

# Dirac spinons intermingled with singlet states in the random kagome antiferromagnet $\text{YCu}_3(\text{OD})_{6+x}\text{Br}_{3-x}$ ( $x = 0.5$ )

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We elucidate the ground state nature and low-energy spin dynamics of the  $s = 1/2$  near-perfect kagome antiferromagnet  $\text{YCu}_3(\text{OD})_{6+x}\text{Br}_{3-x}$  ( $x = 0.5$ ) using  $^{63}\text{Cu}$  nuclear quadrupole resonance (NQR) and muon spin relaxation/rotation ( $\mu\text{SR}$ ) techniques. Our combined  $^{63}\text{Cu}$  NQR and  $\mu\text{SR}$  data, along with the inverse Laplace transform analysis, reveal an inhomogeneous ground state comprising the majority of a gapless spin liquid intermingled with a minor fraction of spin singlets with varying energy gaps. Furthermore, the  $^{63}\text{Cu}$  NQR relaxation rate evinces distinct signatures of Dirac spinons, featuring a power-law dependence of  $1/T_1 \sim T^\eta$  with  $\eta = 1.35$  at temperatures below  $0.13\text{J}$  ( $\cong 8\text{K}$ ). Our results demonstrate that a kagome lattice endowed with specific exchange randomness provides a prominent platform for exploring Dirac fermions in magnetic insulators.

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## I. INTRODUCTION

The  $s = 1/2$  kagome Heisenberg antiferromagnets (KHAFs) have long been considered a holy grail in the quest for quantum spin liquids (QSLs) [1]. Recently, a growing body of numerical calculations supports the existence of either a  $\mathbb{Z}_2$  gapped QSL or a gapless  $U(1)$  Dirac QSL [2–5]. However, perturbations inevitable in real materials cause transitions from the sought-after QSL to neighboring phases such as chiral spin liquids, valence bond solids (VBSs), and magnetically ordered states [6].

From a materials standpoint, the discovery of herbertsmithite  $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$  in 2005 has ignited a surge of research activity [7–10]. Subsequent investigations into its polymorph and variations, including kapellasite, Zn-brochantite, and Zn-barlowite, have uncovered intriguing QSL characteristics such as fractionalized spin excitations and the absence of long-range magnetic order [11–16]. Nonetheless, the presence of Zn-Cu antisite mixing gives rise to a coexistence of spatially varying gapped singlets and a gapless disordered state [17], posing formidable challenges in comprehending the intrinsic kagome physics.

In this context, the recently synthesized  $\text{YCu}_3(\text{OH})_{6+x}\text{X}_{3-x}$  ( $X = \text{halogen}$ ) compounds stand out, primarily owing to their tunable structure, which can vary from a perfect kagome lattice to a distorted structure depending on the composition [18–22]. Unlike herbertsmithite,  $\text{YCu}_3(\text{OH})_{6+x}\text{X}_{3-x}$

lacks intersite mixing due to the substantial difference in the ionic radii of  $\text{Y}^{3+}$  and  $\text{Cu}^{2+}$ . Notably,  $\text{YCu}_3(\text{OH})_{6+x}\text{Br}_{3-x}$  ( $x \sim 0.5$ ) (abbreviated as  $\text{YCu}_3\text{-Br}_{2.5}$ ) defies conventional magnetic ordering down to 50 mK and displays thermodynamic and spectroscopic signatures of Dirac spinons, including a low-temperature  $T^2$  specific heat, spinon excitations with a conical structure, and superlinear magnetic Raman excitations [23–30]. However, thermal conductivity measurements indicate the presence of a spin gap that seems to contradict gapless excitations at the Dirac nodal points [31]. Furthermore, the observed Dirac nodes deviate in their position and number within the Brillouin zone from the predictions of the mean-field KHAF model [2,32]. Exchange randomness, arising from a random occupation of  $\text{Br}^-$  and  $\text{OH}^-$ , may be responsible for such modifications, warranting further exploration.

In this paper, we combine muon spin relaxation/rotation ( $\mu\text{SR}$ ) and  $^{63}\text{Cu}$  nuclear quadrupole resonance (NQR) techniques to examine the purported Dirac spinons in  $\text{YCu}_3\text{-Br}_{2.5}$ . Our local-probe data unveil the prevalence of a Dirac-like QSL state, coexisting with a minority of spin singlets. This observation underscores the persistence of Dirac spinons against exchange randomness. Additionally, the inhomogeneous ground state reveals deeper insights into the seemingly conflicting gapped and gapless behaviors reported among different measurement techniques.

## II. EXPERIMENTAL DETAILS

Single crystals of  $\text{YCu}_3(\text{OD})_{6.5}\text{Br}_{2.5}$  were synthesized by a hydrothermal method as detailed in Ref. [28].

