

Experimental report

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Title: Identification of complex magnetic states in GaV₄Se₈

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Samples: GaV₄Se₈

Instrument	Requested days	Allocated days	From	To
D33	3	3	19/03/2018	22/03/2018

Abstract:

Experimental report for proposal No. DIR-151

Identification of complex magnetic states in GaV₄Se₈

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Introduction

Recently, a new topologically-protected spin structure has been discovered in cubic helimagnets, a lattice of spin vortices, the so-called skyrmion lattice (SkL) [1]. After the first observation of skyrmions using small-angle neutron scattering (SANS), the real-space spin-vortex configuration of each skyrmion was unambiguously confirmed by Lorentz force transmission electron microscopy [2]. In the case of the most studied cubic helimagnets, such as MnSi, (Fe_{0.5}Co_{0.5})Si, Cu₂OSeO₃ (all with the chiral cubic spacegroup P2₁3), a single-q helical ground state is realized due to a competition between the symmetric exchange and the Dzyaloshinskii-Moriya (DM) interaction, where the latter is allowed by the lack of inversion symmetry. The relative strength of these two interactions determines the length of the magnetic q-vector. The SkL, which can be described by three coupled q-vectors connected by a 3-fold symmetry (a triple-q state), is stabilized by finite temperature and magnetic field close to the paramagnetic phase boundary [1]. The present surge of interest in skyrmions is motivated by their use in possible spintronics applications, since these nanoscale spin whirlpools can be detected electrically via the topological Hall-effect [3], and they can be moved by low current densities in metallic skyrmion hosts [4, 5], or low electric fields in the hitherto only known insulating host Cu₂OSeO₃.

Recently, another type of SkL state, the so-called Néel-type or ‘hedgehog’-like SkL was found in the multiferroic GaV₄S₈ [6]. The magnetic building blocks of GaV₄S₈ are tetrahedral clusters of V ions whose unpaired electron with $S=1/2$ spin drives a Jahn-Teller transition, and at $T_s=42$ K the spacegroup symmetry is reduced from cubic ($F\bar{4}3m$) to polar rhombohedral ($R\bar{3}m$) [7]. The axial crystal symmetry in GaV₄S₈ leads to strong easy-axis anisotropy, which stabilizes the ferromagnetic order up to 5 K while the cycloidal (Cyc) and the Néel-type SkL phases persist in the temperature range between 5 K and $T_c=13$ K. On the other hand in the isostructural sister compound GaV₄Se₈ with easy-plane anisotropy, the SkL state found to be stable down to the lowest temperatures, making GaV₄Se₈ unique among all bulk skyrmion host materials [8]. However, in this compound when $H \parallel [111]$ the magnetic phase diagram displays more new phases at low temperature than GaV₄S₈. The primary aim of this experiment was to resolve the magnetic structure in the different magnetically ordered phases of GaV₄Se₈ using polarized SANS. The polarized SANS experiments were carried out at the D33 beamline of the ILL between 19th - 22nd of March, 2018.

Results

First, we tried to measure SANS with longitudinal polarization for $H \parallel [111]$ as described in the proposal, however, this experiment failed for the following reasons. When an electromagnet with a U-shape magnetic core provided the external magnetic field the neutron beam was guided through holes drilled in the core. Since strong enough guide fields could not be generated inside the holes the spin polarization of the beam was lost and we could not measure flipping ratio better than ~ 2 on the detector. We also tried the same experiment with a cryomagnet (‘Blue Charlie’), however, its stray fields were too large and they depolarized the ³He cell sitting in the ‘magic-box’. Even when the cryomagnet was moved to SDI2 =

18000, which is the largest possible distance between the goniometer of the cryostat and the detector, the decay time of the polarization of the ^3He cell was reduced to a few hours, which is comparable to the measurement time necessary for a reasonable signal-to-noise ratio.

As a back-up plan we studied the phase diagram for $\text{H} \parallel [1-10]$ using the electromagnet. In this case the incident beam was propagated perpendicular to the plane of the U shape magnetic core, therefore, sufficient guide fields could be created. The raw experimental results are summarized in Fig. 1. Following a zero-field cooling the ring of intensity and the peaks at $[1-10]$ direction are only visible in the spin-flip (SF) channel in $B = 24 \text{ mT}$ at 12 K . The other four spots corresponding to $[10-1]$ and $[01-1]$ directions still have some intensity in the non-spin-flip (NSF) channel as well since the rhombohedral axis of the corresponding structural domains spans $\sim 35^\circ$ with the field. Qualitatively this result is in agreement with the presence of cycloids in this phase as in the case of GaV_4S_8 [9]. When the field is increased the intensity is moved into two spots with q -vector orthogonal to the field, which is also consistent to have cycloids in the ground state with larger susceptibility perpendicular to the cycloidal plane. When the magnetic field is reduced to 24 mT again (noted by AF = ‘after field’ in Fig. 1) the zero-field cooled state is not recovered indicating a metastable state. A quantitative analysis of the data is in progress.

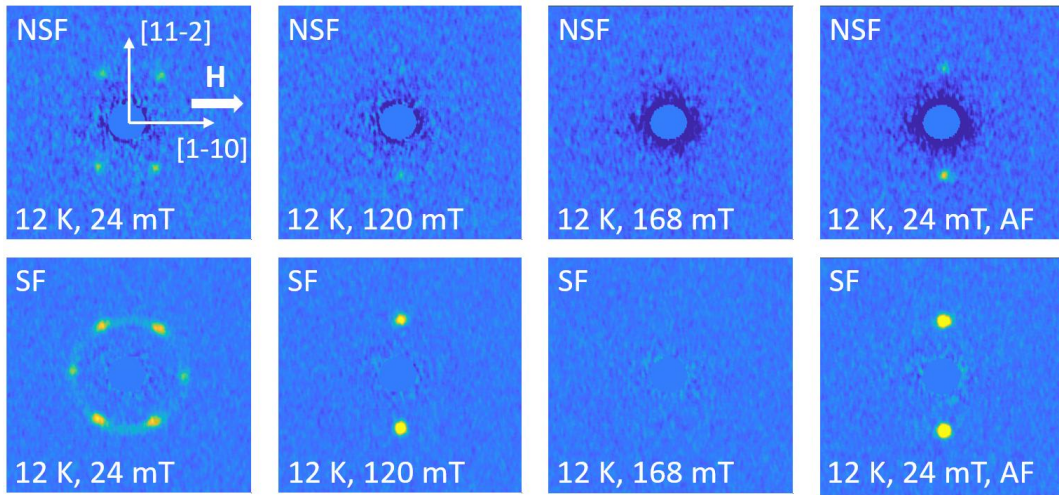


Figure 1 SANS patterns measured both in the non-spin-flip (NSF) and in the spin-flip (SF) channels on the (111) plane with transverse field i.e. $\text{H} \parallel [1-10]$, which set the polarization direction of the incoming neutron beam as well. AF = ‘after field’ corresponds to a low-field measurement carried out after polarizing the spins by 168 mT at 12 K .

Conclusions

We tried to characterize the magnetic phases of GaV_4Se_8 using longitudinal polarization analysis at the D33 SANS instrument, but our attempts failed due to technical issues. In order to complete the main goal of our proposal the stray fields of the cryomagnet needs to be shielded to allow polarized SANS experiments at least up to $\sim 300 \text{ mT}$. The failure of our experiments motivated the D33 team to develop a magnetic shield for the ‘Blue Charlie’ cryomagnet. This improvement will be valuable for the whole community aiming to perform polarized SANS experiment.

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