Field-Induced Instability of a Gapless Spin Liquid with a Spinon Fermi Surface

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The ground state of the quantum kagome antiferromagnet Zn-brochantite, $ZnCu_3(OH)_6SO_4$, which is one of only a few known spin-liquid (SL) realizations in two or three dimensions, has been described as a gapless SL with a spinon Fermi surface. Employing nuclear magnetic resonance in a broad magnetic-field range down to millikelvin temperatures, we show that in applied magnetic fields this enigmatic state is intrinsically unstable against a SL with a full or a partial gap. A similar instability of the gapless Fermi-surface SL was previously encountered in an organic triangular-lattice antiferromagnet, suggesting a common destabilization mechanism that most likely arises from spinon pairing. A salient property of this instability is that an infinitesimal field suffices to induce it, as predicted theoretically for some other types of gapless SLs.

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Fermi-surface instability is one of the central concepts in condensed matter physics, responsible for diverse collective phenomena [1]. In metals, various symmetry-broken phases, e.g., the BCS superconducting, Peierls [2], electronic nematic [3,4] and itinerant antiferromagnetic [5] states occur due to such instabilities. An extension of the Fermi-liquid theory to Mott insulators leads to fermionic quantum spin liquids (SLs) [6]. These are intriguing disordered, yet highly entangled, states of matter that are characterized by effective low-energy charge-neutral fermionic quasiparticles known as spinons, which interact through emergent gauge fields [6–10]. In analogy to the Fermi-surface instabilities in metals, many SLs with different symmetries may be considered as being born out of the SL with a spinon Fermi surface (dubbed a spinon metal) [11,12]. Since this parent state is gapless and thus exposed to perturbations and fluctuations, finding its rare realizations is challenging *per se* [13–16]. Moreover, clarifying the nature of its experimentally observed instabilities [17,18] by confronting numerous theoretical proposals [11,12,19–21] with experimental facts represents an even greater challenge. Clearly, identifying some common origin of such instabilities would be beneficial for obtaining an in-depth understanding of the Fermi-surface instabilities in general.

In this context, Zn-brochantite, $ZnCu_3(OH)_6SO_4$, a representative of the paradigmatic two-dimensional geometrically frustrated quantum kagome antiferromagnet [7], is of particular interest. Around 10 K, well below the average nearest-neighbor exchange interaction $J = 65$ K [15], it exhibits a spinon Fermi-surface SL state with Pauli-like kagome-lattice magnetic susceptibility χ_k and with specific heat c_p increasing linearly with temperature [15]. Unexpectedly, this state progressively transforms when the temperature is lowered as χ_k and c_p/T get gradually enhanced and saturate at 2–3 times larger values below ~0.6 K [15,22]. This state remains stable down to the lowest experimentally accessible temperatures ($T/J \lesssim 3\times 10^{-4}$) [18]. The crossover within the spinon Fermi-surface SL state is likely associated with impurities originating from the 6%–9% Zn-Cu intersite disorder [15] and coupled to the kagome spins [22], which could affect the spinon density of states at the Fermi level by pinning of spinon excitations [18].

Moreover, diverse field-induced instabilities were reported in a few two-dimensional SL candidates, like spin freezing in the archetype quantum kagome antiferromagnet herbertsmithite [23] and the organic triangular antiferromagnet κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ [24], and nontrivial symmetry breaking and/or topological ordering in another organic triangular antiferromagnet $EtMe_3Sb[Pd(dmit)_2]_2$ [17]. In this light, studying the response of the SL with a spinon Fermi surface in Zn-brochantite to the magnetic field could help address the fundamental question about the stability of this gapless state against time-reversal symmetry breaking. Specifically, are gapless excitations of this state also intrinsically unstable against the applied field, as theoretically predicted for some other SLs, like the Dirac $U(1)$ SL on the kagome lattice [25], or the Kitaev SL on the honeycomb [8] and the hyperhoneycomb lattices [26]?