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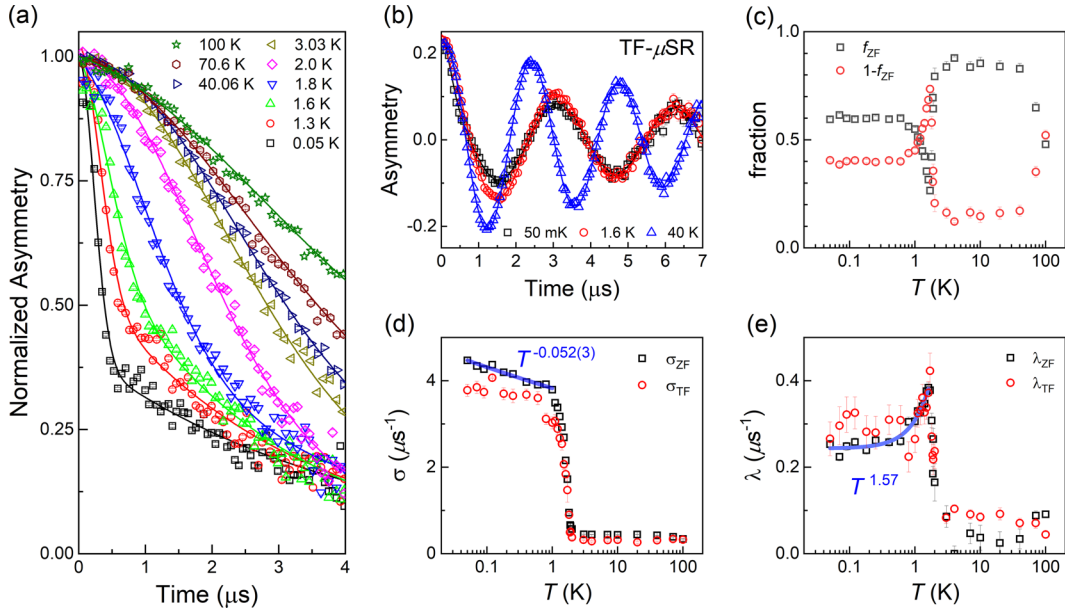


FIG. 1. (a) Temperature dependence of normalized ZF- $\mu$ SR spectra. (b) TF- $\mu$ SR spectra at selected temperatures. (c) Fraction of the Gaussian relaxing component. (d) Muon relaxation rates  $\sigma_{ZF}$  (black squares) and  $\sigma_{TF}$  (red circles) plotted against temperature on a semilogarithmic scale. (e) Semilog plot of  $\lambda_{ZF}$  (black squares) and  $\lambda_{TF}$  (red circles) vs temperature, extracted from the relaxation fitting function detailed in the main text. The solid lines present their power-law dependence below 1 K.

$\mu$ SR experiments were conducted at the M15 and M20 beamlines in TRIUMF (Vancouver, Canada). The single crystals were loaded into a dilution refrigerator for low- $T$  measurements ( $T = 0.05$ –4 K) at the M15 beamline. Higher-temperature measurements ( $T = 1.8$ –100 K) were performed on samples packed by Al-mylar tape using the LAMPF spectrometer at the M20 beamline. Zero-field (ZF) and transverse-field (TF)  $\mu$ SR data at  $T = 1.8$  K were used to quantify the background fraction of signals arising from the silver sample holder in the dilution refrigerator. All obtained  $\mu$ SR data were fitted utilizing the MUSRFIT software package [33].

The  $^{63,65}\text{Cu}$  ( $I = 3/2$ ) NQR spectra were acquired using a standard Hahn-echo sequence with a  $\pi/2$  pulse length of  $\tau_{\pi/2} = 15 \mu\text{s}$ . While tracking all peaks, we varied the frequency point by point with a 0.01 MHz interval. The spin-lattice relaxation rate  $1/T_1$  was measured by the saturation recovery technique with multiple saturation pulses and  $\tau_{\pi/2} = 2$ –3  $\mu\text{s}$ . The nuclear magnetization recovery curves  $M(t)$  for the central transition line with elapsed time  $t$  were fitted to the expression  $[M_\infty - M(t)]/M_\infty = A[e^{-(3t/T_1^{\text{str}})^\beta}]$ , where  $A$  is a fitting parameter and  $\beta$  is the stretched exponent. The  $1/T_1$  difference between  $^{65}\text{Cu}$  and  $^{63}\text{Cu}$  was found to be marginal, approximately 3%. In addition to the stretched fit, we further analyzed  $M(t)$  using the inverse Laplace transform (ILT) method based on Tikhonov regularization [34,35]. Assuming that  $1/T_1$  is spatially varied, the histogram  $P(1/T_1)$ , representing the probability density for the nuclear spins with each relaxation rate, was deduced using the expression  $M(t)/M_\infty = \sum_j P(1/T_{1j})[1 - \exp(-3t/T_{1j})]$  with  $\sum_j P(1/T_{1j}) = 1$ . Here,  $1/T_{1j}$  denotes the  $j$ th value of the distributed  $1/T_1$ . In deriving  $P(1/T_1)$ , we ensured the validity of our fitting procedure by following the

