $\vec{Q}$, we can distinguish between two possible scenarios of exchange coupling across APBs: In the first case there is no exchange coupling across APBs, leading to isolated ferromagnetic antiphase domains (APDs). The magnetic scattering signal of this scenario corresponds to that of ferromagnetic nanoparticles under a magnetic field, resulting in a redistribution of intensity in the SANS signal perpendicular to the magnetic field [3]. In the second scenario the APDs couple antiferromagnetically across APBs, displaying a behaviour similar to that of an elemental antiferromagnet. Here the spins align perpendicular to already small external magnetic fields, corresponding to a redistribution of intensity of the SANS signal along field direction.
The magnetic SANS experiment was carried out at the instrument D33 at the Institute Laue-Langevin (ILL), Grenoble, France. We performed measurements on powder samples of Ni_2MnAl and $Ni_2MnAl_{0.5}Ga_{0.5}$ in different annealing states using unpolarized incident neutrons with a mean wavelength of $\lambda = 5\,\text{Å}$, $\Delta\lambda/\lambda \sim 10\%$. For each sample, we used the same sample-to-detector distances of $1.2\,\text{m}$ for the front detector and $2\,\text{m}$ for the rear one with a corresponding collimation length of $2.8\,\text{m}$. We measured the SANS signal in a temperature range of $50\,\text{K}$ to $500\,\text{K}$ and applied a magnetic field perpendicular to the incident beam in a range of $0\,\text{mT}$ to $640\,\text{mT}$.
To distinguish between the two scenarios we compare the 2D detector images of measurements under zero magnetic field and with a small magnetic field present at temperatures well below the critical temperature of the sample. The obtained 2D detector images of the sample $Ni_2MnAl_{0.5}Ga_{0.5}$ $3\,\text{h}$ annealed, $T_c = 342\,\text{K}$, are shown in figure 1. Figure 1a) depicts the detector image of the measurement at $60\,\text{K}$ under zero magnetic field. Here we observe an isotropic scattering picture from our powder sample, corresponding to an isotropic spin distribution in our sample. The 2D detector image of the measurement at $60\,\text{K}$ with an applied magnetic field of $640\,\text{mT}$ is illustrated in figure 1b). With a magnetic field present we see a distinct redistribution of intensity along the magnetic field direction, indicating spins aligning perpendicular to the magnetic field.
To analyze the data further we radially average the scattering images in the Q-range from 0.06 to $0.1\,\text{Å}^{-1}$. The scattering image of the measurement $Ni_2MnAl_{0.5}Ga_{0.5}$ $10\,\text{d}$ annealed at $60\,\text{K}$ with an applied magnetic field of $640\,\text{mT}$ is shown in figure 2a). Figure 2b) depicts the corresponding

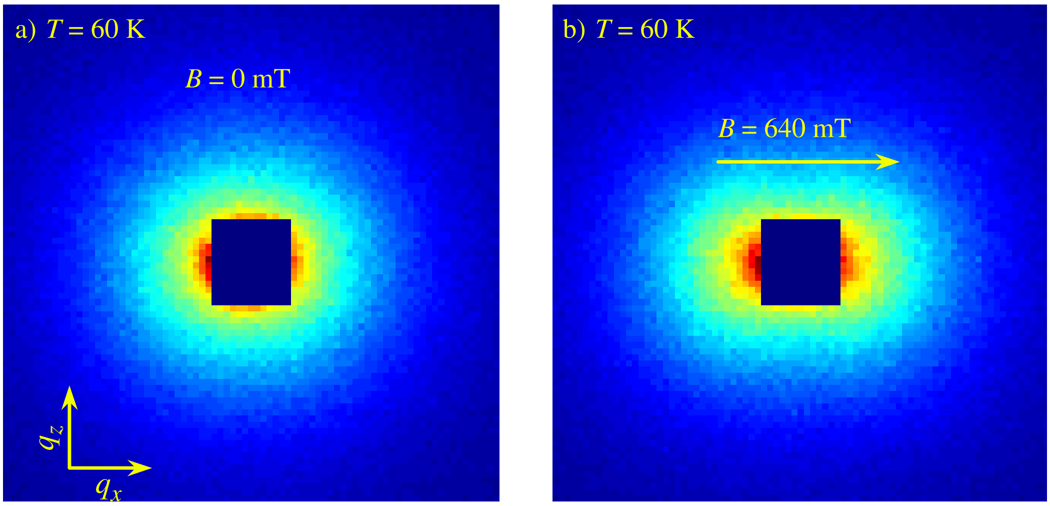

Figure 1: 2D SANS profiles of $Ni_2MnAl_{0.5}Ga_{0.5}$ 3 h annealed at a) 60 K with no field applied and b) 60 K with a horizontally applied magnetic field of 640 mT.

scattering profile depending on the angle β between $\vec{q}$ and the magnetic field $\vec{B}$ normalized by the average intensity in this Q range. For $\vec{q}$ parallel to $\vec{B}$ we observe 13% more intensity than in average, while for $\vec{q}$ perpendicular to $\vec{B}$ we obtain 13% less.

