Experimental report

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Title: Search for new Skyrmion states in Co6.75Zn6.75Mn6.5

Research area: Physics

This proposal is a new proposal

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Samples: Co6.75Zn6.75Mn6.5

Instrument	Requested days	Allocated days	From	To
D33	4	4	26/01/2017	30/01/2017

Abstract:

Magnetic Skyrmions are topological magnetic spin-vortex-like objects that form in chiral magnets. In the recently discovered Skyrmion hosts CoxZnyMnz alloys, the randomness of site occupancies provides a potential landscape with a disorder strength that can be tuned according to the composition. This makes certain CoxZnyMnz alloys as candidates for hosting new Skyrmion states that are only realisable in the presence of certain levels of disorder, such as Skyrmion glass-like states. Here we propose a D33 SANS study of the microscopic magnetic correlations in the alloy Co6.75Zn6.75Mn6.5. Our preliminary zero field SANS data on this compound show the formation of both helical and helical glass-like states, and unusual T-dependent behaviour on cooling. With this proposal our goals are i) to characterise the microscopic magnetic phase diagram of this material under finite magnetic field, and ii) to search for evidence of a Skyrmion glass state. Proving the latter would be important due to the ongoing debate as to whether crystallographic concepts can strictly be explored in Skyrmion states.

Experimental Report 5-42-443: Search for new skyrmion states in $Co_{6.75}Zn_{6.75}Mn_{6.5}$

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Introduction

Magnetic skyrmions are topological magnetic spin-vortex-like objects that can form in various non-centrosymmetric magnets [1]. In all the chiral cubic host systems, such as MnSi, Cu_2OSeO_3 and Co-Zn-Mn alloys, the magnetism is described by a heirarchy of energy scales; i) an effective ferromagnetic exchange J that favours collinear order, ii) a weaker Dzyaloshinskii-Moriya interaction (DMI), D that promotes non-collinear order, iii) a weaker still cubic anisotropy K that determines the direction of magnetic propagation in zero magnetic field. Due to the ubiquitous existence of this energy scale heirarchy in the chiral cubic magnets, their phase diagrams are similarly structured: a multi-domain single-$\mathbf{q}$ (helical) phase is stable for internal fields (B) $0 < B < B_{c1}(T)$, the single-$\mathbf{q}$ domain conical phase for $B_{c1} < B < B_{c2}(T)$, and the skyrmion phase is stable over a narrow interval in finite $\mu_0 H$ and T just below the ordering temperature T_c. In all phases, and in all systems, the magnetic periodicity $\mathbf{q}$ is determined at the mean-field level by $\mathbf{q} = D/2J$, which lies in the nanometric range (~10-100 nm). This makes SANS a crucial tool for exploring chiral magnetism in these systems.

$(Co_{0.5}Zn_{0.5})_{20-x}Mn_x$ ($0 < x < 20$) alloys with the β-Mn structure are a recently discovered, highly tunable host system of chiral magnetism and skyrmions [3]. One end member, $Co_{10}Zn_{10}$ is a chiral helimagnet that hosts a magnetic field-induced skyrmion phase. As shown in Fig. 1, by alloying with increasing amounts of Mn T_c is quickly suppressed, eventually leading to β-Mn itself as another end member, a famous elemental frustrated spin liquid with no long-range spin order [2].

In this D33 experiment we explored the chiral magnetism in crystals of two samples with intermediate Mn concentrations, the title compound $Co_{6.75}Zn_{6.75}Mn_{6.5}$ (T_c=102 K), and also the slightly less Mn-rich $Co_7Zn_7Mn_6$ (T_c=160 K). As seen from Fig. 1, these samples display critical Mn concentrations close to where the long-range helical magnetism (with magnetic field-induced skyrmion state) gives way to frustration-induced spin glass states at higher Mn concentration. Consequently, at these compositions it is possible to study the interplay between disorder generated created by the intrinsic randomness of site occupancies in the crystals, and magnetic frustration that becomes incresingly influential with higher Mn contents. In particular, these alloys are good candidates to host a highly tunable potential landscape for helical and skyrmion domains. This makes these samples possible hosts of new types of chiral magnetic phases that have been anticipated theoretically in the presence of disorder [4].

