Emergent Spin-Gapped Magnetization Plateaus in a Spin-1/2 Perfect Kagome Antiferromagnet

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The two-dimensional spin-1/2 kagome Heisenberg antiferromagnet is believed to host quantum spin liquid (QSL) states with no magnetic order, but its ground state remains largely elusive. An important outstanding question concerns the presence or absence of the 1/9 magnetization plateau, where exotic quantum states, including topological ones, are expected to emerge. Here we report the magnetization of a recently discovered kagome QSL candidate $YCu_3(OH)_{6.5}Br_{2.5}$ up to 57 T. Above 50 T, a clear magnetization plateau at 1/3 of the saturation moment of Cu^{2+} ions is observed, supporting that this material provides an ideal platform for the kagome Heisenberg antiferromagnet. Remarkably, we found another magnetization plateau around 20 T, which is attributed to the 1/9 plateau. The temperature dependence of this plateau reveals the presence of the spin gap. The observation of 1/9 and 1/3 plateaus highlights the emergence of novel states in quantum spin systems.

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Frustrated spin systems offer a rich platform for exotic quantum many-body states, originating from competing interactions and quantum fluctuations. Among such systems, QSLs [1,2] are the most fascinating states, which are one of the most entangled quantum states conceived to date. It is widely believed that long-range quantum entanglement leads to many amazing emergent phenomena, such as fractionalized excitations and topological orders. The spin-1/2 Heisenberg antiferromagnet (AFM) on a 2D kagome lattice serves as a prime example for the QSL. However, despite decades of tremendous research, understanding the nature of the kagome AFM has proved to be one of the most vexing issues in the quantum spin systems.

There are several crucially important but unresolved issues for elucidating the ground state properties of the 2D kagome system. Among them, whether the ground state in zero field is gapped or gapless has been highly controversial [3–9]. The ground state in an external magnetic field has also been largely elusive both theoretically and experimentally. Of particular interest is the magnetization plateaus arising from a field-induced spin gap [10–16]. Specifically, it has been discussed theoretically that in an external magnetic field, the kagome AFM may exhibit a series of spin-gapped phases with magnetization plateaus at 1/9, 1/3, 5/9, and 7/9 of the saturation moment.

Theoretically, some or all of these plateaus are expected to appear as a result of quantum entanglement, rather than simple energetics of classical spins. The 1/3 plateau in the kagome AFM, which is found rather robustly regardless of the theoretical methods used [10–16], has been suggested to appear purely due to quantum mechanical effects, unlike the one of classical origin known for the triangular AFM [17]. In stark contrast to the 1/3 plateau, as for the 1/9 plateau, even its existence is a nontrivial problem. The 1/9 plateau state, if it exists, is expected to be a highly unusual quantum state including a QSL with a topological order [10].

Experimental verification of whether such magnetization plateaus really exist should be the key to exploring the enigmatic ground state phases of the spin-1/2 kagome AFM. However, its elucidation remains a significant challenge because there are no ideal candidate materials for such a system with a QSL ground state. Thus, we may safely conclude that even the existence of the plateaus is still open.

Until now, there are only a few candidate materials for the spin-1/2 kagome AFM. Among them, herbertsmithite $ZnCu_3(OH)_6Cl_2$ [18,19] has been most extensively studied as a canonical candidate for bearing a QSL ground state, because it possesses a perfect kagome structure and does not magnetically order down to 50 mK [20,21]. The spinon continuum has also been reported by neutron scattering experiments [22], supporting the spin fractionalization. However, the observation of the magnetization plateaus in herbertsmithite has been prevented by multiple factors. First, the intrinsic magnetic properties are significantly influenced by orphan spins [23–29] introduced by inevitable