Here we provide the answer to this question by reporting a field-induced instability of the gapless spinon Fermi-surface ground state in Zn-brochantite. This was discovered by employing nuclear magnetic resonance (NMR), which has proven particularly well suited for differentiating between impurity and intrinsic properties of this compound, as the ^2D nuclei dominantly couple to the kagome spins [18]. By performing a series of ^2D NMR spin-lattice relaxation ($1/T_1$) measurements in various applied fields down to millikelvin temperatures, we find that, surprisingly, the critical temperature T_c associated with this instability scales linearly with the applied magnetic field B, yielding $T_c \to 0$ at $B \to 0$. We propose that the field-induced SL state originates from spinon pairing and has a full or a partial excitation gap. The instability is reminiscent of that found in the triangular-lattice gapless spinon Fermi-surface SL candidate $\mathrm{EtMe_3Sb[Pd(dmit)_2]_2}$ and thus appears to be a common feature of such SLs.

In this study, we extend our ^2D NMR $1/T_1$ data previously recorded in a field of 4.69 T between 2 and 300 K [18], down to 50 mK [Fig. 1(a)]. As already established, the power-law decrease of $1/T_1$ below 200 K reveals quantum critical behavior, while the decrease below the broad maximum around 5 K corresponds to the crossover within the SL state of Zn-brochantite [18]. Unexpectedly, by lowering the temperature to the millikelvin range we find a pronounced anomaly in the form of a sharp kink in $1/T_1$ at $T_c = 0.76$ K [Fig. 1(a)], which is obviously not related to the smooth crossover within the SL state. Below T_c, $1/T_1$ quickly drops by several orders of magnitude [Fig. 2(a)]. This is usually a sign of a phase transition and was encountered before in herbertsmithite [23] as well as in organic [17,24] and inorganic [27] triangular antiferromagnets. However, T_c does not seem to correspond to a standard magnetic transition, where a divergence of $1/T_1$ is usually observed.

The first essential question that arises is whether the magnetic state of Zn-brochantite below T_c remains a spin liquid. In order to address it we compare the NMR spectra recorded just above and well below T_c [Fig. 1(b)], as the emergence of frozen moments is generally reflected in broadening of NMR lines. This is indeed the case in κ-$\mathrm{(BEDT\text{-}TTF)_2Cu_2(CN)_3}$ [24] and in herbertsmithite in high magnetic fields [23]. On the other hand, the absence of any spectral broadening in $\mathrm{EtMe_3Sb[Pd(dmit)_2]_2}$ was regarded as evidence of a nontrivial symmetry breaking and/or topological ordering within the SL state [17], while no simultaneous anomaly was observed in any thermodynamic observable [13]. Similarly to this case, the estimated

