Possible Dirac quantum spin liquid in the kagome quantum antiferromagnet
$\text{YC}_{3}(\text{OH})_{6}\text{Br}_{2}[\text{Br}_x(\text{OH})_{1-x}]$

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We studied the magnetic properties of $\text{YC}_{3}(\text{OH})_{6}\text{Br}_{2}[\text{Br}_x(\text{OH})_{1-x}]$ ($x = 0.33$), where $\text{Cu}^{2+}$ ions form two-dimensional kagome layers. There is no magnetic order down to 50 mK while the Curie-Weiss temperature is on the order of -100 K. At zero magnetic field, the low-temperature specific heat shows a $T^5$ dependence. Above 2 T, a linear temperature dependence term in specific heat emerges, and the value of $\gamma = C/T$ increases linearly with the field. Furthermore, the magnetic susceptibility tends to a constant value at $T = 0$. Our results suggest that the magnetic ground state of $\text{YC}_{3}(\text{OH})_{6}\text{Br}_{2}[\text{Br}_x(\text{OH})_{1-x}]$ is consistent with a Dirac quantum-spin-liquid state with a linearly dispersing spinon strongly coupled to an emergent gauge field, which has long been theoretically proposed as a candidate ground state in the two-dimensional kagome Heisenberg antiferromagnetic system.

The two-dimensional (2D) kagome Heisenberg antiferromagnet (KHA) has stimulated great research interest and activities [1-8]. This is because the Lieb-Schultz-Mattis-Oshikawa-Hastings theorems [9,11] forbid KHA systems from having a trivially gapped ground state and therefore increase the possibilities of realizing exotic states such as quantum spin liquids (QSLs) and valence bond solids [7,12-16]. If we consider only the homogeneous nearest-neighbor exchange, the leading candidates for the ground states are gapped $Z_2$ QSL [17-21] and gapless $U(1)$ Dirac QSL [22-27]. Introducing second-neighbor and third-neighbor exchanges or more generic interactions will result in even richer ground states [25-34]. Given these varieties, it is generally expected that a kagome magnetic material exhibiting no magnetic ordering at low temperatures is more likely to acquire a QSL ground state.

Experimentally, the best candidate so far for a kagome QSL is undoubtedly herbertsmithite $[\text{ZnCu}_3(\text{OH})_6\text{Cl}_2]$ [35,36], which consists of perfect kagome planes formed by $\text{Cu}^{2+}$ ions ($S = 1/2$). The central debate for this material, if the ground state is indeed a QSL, is whether the spin system is gapped or gapless [37,39]. While this is supposed to be easy to observe experimentally, the existence of a few percent of magnetic $\text{Cu}^{2+}$ ions on the nonmagnetic Zn$^{2+}$ sites makes the low-energy spectrum and low-temperature thermodynamical properties dominated by these impurity spins [40,42]. Similar issues have also been found for many other KHAs [43,48]. To avoid site disorder, $\text{YC}_{3}(\text{OH})_{6}\text{Br}_{2}[\text{Br}_x(\text{OH})_{1-x}]$ is consistently with a Dirac quantum-spin-liquid state with a linearly dispersing spinon strongly coupled to an emergent gauge field, which has long been theoretically proposed as a candidate ground state in the two-dimensional kagome Heisenberg antiferromagnetic system.
magnetic impurities are found, and their effects on the thermodynamical properties are negligible. The system shows no magnetic ordering down to 50 mK despite the large Curie-Weiss temperature (∼−79 K). The low-temperature specific heat $C$ is proportional to $T^2$ below 0.7 K, while the field-induced $T$-linear component of $C$ is proportional to the magnetic field. These results are consistent with the theoretical expectation of a Dirac QSL [22].

Single crystals of YCu$_3$(OH)$_6$Br$_2$[Br$_2$(OH)$_{1−x}$] (YCu$_3$-H) were grown using the hydrothermal method reported previously [54]. The crystals are hexagonal plates with an in-plane diameter of 0.5 to 1 mm and a thickness of 0.1 to 0.3 mm. All the crystals were ultrasonically cleaned in water before measurements were taken to remove possible impurities attached to the surfaces of the crystals. The deuterated single crystals (YCu$_3$-D) were synthesized using the same method with the corresponding deuterated starting materials and heavy water. All the results are for YCu$_3$-H if not otherwise mentioned. The crystal structure and chemical formula were determined by single-crystal x-ray diffraction (SCXRD). Specific-heat and magnetic-susceptibility measurements were measured on physical property measurement systems (Quantum Design) and magnetic property measurement systems (Quantum Design), respectively. To obtain a good enough signal, we typically used either c-axis-aligned or randomly oriented crystals in these measurements.