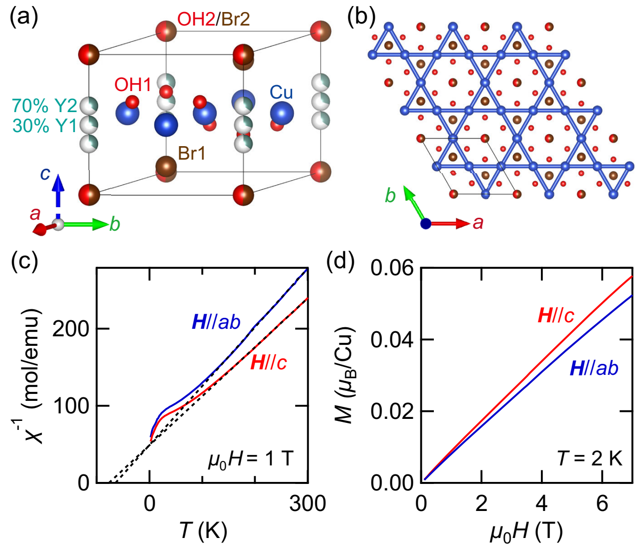

FIG. 1. Crystal structure and magnetic properties of YCOB. (a) Crystal structure of YCOB. The intersite mixing of OH2 and Br2 displaces 70% of Y^{3+} ions from the optimal Y1 position on the kagome plane. (b) The kagome plane of Cu^{2+} in YCOB. YCOB consists of a 2D perfect kagome lattice of Cu^{2+} ions without antisite mixing by other nonmagnetic ions. (c) Temperature dependence of the inverse of the magnetic susceptibility χ^{-1} at 1 T for both $\boldsymbol{H}||c$ and $\boldsymbol{H}||ab$. Linear fits of the high temperature data are denoted by dashed lines. The large negative Curie-Weiss temperatures $\theta_c = -77$ and $\theta_{ab} = -65$ K indicate a predominant antiferromagnetic interaction. (d) Field dependence of magnetization M_{ab} and M_c at 2 K up to 7 T. The small anisotropy $M_c/M_{ab} = 1.1$ is observed.

replacements of Cu^{2+} and Zn^{2+} ions in and between the 2D kagome layers [30,31]. In addition, relatively large exchange interaction in the 2D kagome layers makes it difficult to access the field required to observe the plateaus. In herbertsmithite, an anomaly near 1/3 magnetization has been claimed near 150 T [32], but drawing a definite conclusion is difficult owing to the limitation of experimental resolution. The presence of the 1/3 magnetization plateau has been reported for other kagome AFMs such as volborthite $Cu_3V_2O_7(OH)_2 \cdot 2H_2O$ [33], Cd-kapellasite $CdCu_3(OH)_6(NO_3)_2 \cdot H_2O$ [14], and $Cs_2ATi_3F_{12}$ ($A =$ Li, Na, K) [34,35], but the distortion of a kagome lattice or additional magnetic interactions causing a long-range order may mask the intrinsic nature of the ideal kagome AFM. No signature of the 1/9 plateau has been observed: not only in kagome AFMs but also in any insulating spin-1/2 quantum AFMs.

Recently, a promising candidate of spin-1/2 kagome AFM $YCu_3(OH)_{6.5}Br_{2.5}$ (YCOB) [36–40] has been discovered. YCOB consists of a 2D perfect kagome layer of Cu^{2+} ions [Figs. 1(a) and 1(b)]. Despite large exchange interaction J of 40–80 K, no magnetic order has been observed down to 50 mK [38]. The intersite mixing of OH^- and Br^- pushes 70% of Y^{3+} ions away from their ideal position, introducing bond randomness in the exchange interaction [37]. However, the antisite disorder between magnetic Cu^{2+} and nonmagnetic Y^{3+} is absent due to the very different ionic radii, retaining the perfect kagome lattice of Cu^{2+} intact and free from orphan spins.

In this Letter, we investigate the magnetization process of YCOB up to 57 T. We observe a pronounced magnetization plateau at 1/3 of the Cu^{2+} saturation moment, which is consistent with the expected properties of kagome Heisenberg AFM. The most prominent feature is a distinct plateau at 1/9 of the saturation moment. Furthermore, the temperature dependence of the magnetization curve reveals an opening of the spin gap at the 1/9 plateau. These demonstrate the emergence of unique quantum states in this kagome AFM.

Single crystals of YCOB were grown by a hydrothermal method as reported previously [37]. Multiple single crystals, aligned along the crystal c axis, were collected for magnetization studies. Magnetization experiments up to 7 T were conducted by a magnetic property measurement system (MPMS). For high field magnetization experiments, magnetic fields up to 57 T were generated using a nondestructive pulse magnet installed at the International MegaGauss Science Laboratory of the Institute for Solid State Physics in the University of Tokyo. Magnetization in pulsed fields was measured by the standard induction method using coaxial pick-up coils. The absolute values of the magnetization were calibrated by the low field data measured by MPMS.

