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Dimensional crossover in a spin-liquid-to-helimagnet quantum phase transition

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Neutron scattering is used to study magnetic-field-induced ordering in the quasi-one-dimensional quantum spin-tube compound Sul-Cu_2Cl_4 that in zero field has a nonmagnetic spin-liquid ground state. The experiments reveal an incommensurate chiral high-field phase stabilized by a geometric frustration of the magnetic interactions. The measured critical exponents $\beta \approx 0.235$ and $\nu \approx 0.34$ at $H_c \approx 3.7$ T point to an unusual subcritical scaling regime and may reflect the chiral nature of the quantum critical point.

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Quantum critical points (QCPs) in spin liquids have recently become a forefront issue in magnetism.[1] In particular, phase transitions in gapped quantum-disordered antiferromagnets (AFs) induced by the application of external magnetic fields provide a new way to study a number of fundamental phenomena. For example, some models can be directly mapped onto Bose-Einstein condensation (BEC),[2–4] while fractional magnetization plateaus in certain systems are magnetic analogs of Mott-insulator phases,[4,5] and quenched disorder can lead to the formation of an effective Bose glass.[6] At the same time, the abundance and variety of low-dimensional spin systems enable access to an entire range of previously inaccessible dimensional-crossover phenomena.[7–11] Most importantly, field-induced quantum phase transitions are found in a number of prototypical magnetic materials and can thus be studied experimentally. Neutron scattering turned out to be particularly useful, driving much of the theoretical development.[12–15]

Another current topic in quantum magnetism is that of chirality. Spontaneous breaking of inversion symmetry in classical magnets has been known for many decades and manifests itself in long-range helimagnetic order. Today, theorists seek to understand how chirality can exist in disordered spin liquids[16–18] and how it may be involved in quantum phase transitions and critical behavior.[19,20] In this context, we hereby report an experimental observation of a field-induced quantum phase transition from a disordered spin liquid to an ordered chiral incommensurate state. This phenomenon is studied in the geometrically frustrated Heisenberg $S=1/2$ spin-tube antiferromagnet Sul-Cu_2Cl_4. We observe highly unusual values of the critical exponents that represent dimensional-crossover scaling and may be a signature of chirality at this QCP.

As discussed in detail in Ref. 21, Sul-Cu_2Cl_4, with the chemical formula Cu_2Cl_4-$H_8C_4SO_2$, realizes a rare $S=1/2$ four-leg Heisenberg spin-tube model, the tubes running along the c axis of the triclinic crystal structure.[21] Zero-point quantum spin fluctuations entirely destroy long-range order in this system. The magnetic ground state is a spin liquid with activated susceptibility and specific heat. The spectrum consists of strongly dispersive triplet excitations with an energy gap of $\Delta \approx 0.52$ meV and a spin-wave velocity of $v \approx 14$ meV. Neutron-scattering experiments failed to detect any dispersion of magnetic excitations perpendicular to the tube axis or any splitting of the gap mode in zero field. Based on the experimental resolution of about 0.2 meV full width at half maximum (FWHM), one can place upper bounds on the intertube coupling and anisotropic (non-Heisenberg) interactions: $J_{\perp}, D < 0.05$ meV, respectively. Thus, Sul-Cu_2Cl_4 is an exceptionally isotropic and one-dimensional (1D) system. The conveniently small gap can be overcome by applying a moderate magnetic field.[22,23] A field-induced ordering transition occurs at $H_c \approx 4$ T and manifests itself in a λ specific-heat anomaly and the appearance of nonzero uniform magnetization.[22,23] The directional dependence of the critical field is fully accounted for by the anisotropy of the g-tensor.[22] This gives an even tighter limit on the magnitude of anisotropy: $D < 10^{-3}$ meV.

A crucial feature of Sul-Cu_2Cl_4 is a partial geometric frustration of exchange interactions on the tube rungs.[21] In a classical magnet such frustration is often resolved through the formation of a spiral (helimagnetic) spin structure.[24] The latter has a periodicity defined by the ratio of conflicting exchange constants and therefore totally independent of the periodicity of the underlying crystal lattice. Due to the singlet nature of the ground state in Sul-Cu_2Cl_4, such static helimagnetic order is absent, but *dynamic* incommensurate correlations are preserved. The equal-time correlation function is maximized, and the gap modes have dispersion minima at incommensurate positions, slightly off the AF point: $l_0 = 0.5 \pm \delta$, with $\delta = 0.022(2)$.[21]

