μSR insight into the impurity problem in quantum kagome antiferromagnets

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Impurities, which are unavoidable in real materials, may play an important role in the magnetism of frustrated spin systems with a spin-liquid ground state. We address the impurity issue in quantum kagome antiferromagnets by investigating ZnCu$_3$(OH)$_6$SO$_4$ (Zn-brochantite) by means of muon spin spectroscopy. We show that muons dominantly couple to impurities, originating from Cu-Zn intersite disorder, and that the impurity spins are highly correlated with the kagome spins, allowing us to probe the host kagome physics via a Kondo-like effect. The low-temperature plateau in the impurity susceptibility suggests that the kagome spin-liquid ground state is gapless. The corresponding spin fluctuations exhibit an unconventional spectral density and a non-trivial field dependence.

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The two-dimensional Heisenberg quantum kagome antiferromagnet (QKA), the paradigm of geometrical frustration, has been in the focus of attention for several years [1,2]. Theoretical studies have lately converged on a spin-liquid (SL) ground state [3,8], most likely with a finite gap to magnetic spinon excitations [5–7]. Although early experiments, on the contrary, spoke in favor of a gapped SL [9–12], experimental evidence of a gapless ground state [9–12] has also recently been presented. However, the SL behavior of Zn-brochantite is even more perplexing, as two SL instabilities have recently been found at different temperatures [37]. Here, the impurities may play an important role by pinning spinons at low T’s and thus enabling a spinon-instability mechanism.

We tackle the impurity problem in QKA’s by performing muon spin relaxation and rotation (μSR) measurements on Zn-brochantite. At low T’s, the muons are expected to be dominantly influenced by impurities through the long-range dipolar interaction [37]. Such a scenario was indeed confirmed in herbertsmithite, where, however, the presence of a coupling between the impurity spins and the kagome spins remains unresolved. If the impurities are coupled to the correlated state of the host, like they are in high-$T_c$ superconductors and in many other strongly correlated electron systems [38,39], the muons may indirectly probe subtle correlations of the host state. Moreover, the screening response of spinon excitations to impurities should strongly depend on the particular SL ground state [40], which could then be determined by μSR. Here we demonstrate that in Zn-brochantite muons detect the impurity magnetism at low T’s and that intrinsic correlations between the impurity spins and the kagome spins due to a Kondo-like effect are indeed present. The observed low-T local-susceptibility plateau is consistent with a gapless host SL featuring a spinon Fermi surface [40]. Moreover, this SL state is characterized by unusual spin dynamics with an intriguing magnetic-field dependence.

μSR is a sensitive probe of magnetism that can easily distinguish between static and dynamic local magnetic fields $B$. In our previous μSR study in a small long-
FIG. 1. The $\mu^+$ polarization in Zn-brochantite in a TF of 0.3 T in the laboratory frame of reference (LFR; upper panel) and in the rotating frame of reference (RFR; lower panel), the latter rotating with the Larmor frequency $\nu$ and in the rotating frame of reference (RFR; lower panel), the latter rotating with the Larmor frequency $\nu$ and in the rotating frame of reference. The magnetic field (TF) of 0.3 T, as well as muon spin relaxation measurements in a transverse magnetic field $B_T$ are well described by the model $\pi/\nu t$'s. The extrapolation line yields the dominant contribution to $\chi$. This suggests that the muons mainly detect the magnetism of the impurity spins at high $T$'s, whereas at low $T$'s the diluted impurities yield the dominant contribution to $\chi$. The observed crossover is thus in line with the fact that in Zn-brochantite the bulk magnetic susceptibility $\chi$ is dominated by dense kagome spins at high $T$'s, whereas at low $T$'s the diluted impurities yield the dominant contribution to $\chi$. The observed crossover is thus in line with the fact that in Zn-brochantite the bulk magnetic susceptibility $\chi$ is dominated by dense kagome spins at high $T$'s, whereas at low $T$'s the diluted impurities yield the dominant contribution to $\chi$. The observed crossover is thus in line with the fact that in Zn-brochantite the bulk magnetic susceptibility $\chi$ is dominated by dense kagome spins at high $T$'s, whereas at low $T$'s the diluted impurities yield the dominant contribution to $\chi$. The observed crossover is thus in line with the fact that in Zn-brochantite the bulk magnetic susceptibility $\chi$ is dominated by dense kagome spins at high $T$'s, whereas at low $T$'s the diluted impurities yield the dominant contribution to $\chi$. The observed crossover is thus in line with the fact that in Zn-brochantite the bulk magnetic susceptibility $\chi$ is dominated by dense kagome spins at high $T$'s, whereas at low $T$'s the diluted impurities yield the dominant contribution to $\chi$. The observed crossover is thus in line with the fact that in Zn-brochantite the bulk magnetic susceptibility $\chi$ is dominated by dense kagome spins at high $T$'s, whereas at low $T$'s the diluted impurities yield the dominant contribution to $\chi$. The observed crossover is thus in line with the fact that in Zn-brochantite the bulk magnetic susceptibility $\chi$ is dominated by dense kagome spins at high $T$'s, whereas at low $T$'s the diluted impurities yield the dominant contribution to $\chi$. The observed crossover is thus in line with the fact that in Zn-brochantite the bulk magnetic susceptibility $\chi$ is dominated by dense kagome spins at high $T$'s, whereas at low $T$'s the diluted impurities yield the dominant contribution to $\chi$. The observed crossover is thus in line with the fact that in Zn-brochantite the bulk magnetic susceptibility $\chi$ is dominated by dense kagome spins at high $T$'s, whereas at low $T$'s the diluted impurities yield the dominant contribution to $\chi$.
early with the bulk susceptibility (lower inset in Fig. 2) up to \( \chi \approx 0.3 \text{ cm}^3/\text{mol} \), i.e., down to \( T \approx 5 \text{ K} \), which confirms the dominant coupling of the muons to the kagome spins at high \( T \)'s, in line with the high-\( T \) value of \( \beta \). The corresponding Knight shift