Figure 1(c) depicts the temperature (T) dependence of the inverse of the magnetic susceptibility χ_c^{-1} and χ_{ab}^{-1} at $\mu_0H =$ 1 T for $\boldsymbol{H}||c$ and $\boldsymbol{H}||ab$, respectively. At high temperatures where k_BT well exceeds J, both χ_c^{-1} and χ_{ab}^{-1} increase linearly with T. Linear fits of the high temperature data for $T \geq 150$ K (dashed lines) give the Curie-Weiss temperatures $\theta_c = -77$ K and $\theta_{ab} = -65$ K, indicating a predominant antiferromagnetic interaction. We fit the experimental data with numerical calculations for the ideal kagome Heisenberg AFM [41–43], yielding values of J ranging from 65 to 77 K (see Supplemental Material [44]). Moreover, we analyze the experimental results using calculations incorporating Dzyaloshinskii-Moriya (DM) interaction, second nearest neighbor interaction, and Ising anisotropy [41]. The best fits are obtained for J ranging from 57 to 65 K, close to the values obtained for the ideal kagome Heisenberg AFM. As shown in Fig. 1(d), the field dependence of magnetization M exhibits a small anisotropy $M_c/M_{ab} = 1.1$ (see Supplemental Material [44]).

Figure 2(a) displays the field (H) dependence of M_c at several temperatures. The field derivative of the data dM_c/dH is also shown in Fig. 2(b). We first discuss the high field feature above 50 T highlighted by a dark gray region. At the lowest temperature of 0.6 K, M_c flattens out with H above 50 T, indicating the appearance of a magnetization plateau. The plateau is also confirmed by the sharp reduction of dM_c/dH, which almost vanishes