Figure 1(a) shows the crystal structure of YCu$_3$-H ($x = 0.33$), with detailed information given in Table I, which was obtained from SCXRD. As reported previously for the $x = 0.51$ sample [64], the Cu$^{2+}$ ions form 2D kagome layers [Fig. 1(b)]. Compared to the $x = 0.51$ sample, the occupancy of Y11 increases from 10% to 30% while that of Br2 decreases from 51% to 33%. Moreover, the in-plane and c-axis lattice constants become slightly larger and smaller, respectively, and the Cu-O1-Cu angle for our sample (115.81°) is larger than that for the $x = 0.51$ sample (114.08°). For YCu$_3$-D ($x = 0.32$), the results are almost the same, as shown in Table II. We note that the molecular formula for our sample may be roughly written as Y$_2$Cu$_3$(OH)$_6$Br$_2$, but no superstructure as reported in Refs. [53, 55] has been found here.

Figure 1(c) shows the inverse magnetic susceptibility $\chi^{-1} = H/M$ of YCu$_3$-H as a function of the temperature for the field parallel to the ab plane. A linear fit to the data above 150 K gives the Curie-Weiss temperature $\theta_{CW}$ and effective moment $\mu_{eff}$ as about −79 K and 1.94 $\mu_B$, respectively, which are similar to those for the $x = 0.51$ sample [54]. It should be noted that these values will change when adding a temperature-independent background as a fitting parameter in the Curie-Weiss

### Table I. Fractional atomic coordinates and equivalent isotropic displacement parameters of YCu$_3$(OH)$_6$[Br$_2$(OH)$_{1−x}$] with space group $P\overline{3}m1$ (No. 164): $a = b = 6.6784(2)$Å, $c = 5.9901(3)$Å, $\alpha = \beta = 90^\circ$, $\gamma = 120^\circ$.

| Atom     | $x$     | $y$     | $z$     | Occupancy | $U_{eq}$ (Å$^2$) |
|----------|---------|---------|---------|-----------|-----------------|
| Y11      | 0.0000  | 0.0000  | 0.5000  | 0.301(2)  | 0.0107(4)       |
| Y12      | 0.0000  | 0.0000  | 0.6222(3)| 0.343(12)| 0.0107(4)       |
| Cu       | 0.5000  | 0.5000  | 0.5000  | 1         | 0.0127(3)       |
| Br1      | 0.666667| 0.333333| 0.85645(10)| 1        | 0.0179(3)       |
| O1       | 0.1888(3)| 0.8112(3)| 0.6288(7) | 1        | 0.0298(8)       |
| Br2      | 0.0000  | 0.0000  | 0.0000  | 0.326(6)  | 0.0207(8)       |
| O2       | 0.0000  | 0.0000  | 0.0000  | 0.674(6)  | 0.0207(8)       |

### Table II. Fractional atomic coordinates and equivalent isotropic displacement parameters of YCu$_3$(OD)$_6$[Br$_2$(OH)$_{1−x}$] with the space group of $P\overline{3}m1$ (No. 164): $a = b = 6.6779(3)$Å, $c = 5.9874(4)$Å, $\alpha = \beta = 90^\circ$, $\gamma = 120^\circ$.