methodology outlined in Ref. [35]. Furthermore, we stress that our fitting protocol was successfully applied to disordered Kitaev materials  $\text{H}_3\text{LiIr}_2\text{O}_6$  and hyperkagome antiferromagnet  $\text{Na}_{4-x}\text{Ir}_3\text{O}_8$  [36,37].

### III. RESULTS

#### A. Muon spin relaxation/rotation

Figures 1(a) and 1(b) present the ZF- and TF- $\mu$ SR spectra of  $\text{YCu}_3\text{-Br}_{2.5}$ . With decreasing temperature, the muon spin polarization undergoes rapid relaxation in the initial time window  $t = 0$ –1  $\mu\text{s}$ . Nonetheless, the lack of coherent oscillation down to 50 mK and a 1/3 recovery of the muon polarization at long times precludes the occurrence of static magnetic ordering.

For a quantitative analysis, we modeled the ZF- $\mu$ SR spectra as the sum of a Gaussian and a simple exponential function, along with a background signal  $a_{\text{bkg}}$ :  $P_{ZF}(t) = f_{ZF} \exp(-\frac{1}{2}\sigma_{ZF}^2 t^2) + (1 - f_{ZF}) \exp(-\lambda_{ZF} t) + a_{\text{bkg}}$ . Here,  $\sigma_{ZF}$  and  $\lambda_{ZF}$  are the zero-field muon relaxation rates, and  $f_{ZF}$  is the fraction of the Gaussian relaxing component. Additionally, the TF- $\mu$ SR spectra were fitted using the relaxation function,  $a_{TF} P_{TF}(t) = a_{TF} [f_{TF} \exp(-\frac{1}{2}\sigma_{TF}^2 t^2) + (1 - f_{TF}) \exp(-\lambda_{TF} t)] \cos(2\pi f_\mu t + \phi)$ , where  $a_{TF}$  is the initial asymmetry from the sample,  $f_\mu$  is the muon Larmor precession frequency, and  $\phi$  is the initial phase. Noteworthy is that Gaussian-shaped relaxation has been reported in kagome systems  $\text{SrCo}_8\text{Ga}_4\text{O}_{19}$  [38] and  $\text{YBaCo}_4\text{O}_{7+\delta}$  [39], suggesting the presence of a local field infrequently detected within the fluctuation time of muons. In this vein, the concurring Gaussian and exponential relaxation functions allude to an inhomogeneous mixture of distinctly behaving spins.

The resulting fit parameters are summarized in Figs. 1(c)–1(e), revealing similar trends between the  $\sigma_{ZF}$  ( $\lambda_{ZF}$ ) and  $\sigma_{TF}$  ( $\lambda_{TF}$ ) data. As depicted in Fig. 1(c), the Gaussian and exponential relaxing components are nearly equal at 100 K. As the temperature is lowered, the fraction  $f_{ZF}$  exhibits a steep increase, reaching  $\approx 0.84$  at  $T = 40$  K, followed by a sharp drop to 0.26 at  $T_f = 1.6$  K. Eventually,  $f_{ZF}$  recovers to a value of  $\approx 0.6$  below 1 K. These two anomalies are associated with the low- and high-temperature peaks observed in the specific heat, corresponding to the spin freezing and spin-range spin ordering, respectively [28]. The fraction  $f_{TF}$  displays similar behavior to  $f_{ZF}$  (not shown here).

Evident from Fig. 1(d), both  $\sigma_{ZF}$  and  $\sigma_{TF}$  show a steep increment with lowering temperature through 2 K, indicating the slowdown of spin correlations. For temperatures below 1 K,  $\sigma_{ZF}(T)$  exhibits a weak power-law increase of  $\propto T^{-0.052}$ . We note that the values of  $\sigma_{ZF}(\sigma_{TF}) \sim 4\text{--}4.5 \mu\text{s}^{-1}$  are an order of magnitude larger than  $\lambda_{ZF}(\lambda_{TF}) \sim 0.25\text{--}0.3 \mu\text{s}^{-1}$ . In the fast-fluctuation regime above 2 K, the exchange fluctuation rate is given by  $\nu = \sqrt{z}JS/\hbar \sim 8.25 \times 10^{12} \text{ s}^{-1}$  [40], where  $z = 4$  is the nearest-neighbor coordination number and  $J \sim 60$  K is the exchange interaction [28]. Using the relation  $\lambda = \Delta^2/\nu$  in the motional narrowing limit,  $\lambda_{ZF}(T = 100 \text{ K}) = 0.0912 \mu\text{s}^{-1}$  yields a distribution width of local magnetic fields  $\Delta/\gamma_\mu = 7.20(1) \text{ kG}$ .