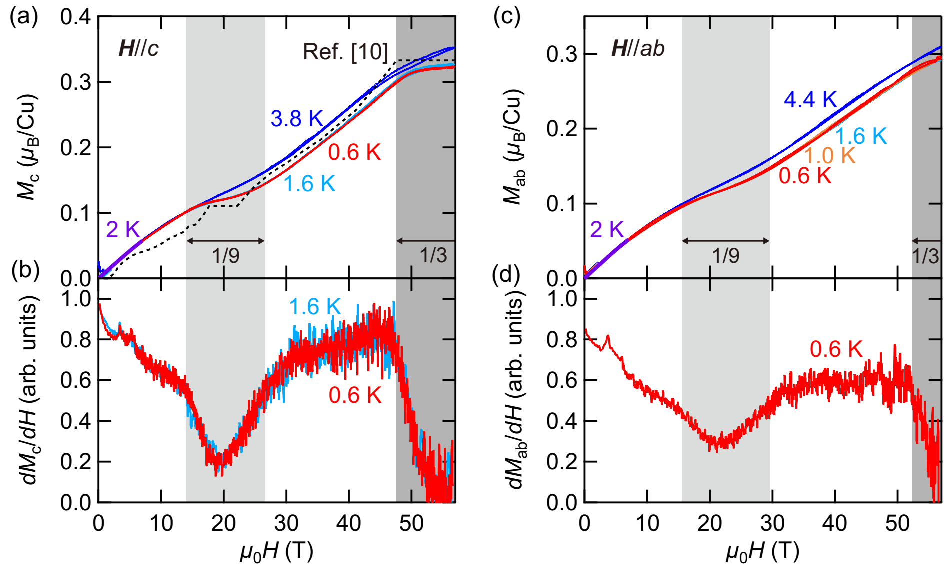

FIG. 2. Magnetization process of YCOB up to 57 T. (a) Magnetization process at 0.6, 1.6, and 3.8 K for $\boldsymbol{H}||c$. At 0.6 K, distinct magnetization plateaus at 1/9 and 1/3 of the Cu^{2+} saturation moment are observed, as highlighted by light and dark gray regions, respectively. The magnetization curve at 0.6 K nearly perfectly overlaps with that at 1.6 K in both plateau regions, which provides evidence for the formation of the spin gap. For comparison, we also plot the magnetization process obtained by numerical calculations from Fig. 1 in Ref. [10]. The best fit $J = 37$ K is obtained by adjusting the center of the 1/9 plateau for the experimental results and the numerical calculations. (b) The field dependence of the derivative dM_c/dH at 0.6 and 1.6 K. dM_c/dH is significantly reduced within the 1/9 and 1/3 plateaus regions. (c) Magnetization process at 0.6, 1.0, 1.6 K, and 3.8 K for $\boldsymbol{H}||ab$. Both 1/9 and 1/3 magnetization plateaus are less pronounced for M_{ab} compared to M_c. (d) The field dependence of the derivative dM_{ab}/dH at 0.6 K. The absolute values of M_c and M_{ab} were calibrated by the MPMS data at 2 K (purple line) in (a) and (c).

around 55 T. Notably, M_c at the plateau is very close to 1/3 of the fully polarized value ($1\mu_B$/Cu), demonstrating the presence of the 1/3 magnetization plateau. This result provides the first compelling evidence for the 1/3 plateau in the kagome AFM candidate with no magnetic order, revealing that this system serves as a good platform for the quest of spin-1/2 kagome Heisenberg AFM.

We now focus on the magnetization behavior below 50 T. The most salient feature is that the slope of the magnetization flattens out around 20 T at low temperatures. This is also evident from the strong reduction of dM_c/dH in Fig. 2(b). Around 20 T, dM_c/dH shows deep minima, approaching zero. However, unlike to the 1/3 plateau, it does not completely fall to zero, indicating that the magnetization curve has a small but finite slope. We will discuss the possible origins of this later. Remarkably, the absolute value of M_c at 20 T is close to one-third of that of the 1/3 plateau, i.e., 1/9 of the fully polarized value. We therefore conclude the emergence of 1/9 plateau around 20 T.

Figures 2(c) and 2(d) depict the H dependence of M_{ab} and dM_{ab}/dH, respectively. Again, both 1/9 and 1/3 magnetization plateaus are observed in M_{ab} around 20 and above 50 T, respectively, as evident from the reductions of dM_{ab}/dH. The plateaus of the M_{ab} curve are less pronounced compared with those of M_c (see also Supplemental Material [44]). The magnetic fields at the center of the 1/9 plateau, determined by the minima of dM/dH, are 19.3 and 21.5 T for $\boldsymbol{H}||c$ and $\boldsymbol{H}||ab$, respectively. This nearly 10% difference is attributed to the anisotropy of the magnetization $M_c/M_{ab} = 1.1$ [see Fig. 1(d)].

Next we discuss the issue of a spin gap at the 1/9 plateau. Both magnetization curves M_c and M_{ab} at 1.6 K in the plateau regime nearly perfectly overlap with those taken at lower temperatures, as shown in Figs. 2(a) and 2(c). This implies the presence of spin gap. In fact, it has been shown theoretically that the magnetization plateau is temperature independent when the temperature is well below the spin gap energy and the magnetization curve overlaps well with that at $T = 0$ [16,45]. The presence of spin gap is further supported by the temperature dependence of $M_c(T)$ and $M_{ab}(T)$ in the plateau regime, as shown in Figs. 3(a) and 3(b), respectively. It is obvious that both $M_c(T)$ and $M_{ab}(T)$ are temperature independent at low temperatures, while they exhibit a sharp upturn at higher temperatures, suggesting the activation type behavior of the magnetization. It should be noted that, for a gapless Dirac spin liquid state [46], dM/dH in the 1/9 plateau region shows a strong temperature dependence, which is inconsistent with the present results. (see Supplemental Material [44]).

The temperature dependence of the magnetization at the 1/9 plateau indicates that the magnitude of the spin gap Δ_s is much larger than 1.6 K. It has been pointed out that the gap size is roughly estimated by the plateau width [47], since the spin gap is closed when the Zeeman energy $g\mu_B H$ reaches the gap size. The plateau width of ∼12 T [a light gray region in Figs. 2(a) and 2(b)] indicates the opening of the spin gap of $\Delta_s \sim 16$ K.