| Atom     | $x$     | $y$     | $z$     | Occupancy | $U_{eq}$ (Å$^2$) |
|----------|---------|---------|---------|-----------|-----------------|
| Y11      | 0.0000  | 0.0000  | 0.5000  | 0.314(2)  | 0.0076(4)       |
| Y12      | 0.0000  | 0.0000  | 0.6227(3)| 0.3428(12)| 0.0076(4)       |
| Cu       | 0.5000  | 0.5000  | 0.5000  | 1         | 0.0099(3)       |
| Br1      | 0.666667| 0.333333| 0.85614(10)| 1        | 0.048(3)        |
| O1       | 0.1890(7)| 0.8110(3)| 0.6298(7) | 1        | 0.0267(8)       |
| Br2      | 0.0000  | 0.0000  | 0.0000  | 0.317(8)  | 0.0152(10)      |
| O2       | 0.0000  | 0.0000  | 0.0000  | 0.683(8)  | 0.0152(10)      |
Figure 2(a) shows the temperature dependence of the specific heat, where no magnetic ordering is found down to 50 mK. A shoulder appears at about 2 K, which is typical for a KHA from numerical calculations [56]. One-to-five-mK anomalies (or a linear function, i.e., $\alpha T^2$, with $\alpha$ being a fitting parameter. We find that no gap function can describe these anisotropy fields, where the nuclear Schottky anomaly becomes less significant. The linear temperature dependence of $C/T$ can thus be directly seen without the need to remove the Schottky contribution, therefore avoiding uncertainties at large fields. Interestingly, the slope clearly decreases with increasing field, which is different from that of YCu3-H. But for both the YCu3-H and YCu3-D samples, we can fit the $C/T$ data with a generic linear function, i.e., $C/T = \gamma + \alpha T$, and show the obtained field dependence of $\gamma$ in Fig. 2(d). We see that above 2 T, $\gamma$ for both samples increases linearly with the field, showing a linear temperature dependence of the specific heat at high fields. This is the second experimental evidence that the material exhibits consistent behavior of a Dirac QSL under magnetic field at low temperature, i.e., $k_B T \ll \mu_B B$ [22].

Figure 3(a) shows the $M - H$ loop for YCu3-H under magnetic field [40]. Below about 0.7 K, $C/T$ shows a linear temperature dependence at all fields with little change in the slope.

Figure 3(c) further shows the results for the YCu3-D sample, where the nuclear Schottky anomaly becomes less significant. The linear temperature dependence of $C/T$ can thus be directly seen without the need to remove the Schottky contribution, therefore avoiding the uncertainties at large fields. Interestingly, the slope clearly decreases with increasing field, which is different from that of YCu3-H. But for both the YCu3-H and YCu3-D samples, we can fit the $C/T$ data with a generic linear function, i.e., $C/T = \gamma + \alpha T$, and show the obtained field dependence of $\gamma$ in Fig. 2(d). We see that above 2 T, $\gamma$ for both samples increases linearly with the field, showing a linear temperature dependence of the specific heat at high fields. This is the second experimental evidence that the material exhibits consistent behavior of a Dirac QSL under magnetic field at low temperature, i.e., $k_B T \ll \mu_B B$ [22].

Figure 3(b) shows the $M - H$ loop for YCu3-D, where no hysteresis is found at all temperatures. At 2 K, the slope $dM/dH$ decreases with increasing field and becomes almost field independent above about 5 T. At 15 K, $M$ linearly depends on $H$ for the whole field range. Similar behaviors are also observed for $H//ab$, as shown in Fig. 3(b). The anisotropy $M_i/M_{ab}$ between $H//c$ and $H//ab$ at high fields is about 1.07 at 2 K and slightly increases with increasing temperature ( $\sim 1.13$ at 15 K).

To further investigate the magnetic susceptibility at lower temperatures, we studied the temperature depen-
ence of the magnetization below 2 K, as shown in Fig. 3(c). No divergent behavior is found, suggesting the absence of free or weakly correlated spins. A slight difference between the zero-field-cooling (ZFC) and field-cooling (FC) processes appears below about 0.8 K. The ratio between this difference $\Delta M$ and the mean value of $M$ is less than 0.6%, suggesting it may not come from the intrinsic properties of the samples. In fact, the measurements of the unwashed YCu3-H samples reveal similar behaviors with $\Delta M/M > 6\%$, strongly demonstrating that the difference between ZFC and FC come from the external magnetic impurities, most of which were attached to the crystal surfaces. Overall, the intrinsic magnetic susceptibility of our samples should tend to a constant when $T$ goes to zero.

The above results demonstrate that the YCu3(OH)$_6$Br$_2$[Br$_x$(OH)$_{1-x}$] system indeed has few magnetic impurities, as expected [54]. As shown in the Supplemental Material [55], the magnetic impurities in our samples are mostly attached to the sample surfaces and can be removed by ultrasonic washing in water. For the washed samples, the existence of a very small amount of impurities has negligible effects on determining the thermodynamical properties as shown by both the specific-heat and magnetic-susceptibility results. This is different from herbertsmithite and many other KHA [40–45, 47, 48], for which the effect of magnetic impurities is very hard to separate from the bulk properties.