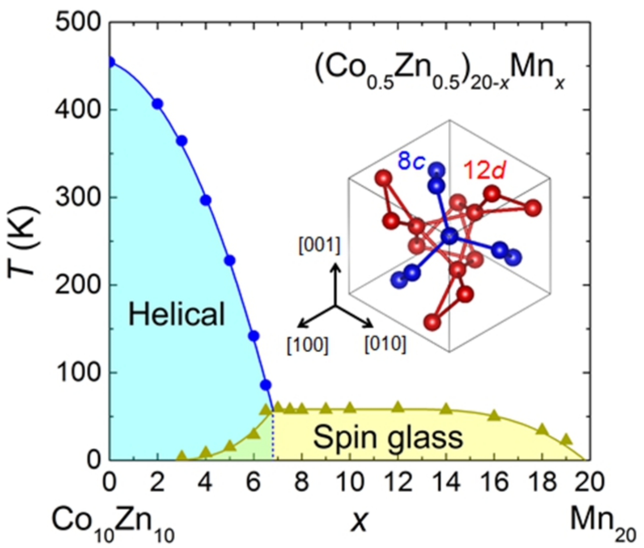

Figure 1: $T - x$ phase diagram of $(Co_{0.5}Zn_{0.5})_{20-x}Mn_x$

Experimental Method

Single crystal samples of $Co_{6.75}Zn_{6.75}Mn_{6.5}$ and $Co_7Zn_7Mn_6$ were prepared by the Bridgman method. The samples were installed into a horizontal field cryomagnet so that the applied magnetic field $\mu_0 H$ was along the cubic [001] axis. The magnet was installed at D33, and two geometries could be studied while maintaining $\mu_0 H \| [001]$; $\mu_0 H \| \mathbf{k}_i$ and $\mu_0 H \perp \mathbf{k}_i$. The former geometry is the standard one for studying the $\mu_0 H$-induced skyrmion phase, in addition to the zero-field helical order that propagates in the [100]-[010] plane. In the second geometry, spin correlations due to the conical order can be studied.

In our SANS experiments we used neutrons with a wavelength of 10 Å, an incoming collimation of 12.8 m and positioned the main 2D detector 13.4 m behind the sample. The SANS measurements in the ordered phases were done in the usual way, by means of rocking scans where the magnet and sample were tilted or rotated together over an angular range that moved the SANS signal through the Ewald sphere at the detector. Measurements were done at selected magnetic fields and temperatures following various zero field cooling (ZFC) or field-cooling (FC), and $\mu_0 H$-sweeping protocols.

Results

Using D33, SANS measurements were performed first on the $Co_{6.75}Zn_{6.75}Mn_{6.5}$ sample. Under ZFC below T_c, the scattered intensity evidenced weakly-correlated magnetism that displayed no azimuthal angle dependence around the direct beam. Fig. 2 thus shows the angularly averaged total scattered SANS intensity as a function of $|\mathbf{q}|$, with a thermally-dependent radial correlation length that clearly shortens as the T falls. These data show an enhancement of the disorder landscape as the T falls, which is assigned to increasingly frustrated magnetic interactions between Mn spins at lower Ts. In addition further $\mu_0 H$-dependent data were collected at T just below T_c, and these data are currently undergoing analysis. At this stage, we decided to forego collecting further data, for example $\mu_0 H$-dependent data at lower T, due to the weakness of the signal and our desire to devote a portion of our beamtime to address urgent scientific questions in the closely related composition $Co_7Zn_7Mn_6$ (see below). Therefore, we aim to complete our SANS study of $Co_{6.75}Zn_{6.75}Mn_{6.5}$ in a future experiment.