The field-induced QCP was studied in two series of neutron-scattering experiments. On the V2-FLEX three-axis spectrometer at HMI we utilized an assembly of 12 fully deuterated Sul-Cu_2Cl_4 single crystals with a total mass of about 1 g. The crystals were coaligned to a triangular mosaic

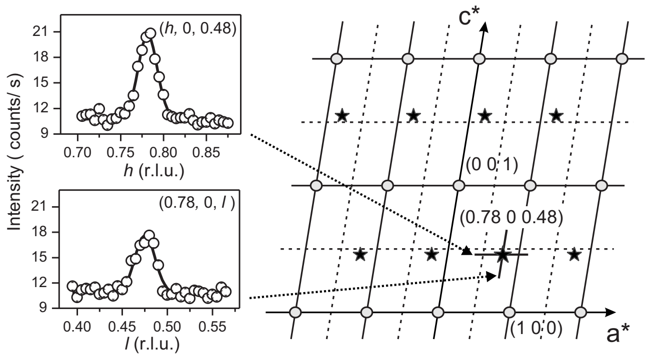

FIG. 1. Neutron-scattering elastic scans along a^* and c^* directions through the (0.78,0,0.48) magnetic peak and reciprocal lattice space of Sul-Cu_2Cl_4 indicating the incommensurate nature of the field-induced long-range magnetic order.

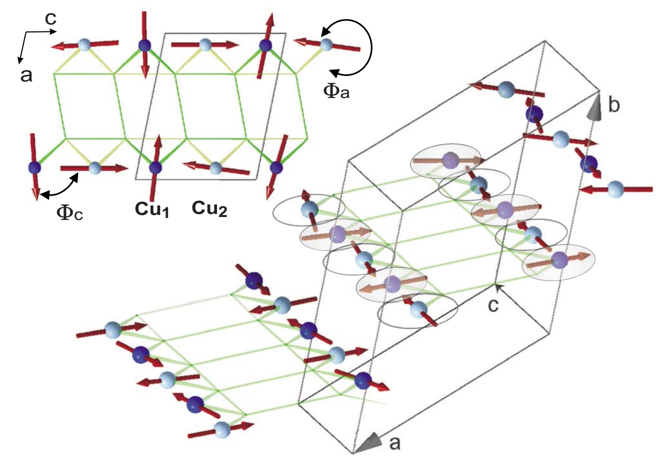

FIG. 2. (Color online) Schematic view of the incommensurate magnetic phase, modeled by a planar helimagnetic structure with the wave vector $k=(-0.22,0,0.48)$.

spread of 1.9° FWHM. The $(h,0,l)$ reciprocal-space plane coincided with the scattering plane of the spectrometer, while the magnetic field, generated by a 14.5 T cryomagnet, was applied along the b axis. The instrument was operated in three-axis mode, using 4.7 Å neutrons selected by and pyrolitic graphite (PG) (002) monochromator and analyzer. Sample temperature was controlled by a ^{3}He-^{4}He dilution refrigerator. A second series of measurements was carried out on the D23 lifting-counter diffractometer at ILL. Diffraction data were taken on a $2.5\times2\times5$ mm^3 deuterated single crystal, whose quality was previously tested using the Orient Express instrument at ILL. The crystal was loaded in a 12 T superconducting magnet $(H\|b)$ equipped with a dilution insert. Diffracted peaks in the $(h,0,l)$ and $(h,1,l)$ scattering planes were measured using monochromatic neutron beams with the wavelengths 1.27 Å (Cu 200) or 2.37 Å (PG 002).

Neutron data collected at $T=130$ mK confirm that the field-induced transition previously seen in bulk measurements represents the onset of long-range magnetic order. A comprehensive search in the $(h,0,l)$ plane detected the emergence of magnetic Bragg peaks beyond $H_c=3.7$ T. The value of the critical field is lower than the value 4.3 T reported in Ref. 22. The discrepancy is accounted for by a lower experimental temperature, a different geometry $(H\|b)$ and the anisotropy of the g-tensor.[23] The observed magnetic reflections are located at incommensurate reciprocal-space positions. As shown in Fig. 1, these peaks can be indexed as $(h\mp\xi,k,l\pm\zeta)$, where $\xi=0.22$, $\zeta=0.48$, and h, k, and l are integers. Peaks corresponding to odd values of l are systematically absent. Further searches took advantage of the lifting counter geometry of D23 venturing outside the $(h,0,l)$ plane but revealed no additional reflections.

