Experimental report

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Title: Possible skyrmion lattice state inthe lacunar spinel GaV4S8

Research area: Physics

This proposal is a new proposal

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Samples: GaV4S8

Instrument	Requested days	Allocated days	From	To
D33	6	5	08/07/2015	13/07/2015

Abstract:

Modulated spin patterns, such as helical or cycloidal structures, can arise in magnets with broken inversion symmetry due to Dzyaloshinskii-Moriya interaction. Recently, these low-symmetry materials have been discovered to even host a spin vortex lattice, the so-called skyrmion lattice (SkL), which attracts great interest owing to its potential applications in spintronics. We are studying the novel non-centrosymmetric compound GaV4S8 which has a rich largely unexplored magnetic phase diagram that may host skyrmions. Using magnetic force microscopy we have already observed stripes (helical) and particle-like (SkL) magnetic structures in the different low field phases. In addition, a test SANS experiment on GaV4S8 confirms the existence of the long-wavelength magnetic order in the bulk. Using D33, the goals of this proposal are i) to systematically characterize the magnetic order over the magnetic phase diagram, and ii) to confirm the existence of a SkL in GaV4S8. The latter can be achieved by the detection of higher-order scattering, which necessitates the high-flux of the ILL. The anticipated results can confirm the realization of a new form of SkL in a new class of materials.

Experimental report for proposal No. 5-42-392
Possible skyrmion lattice state in the lacunar spinel GaV_4S_8

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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, (Fe0.5Co0.5)Si, Cu_2OSeO_3 (all with the chiral cubic spacegroup $P2_13$), 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 Cu_2OSeO_3.

Recently we studied another candidate, the non-centrosymmetric Mott insulator GaV_4S_8, which host magnetization steps in low magnetic fields. The magnetic building blocks of the GaV_4S_8 FCC lattice are tetrahedral clusters of vanadium ions. On each tetrahedron the vanadium *d*-orbitals are strongly hybridized which leads to a ground state with 3-fold degenerate cluster orbitals and S=1/2 spin [6]. The orbital degeneracy in removed at T_s=42 K by a rhombohedral distortion along the <111> directions resulting in 4 structural domains at low temperatures [6]. The Curie-Weiss analysis of the magnetic susceptibility gives dominantly ferromagnetic coupling, and the magnetic order develops below T_c=13 K [7]. According to our magnetic force microscopy (MFM) experiments the out-of-plane component of the magnetization orders into stripes and the triangular lattice of nanoscale dots. We ascribed these structures to magnetic cycloids and a new type of skyrmion lattice the so-called Néel-type skyrmion lattice. To establish the magnetic order in the bulk material we carried out polarized small angle neutron scattering (SANS) experiments at the D33 beamline of the ILL.

Results

As a first step, neutron scattering experiments were performed at 11 K as a function of the strength and the orientation of the magnetic field to study all the different magnetically ordered phases. Typical images are shown in Fig. 1 when the neutron beam and the external field were parallel to each other. On the (111) plane we observed a ring of intensity and six spots with 6-fold symmetry in zero-field. The length of the bulk q-vector agrees well with the wavelength of the magnetic stripes observed in the

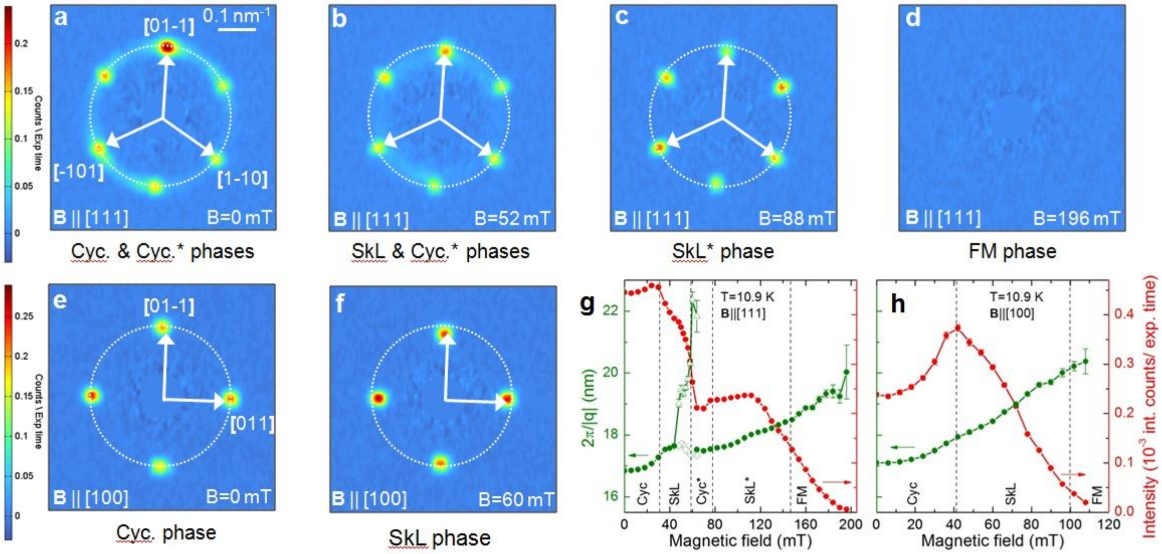

Figure 1 SANS pattern measured in different magnetically ordered phases when B||[111] and B||[100] in panel a)-d) and e)-f), respectively. g)-h) The magnetic field dependence of the wavelength of the modulated order and the scattered intensity. [10]

