ISSN 1674-1056
http://cpb.iphy.ac.cn
http://iopscience.iop.org/cpb
中国物理 B
Chinese Physics
中国物理 Chinese Physics B

Recent progress on magnetic-field studies on quantum-spin-liquid candidates

Zhen Ma(马祯), Kejing Ran(冉柯静), Jinghui Wang(王靖珲), Song Bao(鲍嵩), Zhengwei Cai(蔡正蔚), Shichao Li(李世超), Jinsheng Wen(温锦生)

Citation: Chin. Phys. B . 2018, 27(10): 106101. **doi:** 10.1088/1674-1056/27/10/106101

Journal homepage: http://cpb.iphy.ac.cn; http://iopscience.iop.org/cpb

What follows is a list of articles you may be interested in

Observation of spin glass transition in spinel $LiCoMnO_4$

Chen Hong, Yang Xu, Zhang Pei-Song, Liang Lei, Hong Yuan-Ze, Wei Ying-Jin, Chen Gang, Du Fei, Wang Chun-Zhong
Chin. Phys. B . 2015, 24(12): 127501. **doi:** 10.1088/1674-1056/24/12/127501

Spin frustration and magnetic ordering in triangular lattice antiferromagnet $Ca_3CoNb_2O_9$

Dai Jia, Zhou Ping, Wang Peng-Shuai, Pang Fei, Tim J. Munsie, Graeme M. Luke, Zhang Jin-Shan, Yu Wei-Qiang
Chin. Phys. B . 2015, 24(12): 127508. **doi:** 10.1088/1674-1056/24/12/127508

Double spin-glass-like behavior and antiferromagnetic superexchange interaction between Fe^{3+} ions in α-$Ga_{2-x}Fe_xO_3$ ($0 \le x \le 0.4$)

Lv Yi-Fei, Xiang Jian-Yong, Wen Fu-Sheng, Lv Wei-Ming, Hu Wen-Tao, Liu Zhong-Yuan
Chin. Phys. B . 2015, 24(3): 037502. **doi:** 10.1088/1674-1056/24/3/037502

Asymmetric exchange bias training effect in spin glass (FeAu)/FeNi bilayers

Rui Wen-Bin, He Mao-Cheng, You Biao, Shi Zhong, Zhou Shi-Ming, Xiao Ming-Wen, Gao Yuan, Zhang Wei, Sun Li, Du Jun
Chin. Phys. B . 2014, 23(10): 107502. **doi:** 10.1088/1674-1056/23/10/107502

Spin-correlation function of the fully frustrated Ising model and ± *J* Ising spin glass on the square lattice

M Y Ali, J Poulter
Chin. Phys. B . 2013, 22(6): 067502. **doi:** 10.1088/1674-1056/22/6/067502

TOPICAL REVIEW — Fundamental research under high magnetic fields

Recent progress on magnetic-field studies on quantum-spin-liquid candidates*

Zhen Ma(马祯)[1], Kejing Ran(冉柯静)[1], Jinghui Wang(王靖珲)[1], Song Bao(鲍嵩)[1], Zhengwei Cai(蔡正蔚)[1], Shichao Li(李世超)[1], and Jinsheng Wen(温锦生)[1,2,†]

[1] National Laboratory of Solid State Microstructures and Department of Physics, Nanjing University, Nanjing 210093, China
[2] Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing 210093, China

(Received 25 June 2018; revised manuscript received 25 July 2018; published online 10 September 2018)

Quantum spin liquids (QSLs) represent a novel state of matter in which quantum fluctuations prevent the conventional magnetic order from being established, and the spins remain disordered even at zero temperature. There have been many theoretical developments proposing various QSL states. On the other hand, experimental movement was relatively slow largely due to limitations on the candidate materials and difficulties in the measurements. In recent years, the experimental progress has been accelerated. In this topical review, we give a brief summary of experiments on the QSL candidates under magnetic fields. We arrange our discussions by two categories: i) Geometrically-frustrated systems, including triangular-lattice compounds $YbMgGaO_4$ and $YbZnGaO_4$, κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$, and $EtMe_3Sb[Pd(dmit)_2]_2$, and the kagomé system $ZnCu_3(OH)_6Cl_2$; ii) the Kitaev material α-$RuCl_3$. Among these, we will pay special attention to α-$RuCl_3$, which has been intensively studied by ours and other groups recently. We will present evidence that both supports and rejects the QSL ground state for these materials, based on which we give several perspectives to stimulate further research activities.

Keywords: quantum spin liquids, spin glass, neutron scattering

PACS: 61.05.fg, 75.10.Kt, 75.50.Lk **DOI:** 10.1088/1674-1056/27/10/106101

1. Introduction

Generally, a system tends to lower its symmetry to be in the low-energy state. As a consequence, a magnetic material will break certain symmetry according to Landau's theorem, and the magnetic moments carried by the electron spins will form an ordered pattern at low temperatures.[1,2] Excitations associated with these ordered spins are conventional magnons with spin $S = 1$ (Refs. [3]–[9]). However, in systems with small spin and strong quantum fluctuations, such a conventional order can be avoided, leading to a quantum-spin-"liquid" (QSL) state.[10] Now, it is known that geometrical frustration, a situation where the antiferromagnetic Heisenberg exchange interactions cannot be satisfied simultaneously among different sites in triangular [Fig. 1(a)] or kagomé lattices [Fig. 1(b)], can result in strong quantum fluctuations.[11–13] In 1973, Anderson proposed the resonant-valence-bond (RVB) model on the triangular lattice for the QSL state. It is a superposition of all possible configurations of the singlets formed by any of the two strongly interacting spins.[14–17] The elementary excitations in QSLs are fractionalized quasiparticles, e.g., charge-free $S = 1/2$ spinons, fundamentally different from conventional magnons.[14–17]

The QSL state defined by the RVB model does not have an exact solution. In 2006, Kitaev constructed an exactly-solvable $S = 1/2$ model on the honeycomb lattice [Fig. 1(c)]. This model, as in Eq. (1), is named the Kitaev model.

$$H = -K_x \sum_{x\text{-bonds}} S_i^x S_j^x - K_y \sum_{y\text{-bonds}} S_i^y S_j^y - K_z \sum_{z\text{-bonds}} S_i^z S_j^z. \quad (1)$$

Here, H is the Hamiltonian, and $K_{x,y,z}$ are the nearest-neighbor Kitaev interactions of the x, y, and z bonds on the honeycomb lattice. QSLs defined by this model are termed the Kitaev QSLs. Unlike QSLs arising from geometrical frustration, the Kitaev QSL results from bond-dependent interactions that frustrate spin configurations on a single site.[18] Initially, the Kitaev model was treated as a toy model since the anisotropic spin interactions are unrealistic in a spin-only system. Later, it was suggested that in Mott insulators with strong spin-orbital coupling (SOC), the anisotropic Kitaev interaction may be achievable due to the spatial anisotropy of the orbitals.[19] As such, our discussions on the QSL candidates will be based on two categories: geometrical-frustration-induced ones and Kitaev QSL candidates.

Besides the rich and exotic physics of QSLs, they also hold promising application potentials, for example, quantum computation via braiding the non-Abelian anyons in

*Project supported by the National Natural Science Foundation of China (Grant Nos. 11674157 and 11822405) and the Fundamental Research Funds for the Central Universities (Grant No. 020414380105).

†Corresponding author. E-mail: jwen@nju.edu.cn

© 2018 Chinese Physical Society and IOP Publishing Ltd

these materials.[18,20–23] Furthermore, understanding QSLs may help understand the mechanism of high-temperature superconductivity.[24–28] For these reasons, research on QSL has been surging in the past few decades. There have been many review articles summarizing the progress on QSLs already.[11,21,29–42] In this topical review, we will restrict our discussions to the measurements under magnetic fields only.

In general, an external magnetic field can be detrimental to the QSL phase, as the field may induce symmetry breaking. Ultimately, when the field is strong enough, all the spins will be polarized and the moments will align with the field direction — the system is then a ferromagnet. On the other hand, in the fully polarized state, one can extract the exchange interactions from the spin-wave excitation spectra obtained by inelastic neutron scattering (INS) measurements and understand the magnetic ground state in zero field. Investigating the magnetic excitations under fields with techniques such as nuclear magnetic resonance (NMR), muon spin relaxation (μSR), electron spin resonance, and terahertz spectroscopy also provides key information on the interactions underlying the exotic states of the QSL candidates. Furthermore, studying the field evolution of the thermal transport properties can also provide insights into the QSL physics. Finally, in available systems where the Kitaev physics is relevant, there are other non-Kitaev terms setting in at low temperatures, resulting in an ordered phase instead of the Kitaev QSL. In some of these materials, such as α-$RuCl_3$, applying a magnetic field suppresses the non-Kitaev interactions and drives the system into a possible QSL state. In this article, we will summarize the results from these magnetic-field experiments on the i) Geometrically-frustrated systems, including triangular-lattice compounds $YbMgGaO_4$ and $YbZnGaO_4$, κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$, and $EtMe_3Sb[Pd(dmit)_2]_2$, and the kagomé system $ZnCu_3(OH)_6Cl_2$; ii) the Kitaev material α-$RuCl_3$, which has been subject to intensive investigations by ours and other groups recently. We first present experimental evidence for each of these materials, then form discussions based on these results, and raise several questions in the end.

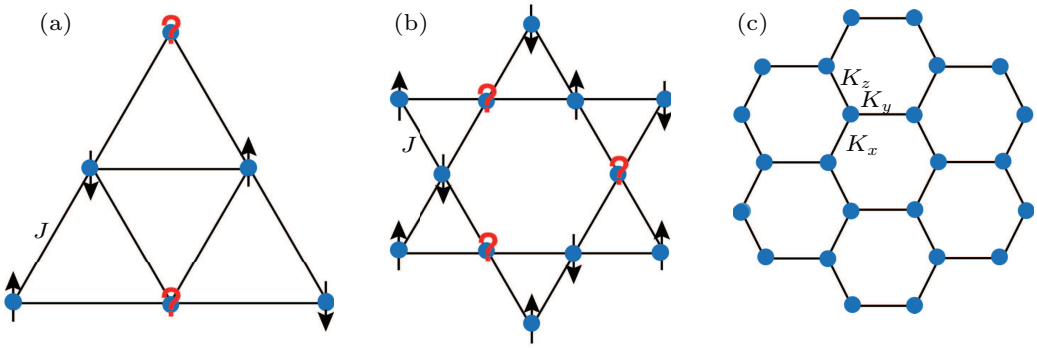


Fig. 1. (color online) Schematics of two-dimensional (a) triangular, (b) kagomé, and (c) honeycomb structures where quantum spin liquids can be realized. Arrows and question marks represent spins and geometrical frustration, respectively. J is the Heisenberg exchange interaction, and $K_{x,y,z}$ are Kitaev interactions along three bonds.