\[
K = \frac{\nu - \nu_0}{\nu_0} = A\chi_\mu
\]

is proportional to the local susceptibility \( \chi_\mu \), where \( A \) is the coupling constant between the electron and the muon magnetic moments. Even though \( A \) is of dipolar origin, one expects \( A \neq 0 \) for powder Cu-based samples, because of a sizable anisotropy of the magnetic response imposed by anisotropic \( g \) factors, which typically span the interval 2.05–2.3 \([12]\). The scaling of \( K \) with \( \chi \) changes around 5 K (Fig. 2) and reveals that the average coupling of the muons to the impurity spins \( A_1 = 36 \text{ mT}/\mu_B \) is somewhat smaller than their coupling to the kagome spins \( A_0 = 44 \text{ mT}/\mu_B \). The observed scaling of \( K \) with \( \chi \) below 5 K, where \( \chi \) is mainly due to impurities \([12]\), and the change in \( \beta \) unquestionably show that at these temperatures the \( \mu \)SR response is indeed dominated by impurities. This is in sharp contrast to NMR measurements \([37]\), where the 2D Knight shift revealed that the magnetic coupling with the impurity spins is much smaller than with the kagome spins [note the low-\( T \) leveling-off of the curve \( K_{\text{NMR}}(\chi) \) in Fig. 3]. This can be explained by the difference in the dominant coupling mechanism of the two probes with electron magnetic moments, i.e., the short-ranged hyperfine coupling in NMR versus the long-ranged dipolar coupling in \( \mu \)SR.

\( K \) saturates below \( \sim 0.5 \text{ K} \), while the saturation of \( \lambda_T \) occurs at a \( \sim 3 \) times higher temperature (Fig. 2). This dichotomy is even more obvious when comparing the variation of \( \lambda_T \) and \( K \) with \( \chi \) (Fig. 4). Both of these parameters reflect the static properties of the \( B_\mu \) distribution: \( K \) the average field value and \( \lambda_T \) the distribution width. In a paramagnet with inhomogeneous broadening due to anisotropic powder averaging, one should find \( \lambda_T \propto K \propto \chi \). However, if antiferromagnetic (AFM) correlations start developing they can affect the shape of the local field distribution. Such an AFM correlated regime can therefore lead to a situation where \( \lambda_T \neq K \propto \chi \). The saturation of \( \lambda_T \) below \( \sim 1.5 \text{ K} \) is thus a sign of quasi-static AFM correlations involving the impurity spins, as they dominate the system’s magnetic response at low \( T \)'s. This temperature corresponds well to the effective Weiss temperature \( \theta^C_{\text{SW}} \approx -1.2 \text{ K} \) of the impurity spins at low \( T \)'s \([12]\). Furthermore, the \( \lambda_T(\chi) \) dependence exhibits an anomalous slope change already at 70 K (Fig. 3), a temperature that is close to the Weiss temperature \( \theta^C_{\text{SW}} = -79 \text{ K} \) of the kagome spins \([12]\). This anomaly is thus likely due to the development of quasi-static spin correlations among the intrinsic kagome spins, which are also reflected in increased 2D NMR line width \([37]\).

The saturation of \( K \) below \( \sim 0.5 \text{ K} \) (Fig. 3), the low-
Two power-law regimes are indicated by solid lines. The inset shows the corresponding polarization curves (symbols) and stretched-exponential fits (solid lines).