We thus conclude that YCu3(OH)$_6$Br$_2$[Br$_x$(OH)$_{1-x}$] is a strong candidate for realizing the Dirac QSL state, where the low-energy spinons form Dirac cones and their interactions are mediated by the emergent gauge fields [22, 23, 57, 58]. The most promising evidence comes from the specific-heat results. First, the specific heat shows a $T^2$ dependence at zero field, which has been predicted for a U(1) Dirac QSL because of the Dirac nodes [22]. Second, the linear temperature dependence of the specific heat is found at high fields, suggesting the appearance of the spinon Fermi surface, which has also been predicted theoretically [22]. It is interesting to note that this linear component shows up only above 2 T, which is again consistent with the condition for the above theoretical argument, i.e., $k_BT \ll \mu_B B$. Moreover, the linear-field-dependence and quadratic-temperature-dependence coefficients for YCu3-H are 0.01 J/mol TK$^2$ and 0.33 J/mol K$^3$, respectively, which gives a ratio that is just about 1/7 of the theoretical predicted value ($\sim 0.21$ T$^{-1}$) [22]. For YCu3-D, the ratio is similar. These results may give a hint of the detailed structure of the Dirac nodes and low-energy spinon excitations and could even imply some kind of gauge fluctuation. It is particularly interesting to point out that the low-temperature specific heats behave differently for the YCu3-H and YCu3-D samples, which suggests that the fine structures of Dirac cones may be tuned. It is also worth noting that although the magnetic susceptibility $\chi$ is expected to show a linear temperature dependence, it tends to a large constant when $T$ goes to zero for our sample. Theoretically, it has been shown that a non-zero $\chi$ at 0 K is indeed possible for a gapless QSL [59, 63]. Our observations therefore provide a promising experimental signature of the material realization of the highly non-trivial phase with emergent matter fields (Dirac spinon) coupled with gauge fields [22, 23, 57, 58, 64, 65], which has been pursued by broad communities ranging from quantum material to high energy.

It is interesting that a Dirac QSL may be realized in our sample but not in YCu3(OH)$_6$Cl$_3$, which exhibits an AFM order at low temperatures [61, 63]. This order is shown to be caused by the large Dzyaloshinskii–Moriya (DM) interaction, which also gives rise to a hump in $C/T$ at 16 K [66]. In our case, the hump is at about 1.5 K, which means that the DM interaction is either an order smaller or even absent since the hump could also come from the low-energy excitations for a KHA [66]. This is also consistent with the negligible magnetic anisotropy shown by the $M - H$ loops. The small DM interaction may be associated with the random distributions of Y and Br$_2$ atoms. We note that the YCu3(OH)$_6$Cl$_{3-x}$ ($x = 1/3$) system has been suggested to show no magnetic order [63], which may be connected to our samples with partial occupancy of Br$_2$ and two sites for Y, as shown in Tables I and II. However, no distortion for Cu is found for YCu3(OH)$_6$Br$_2$[Br$_x$(OH)$_{1-x}$].

In conclusion, we have shown that the magnetic ground state of the YCu3(OH)$_6$Br$_2$[Br$_x$(OH)$_{1-x}$] system may be a Dirac QSL on a 2D kagome lattice using the low-temperature specific-heat measurements. The very small number of magnetic impurities make it possible to directly compare theoretical and experimental results, helping us realize KHA models in a real material. Moreover, the fine structure of Dirac nodes may be tuned by site disorders, which needs to be further studied.

Note added. Recently, we noted that studies on single-crystal YCu3(OH(D))$_{0.5}$Br$_{2.5}$ have also been reported [67].