2. Geometrically-frustrated systems

2.1. $YbMgGaO_4$ and $YbZnGaO_4$

$YbMgGaO_4$ with the quasi-two-dimensional triangular-lattice structure has been reported to be a promising QSL candidate recently.[43–49] It has a negative Curie–Weiss temperature of ~ -4 K[43,44] but does not show a long-range magnetic order down to 30 mK, indicating strong geometrical frustration.[45,46] The magnetic specific heat under different fields for $YbMgGaO_4$ is shown in Fig. 2(a).[43] In zero field, there is a broad hump at 2.4 K, instead of a sharp λ-type peak expected for a well-defined phase transition. The magnetic specific heat is large at low temperatures, suggesting a gapless ground state. By applying magnetic fields up to 9 T, the system becomes a fully-polarized ferromagnetic state and opens a spin gap of ~ 8.26 K.[43,46,50] $YbZnGaO_4$, a compound isostructural to $YbMgGaO_4$, shows very similar behaviors. In Fig. 2(b), the magnetic specific heat results of $YbZnGaO_4$ in zero and 9-T fields are shown. Similarly, the results indicate a gapless state in zero field, and a gapped state with a gap size of ~ 6.18 K in a 9-T field.[51]

In zero field, the magnetic excitation spectra for both $YbMgGaO_4$[45,46] and $YbZnGaO_4$[51] exhibit as a broad "continuum" in INS measurements, as shown in Fig. 3(a).[45] Since QSLs are characterized by spin excitations carrying fractional quantum numbers, e.g., deconfined spinons, such observations of the continuum have been taken to be strong evidence for a QSL state in $YbMgGaO_4$.[45,46] Applying a magnetic field on the sample will force the magnetic moment to align with the field direction, which results in a ferromagnetic state in high fields. Paddison *et al.*[45] carried out INS measurements under a 7.8-T field applied along the c axis and observed the spin-wave spectra resulting from the ferromagnetic state, as shown in Fig. 3(b). In addition, there are still broad and continuum-like excitations, which are believed to be due to

the disorder effect. By fitting the spectra with an anisotropic Heisenberg model plus the Zeeman term to include the field effect, they concluded that the ground state of $YbMgGaO_4$ was a QSL in which next-nearest-neighbor exchange interactions, anisotropy, and disorder all played important roles. In $YbZnGaO_4$, the INS results are also quite similar.[51]

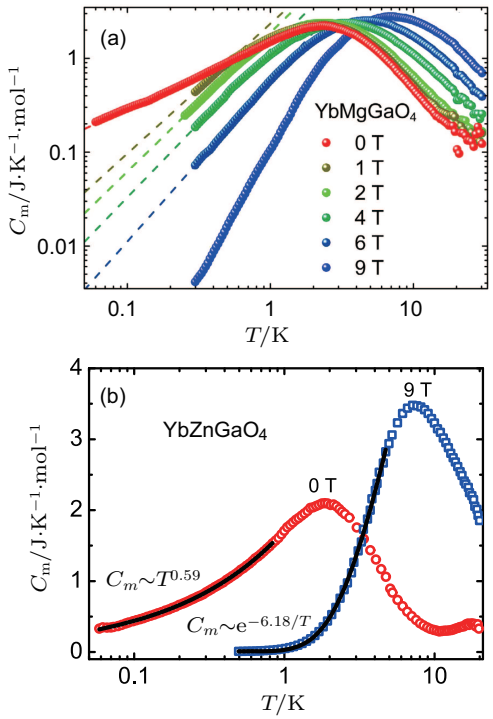


Fig. 2. (color online) (a) Temperature dependence of the magnetic specific heat of $YbMgGaO_4$ under different magnetic fields. Dashed lines indicate power-law fits to the low-temperature data. From Ref. [43]. (b) Magnetic specific heat of $YbZnGaO_4$ measured under zero and 9-T fields. Solid lines are fits to the data. From Ref. [51].

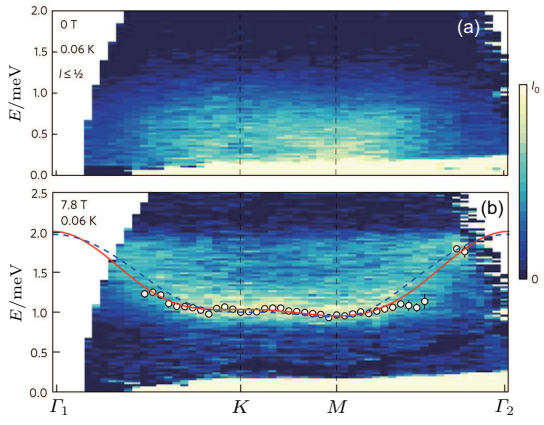


Fig. 3. (color online) Magnetic excitation spectra along high-symmetry directions under (a) zero, and (b) 7.8-T fields applied parallel to the c axis. Curves are fits to the spin-wave excitations using the anisotropic spin model. From Ref. [45].

While results above are all consistent with $YbMgGaO_4$ and $YbZnGaO_4$ being QSLs, there are several issues that make this conclusion questionable: i) The small value of the exchange interaction (~ 0.15 meV) restricts typical INS measurements in the overdamped regime of the excitation spectra,[43–46] so it is not clear whether the broad features both in momentum and energy are intrinsic or due to the large probing energy; ii) since Mg^{2+}/Zn^{2+} and Ga^{3+} in the nonmagnetic layers are randomly distributed,[43,44,52] the disorder effect, which is considered to be detrimental to the QSL phase, is severe;[45,48,53] iii) thermal conductivity (κ) results on both $YbMgGaO_4$[50] and $YbZnGaO_4$[51] as shown in Fig. 4 strongly challenge the idea of these materials being QSLs, as we discuss in the following.

As shown in Fig. 4, in zero field, κ is only half of that of the nonmagnetic reference sample $LuMgGaO_4$, in which only phonons contribute to κ. By fitting the zero-field data with $\kappa/T = \kappa_0/T + nT^{\beta-1}$ where κ_0 and nT^{β} represent non-phonon and phonon contributions, respectively, it is shown that κ_0/T for both $YbMgGaO_4$ and $YbZnGaO_4$ are effectively zero within experimental errors, similar to the case of $LuMgGaO_4$.[50,51] Thermal conductivity can be written as $\kappa = 1/3C_m v_F l$, where C_m, v_F, and l are the specific heat, Fermi velocity, and the mean-free path of the quasiparticles, respectively. By assuming that the κ at 0.1 K is totally contributed by the magnetic excitations, Xu *et al.* estimated l to be 8.6 Å for $YbMgGaO_4$, only about 2.5 times of the spin–spin distance.[50] In contrast, another QSL candidate $EtMe_3Sb[Pd(dmit)_2]_2$ has a high $\kappa_0/T = 0.2$ W·K^{-2}·m^{-1}, which is considered to be evidence for the presence of highly mobile quasiparticles with l of ~ 1000 times of the spin–spin distance.[54]

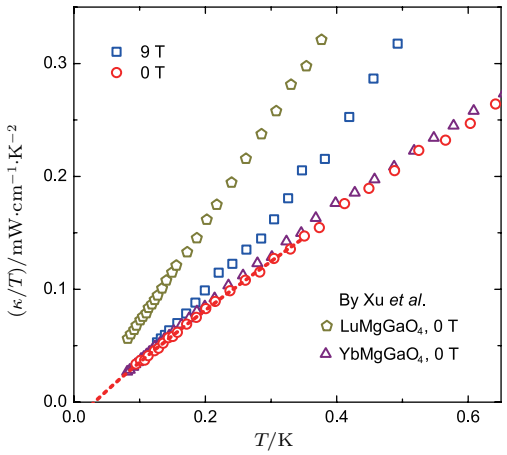


Fig. 4. (color online) The in-plane thermal conductivity of a $YbZnGaO_4$ single crystal under zero and 9-T fields applied parallel to the c axis. For comparison, the thermal conductivity of $YbMgGaO_4$ and $LuMgGaO_4$ reported in Ref. [50] is also plotted. From Ref. [51].

Furthermore, in both $YbMgGaO_4$ and $YbZnGaO_4$, it appears that the role of the magnetic excitations is to scatter off phonons that conduct heat. Therefore, when the magnetic excitations are present, κ is reduced — this explains the the reduction of κ as compared to that of $LuMgGaO_4$.[50,51] This is further manifested in the magnetic-field measurements: in a field of 9 T that opens a gap of 8.26 K and 6.18 K in

$YbMgGaO_4$[50] and $YbZnGaO_4$,[51] respectively, there are almost no magnetic excitations to scatter phonons, so κ increases. Therefore, a gapless QSL does not seem to be an applicable description for $YbMgGaO_4$ and $YbZnGaO_4$, because their significant magnetic excitations, as evidenced by the large magnetic specific heat, should contribute to κ.[55,56] These results, however, can be understood within a disordered-magnet picture, in which the mean-free path of the magnons is reduced with disorder, and they are not expected to conduct heat. We will discuss it further in Section 4.

2.2. κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

Layered organic κ-(BEDT-TTF)$_2X$ are Mott insulators having a two-dimensional triangular lattice, as illustrated in Fig. 1(a). Here, BEDT-TTF (ET) denotes the electron donating molecule and X represents a variety of anions with closed shells.[57–59] Among these, κ-$(ET)_2Cu_2(CN)_3$ is suggested to be a QSL.[60–63] The temperature dependence of magnetic susceptibility features a broad peak, which can be reproduced by the triangular-lattice Heisenberg model with an exchange interaction of $J \sim 250$ K.[60] The NMR spectra show neither a distinct broadening nor splitting down to 32 mK, suggesting the absence of magnetic order down to the temperature of 4 orders of magnitude lower than J.[64] A μSR experiment also confirms this conclusion.[65]

In Fig. 5(a), temperature dependences of the specific heat for κ-$(ET)_2Cu_2(CN)_3$ under various fields are shown together with those of other ET salts.[66] No discernible field effect is observed. From a linear extrapolation of the data in Fig. 5(a) down to zero temperature, a linearly temperature-dependent term, i.e., the electronic coefficient γ in $C_pT^{-1} = \gamma + \beta T^2$ is determined to be a finite value of 20 ± 5 mJ·K^{-2}·mol^{-1}, indicating the presence of gapless magnetic excitations in κ-$(ET)_2Cu_2(CN)_3$ at zero temperature.[66] This result is consistent with Anderson's proposal for the spin-liquid state in two-dimensional materials with the triangular-lattice structure.[14] For other ET salts shown in Fig. 5(a), γ is zero as expected for nonmagnetic insulators. The γ term is considered to be proportional to the spinon density of states, almost field independent.[66]

Thermal conductivity measurements can provide key information on the elementary excitations from the magnetic ground state in a QSL candidate, as κ is sensitive to itinerant excitations such as the spinons. In Fig. 5(b), κ/T versus T^2 for κ-$(ET)_2Cu_2(CN)_3$ is shown.[67] The κ/T has an extremely small value and tends to vanish as temperature decreases to zero. It should be stressed that the vanishing value of κ/T at $T = 0$ immediately indicates the absence of low-lying fermionic excitations,[67] in sharp contrast to the specific heat data suggesting the presence of gapless excitations in Ref. [66]. Moreover, the behavior under a magnetic field up to 10 T perpendicular to the basal plane shows a nearly parallel and small shift from that under zero field. As the zero-field data, the 10-T results also suggest the absence of magnetic excitations at low temperatures. The obvious contradiction of the specific heat and thermal conductivity results leaves the nature of the magnetic ground state of κ-$(ET)_2Cu_2(CN)_3$ an open question, which will be discussed further in Section 4.

