Experimental report

30/11/2023

Proposal: **5-41-1227**

Council: 4/2023

Title: Characterising Exotic Magnetism inEuGa4

Research area: Materials

This proposal is a new proposal

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Samples: EuGa4

Instrument	Requested days	Allocated days	From	To
D33	5	3	17/11/2023	20/11/2023

Abstract:

Magnetic skyrmions show promise as elements in novel spintronics and advanced computing devices. Recent advances in skyrmion material discovery have demonstrated a number of centrosymmetric nanoskyrmion materials which host skyrmions an order of magnitude smaller than those in conventional DMI stabilised skyrmion hosts, suggested to be stabilised by the RKKY interaction. We wish to investigate a novel binary compound, EuGa4, which is suggested to host an exotic phase diagram at low temperatures and high magnetic fields. Comparing this to an isostructural compound EuAl4 will allow us to further understand the stabilising mechanisms which lead to exotic incommensurate magnetic textures in these model nanoskyrmion hosts.

Characterising Exotic Magnetism in $EuGa_4$ – Experimental Report

Experimental Aims:

The proposed experiment aimed to screen the magnetic phase diagram of $EuGa_4$, an end member of the $Eu(Ga_{1-x}Al_x)_4$ series known to host multiple skyrmion phases, before applying for future beamtime to study this material under high pressure. Using small angle neutron scattering, we planned to investigate the potential existence of incommensurate magnetic peaks at wavelengths between 0.15 – 0.3 A and characterise their changes with temperature and magnetic field, if any.

$EuGa_4$:

The aim of this experiment was to search for and characterise the incommensurate magnetic wavevectors in $EuGa_4$. To start with, we aligned on the sample such that the *h* and *k* crystal directions were oriented into the corners of the detector with the c-axis parallel to the magnetic field and neutron beam. At zero magnetic field, we cooled below the transition temperature and performed full reciprocal space maps (within the available reciprocal space range) by rotating the sample stick. By using a small wavelength (3.6 A), we obtained a large amount of reciprocal space in order to capture any incommensurate magnetic wavevectors as we cooled below the transition temperature. As shown in Figure 1.1, no incommensurate magnetic wavevectors were captured at three temperatures below the transition temperature. Similarly, we performed large rocking scans at finite magnetic field at 1 T intervals between 0 – 7 T at 10 K and did not observe any incommensurate wavevectors. These results are indicative of a lack of incommensurate magnetic order within the $EuGa_4$ system.

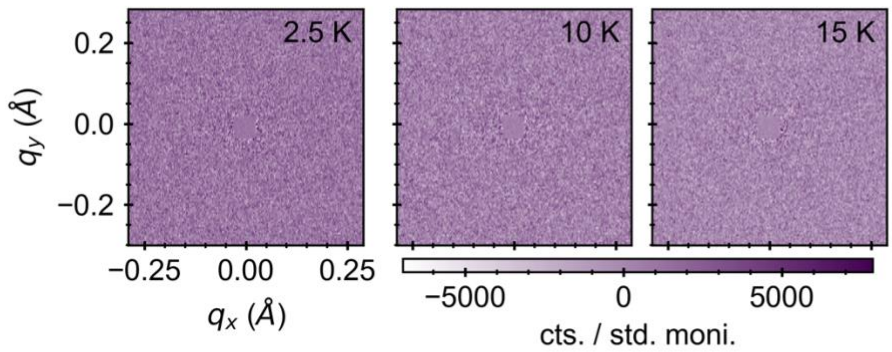

Figure 1.1: *Integrated full reciprocal space scans (background subtracted) of main D33 detector at zero field.*

Although immediately not so interesting, this data will be a useful comparison to our expected data in a future proposal where we expect to induce incommensurate magnetic order through hydrostatic pressure. We are currently performing high pressure magnetometry to uncover the influence of pressure on stabilising complex magnetism within these systems. As a consequence, these SANS results are a clear indicator that in ambient conditions we cannot stabilise complex magnetism such as helical order within $EuGa_4$.

$EuGa_{2.5}Al_{1.5}$ – a potential skyrmion candidate:

Due to the possibility of observing no incommensurate magnetic wavevectors in $EuGa_4$, we discussed with the beamline scientist (prior to the experiment) about bringing a second back-up sample, $EuGa_{2.5}Al_{1.5}$, a related material we have pre-characterised using magnetometry and resonant x-ray scattering. Our previous data had uncovered the nature of the zero field magnetic phases labelled SDW and HEL in Figure x, and their subsequent transitions.