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[1] S. Sachdev, Kagomé and triangular-lattice Heisenberg antiferromagnets: Ordering from quantum fluctuations and quantum-disordered ground states with unconfined bosonic spinons. Phys. Rev. B 45, 12377 (1992)

[2] M. B. Hastings, Dirac structure, RVB, and Goldstone modes in the kagomé antiferromagnet. Phys. Rev. B 63, 014413 (2000)

[3] L. Balents, M. P. A. Fisher, and S. M. Girvin, Fractionalization in an easy-axis Kagome antiferromagnet, Phys. Rev. B 65, 224412 (2002)

[4] L. Balents, Spin liquids in frustrated magnets, Nature 464, 199 (2010)

[5] M. R. Norman, Colloquium: Herbstsmithite and the search for the quantum spin liquid, Rev. Mod. Phys. 88, 041002 (2016)

[6] L. Savary and L. Balents, Quantum spin liquids: A review, Rep. Prog. Phys. 80, 016502 (2017)

[7] Y. Zhou, K. Kanoda, and T.-K. Ng, Quantum spin liquid states, Rev. Mod. Phys. 89 (2017)

[8] C. Broholm, R. J. Cava, S. A. Kivelson, D. G. Nocera, M. R. Norman, and T. Senthil, Quantum spin liquids, Science 367, 263 (2020)

[9] E. Lieb, T. Schultz, and D. Mattis, Two soluble models of an antiferromagnetic chain, Annals of Physics 16, 407 (1961)

[10] M. Oshikawa, Commensurability, excitation gap, and topology in quantum many-particle systems on a periodic lattice, Phys. Rev. Lett. 84, 1535 (2000)

[11] M. B. Hastings, Lieb-Schultz-Mattis in higher dimensions, Phys. Rev. B 69, 104431 (2004)

[12] J. T. Chalker, P. C. W. Holdsworth, and E. F. Shender, Hidden order in a frustrated system: Properties of the heisenberg kagome antiferromagnet, Phys. Rev. Lett. 68, 855 (1992)

[13] I. Ritchey, P. Chandra, and P. Coleman, Spin folding in the two-dimensional Heisenberg kagome antiferromagnet, Phys. Rev. B 47, 15342 (1993)

[14] T. Yildirim and A. B. Harris, Magnetic structure and spin waves in the Kagome jasorite compound $KFe_3(SO_4)_2(OH)_6$. Phys. Rev. B 73, 214446 (2006)

[15] K. Matan, T. Ono, Y. Fukumoto, T. J. Sato, J. Yamaura, M. Yanoo, K. Morita, and H. Tanaka, Pinwheel valence-bond solid and triplet excitations in the two-dimensional deformed kagome lattice, Nature Physics 6, 865 (2010)

[16] R. W. Smaha, W. He, J. M. Jiang, J. Wen, Y.-F. Jiang, J. P. Sheckelton, C. J. Titus, S. G. Wang, Y.-S. Chen, S. J. Teat, A. A. Aczel, Y. Zhao, G. Xu, J. W. Lynn, H.-C. Jiang, and Y. S. Lee, Materializing rival ground states in the barlowite family of kagome magnets: quantum spin liquid, spin ordered, and valence bond crystal states, [np] Quantum Materials 5, 23 (2020)

[17] S. Yan, D. A. Huse, and S. R. White, Spin-liquid ground state of the $s = 1/2$ kagome Heisenberg antiferromagnet, Science 332, 1173 (2011)

[18] S. Depenbrock, I. P. McCulloch, and U. Schollwöck, Nature of the spin-liquid ground state of the $s = 1/2$ Heisenberg model on the kagome lattice, Phys. Rev. Lett. 109, 067201 (2012)

[19] H.-C. Jiang, Z. Wang, and L. Balents, Identifying topological order by entanglement entropy, Nat. Phys. 8, 902 (2012)

[20] F. Yang and H. Yao, Frustrated resonating valence bond states in two dimensions: Classification and short-range correlations, Phys. Rev. Lett. 109, 147209 (2012)

[21] J.-W. Mei, J.-Y. Chen, H. He, and X.-G. Wen, Gapped spin liquid with $Z_2$ topological order for the kagome Heisenberg model, Phys. Rev. B 95, 235107 (2017)

[22] Y. Ran, M. Hermele, P. A. Lee, and X.-G. Wen, Projected-wave-function study of the spin-1/2 kagome antiferromagnet on the kagome lattice, Phys. Rev. Lett. 98, 117205 (2007)

[23] M. Hermele, Y. Ran, P. A. Lee, and X.-G. Wen, Properties of an algebraic spin liquid on the kagome lattice, Phys. Rev. B 77, 224413 (2008)