On the contrary, a discernible peak at  $T_f$  observed in  $\lambda_{ZF}$  and  $\lambda_{TF}$  [see Fig. 1(e)] indicates the partial freezing of local magnetic moments at approximately 1.6 K. On cooling below  $T < T_f$ ,  $\lambda_{ZF}(T)$  follows a  $T^{1.57}$  power-law decrease with a residual relaxation rate  $\lambda_{ZF}(T = 50 \text{ mK}) = 0.25 \mu\text{s}^{-1}$ . Similar behavior has been observed in a range of geometrically frustrated magnets, including  $\text{Dy}_2\text{Ti}_2\text{O}_7$  [41],  $\text{Y}_2\text{Mo}_2\text{O}_7$  with  $\lambda \propto T^{2.1}$  for  $T < 22 \text{ K}$  [42], and  $\text{RbAg}_2\text{Cr}[\text{VO}_4]_2$  with  $\lambda \propto T^{0.9}$  below 20 K [43]. The substantial residual spin dynamics below  $T < T_f$  signifies the persistence of low-energy magnetic fluctuations within the  $\mu\text{SR}$  time window.

Next, we move on to the muon decoupling experiments to further characterize spin-spin correlations. Figures 2(a) and 2(b) present the longitudinal field (LF)- $\mu\text{SR}$  spectra measured at  $T = 50 \text{ mK}$  and 10 K. It is evident that our highest applied LF of 4000 G is insufficient to attain full suppression of muon spin depolarization, which evinces the predominance of dynamically fluctuating spins. The same relaxation function utilized to fit the ZF- $\mu\text{SR}$  data is also used to model the LF- $\mu\text{SR}$  data for temperatures below  $T_f$ .

As shown in Fig. 2(c), the LF dependence of  $\sigma_{LF}(B)$  at  $T = 50 \text{ mK}$  initially decreases rapidly, and then levels out above 2000 G. We find that  $\sigma_{LF}(B, T = 50 \text{ mK})$  is well described by the Redfield equation  $\sigma_{LF}(B) = 2\gamma_\mu^2 \langle B_{\text{loc}}^2 \rangle \nu / (\nu^2 + \gamma_\mu^2 B_{\text{LF}}^2)$ . Here,  $\gamma_\mu$  is the muon gyromagnetic ratio and  $\langle B_{\text{loc}} \rangle$  is the time average of the fluctuating internal field at muon sites. The estimated values of  $\nu = 36.89(1) \mu\text{s}^{-1}$  and  $\langle B_{\text{loc}} \rangle = 105.44(7) \text{ G}$  are roughly five times slower and larger than those of herbertsmithite, which are  $\nu = 150 \text{ MHz}$  and  $\langle B_{\text{loc}} \rangle = 18 \text{ G}$ , respectively [7]. Meanwhile,  $\lambda_{LF}(B)$  [open blue squares in Fig. 2(d)] exhibits a two-stage decrease with increasing LF, suggesting the development of complex spin correlations.

For the  $T = 10 \text{ K}$  LF- $\mu\text{SR}$  spectra, we model them as the relaxation function,  $a_0 P_{LF}(t) = a_0 [f G_{LF}(t) + (1-f) \exp(-\lambda_{LF} t)] + a_{\text{bkg}}$ , where  $a_0$  is the initial asymmetry for

