Dimensional crossover in a spin liquid to helimagnet quantum phase transition

V. O. Garlea and A. Zheludev
Neutron Scattering Sciences Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA.

K. Habicht and M. Meissner
BENSC, Hahn-Meitner Institut, D-14109 Berlin, Germany.

B. Grenier, L.-P. Regnault, and E. Ressouche
CEA-Grenoble, INAC-SPSMS-MDN, 17 rue des Martyrs, 38054 Grenoble Cedex 9, France.

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Neutron scattering is used to study magnetic field induced ordering in the quasi-1D quantum spin-tube compound Sul–Cu$_2$Cl$_4$ that in zero field has a non-magnetic 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 sub-critical scaling regime and may reflect the chiral nature of the quantum critical point.

Quantum critical points (QCPs) in spin liquids have recently become a forefront issue in magnetism. 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), while fractional magnetization plateaus in certain systems are magnetic analogues of Mott-insulator phases and quenched disorder can lead to the formation of an effective Bose glass. 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. 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.

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 and how it may be involved in quantum phase transitions and critical behavior. 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$_2$Cl$_4$. 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$_2$Cl$_4$, with the chemical formula Cu$_2$Cl$_4$-H$_2$CuClSO$_2$, realizes a rare $S = 1/2$ four-leg Heisenberg spin model, the tubes running along the $c$ axis of the triclinic crystal structure. 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 FWHM, one can place upper bounds on the inter-tube coupling and anisotropic (non-Heisenberg) interactions: $J_\perp, D < 0.05$ meV, respectively. Thus, Sul–Cu$_2$Cl$_4$ is an exceptionally isotropic and 1-dimensional system. The conveniently small gap can be overcome by applying a moderate magnetic field. A field-induced ordering transition occurs at $H_c \approx 4$ T and manifests itself in a lambda specific-heat anomaly and the appearance of non-zero uniform magnetization. The directional dependence of the critical field is fully accounted for by the anisotropy of the $g$-tensor. This gives an even tighter limit on the magnitude of anisotropy: $D < 10^{-3}$ meV.

A crucial feature of Sul–Cu$_2$Cl$_4$ is a partial geometric frustration of exchange interactions on the tube rungs. In a classical magnet such frustration is often resolved through the formation of a spiral (helimagnetic) spin structure. 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$_2$Cl$_4$, 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: $l_0 = 0.5 \pm \delta, \delta = 0.02(2)$. The field-induced QCP was studied in two series of neutron scattering experiments. On the V2-FLEX 3-axis spectrometer at HMI we utilized an assembly of 12 fully deuterated Sul–Cu$_2$Cl$_4$ single crystals with a total mass of about 1 g. The crystals were co-aligned to a trian-
ular mosaic spread of 1.9° full width at half maximum (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 3-axis mode, using 4.7 Å neutrons selected by and Pyroritic Graphite PG(002) monochromator and analyzer. Sample temperature was controlled by a \(3.8\) K cryostat equipped with the wave vector \(\mathbf{k}\) of the field-induced transition previously seen in bulk measurements.

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. Such spin configurations survive in quantum spin models, albeit with strongly renormalized periodicities. In our data analysis we therefore postulated a uniform helimagnetic structure for \(\text{Sul–Cu}_2\text{Cl}_4\) 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 \(\phi_a\) and \(\phi_c\) 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 \(\phi_a = 273 ± 3°\), \(\phi_c = 83 ± 9°\) and an ordered moment \(m = 0.044(1) \mu_B\) per site. The obtained solution is unique within the assumed planar-spiral model with 4 spins per unit cell.

To access the character of the phase transition itself, we measured the critical exponent \(\beta\) associated with the magnetic order parameter \(m(\mathbf{H},T)\) and defined as \(m(\mathbf{H},T) \propto (\mathbf{H} - H_c(T))^{\beta(T)}\) for \(H \to H_c\). The field dependences 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 \(\delta H\). Taking the limit \(\delta H \to 0\) 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. 3. The typical error bar on \(H_c\) is 0.02 T. Temperature dependence of the exponent \(\beta\) was empirically fit to a parabola that had zero slope at \(T = 0\) K.
tion of the parabola yielded a value $\beta = 0.235(6)$. Another important critical index is $\nu$ that defines the phase boundary: $H_c(T) - H_c(0) \propto T^{1/\nu}$ at $T \to 0$. From a power-law fit (solid line in Fig. 4(a)) to our $H_c(T)$ data we get $\nu = 0.34(3)$. Overall, the fitting curve agrees well with the results of bulk measurements (open symbols in Fig. 4(a)).

The measured value of the critical exponents are quite unusual. In particular, they are obviously different from those in the well-understood scenario of 3D BEC of magnons, 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 Ni(C$_5$D$_{14}$N$_2$)$_2$N$_3$(PF$_6$) (NDMAP). However, in our case of Sul–Cu$_2$Cl$_4$, the smallest field interval used in the determination of $\beta$ and $H_c$ is $\delta H_{\text{min}} = 0.5$ T. This window defines the energy scale of the slowest relevant fluctuations $\hbar \omega_{\text{min}} = \mu_B \delta H_{\text{min}} \sim 0.1$ 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 indexes of 3D long-range ordering in a material as effectively 1D as Sul–Cu$_2$Cl$_4$. In fact, by the same reasoning as in the previous paragraph, they can not, as $J_\perp < \hbar \omega_{\text{min}}$. The 3D critical indexes manifests themselves only undetectably, close to the transition point. In the absence of residual 3D coupling, the quasi-1D Sul–Cu$_2$Cl$_4$ would remain disordered at $T = 0$ even in an strong applied fields. Instead, it would become a Luttinger spin liquid 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. Though 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-2D XY magnets. As a function of temperature, the 2D system does not order in the usual sense, though the correlation length diverges at the Kosterlitz-Thouless point. The experimentally observable 3D ordering in layered materials is governed by a universal “sub-critical” exponent $\beta = 0.23$, distinct from the true 3D-XY critical index $\beta = 0.35$. The scaling observed in Sul–Cu$_2$Cl$_4$ will correspond to an analogous sub-critical 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 indexes in Sul–Cu$_2$Cl$_4$ are modified by the chiral nature of the ordered state. As famously conjectured by Kawamura, helimagnetic ordering forms sep-

![FIG. 4: (a) The order parameter critical index $\beta$ plotted as a function of temperature. The $T = 0$ K extrapolated value, $\beta \to 0.235$, 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 study (open triangles, Ref. 23). Solid line represents a power-law fit to the neutron data, yielding a critical index $\nu = 0.34(3)$](image-url)
arate chiral universality classes with distinct critical indexes. Though still controversial, this theory has been apparently confirmed experimentally in a number of frustrated triangular-lattice AFs, and may apply to the QCP in Sul–Cu$_2$Cl$_4$. Even more interesting is the possibility that due to strong geometric frustration chirality is already present in the spin liquid phase of Sul–Cu$_2$Cl$_4$. The existence of such chiral spin liquids is now well established for spin ladders with 4-spin exchange as well as for the Kitaev model. Thus one can imagine a scenario where chirality in Sul–Cu$_2$Cl$_4$ 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 sub-critical behavior? Or, does chirality appear at lower fields, before the onset of long range order?

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* Electronic address: garleao@ornl.gov

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