Here we compare the magnetization curves of YCOB with numerical calculations of the ideal kagome-Heisenberg AFM [10]. The fitting parameter is the nearest neighbor exchange constant J. The best fit is obtained by $J = 37$ K, as shown by the dashed line in Fig. 2(a). This value significantly deviates from the value of $J \sim 60$–80 K estimated from the temperature dependence of χ. This suggests the observed onset field of the $1/9$ plateau is lower than that for the ideal kagome Heisenberg AFM. Moreover, the observed width of the $1/9$ plateau is extended compared to that for the ideal kagome Heisenberg AFM. It has been proposed that these discrepancies originate from a moderate distribution of exchange interaction [48]. However, as randomly distributed exchange couplings tend to effectively reduce the field-induced gap by creating in-gap states, it is natural to expect that the randomness acts to reduce the width of the plateau, which is opposite to the scenario suggested in Ref. [48]. Instead, these discrepancies are most likely attributed to the presence of additional magnetic interactions, such as DM interactions that will be discussed below and next-nearest-neighbor exchange interaction in YCOB. Since it is challenging to quantitatively estimate the magnitude of these additional interactions at this stage, examining which specific interactions are responsible for these discrepancies is left for future study.

We next discuss the finite slope of the magnetization curve at the $1/9$ plateau. The perfect overlap of the magnetization curves for $T \leq 1.6$ K [Figs. 2(a) and 2(c)] indicates that the finite slope is intrinsic and not caused by the thermal broadening. In general, finite slopes of the magnetization plateaus appears in the presence of magnetic interactions that do not conserve the quantum number of spin angular momentum along the field direction. We point out that the observed finite slope appears as a result of the DM interaction that does not conserve the spin angular momentum parallel to the magnetic field. In fact, the finite slope of the magnetic plateaus has also been reported in the Shastry-Sutherland magnet $SrCu_2(BO_3)_2$ [49,50], which has been attributed to the DM interaction. It should be noted that the presence of the DM interaction has been reported in the related material $YCu_3(OH)_6Cl_3$ [51]. In addition, both $1/9$ and $1/3$ magnetization plateaus are less pronounced in M_{ab} compared to M_c (Fig. 2), suggesting the presence of anisotropic interaction. It is well known that the DM interaction induces such a magnetic anisotropy.

Here we comment on the effect of bond randomness in YCOB. A comparison of magnetic susceptibility measurements and simple numerical calculations that do not include non-Heisenberg interactions has previously suggested

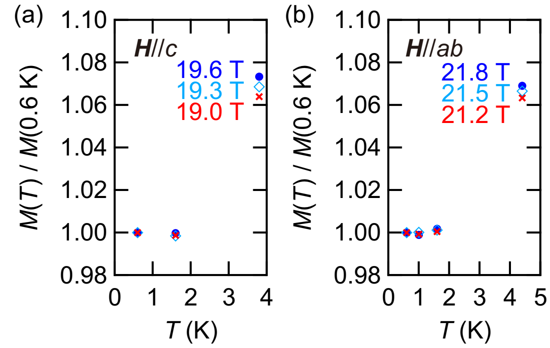

FIG. 3. Temperature dependence of magnetization in the $1/9$ plateau regime. (a) and (b) Temperature dependence of magnetization at several fields in the $1/9$ plateau region for $\boldsymbol{H}||c$ and $\boldsymbol{H}||ab$. The magnetization values are independent of temperature for $T \leq 1.6$ K, indicating the formation of the spin gap.

the fairly large distribution of exchange interactions in YCOB [37]. However, it should be emphasized that arguments based on such simple models should be scrutinized. To begin, in the $1/3$ plateau state of the kagome AFM, the magnetic structure is commensurate with the lattice, regardless of its quantum or classical origin [10,12,14,15]. It is highly unlikely that the $1/3$ plateau is stable even in the presence of strong randomness reported in [37], as disorder usually acts to destabilize such commensurate plateaus. In addition, recent inelastic neutron scattering experiments have reported sharply dispersed spin excitations with Dirac-like linear dispersion [52], which are distinct from the damped spin excitations expected in the presence of large disorder. Therefore, although more quantitative analysis is needed, the amount of disorder is considered to be much smaller than that reported in [37].