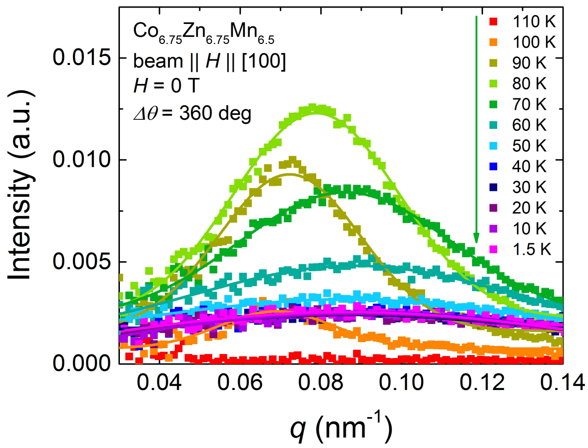

Figure 2: T-dependence of the total scattered SANS intensity versus $|\mathbf{q}|$ during ZFC for $Co_{6.75}Zn_{6.75}Mn_{6.5}$ measured on D33.

Towards obtaining a complete overview of the x-dependence of the magnetic correlations within the vicinity of critical Mn concentrations, we used part of the present beamtime to study the closely related composition $Co_7Zn_7Mn_6$. In this crystal, compelling evidence for a richer phase diagram than the generic one described in the introduction for chiral magnets was obtained from ac susceptibility, cryo-Lorentz transmission electron microscopy (LTEM) and earlier SANS measurements. Here we used our remaining beamtime to collect missing SANS data and complete our recently submitted manuscript [6].

Fig. 3(a) shows the novel magnetic phase diagram for $Co_7Zn_7Mn_6$, with pink symbols denoting data points determined from the SANS measurements done during this beamtime at D33. The overall phase diagram was constructed from $\mu_0 H$-increasing measurements done at fixed T, after an initial ZFC to the target T. A similar phase diagram constructed from down-sweep measurements unveils the various phase boundaries to display a large hysteresis, which is familiar to the phase diagrams determined for more Mn-light compositions [5].

As can be seen in Fig. 3(a), above 100 K, the phase diagram is generically structured as for the other chiral magnets. Below 100 K a crossover region is identified where the correlation length shortens upon cooling, though less dramatically than for $Co_{6.75}Zn_{6.75}Mn_{6.5}$. Instead in $Co_7Zn_7Mn_6$ a pronounced peak in intensity versus $|\mathbf{q}|$ is observed at all Ts upon ZFC, including below the spin glass transition T close to 30 K. The most striking feature to appear in the low T part of the phase diagram is what we term a 'disordered skyrmions' phase, which we propose as to be a

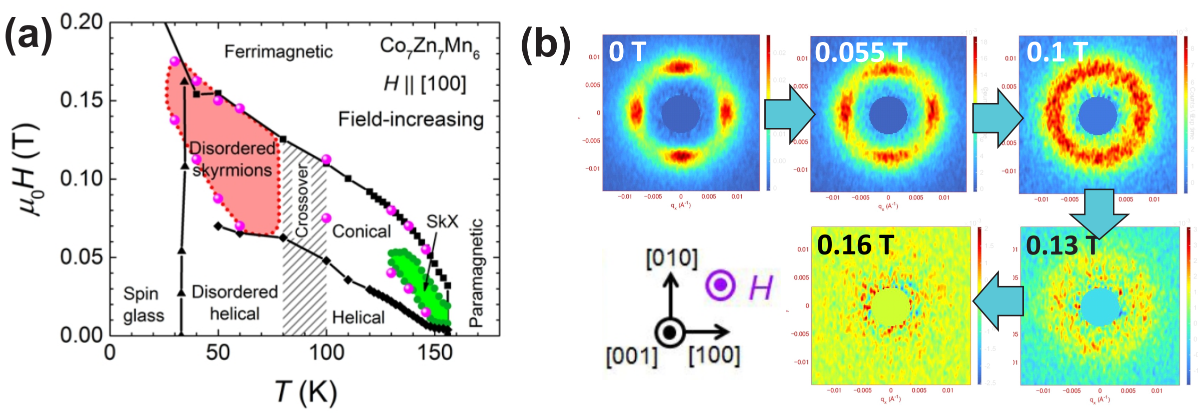

Figure 3: (a) The phase diagram for $Co_7Zn_7Mn_6$ constructed from $\mu_0 H$-increasing measurements after ZFC. The usual skyrmion phase is indicated by green circles. The crossover region around 90 K is indicated by grey hatching. Pink symbols denote phase boundaries or features in the phase diagram determined by SANS. The disordered skyrmions phase is indicated by a red color region. (b) $\mu_0 H$-dependent D33 SANS data obtained during an up-sweep, and at T=60 K after an initial ZFC.

new type of equilibrium skyrmion phase [6].