The structure of the high-field ordered phase was determined from 23 independent magnetic Bragg intensities measured at $T=60$ mK in a field $H=10$ T applied along the b direction. The obtained data set is not sufficient for an unambiguous model-independent determination of the spin arrangement in the incommensurate case. Nevertheless, a unique solution can be derived under just a few additional assumptions. As mentioned above, in classical Heisenberg magnets geometric frustration typically favors a planar helimagnetic state.[24] Such spin configurations survive in quantum spin models,[25] albeit with strongly renormalized periodicities. In our data analysis we therefore postulated a uniform helimagnetic structure for Sul-Cu_2Cl_4 as well. As shown in Fig. 2, all spins were assumed to be confined in the plane perpendicular to the direction of applied field and the period of the spin spiral was chosen to match the observed magnetic propagation vector. The basis of this helical structure is defined by two relative pitch angles, ϕ_a and ϕ_c, between spins coupled along the a and c axes in each unit cell, respectively (Fig. 2). A least-squares fit of this model to the data yields an excellent agreement with $\phi_a=273\pm3°$, $\phi_c=83\pm9°$, and an ordered moment $m=0.044(1)\mu_B$ per site. The obtained solution is unique within the assumed planar-spiral model with four spins per unit cell.

To access the character of the phase transition itself, we measured the critical exponent β associated with the magnetic order parameter $m(H,T)$ and defined as $m(H,T)\propto[H-H_c(T)]^{\beta(T)}$ for $H\to H_c$. The field dependencies of the (0.78,0,0.48) peak intensity, expected to be proportional to $|m|^2$, was measured at several temperatures and is plotted in Fig. 3. As exemplified in the inset of Fig. 3 for the case of $T=130$ mK, power-law fits to the data (Fig. 3, solid lines) were performed over a progressively shrinking field range δH. Taking the limit $\delta H\to0$ at each temperature allows us to zero in on the actual critical region. The resulting $\beta(T)$ and $H_c(T)$ are plotted in solid symbols in Fig. 4. The typical error bar on H_c is 0.02 T. Temperature dependence of the exponent β was empirically fit to a parabola that had zero slope at $T=0$ K. The zero-temperature extrapolation of the parabola yielded a value of $\beta=0.235(6)$. Another important critical index is ν that defines the phase boundary: $H_c(T)-H_c(0)\propto T^{1/\nu}$ at $T\to0$. From a power-law fit [solid line in Fig. 4(b)] to our $H_c(T)$ data we get $\nu=0.34(3)$. Overall, the fitting curve agrees well with the results of bulk measurements[23] [open symbols in Fig. 4(b)].

The measured value of the critical exponents are quite unusual. In particular, they are obviously different from those in the well-understood scenario of three-dimensional (3D)

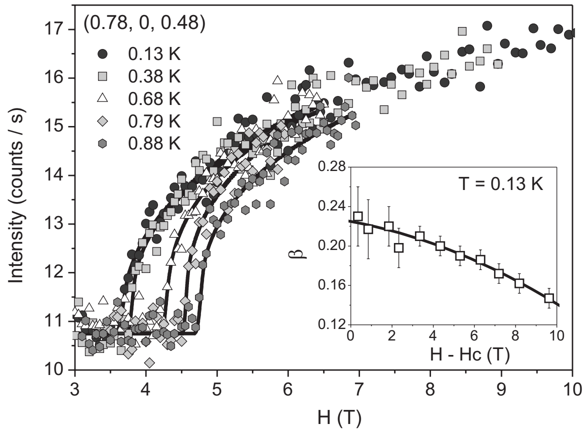

FIG. 3. Field dependence of the (0.78,0,0.48) peak intensity measured at selected temperatures (symbols). The solid lines are power-law fits to the data as described in the text. Inset: the value of the critical index β resulted from fitting the data over a progressively smaller field range $(H-H_c)$ at $T=0.13$ K. The line is a guide for the eyes.