Therefore we wished to use any spare time left after the main investigation to characterise the previously unknown phase 'A' at finite field.

After aligning the sample with the field parallel to the c-axis, along the beam, and rotating the velocity selector to obtain a wavelength of 3.6 A, we were able to detect magnetic peaks in the corners of the main D33 detector. Figure 2.2 shows full, background subtracted rocking scans over each of the three magnetic phases at 2K, 0 T, 0.65 T and 2 T. There is no significant difference in the peak orientation suggesting no macroscopic reorientations of the spin texture as we move from one phase to another. However, when analysing this data further by integrating the intensity of the peaks and tracking the relative q-positions, we observe significant differences between each of the three phases.

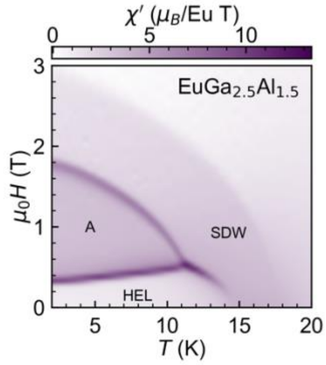

Figure 2.1: *Magnetic phase diagram of* $EuGa_{2.5}Al_{1.5}$ *measured using magnetic susceptibility.*

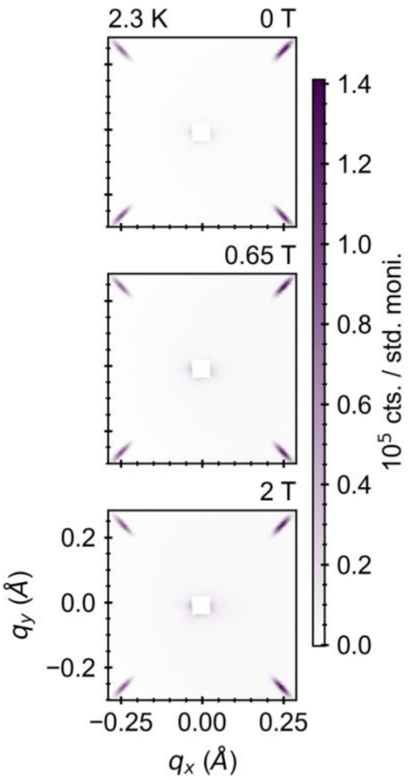

Figure 2.2: *Full, background subtracted rocking scans of each of the three magnetic phases in* $EuGa_{2.5}Al_{1.5}$.

To allow us to collect data across the full phase diagram, we performed rocking scans across only the right hand two peaks. This allowed us to collect both the integrated intensities of potentially two domains of spin texture, while tracking their wavevector positions and discerning higher order Fourier components from multiple scattering events. The integrated intensities and corresponding wavevector positions are shown in Figure 2.3. As demonstrated by the comparison between the SANS data and magnetometry, the phase transitions match very well. With increasing field, we see discontinuous jumps in the intensity matched with small changes in the magnitude of the wavevector as we pass across the transitions. The changes in wavevector are very small, 3% at most, suggesting a relatively stable spin texture with interactions which do not widly change as a function of magnetic field. Similarly, across the temperature range from 2 – 16 K, the wavevectors magnitudes are consistent suggesting the stabilising interactions are almost temperature independent within this temperature range. In contrast, although the scale of the spin texture does not drastically vary with increasing field, the structure factors for each of the three phases are vastly different. There is an overall trend for the intensity to reduce as a function of magnetic field. However between the transition from 'HEL' to 'A', there is a steep drop in intensity over a 0.4 T interval, followed by a small negative gradient before we regain intensity as we transition from 'A' into 'SDW'. A possible explanation for this loss, and subsequent resurgence, of intensity as we cross through the 'A' phase at finite field is the potential formation of higher order Fourier components, indicative of a more complex spin texture. However, for certain we can say that the structure factors of each of the three magnetic phases are significantly different, owing to complex spin textures that change across the boundaries.