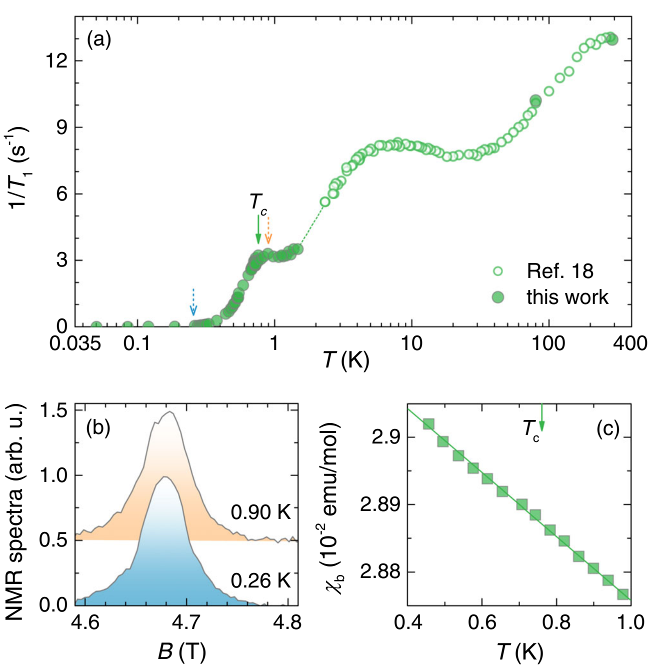

FIG. 1. (a) The temperature dependence of the ^2D NMR spin-lattice relaxation rate $1/T_1$ in the applied field of 4.69 T. The solid arrow indicates the transition temperature $T_c = 0.76$ K, while the dashed arrows indicate the temperatures where the ^2D NMR spectra from panel (b) were recorded. These spectra are normalized and shifted vertically for clarity. (c) The temperature dependence of the bulk susceptibility χ_b in 4.69 T measured by a SQUID magnetometer. Note that the decrease of χ_b with temperature is very small. T_c is indicated by the arrow, while the solid line is a guide to the eye.

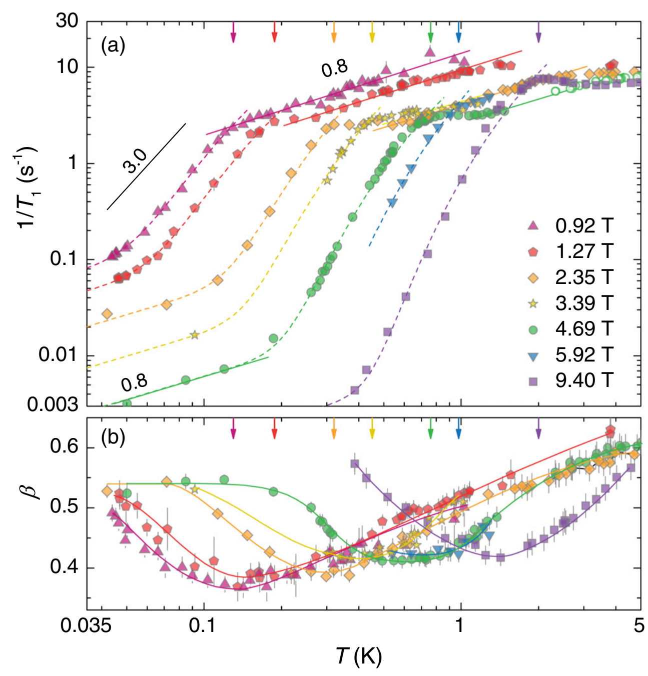

FIG. 2. The temperature dependence of (a) the ^2D NMR spin-lattice relaxation rate $1/T_1$ and (b) the stretching exponent β from the magnetization recovery curves [28] in various applied fields. In panel (a) the solid lines demonstrate power-law dependence (numbers correspond to powers), while the dashed lines show the agreement with the gapped model of Eq. (2). We note that T_c roughly corresponds to a common relaxation rate $1/T_1 = 2.5\ \mathrm{s}^{-1}$ for all except the highest fields. In panel (b) the solid lines are guides to the eye. Arrows in all panels indicate the critical temperatures T_c.

magnetic contributions to the NMR linewidth [28] of 28.8(9) and 28.7(7) mT at 0.26 and 0.90 K, respectively, demonstrate the absence of any magnetic broadening in Zn-brochantite upon crossing $T_c = 0.76$ K. The uncertainty of the linewidths and the hyperfine coupling constant between the ^2D nuclei and kagome spins [18], $A^{\mathrm{iso}} = 34\ \mathrm{mT}/\mu_B$, set a very conservative upper bound of $0.05\mu_B$ on the average frozen moment in the low-temperature state of Zn-brochantite at 4.69 T. No spectral broadening is present even at 9.4 T [28]. We thus arrive at the first important conclusion—the dramatic change of the $1/T_1$ behavior in Zn-brochantite at T_c is not due to bulk spin freezing, but rather suggests a fundamental modification of the excitation spectrum of the SL state.