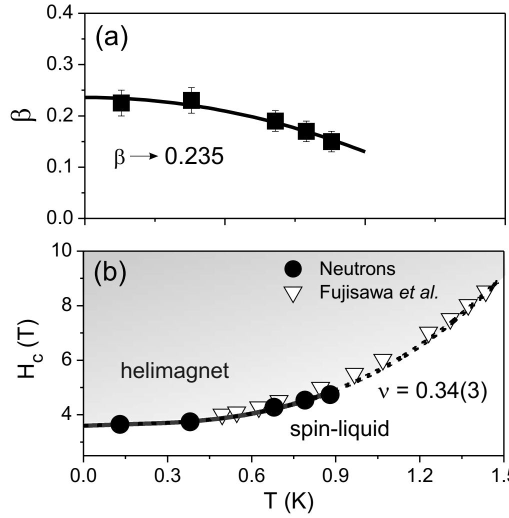

FIG. 4. (a) The order-parameter critical index β plotted as a function of temperature. The $T=0$ K extrapolated value, $\beta \rightarrow 0.235$, is in clear disagreement with that expected for a 3D BEC. (b) Temperature dependence of the transition field, H_c determined by neutron-scattering (solid circles) and specific-heat studies (open triangles, Ref. 23). Solid line represents a power-law fit to the neutron data, yielding a critical index $\nu=0.34(3)$.

BEC of magnons,[2,3] where $\beta=0.5$ and $\nu=2/d=2/3$. The distinction is not entirely unexpected, as a description of the excitations in terms of dilute hardcore bosons may no longer hold in the incommensurate case. In the following paragraphs we shall separately consider three possible reasons for the exotic quantum critical scaling.

First, we can totally rule out the effect of non-Heisenberg terms in the spin Hamiltonian. These play a key role in defining the character of the phase transition in some other spin-gap materials, such as the $S=1$ spin chain compound NDMAP.[12] However, in our case of Sul-Cu_2Cl_4, the smallest field interval used in the determination of β and H_c is $\delta H_{\text{min}}=0.5$ T. This window defines the energy scale of the slowest relevant fluctuations $\hbar\omega_{\text{min}}=g\mu_B\delta H_{\text{min}}\sim 0.1$ meV. Since $D \ll \hbar\omega_{\text{min}}$, any anisotropy terms in the Hamiltonian will manifest themselves only much closer to the critical point than our analysis can approach.

Much more relevant is the question of whether our experiments can access the true critical indices of 3D long-range ordering in a material as effectively 1D as Sul$-Cu_2Cl_4$. In fact, by the same reasoning as in the previous paragraph, they can *not*, as $J_\perp < \hbar\omega_{\text{min}}$. The 3D critical indices manifest themselves only undetectably, close to the transition point. In the absence of residual 3D coupling, the quasi-1D Sul-Cu_2Cl_4 would remain disordered at $T=0$ even in an strong applied fields. Instead, it would become a Luttinger spin liquid,[26] with a divergent correlation length but no static long-range order.

Dimensional crossover at the field-induced QCP in the relevant quasi-1D case was recently studied in the context of the NMR spin relaxation rate $1/T_1$ in the *disordered* state.[11] Although the critical exponents associated with the *ordered* phase and measured in this work have not yet been investigated theoretically, one can draw some analogies with the thermodynamic phase transition in classical quasi-two-dimensional (2D) XY magnets. As a function of temperature, the 2D system does not order in the usual sense although the correlation length diverges at the Kosterlitz-Thouless point.[27] The experimentally observable 3D ordering in layered materials is governed by a universal "subcritical" exponent $\beta=0.23$,[28] distinct from the true 3D-*XY* critical index $\beta=0.35$.[29] The scaling observed in Sul-Cu_2Cl_4 will correspond to an analogous subcritical regime, but whether or not the exponents are universal is yet to be established.

The third and most intriguing consideration is that the (sub)critical indices in Sul-Cu_2Cl_4 are modified by the chiral nature of the ordered state. As famously conjectured by Kawamura,[30] helimagnetic ordering forms separate chiral universality classes with distinct critical indices. Although still controversial, this theory has been apparently confirmed experimentally in a number of frustrated triangular-lattice AFs (Ref. 31) and may apply to the QCP in Sul-Cu_2Cl_4. Even more interesting is the possibility that due to strong geometric frustration chirality is already present in the spin-liquid phase of Sul-Cu_2Cl_4. The existence of such chiral spin liquids is now well established for spin ladders with four-spin exchange,[16] as well as for the Kitaev model.[17] Thus one can imagine a scenario where chirality in Sul-Cu_2Cl_4 is present in zero field or appears in a separate phase transition at $H < H_c$.

In summary, we have observed a field-induced QCP that separates a gapped spin-liquid state from an incommensurate chiral helimagnetic state in a quasi-1D frustrated quantum AF. The highly unusual values of the order-parameter critical exponents pose three important questions to be answered by theorists. Is this QCP characterized by an extended subcritical scaling regime in the *ordered* state and is this scaling universal? Does the chirality of the ordered state qualitatively alter the critical and/or subcritical behavior? Or does chirality appear at lower fields before the onset of long-range order?

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