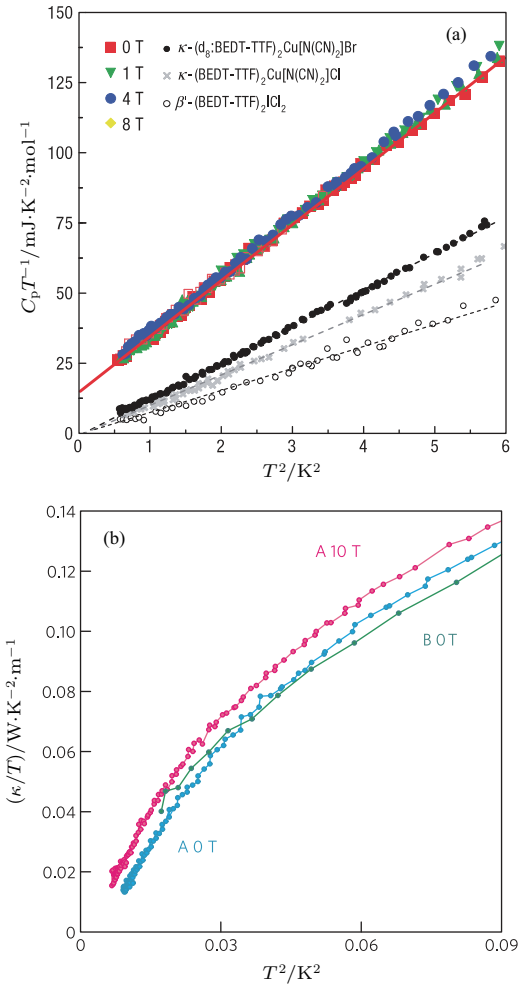


Fig. 5. (color online) (a) Specific heat of κ-$(ET)_2Cu_2(CN)_3$ under different fields up to 8 T in comparison with those of other ET-based salts. From Ref. [66]. (b) Low-temperature thermal conductivity κ of κ-$(ET)_2Cu_2(CN)_3$ (samples A and B). Sample A was investigated under a 10-T field applied along the c axis. From Ref. [67].

2.3. EtMe$_3$Sb[Pd(dmit)$_2$]$_2$

$EtMe_3Sb[Pd(dmit)_2]_2$ is a member of the $A[Pd(dmit)_2]_2$ family, a layered system composed of insulating A and conducting $Pd(dmit)_2$ layers. Here, $A^+ = Et_xMe_{4-x}Z^+$ (Et = C_2H_5, Me = CH_3, Z = N, P, As, and $x = 0$, 1, 2), and dmit is a 1,3-dithiole-2-thione-4,5-dithiolate.[68,69] Spins are on the two-dimensional triangular lattice formed by the $Pd(dmit)_2$ molecules. Although the magnetic susceptibility shows a broad peak around 50 K, no anomaly indicative of magnetic order is observed down to 2 K.[29,68] ^{13}C NMR measurements down to 20 mK also indicate the absence of long-range mag-

netic order at this temperature.[70]

Figure 6(a) shows the specific heat of $EtMe_3Sb[Pd(dmit)_2]_2$ in comparison with the results of κ-$(ET)_2Cu_2(CN)_3$[66] and $EtMe_3P[Pd(dmit)_2]_2$. As in the QSL candidate κ-$(ET)_2Cu_2(CN)_3$, there is no sharp peak indicative of long-range order over the entire temperature range measured. The specific heat of the QSL candidates $EtMe_3Sb[Pd(dmit)_2]_2$ and κ-$(ET)_2Cu_2(CN)_3$ are larger than that of $EtMe_3P[Pd(dmit)_2]_2$ with the nonmagnetic ground state of a valence-bond-solid type order occurring at 25 K.[69,72,73] This suggests that the magnetic entropy survives at low temperatures owing to the fluctuations of correlated spins in the QSL candidates.[71] The low-temperature specific heat for $EtMe_3Sb[Pd(dmit)_2]_2$ is plotted as C_pT^{-1} versus T^2 in comparison with those of other compounds in Fig. 6(b). The finite electronic specific heat coefficient $\gamma = 19.9$ mJ·K^{-2}·mol^{-1} suggests the presence of gapless excitations at zero temperature.[71] The value of γ is almost field independent, consistent with the presence of a spinon Fermi surface.[71] On the other hand, the non-QSL candidates have γ that is effectively zero.

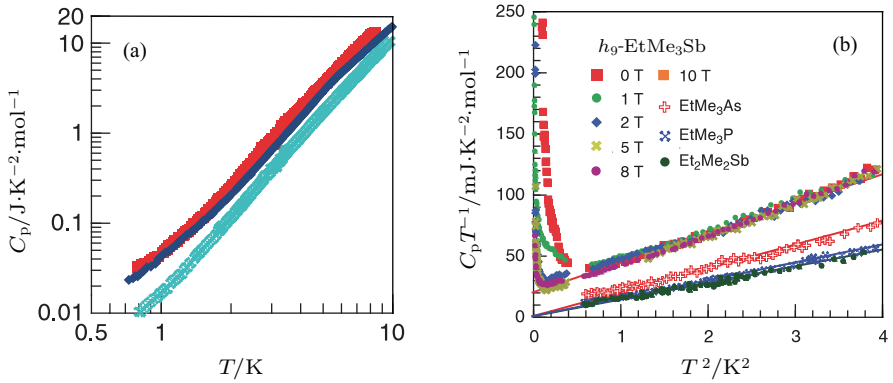


Fig. 6. (color online) (a) Specific heat data of $EtMe_3Sb[Pd(dmit)_2]_2$ (red squares) plotted on a logarithmic scale. The data of $EtMe_3P[Pd(dmit)_2]_2$ (cyan crosses) and κ-$(ET)_2Cu_2(CN)_3$ (blue diamonds) are also plotted for comparison. (b) Low-temperature specific heat of $EtMe_3Sb[Pd(dmit)_2]_2$ under different fields up to 10 T. The data for other nonmagnetic systems are plotted together for comparison. From Ref. [71].

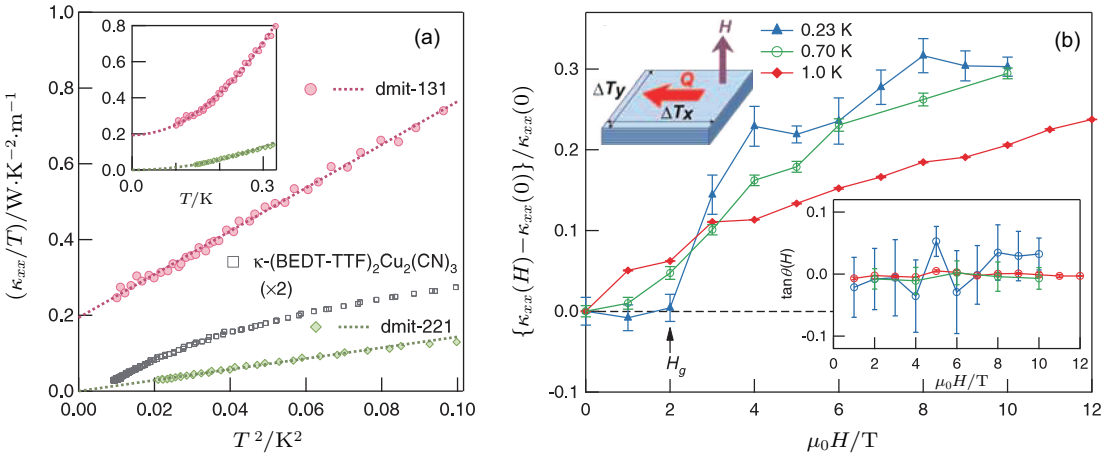


Fig. 7. (color online) (a) Low-temperature plot of κ_{xx}/T as a function of T^2 of dmit-131 (circles), dmit-221 (diamonds), and κ-$(ET)_2Cu_2(CN)_3$ (squares). The inset shows κ/T plotted as a function of T. (b) Field dependence of κ normalized by the zero-field value, $[\kappa_{xx}(H)-\kappa_{xx}(0)]/\kappa_{xx}(0)$ of dmit-131 at low temperatures. Upper left inset shows a schematic of the measurement setup. The lower left inset shows thermal-Hall angle $\tan\theta(H) = \kappa_{xy}/(\kappa_{xx}-\kappa_{xx}^{ph})$ as a function of the field value μ_0H at 0.23 K (triangles), 0.70 K (circles), and 1.0 K (diamonds). κ_{xy} and κ_{xx}^{ph} are the thermal-Hall conductivity and phonon-contributed thermal conductivity, respectively. From Ref. [54].

In Fig. 7(a), the longitudinal thermal conductivity κ_{xx} over temperature T of $EtMe_3Sb[Pd(dmit)_2]_2$ is plotted against T^2 in comparison with those of another QSL candidate κ-$(ET)_2Cu_2(CN)_3$ and a nonmagnetic compound $Et_2Me_2Sb[Pd(dmit)_2]_2$.[54] Interestingly, in contrast to the latter two, where κ_{xx}/T is 0 as T approaches 0, the residual κ_{xx}/T for $EtMe_3Sb[Pd(dmit)_2]_2$ is 0.2 W·K^{-2}·m^{-1}.[54] The presence of large residual κ_{xx} is also confirmed in the inset of Fig. 7(a) in which κ_{xx}/T is plotted as a function of T. This result is consistent with the observation of gapless magnetic excitations in the specific heat measurement. By using $\kappa_{xx} = 1/3C_m v_F l$, it is suggested that there are highly mobilized magnetic excitations with a mean-free path l of $\sim$ 1000 times of the spin–spin distance.[54] Remarkably, this is a rare example where thermal conductivity measurements show a finite κ_{xx}/T at zero temperature. The results indicate the presence of gapless magnetic

excitations consistent with the specific heat data.[71]

The magnetic-field dependence of κ_{xx} is shown in Fig. 7(b).[54] At the lowest temperature, $\kappa_{xx}(H)$ under low fields is insensitive to the magnetic field strength $\mu_0 H$ but displays a steep increase above a characteristic magnetic field of 2 T. This behavior is less profound with increasing temperature. At 1 K, κ_{xx} becomes linear. The observed field dependence is interpreted as the presence of spin-gap-like excitations at low temperatures, along with the gapless excitations inferred from the residual κ_{xx}/T and finite γ.[54,71] The gap behavior is also suggested from the zero-field thermal conductivity measurement where κ_{xx}/T versus T shows a broad peak at ~ 1 K.[54]

2.4. $ZnCu_3(OH)_6Cl_2$

$ZnCu_3(OH)_6Cl_2$ known as Herbertsmithite has a three-dimensional rhombohedral structure and consists of two-dimensional kagomé-lattice planes of spin-1/2 Cu^{2+} ions separated by nonmagnetic layers formed by Zn^{2+}.[74] Geometrical frustration on such a structurally perfect kagomé lattice is expected to be strong and many interesting phenomena may emerge.[35,75] Because large-size single crystals are available for this material,[76] it has been heavily studied by various experimental techniques. Previous measurement results[77–79] of polycrystalline Herbertsmithite suggest that there is no static magnetic order nor spin freezing down to 50 mK despite the large Curie–Weiss temperature of -314 K.[74] Specific heat shows no sharp λ-type peak down to ~ 100 mK.[77] When applying a magnetic field to the sample, the specific heat is changed rapidly as shown in Fig. 8 (Ref. [77]). Under a 14-T field, the low-temperature specific heat is largely reduced, because magnetic excitations are gapped, leaving only phonons contributing to the specific heat. On the other hand, at high temperatures where phonons dominate the specific heat, there is hardly any field effect.