[24] Y. Iqbal, F. Becca, and D. Poilblanc, Projected wave function study of $Z_2$ spin liquids on the kagome lattice for the spin-1/2 quantum Heisenberg antiferromagnet, Phys. Rev. B 84, 020407 (2011)

[25] Y. Iqbal, D. Poilblanc, and F. Becca, Vanishing spin gap in a competing spin-liquid phase in the kagome heisenberg antiferromagnet, Phys. Rev. B 89, 020407 (2014)

[26] Y.-C. He, M. P. Zaletel, M. Oshikawa, and F. Pollmann, Signatures of dirac cones in a dmrg study of the kagome heisenberg model, Phys. Rev. X 7, 031020 (2017)

[27] H. J. Liao, Z. Y. Xie, J. Chen, Z. Y. Liu, H. D. Xue, R. Z. Huang, B. Normand, and T. Xiang, Gapless spin-liquid ground state in the $s = 1/2$ kagome antiferromagnet, Phys. Rev. Lett. 118, 137202 (2017)

[28] Y.-C. He, D. N. Sheng, and Y. Chen, Chiral spin liquid in a frustrated anisotropic kagome heisenberg model, Phys. Rev. Lett. 112, 137202 (2014)

[29] S.-S. Gong, W. Zhu, L. Balents, and D. N. Sheng, Global phase diagram of competing ordered and quantum spin-liquid phases on the kagome lattice, Phys. Rev. B 91, 075112 (2015)

[30] S.-S. Gong, W. Zhu, K. Yang, O. A. Starykh, D. N. Sheng, and L. Balents, Emergent quasi-one-dimensionality in a kagome magnet: A simple route to complexity, Phys. Rev. B 94, 035154 (2016)

[31] Y.-C. Wang, X.-F. Zhang, F. Pollmann, M. Cheng, and Z. Y. Meng, Quantum spin liquid with even ising gauge field structure on kagome lattice, Phys. Rev. Lett. 121, 057202 (2018)

[32] G.-Y. Sun, Y.-C. Wang, C. Fang, Y. Qi, M. Cheng, and Z. Y. Meng, Dynamical signature of symmetry fractionalization in frustrated magnets, Phys. Rev. Lett. 121, 077201 (2018)

[33] C.-Y. Lee, B. Normand, and Y.-J. Kao, Gapless spin liquid in the kagome heisenberg antiferromagnet with dzyaloshinskii-moriya interactions, Phys. Rev. B 98, 224414 (2018)

[34] P. Prelovšek, M. Gomilšek, T. Arh, and A. Zorko, Dynamical spin correlations of the kagome antiferromagnet, Phys. Rev. B 103, 014431 (2021)

[35] M. P. Shores, E. A. Nytko, B. M. Bartlett, and D. G. Nocera, A structurally perfect $s = 1/2$ kagomi3 antiferromagnet, J. Am. Chem. Soc. 127, 13462 (2005)

[36] T. H. Han, J. S. Helton, S. Chu, D. G. Nocera, J. A. Rodriguez-Rivera, C. Broholm, and Y. S. Lee, Fraction-
alized excitations in the spin-liquid state of a kagome-lattice antiferromagnet. Nature 492, 406 (2012)

[37] M. Fu, T. Imai, T.-H. Han, and Y. S. Lee, Evidence for a gapped spin-liquid ground state in a kagome heisenberg antiferromagnet. Science 350, 655 (2015)

[38] T.-H. Han, M. R. Norman, J.-J. Wen, J. A. Rodriguez-Rivera, J. S. Helton, C. Broholm, and Y. S. Lee, Correlated impurities and intrinsic spin-liquid physics in the kagome material herbertsmithite. Phys. Rev. B 94, 060409 (2016)

[39] P. Khuntia, M. Velazquez, Q. Barthélemy, F. Bert, E. Kermarrec, A. Legros, B. Bernu, A. Z. L. Messio, and P. Mendels, Gapless ground state in the archetypal quantum kagome antiferromagnet ZnCu$_3$(OH)$_6$Cl$_2$. Nat. Phys. 16, 469 (2020)

[40] M. A. de Vries, K. V. Kamenev, W. A. Kockelmann, J. Sanchez-Benitez, and A. Harrison, Magnetic ground state of an experimental $S = 1/2$ kagome antiferromagnet. Phys. Rev. Lett. 100, 157205 (2008)