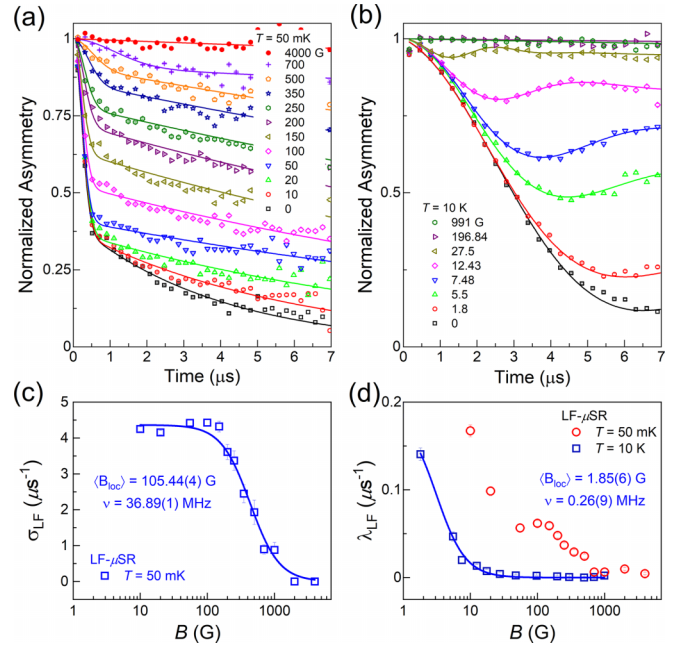


FIG. 2. Longitudinal field dependence of normalized LF- $\mu\text{SR}$  spectra at (a)  $T = 50 \text{ mK}$  and (b)  $T = 10 \text{ K}$ . The muon relaxation rate  $\sigma_{LF}$  (c) at  $T = 50 \text{ mK}$  and  $\lambda_{LF}$  (d) at  $T = 50 \text{ mK}$  (open blue squares)  $T = 10 \text{ K}$  (pink circles) as a function of applied longitudinal fields. The solid lines are the fit using the Redfield formula.

a LF configuration. Here,  $G_{LF}(t)$  describes a system having static magnetic moments under LF conditions [44], given by

$$G_{LF}(t) = 1 - \frac{2\sigma^2}{(2\pi\nu)^2} \left[ 1 - \exp\left(-\frac{1}{2}\sigma^2 t^2\right) \cos(2\pi\nu t) \right] + \frac{2\sigma^4}{(2\pi\nu)^3} \int_0^t \exp\left(-\frac{1}{2}\sigma^2 \tau^2\right) \sigma(2\pi\nu\tau) d\tau.$$

By analyzing  $\lambda_{LF}(B, T = 10 \text{ K})$  based on the Redfield formula, we obtain  $\nu = 0.26(9) \text{ MHz}$  and  $\langle B_{\text{loc}} \rangle = 1.85(6) \text{ G}$ .

## B. Nuclear quadrupole resonance

We turn to the zero-field  $^{63,65}\text{Cu}$  NQR spectra. As exhibited in Fig. 3(a), two distinct lines are seen at  $^{65}\nu_Q = 7.2(1) \text{ MHz}$  and  $^{63}\nu_Q = 7.7(5) \text{ MHz}$  whose frequency ratio matches that of nuclear quadrupole moments ( $Q$ ):  $^{63}\nu_Q / ^{65}\nu_Q = ^{63}Q / ^{65}Q \approx 1.08$  [45]. With decreasing temperature, the  $^{63,65}\text{Cu}$  NQR line shapes experience a power-law  $T^{-0.33}$  broadening above 1 K and  $T^{-0.80}$  below 1 K [see pink circles in Fig. 3(b)], suggesting enhanced magnetic correlations through  $T_f$ . Nonetheless, no apparent NQR shift is observable in their spectral lines, possibly due to the broader linewidth of the  $^{63,65}\text{Cu}$  NQR compared to the thermal shift of  $^{63,65}\nu_Q$ .

We next discuss the  $^{63}\text{Cu}$  nuclear spin-lattice relaxation rate  $1/T_1$ , which pertains to low-energy spin excitations integrated over  $\mathbf{q}$  at the NQR frequency  $\omega_0$ . In determining  $1/T_1$ , we attempted to analyze  $M(t)$  using two alternative methods: a conventional stretched exponential fit versus an ILT fit. Figure 3(c) compares the representative fits at  $T = 0.5 \text{ K}$ , showing that the ILT fit (solid line) provides a somewhat better description of  $M(t)$  than the stretched fit (dotted line).