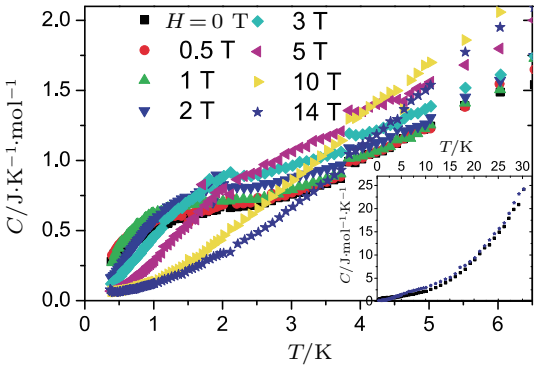


Fig. 8. (color online) Specific heat of Herbertsmithite measured under several different magnetic fields up to 14 T. Inset: Specific heat plotted over a wider temperature range under zero (squares) and 14-T (stars) fields. From Ref. [77].

The availability of large single crystals of Herbertsmithite[76] makes it possible to carry out INS measurements, which reveal a broad continuum expected for a QSL state.[82] The INS results show no spin gap down to 0.25 meV. However, previous calculation results by the density matrix renormalizaton group indicate that the ground state of the Heisenberg model on a kagomé lattice is a fully gapped QSL.[83] To understand the magnetic ground state of Herbertsmithite, Asaba *et al.*[84] performed magnetization measurements on a single crystal using torque magnetometry with an intense magnetic field up to 31 T. Based on the observation that the effective magnetic susceptibility under high fields is independent of the temperature within the low-temperature range between 20 mK and 5 K, they considered the ground state to be gapless, consistent with the INS results.[82]

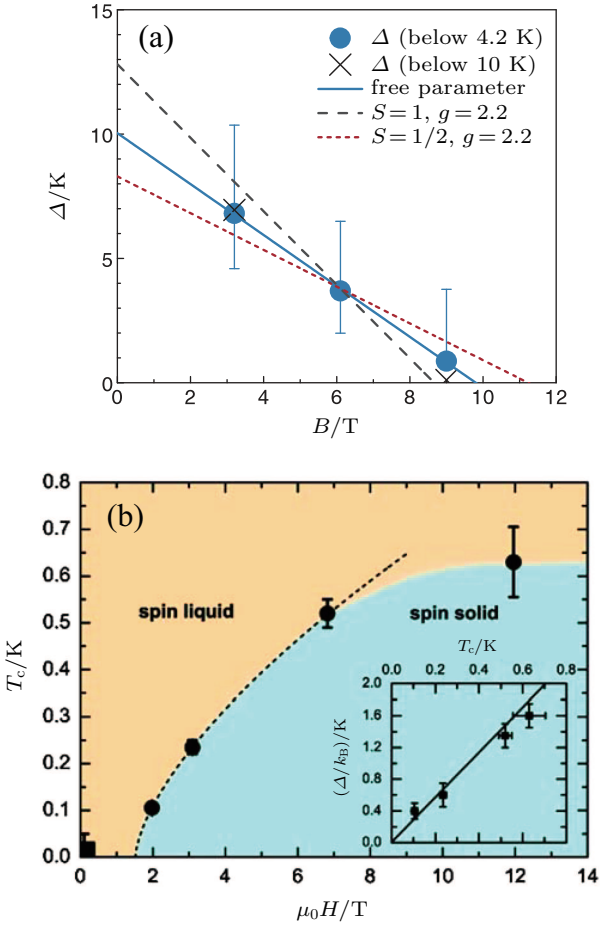


Fig. 9. (color online) (a) Spin excitation gap Δ of Herbertsmithite obtained by fitting NMR spectra with a temperature range up to 4.2 K (circles) and 10 K (crosses). Dashed and dotted lines represent the best fits under the constraint of $S = 1$ and 1/2, respectively. The solid line represents the best free-parameter fit. From Ref. [80]. (b) Transition temperature T_c versus magnetic field phase diagram of Herbertsmithite. The inset plots Δ as a function of T_c. From Ref. [81].

However, as shown in Fig. 9(a), NMR measurements on single crystals of Herbertsmithite reveal a finite gap value 0.86 ± 0.26 meV by extrapolating the fitted values of the gap to zero field.[80] This value is close to a later INS measurement which identifies a spin gap of 0.7 meV by modeling the momentum-integrated data as the sum of the contributions from a damped harmonic oscillator and the gapped

excitations.[85] We note that a more recent calculation employing the tensor network shows that the ground state is a gapless QSL.[86] Concerning the nature of the ground state for this kagomé compound, it turns out that no consensus has been reached so far.

Moreover, NMR measurements on polycrystalline samples identify a quantum critical point at a critical field B_c of 1.53 T, at which the system is believed to evolve from a spin liquid to a solid, as illustrated in Fig. 9(b).[81] The high-field, low-temperature spin-solid state is featured by frozen spins with slow relaxations. As shown in the inset of Fig. 9(b), the gap size in the spin-solid state is roughly proportional to the critical temperature T_c.[81]

3. Kitaev materials — α-$RuCl_3$

Different from aforementioned QSL candidates, which have either triangular or kagomé lattice where antiferromagnetic exchange interactions are geometrically frustrated, the Kitaev QSL has a honeycomb lattice where the frustration on a single site arises from the bond-dependent spin anisotropy.[18] Possible realization of such an exotic state has been suggested in SOC-assisted Mott insulators such as Na_2IrO_3,[19,42,87–91] and Li_2IrO_3.[91–95] These materials have the honeycomb lattice as shown in Fig. 1(c). Due to the combination of the cubic crystal electric field, strong SOC, and electronic correlations, the ground state is Krammers doublets with an effective spin of 1/2 (Refs. [96]–[98]). Furthermore, because of the strong SOC, the effective spin is expected to be anisotropic due to the spatial anisotropy of the 5d orbitals of Ir^{4+}. Therefore, the bond-dependent anisotropic Kitaev interaction may be realized on the honeycomb lattice.[18] However, it is found that they are not QSLs but are instead magnetically ordered at low temperatures.[42,91–95,98–101] Nevertheless, subsequent experimental and theoretical works suggest that the magnetic orders in these materials are unconventional, signifying the presence of notable Kitaev interaction. For instance, it is suggested that the zigzag magnetic order in Na_2IrO_3 can be understood within a Heisenberg–Kitaev model.[89,102] In the phase diagram constructed using this model, there are regions where the Heisenberg interaction is small and the Kitaev interaction is dominant, leading to the Kitaev QSL phase. Because of this, finding Kitaev QSLs in related materials is still encouraging. In the following, we will discuss another Kitaev material α-$RuCl_3$, which has been the focus of recent research due to the availability of high-quality single crystals and the feasibility of neutron scattering measurements.

α-$RuCl_3$ has two-dimensional honeycomb layers formed by the 4d Ru^{3+} ions.[103–108] In fact, realization of the Kitaev interaction in materials with 4d electrons does not sound very promising in the beginning, because their SOCs are smaller compared to those of the 5d systems. However, although the absolute value of the SOC in $RuCl_3$ is smaller, the almost $-90°$ bond angles of the Cl–Ru–Cl bonds of the edge-shared $RuCl_6$ octahedra makes the cubic crystal electric field win and the SOC become a dominant effect.[103–112] Thus, similar to iridates, α-$RuCl_3$ is also an SOC-assisted Mott insulator with an effective spin of 1/2, and the strong spatial anisotropy of the 4d orbitals combined with the SOC makes the bond-dependent Kitaev interaction significant.[18,103,104,113] However, similar to Na_2IrO_3, the ground state of α-$RuCl_3$ is not a Kitaev QSL, but a zigzag magnetic order state instead.[103–108] It has been proposed that the zigzag order is an indication for the presence of the Kitaev interaction in this system.[19,87,105–108] Moreover, INS results indicate that the ground state of α-$RuCl_3$ may be proximate to the Kitaev QSL phase,[114] and both INS[114–117] and Ramman studies[118,119] observe broad continuous magnetic excitations that can be associated with fractionalized excitations resulting from the Kitaev QSL phase.

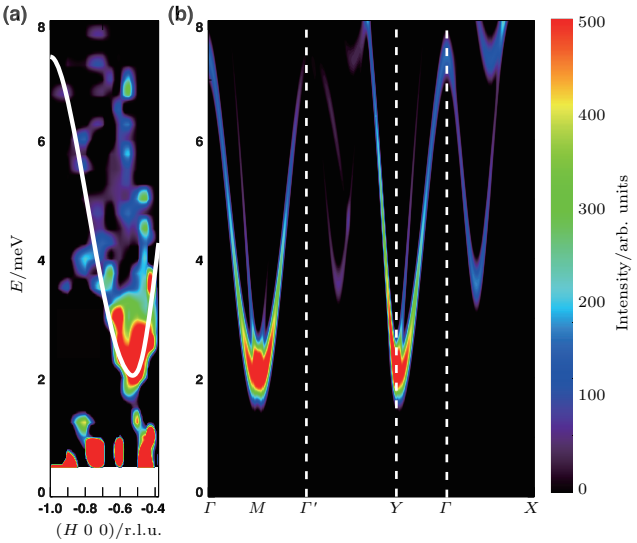


Fig. 10. (color online) (a) Magnetic dispersion along the [100] direction obtained by INS experiments. The solid line is the calculated dispersion as shown in panel (b). (b) Calculated spin-wave spectra along high-symmetry paths. From Ref. [115].