The $1/3$ plateau in the kagome AFM has been discussed as a quantum mechanical state rather than a classical one. In fact, for the $1/3$ plateau, numerical calculations reveal that a crystalline state of entangled magnons living on the individual hexagons gives lower energy than a classical up-up-down spin state [10,12,14,15]. In such a magnon crystal, six spins on each hexagonal plaquette form an entangled state with three resonant magnons, which then crystallize into a $\sqrt{3} \times \sqrt{3}$ superstructure [Figs. 4(a) and 4(b)]. This picture is robustly inferred from a similar state with one magnon on each hexagonal plaquette [Fig. 4(d)] obtained as the rigorous ground state at the $7/9$ plateau [53,54], and generalizes to the $5/9$ plateau as well [Fig. 4(c)]. Therefore, a crystal of emergent highly entangled magnon complexes is anticipated in the observed $1/3$ plateau state.

On the other hand, the $1/9$ plateau is hardly accounted for by this magnon crystal picture, suggesting even more exotic states. One intriguing scenario is a QSL state exhibiting a $\mathbb{Z}_3$ topological order, as observed in density matrix

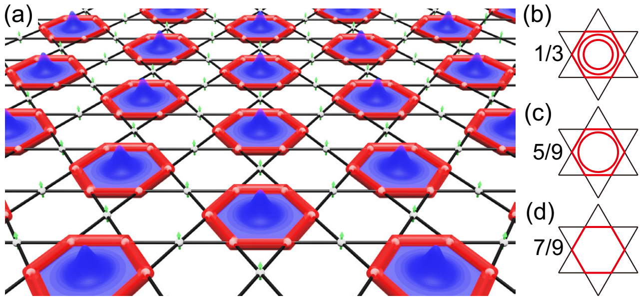

FIG. 4. Crystal state of localized magnons. (a) Magnon crystal state at $1/3$, $5/9$, and $7/9$ magnetization plateaus. Six entangled spins (red hexagons) form the extended magnons (blue) within the hexagon of the kagome lattice, that crystallizes into a $\sqrt{3} \times \sqrt{3}$ superstructure. The other spins (green arrows) point in the field direction. (b)–(d) The hexagonal magnons at $1/3$, $5/9$, and $7/9$ plateaus. Three, two, or one magnons (red hexagons and circles) are localized on each hexagon at $1/3$, $5/9$, and $7/9$ plateaus, respectively.

renormalization group simulations [10]. Another potential explanation is valence bond crystals suggested by tensor network studies [11,13], the latter of which concludes an hourglass $\sqrt{3} \times \sqrt{3}$ superstructure with gapless nonmagnetic excitations. Thus, examining the low energy excitations and the spatial symmetry breaking is pivotal in distinguishing these states.

In summary, by investigating the magnetization process of the recently discovered QSL candidate $YCu_3(OH)_{6.5}Br_{2.5}$ with a perfect kagome structure, we find distinct magnetization plateaus at $1/9$ and $1/3$ of the saturation moment of Cu^{2+}. The temperature dependence of magnetization curves provides evidence for the formation of the spin gap in these plateau states. The observation of both $1/3$ and $1/9$ magnetization plateaus in the spin-1/2 2D quantum magnet demonstrates that the present system provides an ideal platform for exploring strongly correlated exotic quantum states of matter.

Note added.—Recently, we became aware of [46] that reported magnetic oscillations in the vicinity of the $1/9$ plateau in a similar material. In contrast to the present results, their work attributed the $1/9$ plateau state to a gapless Dirac QSL. The magnetization process in a similar material was reported [48]. The observation of the $1/9$ plateau is consistent with this study.

