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Tilted Spirals and Low-temperature Skyrmions in Cu₂OSeO₃

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The B20 cubic chiral magnets are among the most studied skyrmion hosting systems and share a universal magnetic phase diagram of helical spiral, conical spiral, as well as the skyrmion lattice phase [1]. The latter appears in a narrow region of the phase diagram, the so-called A-phase, just below the magnetic ordering temperature. The Mott insulator Cu_2OSeO_3 however, marks a notable exception to this universality

[2-6]. In this system, Small Angle Neutron Scattering (SANS) has shown that at low temperatures and at sufficiently high magnetic fields applied along the easy (001) crystallographic direction, a low-temperature skyrmion phase sets in. In addition the propagation vector of the spiral state does not follow the magnetic field direction but tilts away from it, leading to a multidomain tilted spiral state.



Figure 1. SANS patterns recorded at T=2K for selected magnetic fields applied along the (001) easy crystallographic direction showing the field dependence of the conical (C), helical (H), tilted spiral (TS) and skyrmionic (SK) peaks. The red arrow in the 57 mT panel indicates the direction of the applied magnetic field, while k_i indicates the direction of the incoming neutron beam.

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Figure 2. Contour plots at selected temperatures of the scattered intensity recorded between Q=0.05 and 0.013 nm⁻¹ as a function of the azimuthal angle ϕ (defined in the 48 mT panel of Figure 1), illustrating the change of behavior around 18K.

These phenomena occur at the same areas of the phase diagram and whereas they can been attributed to the interplay of competing energies it is not clear why they appear when they are least expected, at low temperatures, where thermal spin fluctuations are suppressed, and at magnetic fields strong enough to align all spirals along their direction. In order to address this point we performed a systematic SANS study of the evolution of these phases as a function of temperature and magnetic field [7].

The characteristic patterns at T=2K shown in Figure 1 bear the signature of all low temperature phases. At the high field limit, at ± 55 mT, the sample is at the field polarized state, the magnetic moments are aligned along the magnetic field and the SANS intensity vanishes. At the low field limit, at ± 7 mT the sample is in the so-called helical phase, where the orientation of the spirals is imposed by cubic anisotropy and we only observe the (001) Bragg peaks. For intermediate magnetic field strengths, however, in the so-called conical phase where the orientation of the spirals should be imposed by the magnetic field, which is also along (001), additional intensity appears characteristic of the multidomain tilted spiral (TS1 and TS2) and the skyrmionic (Sk) scattering.

These experimental findings are summarized by the contour plots of the scattered intensity versus magnetic field and azimuthal angle (φ) shown for selected temperatures in Figure 2. At T=2 K the tilted spiral scattering appears in an abrupt, step-wise, manner and is well separated from the conical/helical peaks that are centered at $\varphi = \pm 90^{\circ}$. As the temperature increases, the angular separation between the conical/helical and the tilted spiral peaks decreases and the transition becomes gradual. Already at 18K it is almost impossible to separate the peaks from each other and at higher temperatures we only observe a broadening of the helical/conical peaks, an effect that spans the whole conical phase and persists up to above 35K, the highest temperature where our measurements have been performed.

The main features of these experimental findings can be captured by a model based of the phenomenological theory introduced by Dzyaloshinskii [8]. Here however we go a step further toward an almost quantitative comparison between model and experiment, which leads us to the conclusion that the anisotropy constants, the drivers behind the observed behavior, exhibit a pronounced temperature dependence. This explains the change of behavior observed around T ~ 18 K, a temperature which separates the high temperature (low cubic anisotropy) from the low temperature (high cubic anisotropy) behavior. For T < 18 K, the cubic anisotropy exceeds a critical value and is therefore strong enough to induce the observed abrupt appearance of the tilted spirals and at the same time enhance the stability of skyrmions.

The approach we have adopted allows for quantitative comparisons between experiment and theory and can provide a strategy for an in-depth understanding of chiral magnets in view of tailoring their properties for future applications.

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