The next obvious questions are what triggers the instability at T_c and whether it is intrinsic to the kagome spins. In order to address them, we performed additional NMR measurements in various magnetic fields between 0.92 and 9.4 T (Fig. 2). We find [28] that T_c increases steadily with field. It also roughly coincides with the temperature where the $1/T_1$ distribution in each field is the broadest—the stretching exponent β, characterizing the distribution of relaxation times governing the magnetization recovery in the NMR experiment [28], exhibits broad minima [Fig. 2(b)]. Up to $B \sim 5$ T, T_c scales almost linearly with the applied field (with $T_c \to 0$ when $B \to 0$), while at higher fields a cubic correction term is needed and the transition temperature obeys a phenomenological expression $T_c = aB + bB^3$ (Fig. 3). This extra term could be due to the proximity of T_c to the crossover temperature regime of the SL state at higher fields, where the free-spinon

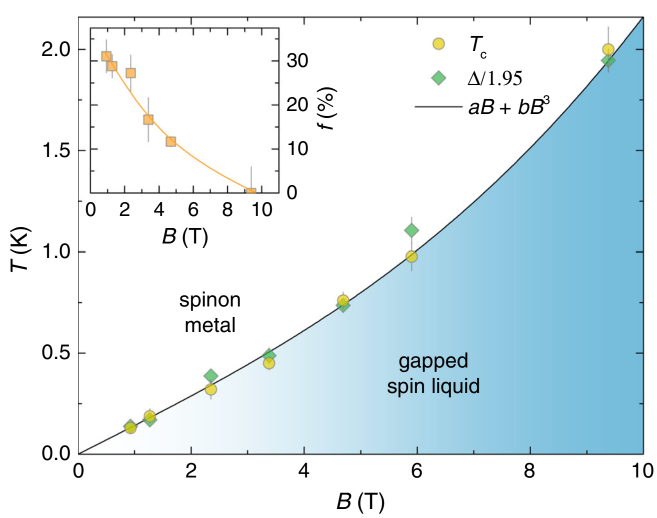

FIG. 3. The phase diagram of Zn-brochantite with the measured critical temperatures T_c and gaps Δ extracted from the low-T $1/T_1$ NMR data. The line shows the $T_c = aB + bB^3$ model fit of the phase boundary between two distinct spin liquid states. The inset shows the field evolution of the fraction f of the residual density of states at the Fermi level [Eq. (2)] below T_c. The line in the inset is a guide to the eye.

density of states is changing [18]. Alternatively, the crossover regime itself could be shifted to higher temperatures at higher fields as the additional Zeeman energy could stabilize spinon pinning, and the extra cubic term could be intrinsic to the instability. In any case, the transition at T_c cannot be accounted for by the polarization of impurities in the applied field. This conclusion is based on the fact that the slope of the $T_c(B)$ curve is very small (it corresponds to an effective g factor of 0.2), its shape at higher fields is convex, and the bulk magnetic susceptibility χ_b in 4.69 T shows no significant anomaly at $T_c = 0.76$ K [Fig. 1(c)] despite a large impurity contribution at low temperatures [15]. We thus reach the next important conclusion that the transition observed in Zn-brochantite at T_c reflects an intrinsic instability within the SL phase induced by the applied magnetic field.

Therefore, we turn to a more elaborate analysis of the NMR relaxation rates. Just above T_c, the power law

$$1/T_1 = cT^\eta \tag{1}$$

with $\eta = 0.8$ fits the experiment [Fig. 2(a)], except for the highest field. This power is very similar to $\eta = 0.73(5)$ found in herbertsmithite above 1 K [23,33]. On the other hand, this dependence is also rather close to the Korringa relation $1/T_1 \propto T$, which holds for free fermions [34,35]. In the picture of gapless $U(1)$ spinons with a Fermi surface, deviations towards $\eta < 1$ are expected [17] due to the coupling of spinons with emergent gauge field fluctuations [21]. The $1/T_1$ data sets above T_c thus agree with the low-temperature zero-field SL state of Zn-brochantite being a gapless SL with a Fermi surface [22]. Moreover, the field-dependent parameter c, which is in such a SL proportional to the squared spinon density of states at the Fermi level (like in an ordinary metal [34]), decreases with increasing field. This is in line with the recently developed picture of spinon metals, where the spectral weight in the dynamic spin structure factor is progressively shifted away from zero energy by an increasing applied field [36].