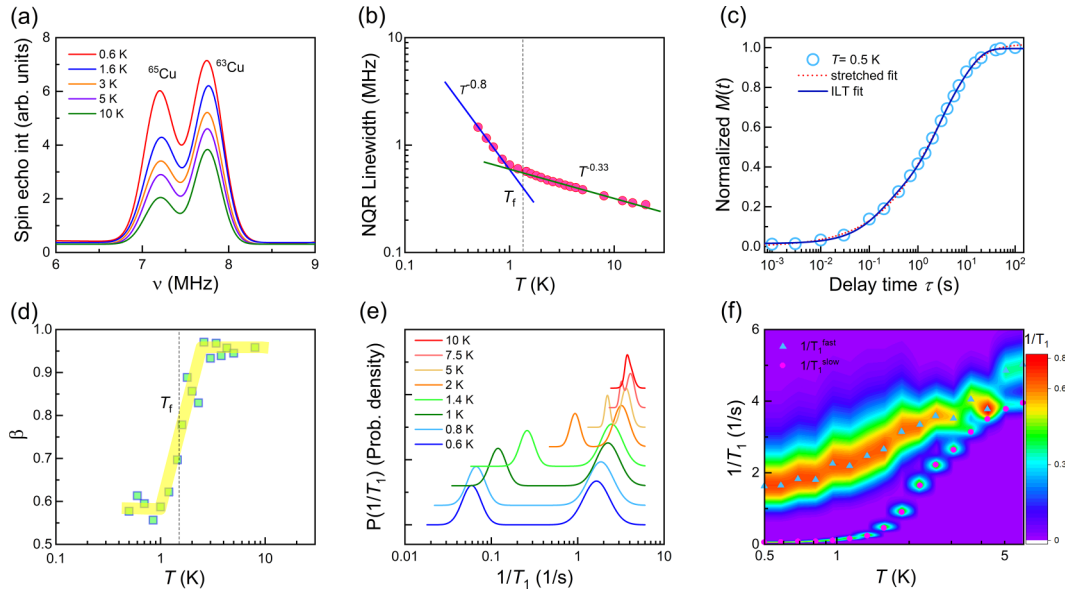


FIG. 3. (a)  $^{63,65}\text{Cu}$  NQR spectra of  $\text{YCu}_3(\text{OD})_{6.5}\text{Br}_{2.5}$  at selected temperatures. (b) Temperature dependence of the NQR linewidth on a semilogarithmic scale. The vertical dashed line indicates the spin-freezing temperature  $T_f$ . The solid lines depict power-law behaviors. (c) Stretched (dotted line) and ILT (solid line) fits of normalized  $M(t)$  at  $T = 0.5$  K. (d) Semilog plot of the stretched exponent  $\beta$  vs temperature. The shaded thick line is a guide to the eye. (e) Distribution function  $P(1/T_1)$  plotted against temperature in a semilog scale. (f) Color contour plot of  $P(1/T_1)$  in the temperature- $1/T_1$  plane. The triangles and circles represent the center positions of the two separate distributions.

As plotted in Fig. 3(d), on cooling through  $T_f$ , the stretched exponent  $\beta$  undergoes a strong reduction from  $\approx 0.96$  to  $0.57$ , implying the development of a significant distribution of  $1/T_1$  due to spin freezing.

In Fig. 3(e), we show a semilog plot of the probability density function  $P(1/T_1)$  vs  $1/T_1$ , extracted from the ILT analysis of  $M(t)$ . The corresponding color contour plot of  $P(1/T_1)$  vs  $T$  is further displayed in Fig. 3(f). Here,  $1/T_1$  on the y axis is presented in a linear scale, in contrast to the logarithmic scale used on the x axis in Fig. 3(e). Notably, a two-peak structure evolves progressively from an initially single line at  $\sim 4$  s $^{-1}$  below  $T = 10$  K. By deconvoluting the  $P(1/T_1)$  histogram into two Gaussian profiles, we trace separately the thermal location of the peaks: the slower  $T_1^{\text{slow}}$  (pink circles) and faster  $T_1^{\text{fast}}$  (green triangles) components. Remarkably,  $T_1^{\text{slow}}$  is almost two orders of magnitude smaller than  $T_1^{\text{fast}}$ .

To shed light on the underlying nature of spin dynamics, the center positions of the slower and slower relaxation components are plotted as a function of temperature in Fig. 4(a).  $T_1^{\text{fast}}(T)$  (green triangles) exhibits a  $T^{1.35}$  power-law decrease as  $T \rightarrow 0$  K from 8 K, which is characteristic of a gapless QSL ground state. Conversely, the notable suppression of  $T_1^{\text{slow}}(T)$  (pink circles) below 8 K suggests the emergence of gapped excitations. The absence of activation behavior means varying spin gaps rather than a well-defined gap energy (not shown here). Further, the lacking constant gap is not compatible with a chiral spin-liquid scenario [6]. To validate the extracted  $P(1/T_1)$  through the ILT procedure, we compare  $T_1^{\text{str}}$  (blue stars) with the center of gravity of  $P(1/T_1)$  [crosses in Fig. 4(a)]. The near-perfect overlap between these values lends strong support to the existence of two-component

spin dynamics, which cannot be adequately captured by the stretched exponential analysis.

The temperature dependence of  $1/T_2$  is shown in the left y axis of Fig. 4(b).  $1/T_2$  (pink circles) exhibits a weak power-law reduction with  $1/T_2 \sim T^{0.15}$  in the  $T = 1$ – $5$  K range and undergoes a steeper decrease below 1 K. Notably,  $1/T_2$  mirrors the thermal evolution of the NQR linewidth [see Fig. 3(b)], although the power-law temperature window is reduced down to 5 K. The right y axis of Fig. 4(b) presents the singlet fraction ( $f_s$ ) of  $\text{Cu}^{2+}$  sites obtained by the integration of the  $P(1/T_1)$  curve [data shown in Fig. 3(e)]. On cooling,  $f_s(T)$  shows a steady decrease from the initial value of  $f_s = 0.31$  down to 2 K, and then undergoes a rapid drop through  $T_f$ , and eventually flattens out with  $f_s \approx 0.02$  below 1 K.