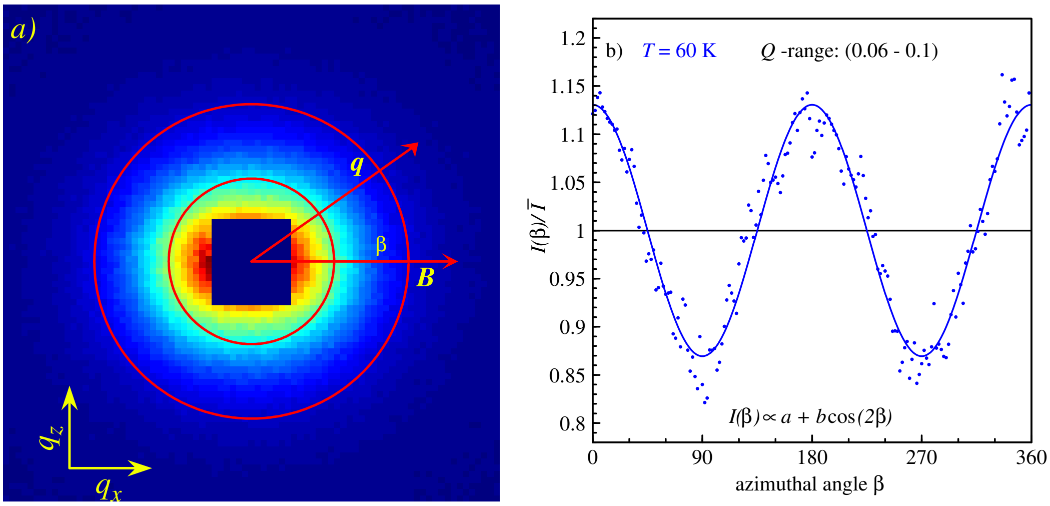

Figure 2: a) SANS signal of the $Ni_2MnAl_{0.5}Ga_{0.5}$ sample 10 d annealed measured at 60 K with a B-field of 640 mT applied and b) the scattering profile depending on the angle between $\vec{q}$ and magnetic field.

In order to quantify the anisotropy of the scattering profiles we fit them with the function $a + b\cos(2\beta)$. By taking the ratio of the fit parameters b/a we obtain a measure for the anisotropy of the signal. Figure 3 depicts the ratio of the system $Ni_2MnAl_{0.5}Ga_{0.5}$ in three annealing states 30 min, 3 h and 10 d in a temperature range of 50 K to 450 K with an applied magnetic field of 640 mT. Increasing the temperature from 50 K towards the transition temperature T_c the anisotropy decreases slowly. Approaching T_c the ratio switches from $b/a > 0$ to $b/a < 0$, indicating a change in the predominant spin orientation, turning them from perpendicular into parallel alignment to the B-field. Increasing the temperature further the ratio converges to 0, corresponding to an isotropic scattering signal resulting from the structural background.

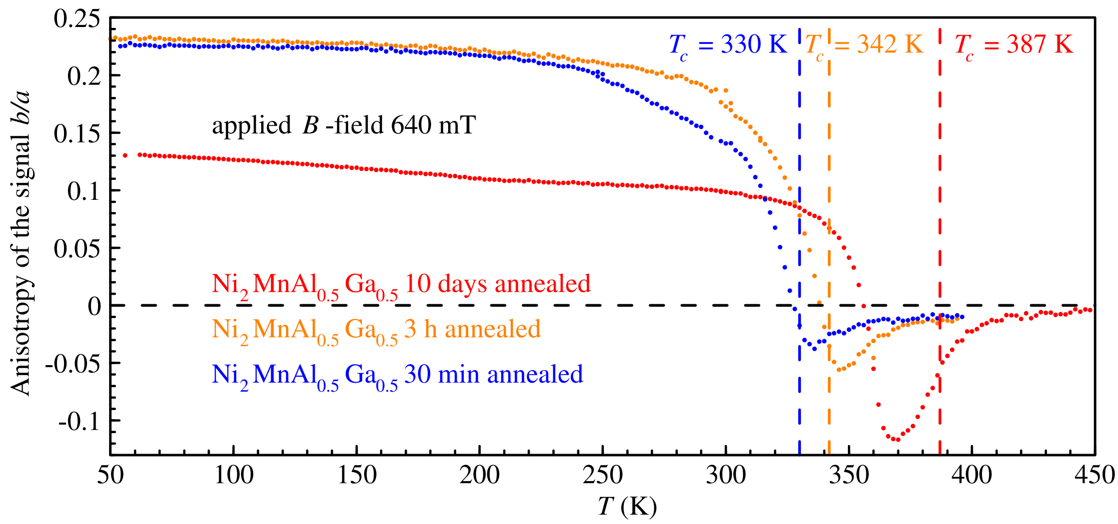

Figure 3: Temperature dependent ratio of the fit parameters b/a of the samples $Ni_2MnAl_{0.5}Ga_{0.5}$ 30 min, 3 h and 10d annealed with a B-field of 640 mT applied. The critical temperatures T_c of the samples were determined in differential scanning calorimetry measurements.

We observe this redistribution of intensity for samples of both alloy systems in all annealing states. From the presented results it can be clearly seen that we obtain signals from our powder samples similar to the temperature dependent response of an antiferromagnet in a magnetic field. This supports our model of antiferromagnetically coupled APDs.
More detailed analysis of these and further scattering patterns at different applied magnetic fields and temperatures is currently ongoing.

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[3] W. Wagner, A. Wiedenmann, W. Petry, A. Geibel, H. Gleiter, *J. Mater. Res* **6**, 2305 (1991)