[41] D. E. Freedman, T. H. Han, A. Prodi, P. Müller, Q.-Z. Huang, Y.-S. Chen, S. M. Webb, Y. S. Lee, T. M. McQueen, and D. G. Nocera, Site specific x-ray anomalous dispersion of the geometrically frustrated kagomé magnet, herbertsmithite, ZnCu$_3$(OH)$_6$Cl$_2$. J. Am. Chem. Soc. 132, 16185 (2010)

[42] Y. Y. Huang, Y. Xu, L. Wang, C. C. Zhao, C. P. Tu, J. M. Ni, L. S. Wang, B. L. Pan, Y. Fu, Z. Hao, C. Liu, J.-W. Mei, and S. Y. Li, Heat Transport in Herbertsmithite: Can a Quantum Spin Liquid Survive Disorder? arXiv e-prints, arXiv:2105.14749 (2021), arXiv:2105.14749 [cond-mat.str-el]

[43] E. Kermarrec, P. Mendels, F. Bert, R. H. Colman, A. S. Wills, P. Strobel, P. Bonville, A. Hillier, and A. Amato, Spin-liquid ground state in the frustrated kagome antiferromagnet MgCu$_3$(OH)$_6$Cl$_2$. Phys. Rev. B 84, 100401 (2011)

[44] Y. Li, B. Pan, S. Li, W. Tong, L. Ling, Z. Yang, J. Wang, Z. Chen, Z. Wu, and Q. Zhang, Gapless quantum spin liquid in the $S = 1/2$ anisotropic kagomé antiferromagnet ZnCu$_3$(OH)$_6$SO$_4$. New J. Phys. 16, 093011 (2014)

[45] Z. Feng, Z. Li, X. Meng, W. Yi, Y. Wei, J. Zhang, Y.-C. Wang, W. Jiang, Z. Liu, S. Li, F. Liu, J. Luo, S. Li, G. qing Zheng, Z. Y. Meng, J.-W. Mei, and Y. Shi, Gapped spin-1/2 spinon excitations in a new kagome quantum spin liquid compound Cu$_3$Zn(OH)$_6$FBr. Chin. Phys. Lett. 34, 075702 (2017)

[46] Y. Wei, Z. Feng, W. Lohstroh, D. H. Yu, D. Le, C. dela Cruz, W. Yi, Z. F. Ding, J. Zhang, C. Tan, L. Shu, Y.-C. Wang, H.-Q. Wu, J. Luo, J.-W. Mei, F. Yang, X.-L. Sheng, W. Li, Y. Qi, Z. Y. Meng, Y. Shi, and S. Li, Evidence for the topological order in a kagome antiferromagnet, arXiv e-prints, arXiv:1710.02991 (2017), arXiv:1710.02991 [cond-mat.str-el]

[47] Z. Feng, W. Yi, K. Zhu, Y. Wei, S. Miao, J. Ma, J. Luo, S. Li, Z. Y. Meng, and Y. Shi, From clarbingullite to a new spin liquid candidate Cu$_3$Zn(OH)$_6$FCl. Chin. Phys. Lett. 36, 017502 (2018)

[48] Y. Fu, M.-L. Lin, L. Wang, Q. Liu, L. Huang, W. Jiang, Z. Hao, C. Liu, H. Zhang, X. Shi, J. Zhang, J. Dai, D. Yu, F. Ye, P. A. Lee, P.-H. Tan, and J.-W. Mei, Dynamic fingerprint of fractionalized excitations in single-crystalline Cu$_3$Zn(OH)$_6$FBr. Nature Communications 12, 3048 (2021)

[49] W. Sun, Y.-X. Huang, S. Nokhrin, Y. Pan, and J.-X. Mi, Perfect kagomé lattices in YCu$_3$(OH)$_6$Cl$_3$: A new candidate for the quantum spin liquid state. J. Mater. Chem. C 4, 8772 (2016)

[50] P. Puphal, M. Bolte, D. Sheptyakov, A. Pustogow, K. Klimt, M. Dressel, M. Baenitz, and C. Krellner, Strong magnetic frustration in Y$_3$Cu$_9$(OH)$_6$Cl$_3$: a distorted kagome antiferromagnet. J. Mater. Chem. C 5, 2629 (2017)