By analyzing the INS spectra, magnetic interactions governing the ground state can be extracted. In Fig. 10(a), we show the spin-wave excitation spectra resulting from the zigzag order state.[115] Instead of using the widely considered Heisenberg–Kitaev (HK) model[19,87] to fit the spectra, a K–Γ effective-spin model is used, where K and Γ represent Kitaev and off-diagonal exchange interactions, respectively. This minimal model describing the ground state of α-$RuCl_3$ is first proposed by Wang *et al.* in Ref. [120]. They recognize the importance of the off-diagonal interactions, but on the other hand, they find that the Heisenberg interactions are at least an order of magnitude smaller than either K or Γ in the parameter range relevant to this material. Fits to the INS

spectra with this model yield a ferromagnetic K of -6.8 meV and a Γ of 9.5 meV,[115] very close to the calculated results of $K = -5.5$ meV and $\Gamma = 7.6$ meV reported in Ref. [120]. These results unambiguously demonstrate that the Kitaev interaction is large and exists in real material.

As mentioned above, the ground state of α-$RuCl_3$ is the zigzag order state instead of a Kitaev QSL. Nevertheless, the magnetic order is rather fragile, with an ordered moment of ~ 0.4 μ_B and an ordering temperature of ~ 8 K.[103–108,115] Such a fragile order can be fully suppressed by either an in-plane magnetic field[22,106,121] or pressure.[123] How do the magnetic excitations behave in the high-field state? Is the high-field disordered state a QSL? If the high-field state is a QSL, what is the relationship between this phase and the long-sought Kitaev QSL? To answer these questions, measurements utilizing various experimental techniques have been carried out.[106,121,122,124–133] Some of these results are discussed in the following.

By following the magnetic-field dependence of the magnetization and specific heat, it is found that the zigzag order is gradually suppressed, and the system becomes a magnetically disordered state at ~ 7.5 T,[124] consistent with earlier reports on the field effect.[106,121,122] NMR spectra on high-quality single crystals also indicate that there is a quantum critical point at $B_c \sim 7.5$ T.[124] Above B_c, the spin-lattice relaxation rate $1/T_1$ of ^{35}Cl shows a power-law behavior as $1/T_1 \sim T^{\alpha}$. In a field range between 8 T and 16 T, $\alpha \approx 3$, suggesting a field-induced QSL featuring Dirac nodal-like spin excitations. A phase diagram summarizing these results is shown in Fig. 11(a). Intensive research on the high-field state utilizing various techniques such as magnetization,[106,121,122,124,132,135] specific heat,[106,124,126,134,135] magnetodielectric,[128] neutron diffraction,[122,126,132] NMR,[124,134] magnetic torque,[127] thermal conductivity,[127,133,135] terahertz spectroscopy,[129,130] and electron spin resonance,[131] has resulted in many somewhat similar phase diagrams, some of which are shown in Fig. 11.

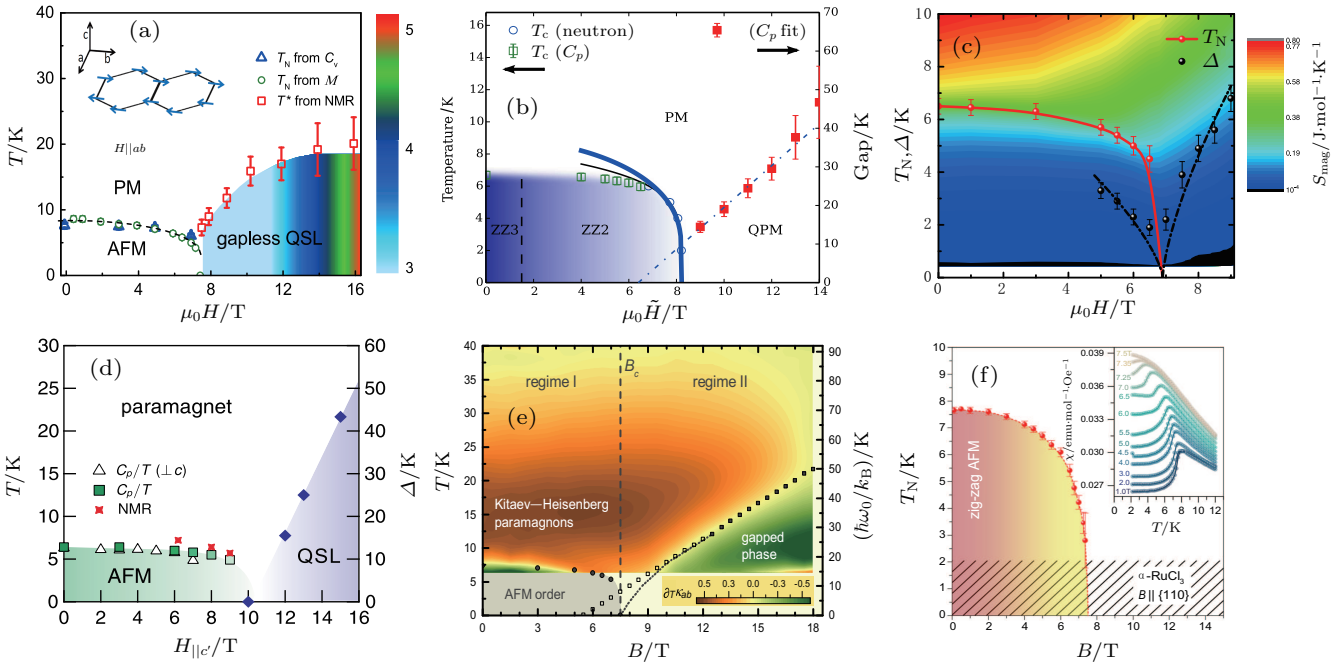


Fig. 11. (color online) Magnetic phase diagram of α-$RuCl_3$ obtained from various measurements. (a) PM and AFM represent paramagnet and zigzag order, respectively. The low field region of the phase diagram is constructed using magnetization and specific heat data, and the high field region is using NMR data. The magnetic field is applied in the a–b plane. The contour map indicates the exponent of the temperature dependence of the spin-lattice relaxation rate. The inset illustrates the zigzag order in the low-field state. From Ref. [124]. (b) ZZ and QPM represent zigzag order and quantum paramagnet, respectively. The phase boundary between ZZ2 and PM is the transition temperature obtained from specific heat and neutron diffraction measurements. The thick solid line is a fit with the transverse-field Ising model, and the thin solid line is a power-law fit. The dashed line is a power-law fit to the gap size Δ. From Ref. [126]. (c) Transition temperatures and gap values obtained from specific heat measurements. The solid line is a guide to the eye. Dashed lines are the fits of the gap function. The magnetic entropy is shown in a color scale. From Ref. [125]. (d) Phase diagram obtained from specific heat and NMR measurements along with the field dependence of the spin gap Δ extracted from the nuclear-spin relaxation rate (right axis). From Ref. [134]. (e) False-color representation of the T derivative of the ab-plane thermal conductivity (κ_{ab}) together with the gap values (solid squares) extracted from the phononic fits. The color scale is in units of W/K^2·m. From Ref. [133]. (f) Magnetic transition temperature as a function of field obtained from susceptibility measurements with field applied along the [110] direction. The inset shows the susceptibility data. From Ref. [132].

On one hand, these phase diagrams all show that the zigzag magnetic order is gradually suppressed by an in-plane magnetic field, and the system reaches a quantum critical point around $B_c \approx 7.5$ T. Furthermore, accumulating evidence suggests that the high-field disordered state above B_c is a QSL. In particular, Banerjee *et al*. have carried out INS measurements to examine the magnetic-field evolution of the magnetic excitations, and some of the results are shown in Fig. 12.[132] They find that the spin-

wave excitations associated with the zigzag order near the M point is suppressed with the field. As shown in Fig. 12(e), above B_c, at $\mu_0 H = 8$ T, excitations near the M point are completely gone, and only excitations near the Γ point remain, similar to the results under zero field above the ordering temperature, as shown in Fig. 12(f). By comparing with calculations, they suggest that the excitations under high fields evidence a field-induced Kitaev QSL state.[132]

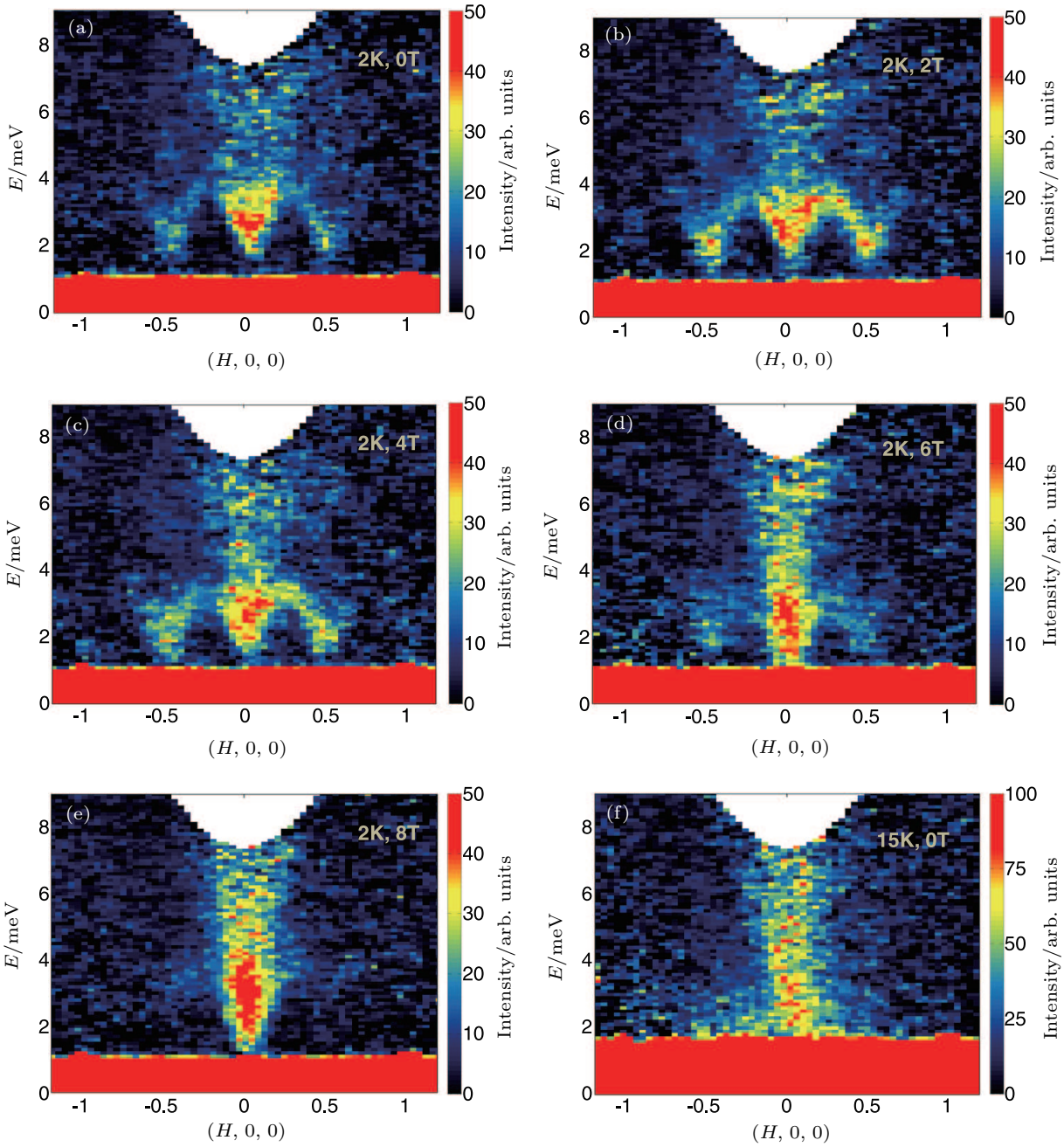


Fig. 12. (color online) (a)–(e) Magnetic-field evolution of the INS spectra along the [100] direction, measured at $T = 2$ K with an external magnetic field applied in the a–b plane. (f) The zero-field data at 15 K, above the ordering temperature. From Ref. [132].