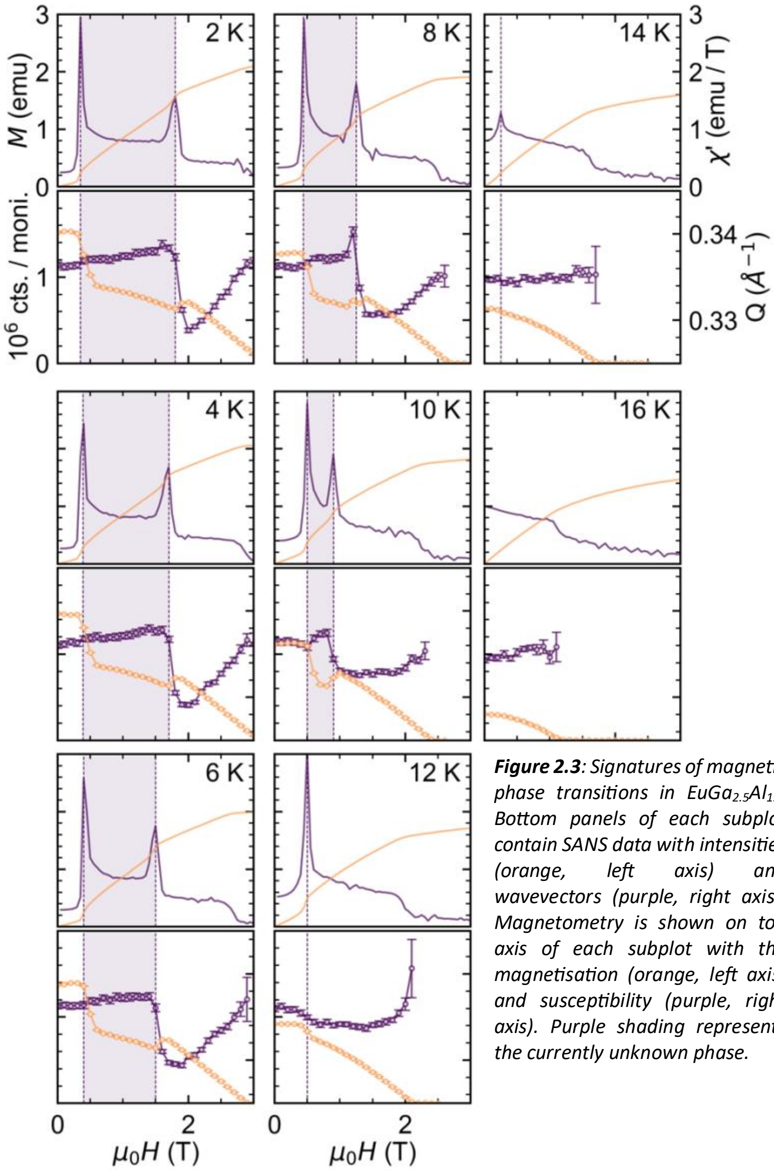

***Figure 2.3**: Signatures of magnetic phase transitions in $EuGa_{2.5}Al_{1.5}$. Bottom panels of each subplot contain SANS data with intensities (orange, left axis) and wavevectors (purple, right axis). Magnetometry is shown on top axis of each subplot with the magnetisation (orange, left axis) and susceptibility (purple, right axis). Purple shading represents the currently unknown phase.*

With the remaining time, we aimed to fully discern the existence of higher order Fourier components within the wavevector range D33 allows by performing Renninger scans. This method works by rocking the Ewald sphere such that the condition for multiple scattering is no longer satisfied meaning that any remaining intensity at the higher order positions should only come from higher order Fourier components of the spin texture.

The results (not shown here) indicate that there are no higher order magnetic Fourier components observed at the $Q_1 + Q_2$ position. However, this is not to say that higher order components do not exist at larger magnitudes, as previously reported in similar centrosymmetric skyrmion hosts. The reduction in intensity simply suggests that we have a more complex structure factor, which requires more study to discern the real space magnetic texture.

Conclusions:

To conclude, during this three day beamtime, we have investigated two compositions within the $Eu(Ga_{1-x}Al_x)_4$ series, $EuGa_4$, and $EuGa_{2.5}Al_{1.5}$. The first has shown a lack of incommensurate magnetic wavevectors across the entirety of the phase diagram, suggesting no magnetic interactions present which can stabilise complex incommensurate order. In contrast, the second composition shows a vastly different magnetic phase diagram, which is matched in the structure factors collected within the SANS data. The large 'A' region shows particularly interesting phenomena suggestive of a potential multi-q or skyrmion phase, which with further analysis including resistivity measurements and polarised neutron scattering, could be the largest region of skyrmion stability across the composition series, highlighting the importance of a fine balance of magnetic interactions in the stability of magnetic skyrmion states.