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[1] S. Sachdev, Phys. Rev. B **45**, 12377 (1992).
[2] L. Balents, Nature (London) **464**, 199 (2010).
[3] Y. Ran, M. Hermele, P. A. Lee, and X.-G. Wen, Phys. Rev. Lett. **98**, 117205 (2007).
[4] S. Yan, D. A. Huse, and S. R. White, Science **332**, 1173 (2011).
[5] H.-C. Jiang, Z. Wang, and L. Balents, Nat. Phys. **8**, 902 (2012).
[6] S. Depenbrock, I. P. McCulloch, and U. Schollwöck, Phys. Rev. Lett. **109**, 067201 (2012).
[7] Y. Iqbal, F. Becca, S. Sorella, and D. Poilblanc, Phys. Rev. B **87**, 060405(R) (2013).
[8] Y.-C. He, M. P. Zaletel, M. Oshikawa, and F. Pollmann, Phys. Rev. X **7**, 031020 (2017).
[9] A. M. Läuchli, J. Sudan, and R. Moessner, Phys. Rev. B **100**, 155142 (2019).
[10] S. Nishimoto, N. Shibata, and C. Hotta, Nat. Commun. **4**, 2287 (2013).
[11] T. Picot, M. Ziegler, R. Orús, and D. Poilblanc, Phys. Rev. B **93**, 060407(R) (2016).
[12] X. Chen, S.-J. Ran, T. Liu, C. Peng, Y.-Z. Huang, and G. Su, Sci. Bull. **63**, 1545 (2018).
[13] D. Z. Fang, N. Xi, S.-J. Ran, and G. Su, Phys. Rev. B **107**, L220401 (2023).
[14] R. Okuma, D. Nakamura, T. Okubo, A. Miyake, A. Matsuo, K. Kindo, M. Tokunaga, N. Kawashima, S. Takeyama, and Z. Hiroi, Nat. Commun. **10**, 1229 (2019).
[15] S. Capponi, O. Derzhko, A. Honecker, A. M. Läuchli, and J. Richter, Phys. Rev. B **88**, 144416 (2013).
[16] J. Schnack, J. Schulenburg, and J. Richter, Phys. Rev. B **98**, 094423 (2018).
[17] A. Chubukov and D. Golosov, J. Phys. Condens. Matter **3**, 69 (1991).
[18] P. Mendels and F. Bert, J. Phys. Soc. Jpn. **79**, 011001 (2010).
[19] M. R. Norman, Rev. Mod. Phys. **88**, 041002 (2016).
[20] P. Mendels, F. Bert, M. A. de Vries, A. Olariu, A. Harrison, F. Duc, J. C. Trombe, J. S. Lord, A. Amato, and C. Baines, Phys. Rev. Lett. **98**, 077204 (2007).
[21] J. S. Helton, K. Matan, M. P. Shores, E. A. Nytko, B. M. Bartlett, Y. Yoshida, Y. Takano, A. Suslov, Y. Qiu, J.-H. Chung, D. G. Nocera, and Y. S. Lee, Phys. Rev. Lett. **98**, 107204 (2007).
[22] T.-H. Han, J. S. Helton, S. Chu, D. G. Nocera, J. A. Rodriguez-Rivera, C. Broholm, and Y. S. Lee, Nature (London) **492**, 406 (2012).
[23] A. Olariu, P. Mendels, F. Bert, F. Duc, J. C. Trombe, M. A. de Vries, and A. Harrison, Phys. Rev. Lett. **100**, 087202 (2008).
[24] T. Imai, E. A. Nytko, B. M. Bartlett, M. P. Shores, and D. G. Nocera, Phys. Rev. Lett. **100**, 077203 (2008).
[25] G. Nilsen, M. De Vries, J. R. Stewart, A. Harrison, and H. Rønnow, J. Phys. Condens. Matter **25**, 106001 (2013).
[26] T.-H. Han, M. R. Norman, J.-J. Wen, J. A. Rodriguez-Rivera, J. S. Helton, C. Broholm, and Y. S. Lee, Phys. Rev. B **94**, 060409(R) (2016).
[27] M. Fu, T. Imai, T.-H. Han, and Y. S. Lee, Science **350**, 655 (2015).