On the other hand, the nature of the field-induced QSL phase, in particular, whether the low-energy magnetic excitations associated with this state are finite,[125,126,130–132,134] is still under debate. To resolve this issue, Yu *et al.*[135] have performed ultralow-temperature thermal conductivity measurements down to 80 mK under magnetic fields, and the results are summarized in Fig. 13. As shown in Fig. 13(a), κ/T increases with field and then decreases to a minimum at the critical field B_c. Above B_c, κ/T increases with field again. These results clearly show that there is a quantum critical point at B_c, consistent with other works.[124–127,130,131,134] By examining the residual κ at zero temperature, it is found that κ_0 is effectively zero in the whole field range probed [Fig. 13(b)]. In the low-field range below B_c, there is a spin-anisotropy gap about 2 meV associated with the zigzag order.[114,115,132] Therefore, absence of the thermal conductivity at zero temperature is ex-

pected. If the high-field state is also fully gapped as suggested in Refs. [125], [126], [130]–[132], [134], these thermal conductivity results are easily explainable. However, as shown in Fig. 11(a), a gapless QSL state near B_c is suggested.[124] We here provide one possible solution to reconcile this discrepancy. According to the report, this gapless state is in fact featured by Dirac-like excitations with gap nodes in the momentum space.[124] In this case, the magnetic density of states, represented as C_m in $\kappa = 1/3C_m v_F l$ is small. At present, the estimated magnetic specific heat C_m has big uncertainties due to the lack of a proper reference sample to subtract the phonon contributions.[106,124–126,134,135] If the real C_m is small, then the gapless state with nodal excitations proposed in Ref. [124] can also be consistent with the thermal conductivity results.[135]

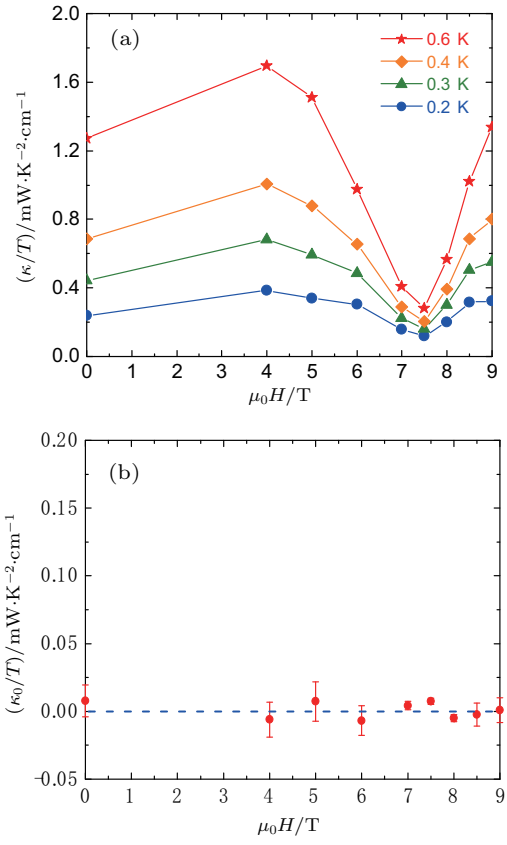


Fig. 13. (color online) (a) Field dependence of κ/T at various temperatures. The minimum of κ/T at ~7.5 T corresponds to B_c, where the zigzag magnetic order disappears. (b) Field dependence of the residual linear term κ_0/T. From Ref. [135].

4. Discussions

As partially reflected from the discussions above, research on the QSL candidates has been quite dynamic. A lot of progress has been made already in recent years. However, it still lacks an ideal QSL candidate so far. Quite often, the spin-"liquid" behavior may have some other origins than quantum fluctuations. Below, we will show some examples.

There is accumulating evidence suggesting $YbMgGaO_4$ to be a promising candidate as a gapless QSL.[43–49] However, the report of no positive contributions from the magnetic excitations to the thermal conductivity is difficult to be reconciled with the gapless QSL picture.[50] One possibility is that the severe disorder effect caused by the random mixing of Mg^{2+} and Ga^{3+} makes the otherwise itinerant spinons localized and thus not conduct heat.[43,44,48,50,52] However, the disorder is considered to be detrimental to the QSL phase for this compound.[53] Ma *et al.*[51] have carried out measurements on $YbZnGaO_4$, a sister compound of $YbMgGaO_4$, utilizing various techniques, including d.c. susceptibility, specific heat, INS, and ultralow-temperature thermal conductivity. They have found that a spin-glass phase can explain the experimental observations in $YbZnGaO_4$: including no long-range magnetic order, prominent broad excitation continua observed by INS, and absence of magnetic thermal conductivity. By analogy, they suggest the spin-glass phase is also applicable to $YbMgGaO_4$.

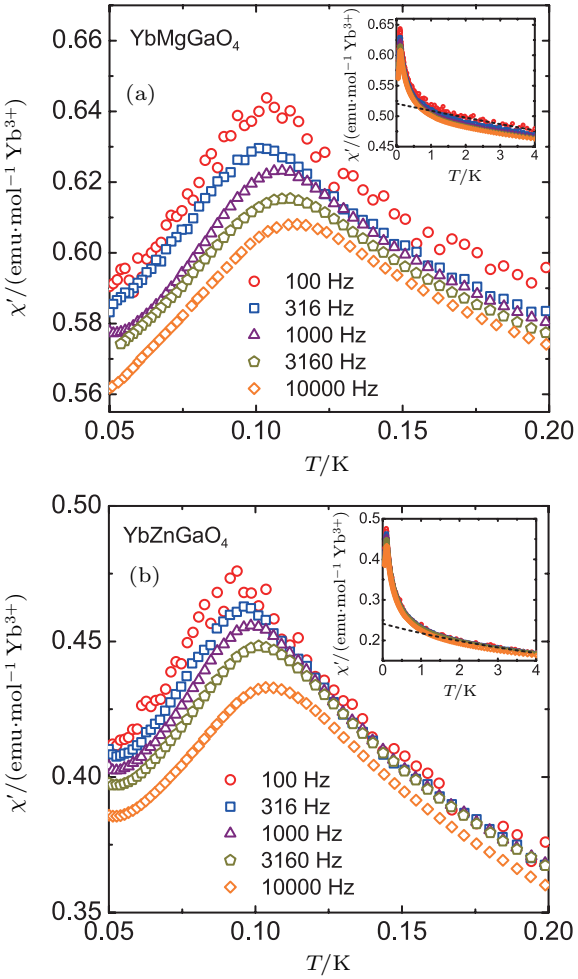


Fig. 14. (color online) (a) and (b) Temperature dependence of the a.c. susceptibility (χ') for $YbMgGaO_4$ and $YbZnGaO_4$, respectively. In the insets, χ' in an extended temperature range up to 4 K are plotted. Dashed lines indicate the Curie–Weiss fits for the 100-Hz data. From Ref. [51].

The spin-glass phase, with frozen, short-range correlations below the freezing temperature T_f,[136–138] can be identified from the a.c. susceptibility. Ma *et al.* have performed

such measurements on both $YbMgGaO_4$ and $YbZnGaO_4$ with temperatures spanning about 3 decades, ranging from 0.05 K to 4 K. Some of the results are shown in Fig. 14. In both compounds, they observe strong frequency-dependent peaks below 0.1 K, evidencing a broad distribution of the spin relaxation times around T_f, typical for a spin glass.[136–142] They consider disorder and frustration to give rise to the spin-glass phase.

However, a μSR study on $YbMgGaO_4$[47] shows that there is no signature of spin freezing down to 0.07 K, which is already below the T_f reported in the a.c. susceptibility measurements.[51] One possible origin of this discrepancy is that these two techniques cover different time scales: the former and latter probes are sensitive to fluctuations with frequencies larger and smaller than 10^4 Hz, respectively.[143] Furthermore, as estimated from the INS results, the portions of the spectral weight in the elastic channel over the total weight, are 16% and 13% for $YbMgGaO_4$ and $YbZnGaO_4$, respectively.[45,51] These roughly represent the portions of moments that have been frozen. All these together may cause difficulties to detect the spin freezing by μSR.

Another important feature for a spin glass is that in the d.c. susceptibility measurements, there should be a cusp at T_f, where the zero-field-cooling and field-cooling susceptibility begin to separate from each other. At present, there are no such data available for $YbMgGaO_4$ and $YbZnGaO_4$, so performing d.c. susceptibility down to temperatures below 0.1 K will be useful to further clarify the ground state of these compounds.

For an organic compound such as κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ and EtMe$_3$Sb[Pd(dmit)$_2$]$_2$, disorder effect is expected to be significant, and it is unclear whether the QSL phase can survive in the presence of strong disorder.[11,21,29–32,144–146] Moreover, as we discuss in Subsection 2.2. for κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, the specific heat indicates a gapless ground state. On the other hand, thermal conductivity measurements reveal no contributions from the magnetic excitations, inconsistent with the gapless QSL state. How to reconcile these contradicting results? Is it because the disorder effect makes the spinons localized and thus not conduct heat? Or the ground state is not a QSL at all? At the current stage, we believe these are open questions, calling for further investigations.

For the most heavily studied kagomé compound, $ZnCu_3(OH)_6Cl_2$, disorder also plays an important role. In particular, there are 5%–15% excess Cu^{2+} replacing the nonmagnetic Zn^{2+}, which induces randomness in the magnetic exchange coupling.[76,147,148] It is believed that such disorder can be accountable for many experimental observations.[76,84,147,149–155] As an example, by considering the Cu impurities, Han *et al.*[85] estimate a spin gap of 0.7 meV in the kagomé layer, close to that obtained from the NMR results.[80]

For QSL candidates, frustration is strong. In the presence of strong disorder, the spin-glass phase is often observed, as disorder and frustration are two important ingredients for a spin glass.[136–138,156] A spin glass mimics a QSL in many aspects — it maintains short-range spin–spin correlations, so in the susceptibility, specific heat, and neutron diffraction measurements, it lacks the signature of a long-range magnetic order; moreover, as demonstrated in Ref. [51], a spin-glass phase can also produce the continuous INS spectra, which is arguably the strongest evidence for a QSL so far. Therefore, in the quest for QSLs, the spin-glass phase which can give rise to spin-liquid-like features must be excluded first before labeling the candidate as a QSL.