Before proceeding, we discuss the differences in the temperature dependencies of the muon relaxation rate ( $\lambda$ ) and the  $^{63,65}\text{Cu}$   $1/T_1$ . In sharp contrast to  $\lambda$  [see Fig. 1(e)],  $1/T_1$  shows no signatures of spin freezing at  $T_f$ . One possible explanation is that the temporal structure of spin fluctuations hinges on the time domain probed by NMR and  $\mu\text{SR}$  in the presence of magnetic inhomogeneities. Another possibility involves the modified form factor of the implanted muons. In magnetic oxides, the positive muon typically resides near the negatively charged oxygen ions. In  $\text{YCu}_3\text{-Br}_{2.5}$ , the  $\text{Br}_2/\text{O}_2$  sites are randomly occupied, unlike the O1 site, leading to variations in the form factor. Therefore, the on-site  $^{63,65}\text{Cu}$  nuclei serve as a better probe of intrinsic spin dynamics. Altogether, our ILT analysis of  $^{63}\text{Cu}$  NQR reveals that the ground state of  $\text{YCu}_3\text{-Br}_{2.5}$  is predominantly characterized by a gapless quantum disordered state, intermingled with a few percentages of random singlets. A mixture of these two



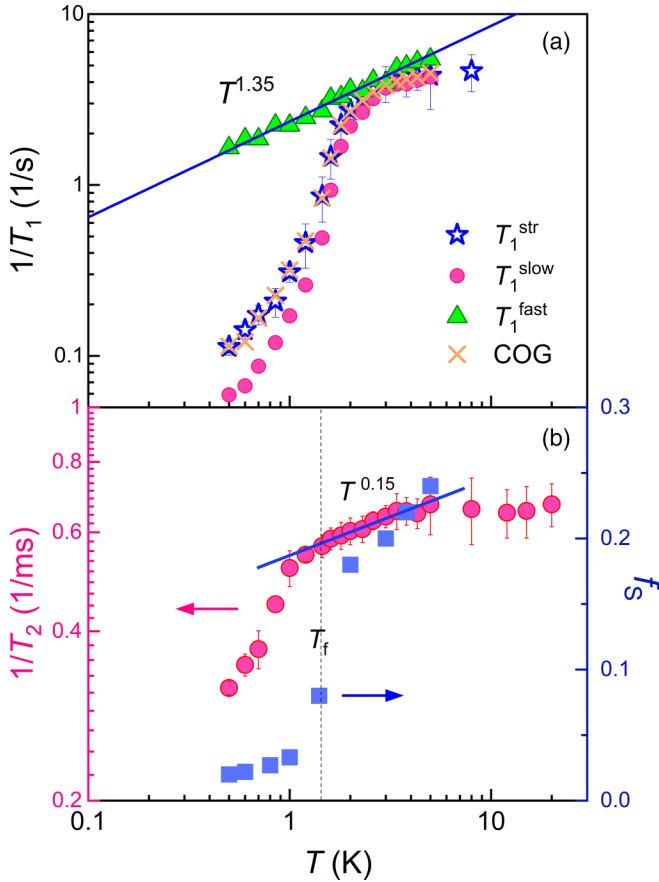


FIG. 4. (a) Temperature dependence of  $1/T_1^{\text{slow}}$  (pink circles),  $1/T_1^{\text{fast}}$  (green triangles),  $1/T_1^{\text{str}}$  (blue stars), and the center of gravity (COG; crosses). (b) Temperature dependence of the spin-spin relaxation rate  $1/T_2$  (left y axis) and the singlet fraction  $f_s$  of Cu sites (right y axis) plotted on a log-log scale. The solid lines represent the power-law dependence of the  $^{63,65}\text{Cu}$  nuclear relaxation rates. The vertical dashed line marks the spin-freezing temperature  $T_f$ .

distinct states appears to occur inevitably in the presence of exchange randomness [46,47].