The sublinear field dependence of $1/\lambda_L$ for $B < B_c$ is rather unusual. The conventional exponential decay of the local-field autocorrelation function $S(t) \propto e^{-t/\tau}$, characteristic of a single-correlation-time ($\tau$) Markovian local-field evolution, yields a Lorentzian spectral density $S(\omega)$, where $1/\lambda_L \propto S(\omega)^{-1} \propto (\gamma_e B)^2$ [42]. The high-field value $p = 2.5(4)$ is indeed close to 2. The experimentally observed $p = 0.2$ at low fields, however, suggests a more exotic spectral density $S(\omega) \propto \omega^{-p} = \omega^{-0.2}$, where temporal correlations decay algebraically as $S(t) \propto t^{-(1-p)} = t^{-0.8}$ [43]. A similar situation with $p \leq 1$ was observed on a few occasions in different SL phases of spin chains [10], pyrochlores [47], and QKA’s [32, 48].

$S(\omega) \propto \omega^{-0.2}$ could be due to a distribution of correlation times of the impurity spins. The impurities could still be decoupled from the SL properties of the kagome spins and the power-law $S(\omega)$ could be due to a presence of impurity-impurity couplings with a distribution of strengths arising from the randomness of impurity positions. However, the drastic change of the spectral density at $B_c$ strongly suggests a more involved scenario.

The sudden sizable increase of $p$ at $B_c \sim 0.4$ T is rather intriguing. It cannot be accounted for by field-induced polarization of the impurity spins, which would yield gradual changes. It rather strengthens the conclusion from the saturation of the muon Knight shift that the impurities are coupled to the SL phase of the kagome spins. The properties of the SL phase obviously abruptly change at $B_c$. Therefore, in addition to the well documented $T$-induced instabilities in Zn-brochantite [37], the low-$T$ SL state could also exhibit a field-induced instability at the critical field $B_c$. This calls for future in-depth studies, as field-induced instability appears to be a common feature of frustrated antiferromagnets [43, 50], yet its origin remains unknown.

The correlations between the impurity and the kagome spins in the SL state can be understood as a Kondo-like effect, which can emerge on frustrated spin lattices from either fermionic or bosonic spinon excitations [40, 61, 52]. The resulting screening generally leads to Curie-Weiss behavior of impurity susceptibility at $T \gtrsim T_K$, where the Kondo temperature $T_K$ is of the order of $\Theta_{\text{CW}}^\text{low}$ [54]. Therefore, the observed low-$T$ Curie-Weiss behavior does not require impurity-impurity interactions. Below $T \sim T_K$ the Curie-Weiss behavior should break down [54], which nicely corresponds to the onset of the plateau in the impurity susceptibility $\chi_\mu$ at $\sim 0.5$ K. Moreover, the Kondo effect can help to differentiate between different candidate SL ground states. In particular, the finite $\chi_\mu$ at $T \to 0$ observed in Zn-brochantite is consistent with predictions for a gapless SL with a spinon Fermi surface [40, 53], while it contradicts the divergent behavior expected for the gapless Dirac $U(1)$ SL’s and the exponentially suppressed impurity response of gapped $Z_2$ SL’s [40]. Our observations are thus in accord with previous hints about the nature of the SL in Zn-brochantite from bulk heat-capacity measurements [12].

The evidently gapless SL in Zn-brochantite is at odds with the apparently gapped SL in herbertsmithite [13]. Moreover, the experimentally detected correlations between the impurity and kagome spins in the SL state of Zn-brochantite stand in contrast to the recent inelastic neutron scattering results on herbertsmithite [14], which advocated that a diluted impurity lattice with random-bond Heisenberg interactions may be effectively decoupled from the intrinsic kagome physics at $T/J \sim 1/100$. The differences between the two compounds may have various origins; the perturbing magnetic anisotropies are not necessarily the same, the position of the Zn (impurity) site is different [12] and the kagome lattice in Zn-brochantite is slightly distorted [12].

In conclusion, by performing $\mu$SR on Zn-brochantite we have shown that this technique can comprehensively addresses the impurity magnetism that generally dominates the bulk response of QKA representatives at low $T$’s. Our results reveal that the low-$T$ kagome-lattice SL state in the investigated compound is reflected in the magnetic behavior of the impurity spins, implying strong correlations between the impurity and the kagome spins. Our findings thus suggest a Kondo-like description of impurity spins on the kagome lattice, a description that is also applicable to other...
quantum SL systems, e.g., the organic triangular-lattice antiferromagnets \cite{Clark2012} and the Kitaev quantum SL's \cite{Yi2008}. Based on theoretical predictions for the Kondo effect in SL's, details of the observed impurity behavior enable us to confirm a gapless SL with a spinon Fermi surface as the ground state of Zn-brochantite. Impurities thus indeed prove to be salient local probes and may turn out to be essential for finally fully understanding the ground state of the QKA.

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