Based on discussions above, we now give several perspectives:

(i) Although great progress has been made in theory,[11,21,29–40] it still lacks a proposal for the defining feature of a QSL that can be detected directly from experiments. At present, observations of the continuous magnetic excitation spectra in INS measurements have often been taken to be the most reliable evidence for a QSL.[45,46,82] However, this is necessary but not sufficient evidence for the fractionalized excitations.[51] A feasible direct proposal to identify a QSL should greatly boost this field.

(ii) As we discussed above, there appear to be no ideal QSLs so far. Materials wise, does there exist a QSL candidate with large magnetic exchange interactions, little disorder, and minimal extra interactions that produce the static magnetic order? In the past, most attention had been paid to materials with a triangular or kagomé lattice where strong geometrical frustration is present.[11] Now, studying the SOC-assisted Mott insulators with anisotropic bond-dependent Kitaev interactions on the honeycomb lattice may offer new possibilities.[18,19,105] For instance, very recently, $H_3LiIr_2O_6$ has been suggested to be a Kitaev QSL.[157]

(iii) According to Anderson's proposal, high-temperature superconductivity can emerge from QSLs.[25–27] There have been some successes in making QSL candidates superconducting by applying pressures to some organic compounds.[33,61,69,158,159] However, another more common route to achieve superconductivity — via chemical doping, has not been successful so far.[160] Is it because there is no ideal QSL candidate so far? Will chemically doping an ideal QSL eventually lead to high-temperature superconductivity as predicted? In this aspect, recent advances in doping using electric-field gating may offer some assistance.[161–166]

5. Summary

To summarize, we review the recent progress on QSLs, especially on the magnetic-field measurements on several QSL candidates, including the geometrically-frustrated triangular and kagomé compounds, including $YbMgGaO_4$, $YbZnGaO_4$, κ-$(ET)_2Cu_2(CN)_3$, $EtMe_3Sb[Pd(dmit)_2]_2$, and $ZnCu_3(OH)_6Cl_2$, and the Kitaev material α-$RuCl_3$ with the honeycomb lattice. While there are many experimental evidences showing that they are promising candidates for QSLs, there are also some evidences that may be used to argue against the QSL picture. As such, we provide several perspectives hoping to stimulate further investigations. We anticipate that continuous efforts will be paid off by the discovery of more fascinating physics and ideal candidate materials.

Before ending this review, we note that there are many other materials that have been proposed to be QSLs. We list a few examples below:

(I) $Na_4Ir_3O_8$ is a widely studied QSL candidate with the hyperkagomé lattice.[167–170] In the initial report[167] that suggested it to a QSL, spin freezing indicative of a spin-glass phase at $T_f = 6$ K was observed. The frozen moments were estimated to be less than 10% of the total moments and were thus ignored. Later on, both μSR[169] and NMR measurements[171] showed that the spins are frozen and maintain short-range correlations in the ground state.

(II) Kagomé compounds $ZnCu_3(OH)_6SO_4$[172] and Zn-substituted barlowite $Cu_3Zn(OH)_6FBr$,[173,174] and a hyperkagomé material $PbCuTe_2O_6$.[175,176] In $Cu_3Zn(OH)_6FBr$, it has been shown that the magnetic field dependence of the gap extracted from the NMR data is consistent with that given by fractionalized spin-1/2 spinon excitations.[173]

(III) A triangular spin-1 material $Ba_3NiSb_2O_9$.[177]

(IV) $Ca_{10}Cr_7O_{28}$, a system with complex structure, and more interestingly, with ferromagnetic interactions.[178–180] In this compound, although Balz *et al*. found that there is no static magnetic order in the μSR measurements, they observed frequency dependent peaks in the a.c. susceptibility, which is characteristic of a spin glass.[178] However, they argued that the spin-glass phase could be ruled out by doing the Cole–Cole analysis for the a.c. susceptibility data.[178]

(V) Very recently, a protypical charge-density-wave compound 1T-TaS_2 with the David-star structure has attracted a lot of attention due the possibility of realizing the QSL state.[181–185]

In this review, we did not discuss these materials in detail due to the limited space. Readers who are interested in them can refer to the above references and the references therein.

Acknowledgements

We would like to thank our colleagues and collaborators Jian-Xin Li, Shun-Li Yu, Jun-Ming Liu, Shi-Yan Li, Wei-Qiang Yu, Xin Lu, P Čermák, A Schneidewind, Guochu Deng, S Danilkin, S Yano, and J S Gardner, who made this work possible.

References

[1] Étienne D T d L, Gignoux D and Schlenker M 2005 *Magnetism: Fundamentals* (Berlin: Springer)
[2] White R M 2007 *Quantum Theory of Magnetism: Magnetic Properties of Materials* (Berlin: Springer)
[3] Néel L 1971 *Science* **174** 985
[4] Anderson P W 1959 *Phys. Rev.* **115** 2
[5] White S R and Chernyshev A L 2007 *Phys. Rev. Lett.* **99** 127004
[6] Capriotti L, Trumper A E and Sorella S 1999 *Phys. Rev. Lett.* **82** 3899
[7] Singh R R P and Huse D A 1992 *Phys. Rev. Lett.* **68** 1766
[8] Bernu B, Lecheminant P, Lhuillier C and Pierre L 1994 *Phys. Rev. B* **50** 10048
[9] Bernu B, Lhuillier C and Pierre L 1992 *Phys. Rev. Lett.* **69** 2590
[10] Pomeranchuk I Y 1941 *Zh. Eksp. Teor. Fiz.* **11** 226
[11] Balents L 2010 *Nature* **464** 199
[12] Lee P A 2008 *Science* **321** 1306
[13] Wen X G 2002 *Phys. Rev. B* **65** 165113
[14] Anderson P 1973 *Mater. Res. Bull.* **8** 153
[15] Moessner R and Sondhi S L 2002 *Progr. Theor. Phys. Supplement* **145** 37
[16] Liang S, Doucot B and Anderson P W 1988 *Phys. Rev. Lett.* **61** 365
[17] Fazekas P and Anderson P W 1974 *Philosophical Magazine* **30** 423
[18] Kitaev A 2006 *Ann. Phys.* **321** 2
[19] Jackeli G and Khaliullin G 2009 *Phys. Rev. Lett.* **102** 017205
[20] Kitaev A Y 2003 *Ann. Phys.* **303** 2
[21] Savary L and Balents L 2017 *Reports on Progress in Physics* **80** 016502
[22] Nayak C, Simon S H, Stern A, Freedman M and Das Sarma S 2008 *Rev. Mod. Phys.* **80** 1083
[23] Barkeshli M, Berg E and Kivelson S 2014 *Science* **346** 722
[24] Bednorz J G and Müller K A 1986 *Z. Phys. B* **64** 189
[25] Anderson P W 1987 *Science* **235** 1196
[26] Anderson P W, Baskaran G, Zou Z and Hsu T 1987 *Phys. Rev. Lett.* **58** 2790
[27] Baskaran G, Zou Z and Anderson P 1987 *Solid State Commun.* **63** 973
[28] Kivelson S A, Rokhsar D S and Sethna J P 1987 *Phys. Rev. B* **35** 8865
[29] Kanoda K and Kato R 2011 *Ann. Rev. Condens. Matter Phys.* **2** 167
[30] Witczak-Krempa W, Chen G, Kim Y B and Balents L 2014 *Ann. Rev. Condens. Matter Phys.* **5** 57
[31] Zhou Y, Kanoda K and Ng T K 2017 *Rev. Mod. Phys.* **89** 025003
[32] Starykh O A 2015 *Rep. Progr. Phys.* **78** 052502
[33] Powell B J and McKenzie R H 2011 *Rep. Progr. Phys.* **74** 056501
[34] Kaplan T A and Menyuk N 2007 *Philosophical Magazine* **87** 3711
[35] Norman M R 2016 *Rev. Mod. Phys.* **88** 041002
[36] Rau J G, Lee E K H and Kee H Y 2016 *Ann. Rev. Condens. Matter Phys.* **7** 195
[37] Gardner J S, Gingras M J P and Greedan J E 2010 *Rev. Mod. Phys.* **82** 53
[38] Castelnovo C, Moessner R and Sondhi S L 2012 *Ann. Rev. Condens. Matter Phys.* **3** 35
[39] Mendels P and Bert F 2011 *J. Phys.: Conf. Ser.* **320** 012004
[40] Mendels P and Bert F 2010 *J. Phys. Soc. Jpn.* **79** 011001
[41] Imai T and Lee Y S 2016 *Physics Today* **69** 30
[42] Winter S M, Tsirlin A A, Daghofer M, van den Brink J, Singh Y, Gegenwart P and Valentí R 2017 *J. Physics: Condens. Matter* **29** 493002
[43] Li Y, Liao H, Zhang Z, Li S, Jin F, Ling L, Zhang L, Zou Y, Pi L, Yang Z, Wang J, Wu Z and Zhang Q 2015 *Sci. Rep.* **5** 16419
[44] Li Y, Chen G, Tong W, Pi L, Liu J, Yang Z, Wang X and Zhang Q 2015 *Phys. Rev. Lett.* **115** 167203
[45] Paddison J A M, Daum M, Dun Z, Ehlers G, Liu Y, Stone M B, Zhou H and Mourigal M 2017 *Nat. Phys.* **13** 117

[46] Shen Y, Li Y D, Wo H, Li Y, Shen S, Pan B, Wang Q, Walker H C, Steffens P, Boehm M, Hao Y, Quintero-Castro D L, Harriger L W, Frontzek M D, Hao L, Meng S, Zhang Q, Chen G and Zhao J 2016 *Nature* **540** 559

[47] Li Y, Adroja D, Biswas P K, Baker P J, Zhang Q, Liu J, Tsirlin A A, Gegenwart P and Zhang Q 2016 *Phys. Rev. Lett.* **117** 097201

[48] Li Y, Adroja D, Bewley R I, Voneshen D, Tsirlin A A, Gegenwart P and Zhang Q 2017 *Phys. Rev. Lett.* **118** 107202

[49] Li Y, Adroja D, Voneshen D, Bewley R I, Zhang Q, Tsirlin A A and Gegenwart P 2017 *Nat. Commun.* **8** 15814

[50] Xu Y, Zhang J, Li Y S, Yu Y J, Hong X C, Zhang Q M and Li S Y 2016 *Phys. Rev. Lett.* **117** 267202

[51] Ma Z, Wang J, Dong Z Y, Zhang J, Li S, Zheng S H, Yu Y, Wang W, Che L, Ran K, Bao S, Cai Z, Čermaḱ P, Schneidewind A, Yano S, Gardner J S, Lu X, Yu S L, Liu J M, Li S, Li J X and Wen J 2018 *Phys. Rev. Lett.* **120** 087201