The significantly altered temperature dependence of $1/T_1$ just below T_c suggests an abrupt change of spin excitations within the spin liquid and, consequently, a large change of the spinon Fermi surface. However, at the lowest temperatures the sublinear dependence of $1/T_1$ ($\eta \simeq 0.8$) is recovered [Fig. 2(a)]. The field dependence of this relaxation is incompatible with an impurity scenario [28]. Moreover, the power η is found to be the same as above T_c. Therefore, we propose that this relaxation at the lowest temperatures is due to a residual density of states of gapless spinons at the Fermi surface. This is further supported by the decreasing relative width of the T_1 distribution at the lowest temperatures—the stretching exponent increases back to $\beta \sim 0.5$ [Fig. 2(b)], the same value that characterizes the data above T_c [28].

The coexistence of two relaxation mechanisms below T_c could, in principle, be due to an inhomogeneous phase (e.g., induced by disorder, structural imperfections, etc.), where regions of fully gapped and gapless spinons would coexist in real space [11]. However, this is not the case, as the magnetization recovery curves in the T_1 experiment should then exhibit a characteristic two-step shape at $T < T_c$, where the relaxation times of the two phases differ significantly. On the contrary, the experimental curves are smooth at all temperatures [28]. Therefore, a plausible explanation for the presence of two relaxation mechanisms below T_c is that the gap is opened only in certain regions of the momentum space [11,19]. Then, an effective two-channel relaxation can appear even in a spatially homogeneous state, because $1/T_1$ averages the dynamic spin structure factor over the entire momentum space [37], as $(T_1T)^{-1} \propto \sum_{\mathbf{q}} A_{\mathbf{q}}^2 \chi''(\mathbf{q}, \omega_{\mathrm{NMR}})/\omega_{\mathrm{NMR}}$, with $\chi''(\mathbf{q}, \omega_{\mathrm{NMR}})$ being the imaginary part of the magnetic susceptibility at the wave vector $\mathbf{q}$ and NMR frequency ω_{NMR}.

The $1/T_1$ data at $T < T_c$ can be accounted for by an extended thermal-activation model [38]

$$\frac{1}{T_1} = dT\mathrm{e}^{-\Delta/T} + f^2 cT^{\eta}, \tag{2}$$

where d is a field-dependent parameter related to the relaxation mechanism induced by the opening of the gap Δ, c is determined from the data above T_c using Eq. (1), and $f < 1$ denotes the fraction of the residual density of states at the Fermi level with respect to the full density of states above T_c [Fig. 2(a)]. At the lowest field of 0.93 T, the residual gapless-spinon density of states at $T < T_c$ is $f \sim 30\%$ of the density of states above T_c. With increasing field f decreases, reaching zero around 10 T (inset in Fig. 3). An important conclusion is thus that the spinon instability at T_c affects the majority of spinons near the Fermi surface. The extracted excitation gap scales linearly with the transition temperature (Fig. 3), $2\Delta/T_c = 3.9(1)$, similarly as found in herbertsmithite [23] and close to the characteristic scaling $2\Delta/T_c = 3.5$ of the BCS state.