To support the allocation of this field-induced phase as hosting disordered skyrmions, we collected D33 SANS data in both experimental geometries. Fig. 3(b) shows typical $\mu_0 H$-dependent data obtained on an up-sweep at T=60 K in the $\mu_0 H \| \mathbf{k}_i$ geometry, and after an initial ZFC. The SANS pattern obtained in zero magnetic field shows four broad peaks; this is interpreted in terms of two domains of a disordered helical structure with propagation vectors aligned approximately with the cubic axes as expected. Upon increasing the $\mu_0 H$ to 0.1 T the magnetic scattering distribution transforms to form a ring-shape, before fading away entirely at higher $\mu_0 H$ when entering the induced-ferromagnetic phase. At 0.1 T, a ring-like pattern was also observed in the $\mu_0 H \perp \mathbf{k}_i$ geometry on the up-sweep measurements, indicating that in the disordered skyrmions phase the scattering intensity forms a spherical shell in reciprocal space. Crucially, this spherical distribution of intensity can not be explained in terms of trivial helical or conical spin correlations which would have $\mathbf{q} \| \langle 100 \rangle$ or $\mathbf{q} \| \mu_0 H$, respectively. This motivates us to propose the intensity distribution as due to the new-type of three-dimensionally disordered skyrmion state [6].

Important supporting data towards this interpretation is provided by LTEM of a nanometrically thin plate sample of $Co_7Zn_7Mn_6$. In the same parameter range as evidenced by SANS, a disordered magnetic phase is observed that hosts closed topological magnetic objects, and which is consistent with the proposed disordered skyrmion phase observed in the bulk by SANS. In terms of the physical interpretation, the important point is that this topological disordered skyrmion phase is stabilised as an equilibrium phase at Ts well below the ordering temperature, T_c. Since conventional skrymion phases are always stabilised at high Ts, just below T_c, it is commonly accepted that thermal fluctuations are a key factor for skyrmion phase stability. The observation of a new disordered skyrmion phase here in $Co_7Zn_7Mn_6$ at low T hints that thermal fluctuations less crucial for the stability mechanism, calling for a new idea. We propose that in this case the stability mechanism relies upon magnetic frustration-induced fluctuations. [6] The question of the stability mechanism can be clarified more rigorously by further theoretical and experimental studies. Nevertheless, from our experiments we evidence $Co_7Zn_7Mn_6$ to be a hitherto unique system that hosts two parametrically disconnected equilibrium skyrmion phases [Fig. 3(a)].

Summary

To summarize our experiment, we used D33 to perform SANS measurements on two chiral magnets, $Co_{6.75}Zn_{6.75}Mn_{6.5}$ and $Co_7Zn_7Mn_6$. In $Co_{6.75}Zn_{6.75}Mn_{6.5}$ our data evidence the formation of a very weakly-correlated magnetic state at low T, the magnetic-field dependent properties of which remain to be clarified in a future experiment. For a crystal with a slightly different composition $Co_7Zn_7Mn_6$, we collected sufficient SANS data to evidence the formation of a new type of disordered skyrmions phase, which we propose to be stabilised by magnetic frustration-induced fluctuations [6]. The completion of the SANS study of $Co_{6.75}Zn_{6.75}Mn_{6.5}$ is thus clearly necessary for exploring these ideas experimentally, since the magnetic frustration is more pronounced at this composition compared with $Co_7Zn_7Mn_6$, despite the relatively small change in the sample composition. The consequence of this on the possible topological disordered magnetic phases remain to be identified, and present as an avenue for our further exploration.

References

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