MFM measurements. When the magnetic field was increased and the magnetic order becomes ferromagnetic in the structural domain with its rhomboherdal axis parallel to the field (later referred as unique domain), the ring of intensity disappears, but the six spots, which correspond to the modulated order in the other three domains (noted by * in Fig. 1), remain stable. By further increasing the field the modulated orders were destroyed in all the structural domains, and no scattered intensity was observed. The ring of intensity directly observed for the unique domain rocks for few degree rotations of the sample. The correlation length measured from the rocking curves is about ~700 nm along the rhombohedral axis, while the orientation of the modulation vector is not well defined within the rhombohedral plane. The six spots are likely to be caused by the intersection of the (111) plane and the intensity rings in the other thee domains. The 4-fold pattern observed in the (100) plane also indicates that all the 4 possible magnetic domains are present in the sample. The changes in the magnetic field dependence of the scattered intensity and in the q-vector of the ordering coincide with the phase boundaries detected by magnetization and specific heat measurements.

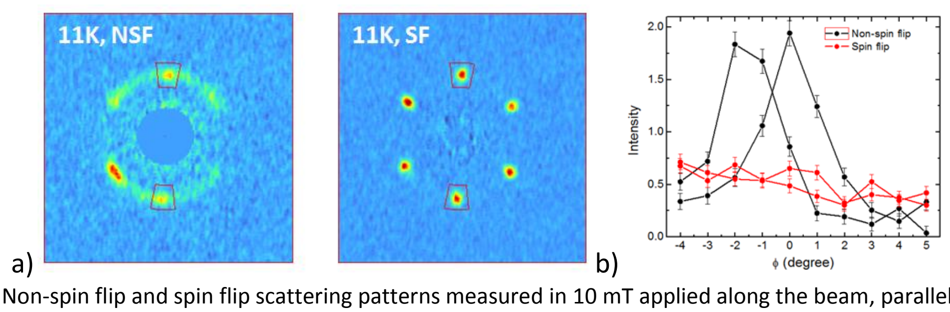

Figure 2 a) Non-spin flip and spin flip scattering patterns measured in 10 mT applied along the beam, parallel to [111]. b) Rocking curves integrated for the red sections.

Next we performed polarized SANS experiments whose key results are shown in Fig. 2 and Fig. 3. When the neutron beam and its polarization are along the rhombohedral axis, [111], the intensity ring of the unique domain appears only in the non-spin flip channel. This strict selection rule implies cycloidal correlations instead of magnetic helices which is also consistent with the polar symmetry of the lattice. When the beam polarization is along the [110] direction the 4 side spots appears beside the 2 on the north/south poles. These 4 spots are caused by 2 domains with ferroelectric polarization along [1-1-1] and [-11-1], thus, their rhombohedral axes are perpendicular to the beam. The 4 spots give only spin-flip scattering, thus, the modulated magnetization cannot have component along the beam, which consistent with the cycloidal correlations.

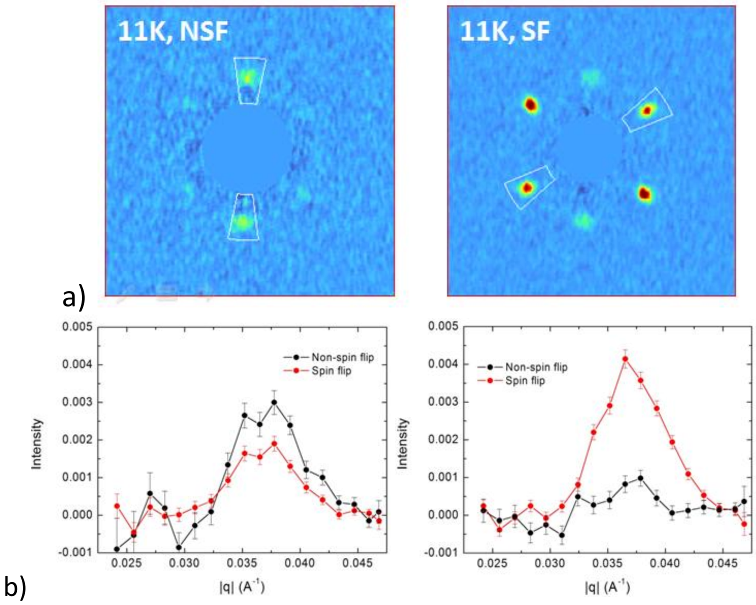

Figure 3 a) Non-spin flip and spin flip scattering patterns measured in 40 mT applied along the beam, parallel to [110]. b) Radial intensity distribution for the white bins pairs as shown in panel a).

We also traced the coherence peak which is a unique signature of multi-q, triangular lattice order of skyrmions. However, we only observed weak multiple scattering at the (1,1) position, which were identified using Renninger scans. We concluded that although spin-anisotropy is important to describe e.g. the magnetization curves, magnetic excitations of GaV4S8 it does not distort the cycloidal order considerably.

Conclusions

We have studied the modulated magnetic phases of GaV4S8 using SANS. These experiments confirmed that the magnetic patterns observed on the surface of the samples by MFM also exist in the bulk material. The direction of the magnetic q-vector is not well defined within the rhombohedral domains, but its length is fixed and agrees with the MFM data. Polarized SANS allowed us to conclude that the magnetic pattern is consistent with cycloidal order which indicates a DM pattern that supports Néel-type skyrmions. Parts of the results have published in Ref. 10.

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