[28] P. Khuntia, M. Velazquez, Q. Barthélemy, F. Bert, E. Kermarrec, A. Legros, B. Bernu, L. Messio, A. Zorko, and P. Mendels, Nat. Phys. **16**, 469 (2020).
[29] J. Wang, W. Yuan, P. M. Singer, R. W. Smaha, W. He, J. Wen, Y. S. Lee, and T. Imai, Nat. Phys. **17**, 1109 (2021).
[30] M. P. Shores, E. A. Nytko, B. M. Bartlett, and D. G. Nocera, J. Am. Chem. Soc. **127**, 13462 (2005).
[31] D. E. Freedman, T. H. Han, A. Prodi, P. Muller, Q.-Z. Huang, Y.-S. Chen, S. M. Webb, Y. S. Lee, T. M. McQueen, and D. G. Nocera, J. Am. Chem. Soc. **132**, 16185 (2010).
[32] R. Okuma, D. Nakamura, and S. Takeyama, Phys. Rev. B **102**, 104429 (2020).
[33] H. Ishikawa, M. Yoshida, K. Nawa, M. Jeong, S. Krämer, M. Horvatić, C. Berthier, M. Takigawa, M. Akaki, A. Miyake, M. Tokunaga, K. Kindo, J. Yamaura, Y. Okamoto, and Z. Hiroi, Phys. Rev. Lett. **114**, 227202 (2015).
[34] M. Goto, H. Ueda, C. Michioka, A. Matsuo, K. Kindo, and K. Yoshimura, Phys. Rev. B **94**, 104432 (2016).
[35] R. Shirakami, H. Ueda, H. O. Jeschke, H. Nakano, S. Kobayashi, A. Matsuo, T. Sakai, N. Katayama, H. Sawa, K. Kindo, C. Michioka, and K. Yoshimura, Phys. Rev. B **100**, 174401 (2019).
[36] X.-H. Chen, Y.-X. Huang, Y. Pan, and J.-X. Mi, J. Magn. Magn. Mater. **512**, 167066 (2020).
[37] J. Liu, L. Yuan, X. Li, B. Li, K. Zhao, H. Liao, and Y. Li, Phys. Rev. B **105**, 024418 (2022).
[38] Z. Zeng, X. Ma, S. Wu, H.-F. Li, Z. Tao, X. Lu, X. H. Chen, J.-X. Mi, S.-J. Song, G.-H. Cao, G. Che, K. Li, G. Li, H. Luo, Z. Y. Meng, and S. Li, Phys. Rev. B **105**, L121109 (2022).
[39] X. Hong, M. Behnami, L. Yuan, B. Li, W. Brenig, B. Büchner, Y. Li, and C. Hess, Phys. Rev. B **106**, L220406 (2022).
[40] F. Lu, L. Yuan, J. Zhang, B. Li, Y. Luo, and Y. Li, Commun. Phys. **5**, 272 (2022).
[41] B. Bernu, L. Pierre, K. Essafi, and L. Messio, Phys. Rev. B **101**, 140403(R) (2020).
[42] C. Hotta and K. Asano, Phys. Rev. B **98**, 140405(R) (2018).
[43] M. Rigol and R. R. P. Singh, Phys. Rev. Lett. **98**, 207204 (2007).
[44] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.132.226701 for additional data.
[45] H. Schlüter, J. Richter, and J. Schnack, J. Phys. Soc. Jpn. **91**, 094711 (2022).
[46] G. Zheng, Y. Zhu, K.-W. Chen, B. Kang, D. Zhang, K. Jenkins, A. Chan, Z. Zeng, A. Xu, O. A. Valenzuela, J. Blawat, J. Singleton, P. A. Lee, S. Li, and L. Li, arXiv:2310.07989.
[47] K. Totsuka, Phys. Rev. B **57**, 3454 (1998).
[48] S. Jeon, D. Wulferding, Y. Choi, S. Lee, K. Nam, K. H. Kim, M. Lee, T.-H. Jang, J.-H. Park, S. Lee, S. Choi, C. Lee, H. Nojiri, and K.-Y. Choi, Nat. Phys. **20**, 435 (2024).
[49] K. Onizuka, H. Kageyama, Y. Narumi, K. Kindo, Y. Ueda, and T. Goto, J. Phys. Soc. Jpn. **69**, 1016 (2000).
[50] T. Chen, C. Guo, P. Sengupta, and D. Poletti, Phys. Rev. B **101**, 064417 (2020).
[51] 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, Phys. Rev. Lett. **125**, 027203 (2020).
[52] Z. Zeng, C. Zhou, H. Zhou, L. Han, R. Chi, K. Li, M. Kofu, K. Nakajima, Y. Wei, W. Zhang, D. G. Mazzone, Z. Y. Meng, and S. Li, arXiv:2310.11646.
[53] J. Schulenburg, A. Honecker, J. Schnack, J. Richter, and H.-J. Schmidt, Phys. Rev. Lett. **88**, 167207 (2002).
[54] J. Schnack, J. Schulenburg, A. Honecker, and J. Richter, Phys. Rev. Lett. **125**, 117207 (2020).