#### IV. DISCUSSION AND CONCLUSION

With the help of  $\mu\text{SR}$  and  $^{63}\text{Cu}$  NQR techniques, we decipher the inhomogeneous nature of the alleged QSL ground state in  $\text{YCu}_3\text{-Br}_{2.5}$ , which holds remarkable similarities with Zn-barlowite and herbertsmithite [17]. In both  $\text{YCu}_3\text{-Br}_{2.5}$  and herbertsmithite, we identify the characteristic temperature  $T^* = 0.13\text{--}0.16J$ , below which heterogeneous magnetic correlations emerge. Despite these apparent resemblances, there exists a fundamental disparity between  $\text{YCu}_3\text{-Br}_{2.5}$  and herbertsmithite. In  $\text{YCu}_3\text{-Br}_{2.5}$ , unlike herbertsmithite, the gapless component obeys a power-law behavior of  $1/T_1 \sim T^\eta$ , which is reminiscent of Dirac fermions predicted in gapless QSL [2,3]. Furthermore, the singlet fraction diminishes with small residual fraction as  $T \rightarrow 0$  K, opposite to the steady growth of the singlet fraction of Cu sites observed for Zn-barlowite and herbertsmithite [17].

This contrasting behavior relies on the distinct species of quenched disorders. In Zn-barlowite and herbertsmithite,

antisite cation mixing and dangling bonds on kagome lattices generate abundant low-lying excitations, which may cover a small spectral weight emanating from the Dirac nodal points. In this situation, random-singlet-type states become dominant over the hypothesized Dirac spin liquid. As to  $\text{YCu}_3\text{-Br}_{2.5}$ , the exchange randomness of three different  $J$ s as a leading perturbation enlarges the Brillouin zone and brings about gapped excitations of an order of  $\sim 10$  K [25,28]. In this case, the alleged Dirac spinons lying below 10 K may be hardly affected by the sizable bond randomness.

We further discuss the observed  $^{63}\text{Cu}$  NQR results in light of mean-field calculations for the isotropic KHAF model [3]. According to the mean-field KHAF model, the Dirac spinon is well defined in the energy range below the spinon bandwidth  $W \approx 0.25J$  ( $\approx 15$  K). Notably, a recent INS study confirmed the Dirac-like linear dispersion up to 7 K [27]. Consistently with the INS results, the onset temperature ( $\sim 0.13J \approx 8$  K) of the power-law  $1/T_1$  amounts to half of the energy scale of  $W$ . Admittedly, the  $1/T_1 \sim T^{1.35}$  behavior alone is insufficient to confirm the presence of Dirac spinons. Nonetheless, the Dirac conical excitations observed through INS [27] suggest that the near-linear  $1/T_1$  dependence may originate from Dirac spinons. Unlike  $\lambda(T)$  in Fig. 1(e) and  $1/T_2$  in Fig. 4(b), the power-law dependence of  $1/T_1$  is barely affected by the weak spin freezing at  $T_f$ . This implies the robustness of Dirac spinon excitations against the small perturbation of  $T_f/W \sim 0.1$ , albeit not in an entirely pristine form.

Secondly, our  $^{63}\text{Cu}$  NQR data yield the exponent  $\eta_{\text{exp}} = 1.35$  [see Fig. 4(a)], associated with the scaling dimension through the relation  $\eta_{N(m)} = 2\Delta_{N(m)} - 1$  [3]. Here,  $\Delta_N \approx 1.73$  represents the correlations of VBS order, while  $\Delta_m \approx 1.06$  is  $\mathbf{q} = \mathbf{0}$  magnetic correlations. The experimental value of  $\Delta_{\text{exp}} \approx 1.18$  means that  $\text{YCu}_3\text{-Br}_{2.5}$  is on the verge of the  $\mathbf{q} = \mathbf{0}$  magnetic instability. Actually, our  $\mu\text{SR}$  data detect the weak freezing of magnetic moments in this material [see Fig. 1(e)].

The prevailing Dirac spinons detected by  $^{63}\text{Cu}$  NQR, coexisting with a small fraction of singlet states, align well with various experimental observations such as the  $T^2$  specific heat, Dirac conical excitations by INS, and superlinear  $\omega$  dependence of Raman spinon scattering [23–28,30]. On the other hand, the semimetallic nature of putative Dirac spinon excitations and the presence of random singlets account for the lack of itinerant quasiparticles suggested by thermal conductivity measurements [31], as the density of states of the low-energy quasiparticles is insufficient to carry heat. All in all, our  $\mu\text{SR}$  and NQR results reveal an inhomogeneous ground state that behaves more intriguingly than a simple spin gap or gapless QSL.

To conclude, we have thoroughly investigated the intricate ground state and its low-energy spin dynamics of an  $s = 1/2$  Cu-based kagome compound  $\text{YCu}_3\text{-Br}_{2.5}$  subject to exchange randomness. Our magnetic resonance measurements uncover that a heterogeneous ground state is overwhelmingly dominated by the much sought-after Dirac spinons. The near realization of these elusive Dirac spinons in random kagome lattices raises the question of their relevance to a Dirac QSL predicted in the disorder-free, isotropic KAHF model.

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