Having established that the instability observed in Zn-brochantite at T_c is intrinsic and pertinent to the SL state, the important question of the underlying instability mechanism arises. Two scenarios based on the current theoretical understanding of the spinon-metal phases in two dimensions could, in principle, be possible. An instability that gaps out a part or the whole Fermi surface is expected in the vicinity of the Mott transition where charge fluctuations are strong [42,43]. Since in Zn-brochantite $U/t \sim 60$ [28] (t and U are the Hubbard hopping and Coulomb repulsion, respectively), while the Mott metal-insulator transition occurs already at $U/t \gtrsim 10$ [44], the system is positioned deep in the insulating phase. Therefore, we propose the second possible scenario, which is based on a spinon-pairing instability [11,19–21].

The $U(1)$ SLs with spinon Fermi surfaces are found as root states of many $\mathbb{Z}_2$ fermionic SLs, when the spinon pairing amplitudes in the $\mathbb{Z}_2$ states are turned off [45]. If spinon pairing that arises from the gauge-field mediated attractive interactions between spinons is considered, such states are susceptible to various pairing instabilities [11,19,21]. A paired-spinon state breaks the $U(1)$ gauge symmetry down to $\mathbb{Z}_2$ and opens an energy gap below the pairing temperature T_c [11,20]. The gap can either be full or partial, the latter corresponding to pairing in a non-symmetric channel [19–21]. There are various possible types of the attractive interaction between spinons that lead to pairing instabilities [11,19,21]. If the spinon pairing were of the singlet BCS type, the paired state should be destroyed above a critical external magnetic field [21]. However, this is not the case for spin-triplet BCS pairing [11] or a more exotic Amperean-type pairing with a spatially modulated amplitude [19]. The latter scenarios are therefore more likely to be realized in Zn-brochantite, as our results suggest that the applied field stabilizes the gapped SL state and acts against thermal fluctuations which tend to prefer the gapless SL (Fig. 3).

Zn-brochantite behaves very similarly to the organic triangular-lattice compound $\mathrm{EtMe_3Sb[Pd(dmit)_2]_2}$, as both seem to exhibit a gapless quantum-critical spinon-metal ground state in zero magnetic field [13] that undergoes an unconventional field-induced instability at low temperatures [17]. In both systems, a gap develops in the excitation spectrum below a field-dependent critical temperature, even though the structure of the gap (full or nodal) may be different due to the different lattices of the two compounds. This instability, which is obviously not a regular thermodynamic transition into a frozen spin state, but rather suggests that the magnetic field strongly affects the spinon excitations, thus seems to be a general characteristic of SLs with spinon Fermi surfaces. Therefore, it would be interesting to check for its presence in other physically different spinon-metal ground-state candidates, one of them possibly being the rare-earth based triangular-lattice antiferromagnet $\mathrm{YbMgGaO_4}$ [16,46–48].

Another interesting aspect of the discovered field-induced instability in Zn-brochantite is that an infinitesimal field apparently suffices to destabilize the zero-field spinon Fermi-surface SL ground state. This is compatible with its gapless nature and stands in contrast to gapped SLs where a finite critical field is generally expected at zero temperature [13]. In this respect, the SL with a spinon Fermi surface responds to the applied field in a manner similar to that theoretically predicted for some other gapless SLs; e.g., in an infinitesimal magnetic field, the gapless Dirac $U(1)$ SL on the kagome lattice should become unstable towards spontaneous spin ordering with gapped excitations [25] and the gapless Majorana fermions of the Kitaev model on the honeycomb lattice become gapped [8,49].

Beyond the intricate physics of spin liquids, our finding of an intrinsic field-induced instability of the gapless Fermi-surface SL realized in Zn-brochantite should turn out to be relevant in a broader context of the Fermi-surface instabilities. It is important to bear in mind that many aspects of pair condensation in such SLs in Mott insulators are *universal*, as they are shared by electron pairing in superconductors [11], electron pairing in metals mediated by order-parameter fluctuations near quantum critical points, and composite-fermion pairing in quantum Hall fluids [21]. It is even possible that in some systems the same fermionic pairing occurs in both SL and superconducting phases [50].

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