[51] A. Zorko, M. Pregelj, M. Klanjšek, M. Gomilšek, Z. Jagličič, J. S. Lord, J. A. T. Verezhak, T. Shang, W. Sun, and J.-X. Mi, Coexistence of magnetic order and persistent spin dynamics in a quantum kagome antiferromagnet with no intersite mixing. Phys. Rev. B 99, 214441 (2019)

[52] A. Zorko, M. Pregelj, M. Gomilšek, M. Klanjšek, O. Zashiko, W. Sun, and J.-X. Mi, Negative-vector-chirality $120^\circ$ spin structure in the defect- and distortion-free quantum kagome antiferromagnet YCu$_3$(OH)$_6$Cl$_3$. Phys. Rev. B 100, 144420 (2019)

[53] Q. Barthélemy, P. Puphal, K. M. Zoch, C. Krellner, H. Luetkens, C. Baines, D. Sheptyakov, E. Kermarrec, P. Mendels, and F. Bert, Local study of the insulating quantum antiferromagnets YCu$_3$(OH)$_6$Cl$_3$; $x=0, 1/3$. Phys. Rev. Materials 3, 074401 (2019)

[54] X.-H. Chen, Y.-X. Huang, Y. Pan, and J.-X. Mia, Quantum spin liquid candidate YCu$_3$(OH)$_6$Br$_2$[Br$_2$(OH)$_4$]; $x \approx 0.51$: With an almost perfect kagomé layer. J. Magn. Magn. Mater. 512, 160666 (2020)

[55] See supplementary materials.

[56] J. Schnack, J. Schublenz, and J. Richter, Magnetism of the $N = 42$ kagome lattice antiferromagnet, Phys. Rev. B 98, 094423 (2018)

[57] D. H. Kim, P. A. Lee, and X.-G. Wen, Massless dirac fermions, gauge fields, and underdoped cuprates, Phys. Rev. Lett. 79, 2109 (1997)

[58] X. Y. Xu, Y. Qi, L. Zhang, F. F. Assaad, C. Xu, and Z. Y. Meng, Monte carlo study of lattice compact quantum electrodynamics with fermionic matter: The parent state of quantum phases, Phys. Rev. X 9, 021022 (2019)

[59] M. E. Zhitomirsky, Field-induced transitions in a kagomé antiferromagnet. Phys. Rev. Lett. 88, 057204 (2002)

[60] Y. Zhou, P. A. Lee, T.-K. Ng, and F.-C. Zhang, Na$_4$Ir$_3$O$_8$ as a 3D spin liquid with fermionic spinons, Phys. Rev. Lett. 101, 197201 (2008)

[61] T. Sakaia and H. Nakano, Gapless spin excitations in the $s = 1/2$ kagome- and triangular-lattice heisenberg antiferromagnets, Physica B 536, 85 (2018)

[62] X. Chen, S.-J. Ran, T. Liu, C. Peng, Y.-Z. Huang, and G. Su, Thermodynamics of spin-1/2 kagomé heisenberg antiferromagnet: algebraic paramagnetic liquid and finite-temperature phase diagram, Sci. Bull. 63, 1545 (2018)

[63] B. Bernu, L. Pierre, K. Essafi, and L. Messio, Effect of perturbations on the kagome $s = \frac{1}{2}$ antiferromagnet at all temperatures, Phys. Rev. B 101, 140403 (2020)

[64] F. F. Assaad and T. Grover, Simple fermionic model of deconfined phases and phase transitions, Phys. Rev. X 6, 041049 (2016)

[65] S. Gazit, M. Randeria, and A. Vishwanath, Emergent Dirac fermions and broken symmetries in confined and deconfined phases of $Z_2$ gauge theories, Nat. Phys. 13, 484 (2017)

[66] T. Arh, M. Gomilšek, P. Prelovšek, M. Pregelj,
M. Klanjšek, A. Ozarowski, S. J. Clark, T. Lancaster, W. Sun, J.-X. Mi, and A. Zorko, Origin of magnetic ordering in a structurally perfect quantum kagome antiferromagnet, Phys. Rev. Lett. 125, 027203 (2020).

[67] J. Liu, L. Yuan, X. Li, B. Li, K. Zhao, H. Liao, and Y. Li, Gapless spin liquid behavior in a kagome heisenberg antiferromagnet with randomly distributed hexagons of alternate bonds, Phys. Rev. B 105, 024418 (2022).