[52] Balents L 2016 *Nature* **540** 534

[53] Zhu Z, Maksimov P A, White S R and Chernyshev A L 2017 *Phys. Rev. Lett.* **119** 157201

[54] Yamashita M, Nakata N, Senshu Y, Nagata M, Yamamoto H M, Kato R, Shibauchi T and Matsuda Y 2010 *Science* **328** 1246

[55] Nave C P and Lee P A 2007 *Phys. Rev. B* **76** 235124

[56] Motrunich O I 2005 *Phys. Rev. B* **72** 045105

[57] Kino H and Fukuyama H 1995 *J. Phys. Soc. Jpn.* **64** 2726

[58] Shimizu Y, Miyagawa K, Kanoda K, Maesato M and Saito G 2005 *Progress of Theoretical Physics Supplement* **159** 52

[59] Komatsu T, Matsukawa N, Inoue T and Saito G 1996 *J. Phys. Soc. Jpn.* **65** 1340

[60] Shimizu Y, Miyagawa K, Kanoda K, Maesato M and Saito G 2003 *Phys. Rev. Lett.* **91** 107001

[61] Kurosaki Y, Shimizu Y, Miyagawa K, Kanoda K and Saito G 2005 *Phys. Rev. Lett.* **95** 177001

[62] Ohira S, Shimizu Y, Kanoda K and Saito G 2006 *J. Low Temp. Phys.* **142** 153

[63] Furukawa T, Kobashi K, Kurosaki Y, Miyagawa K and Kanoda K 2018 *Nat. Commun.* **9** 307

[64] Miyagawa K, Kanoda K and Kawamoto A 2004 *Chem. Rev.* **104** 5635

[65] Pratt F L, Baker P J, Blundell S J, Lancaster T, Ohira-Kawamura S, Baines C, Shimizu Y, Kanoda K, Watanabe I and Saito G 2011 *Nature* **471** 612

[66] Yamashita S, Nakazawa Y, Oguni M, Oshima Y, Nojiri H, Shimizu Y, Miyagawa K and Kanoda K 2008 *Nat. Phys.* **4** 459

[67] Yamashita M, Nakata N, Kasahara Y, Sasaki T, Yoneyama N, Kobayashi N, Fujimoto S, Shibauchi T and Matsuda Y 2009 *Nat. Phys.* **5** 44

[68] Kato R 2014 *Bull. Chem. Soc. Jpn.* **87** 355

[69] Shimizu Y, Akimoto H, Tsujii H, Tajima A and Kato R 2007 *Phys. Rev. Lett.* **99** 256403

[70] Itou T, Oyamada A, Maegawa S and Kato R 2010 *Nat. Phys.* **6** 673

[71] Yamashita S, Yamamoto T, Nakazawa Y, Tamura M and Kato R 2011 *Nat. Commun.* **2** 275

[72] Shimizu Y, Akimoto H, Tsujii H, Tajima A and Kato R 2007 *J. Phys. Condens. Matter* **19** 145240

[73] Tamura M, Nakao A and Kato R 2006 *J. Phys. Soc. Jpn.* **75** 093701

[74] Nytko E A, Helton J S, Müller P and Nocera D G 2008 *J. Am. Chem. Soc.* **130** 2922

[75] Shores M P, Nytko E A, Bartlett B M, and Nocera D G 2005 *J. Am. Chem. Soc.* **127** 13462

[76] Han T H, Helton J S, Chu S, Prodi A, Singh D K, Mazzoli C, Müller P, Nocera D G and Lee Y S 2011 *Phys. Rev. B* **83** 100402

[77] Helton J S, Matan K, Shores M P, Nytko E A, Bartlett B M, Yoshida Y, Takano Y, Suslov A, Qiu Y, Chung J H, Nocera D G and Lee Y S 2007 *Phys. Rev. Lett.* **98** 107204

[78] Mendels P, Bert F, de Vries M A, Olariu A, Harrison A, Duc F, Trombe J C, Lord J S, Amato A and Baines C 2007 *Phys. Rev. Lett.* **98** 077204

[79] de Vries M A, Stewart J R, Deen P P, Piatek J O, Nilsen G J, Rønnow H M and Harrison A 2009 *Phys. Rev. Lett.* **103** 237201

[80] Fu M, Imai T, Han T H and Lee Y S 2015 *Science* **350** 655

[81] Jeong M, Bert F, Mendels P, Duc F, Trombe J C, de Vries M A and Harrison A 2011 *Phys. Rev. Lett.* **107** 237201

[82] Han T H, Helton J S, Chu S, Nocera D G, Rodriguez-Rivera J A, Broholm C and Lee Y S 2012 *Nature* **492** 406

[83] Yan S, Huse D A and White S R 2011 *Science* **332** 1173

[84] Asaba T, Han T H, Lawson B J, Yu F, Tinsman C, Xiang Z, Li G, Lee Y S and Li L 2014 *Phys. Rev. B* **90** 064417

[85] Han T H, Norman M R, Wen J J, Rodriguez-Rivera J A, Helton J S, Broholm C and Lee Y S 2016 *Phys. Rev. B* **94** 060409

[86] Liao H J, Xie Z Y, Chen J, Liu Z Y, Xie H D, Huang R Z, Normand B and Xiang T 2017 *Phys. Rev. Lett.* **118** 137202

[87] Chaloupka J, Jackeli G and Khaliullin G 2010 *Phys. Rev. Lett.* **105** 027204

[88] Shitade A, Katsura H, Kuneš J, Qi X L, Zhang S C and Nagaosa N 2009 *Phys. Rev. Lett.* **102** 256403

[89] Chaloupka J, Jackeli G and Khaliullin G 2013 *Phys. Rev. Lett.* **110** 097204

[90] Yamaji Y, Suzuki T, Yamada T, Suga S i, Kawashima N and Imada M 2016 *Phys. Rev. B* **93** 174425

[91] Mehlawat K, Thamizhavel A and Singh Y 2017 *Phys. Rev. B* **95** 144406

[92] Modic K A, Smidt T E, Kimchi I, Breznay N P, Biffin A, Choi S, Johnson R D, Coldea R, Watkins-Curry P, McCandless G T, Chan J Y, Gandara F, Islam Z, Vishwanath A, Shekhter A, McDonald R D and Analytis J G 2014 *Nat. Commun.* **5** 4203

[93] Takayama T, Kato A, Dinnebier R, Nuss J, Kono H, Veiga L S I, Fabbris G, Haskel D and Takagi H 2015 *Phys. Rev. Lett.* **114** 077202

[94] Williams S C, Johnson R D, Freund F, Choi S, Jesche A, Kimchi I, Manni S, Bombardi A, Manuel P, Gegenwart P and Coldea R 2016 *Phys. Rev. B* **93** 195158

[95] Glamazda A, Lemmens P, Do S H, Choi Y S and Choi K Y 2016 *Nat. Commun.* **7** 12286

[96] Gretarsson H, Clancy J P, Liu X, Hill J P, Bozin E, Singh Y, Manni S, Gegenwart P, Kim J, Said A H, Casa D, Gog T, Upton M H, Kim H S, Yu J, Katukuri V M, Hozoi L, van den Brink J and Kim Y J 2013 *Phys. Rev. Lett.* **110** 076402

[97] Comin R, Levy G, Ludbrook B, Zhu Z H, Veenstra C N, Rosen J A, Singh Y, Gegenwart P, Stricker D, Hancock J N, van der Marel D, Elfimov I S and Damascelli A 2012 *Phys. Rev. Lett.* **109** 266406

[98] Singh Y and Gegenwart P 2010 *Phys. Rev. B* **82** 064412

[99] Liu X, Berlijn T, Yin W G, Ku W, Tsvelik A, Kim Y J, Gretarsson H, Singh Y, Gegenwart P and Hill J P 2011 *Phys. Rev. B* **83** 220403

[100] Choi S K, Coldea R, Kolmogorov A N, Lancaster T, Mazin I I, Blundell S J, Radaelli P G, Singh Y, Gegenwart P, Choi K R, Cheong S W, Baker P J, Stock C and Taylor J 2012 *Phys. Rev. Lett.* **108** 127204

[101] Ye F, Chi S, Cao H, Chakoumakos B C, Fernandez-Baca J A, Custelcean R, Qi T F, Korneta O B and Cao G 2012 *Phys. Rev. B* **85** 180403

[102] Singh Y, Manni S, Reuther J, Berlijn T, Thomale R, Ku W, Trebst S and Gegenwart P 2012 *Phys. Rev. Lett.* **108** 127203

[103] Fletcher J M, Gardner W E, Hooper E W, Hyde K R, Moore F H and Woodhead J L 1963 *Nature* **199** 1089

[104] Fletcher J M, Gardner W E, Fox A C and Topping G 1967 *J. Chem. Soc. A* 1038

[105] Plumb K W, Clancy J P, Sandilands L J, Shankar V V, Hu Y F, Burch K S, Kee H Y and Kim Y J 2014 *Phys. Rev. B* **90** 041112

[106] Kubota Y, Tanaka H, Ono T, Narumi Y and Kindo K 2015 *Phys. Rev. B* **91** 094422

[107] Sears J A, Songvilay M, Plumb K W, Clancy J P, Qiu Y, Zhao Y, Parshall D and Kim Y J 2015 *Phys. Rev. B* **91** 144420

[108] Cao H B, Banerjee A, Yan J Q, Bridges C A, Lumsden M D, Mandrus D G, Tennant D A, Chakoumakos B C and Nagler S E 2016 *Phys. Rev. B* **93** 134423

[109] Guizzetti G, Reguzzoni E and Pollini I 1979 *Phys. Lett. A* **70** 34

[110] Binotto L, Pollini I and Spinolo G 1971 *Physica Status Solidi* **44** 245

[111] Pollini I 1996 *Phys. Rev. B* **53** 12769

[112] Pollini I 1994 *Phys. Rev. B* **50** 2095

[113] Hou Y S, Xiang H J and Gong X G 2017 *Phys. Rev. B* **96** 054410

[114] Banerjee A, Bridges C, Yan J, Aczel A, Li L, Stone M, Granroth G, Lumsden M, Yiu Y, Knolle J, *et al.* 2016 *Nat. Mater.* **15** 733

[115] Ran K, Wang J, Wang W, Dong Z Y, Ren X, Bao S, Li S, Ma Z, Gan Y, Zhang Y, Park J T, Deng G, Danilkin S, Yu S L, Li J X and Wen J 2017 *Phys. Rev. Lett.* **118** 107203

[116] Banerjee A, Yan J, Knolle J, Bridges C A, Stone M B, Lumsden M D, Mandrus D G, Tennant D A, Moessner R and Nagler S E 2017 *Science* **356** 1055

[117] Do S H, Park S Y, Yoshitake J, Nasu J, Motome Y, Kwon Y S, Adroja D T, Voneshen D J, Kim K, Jang T H, Park J H, Choi K Y and Ji S 2017 *Nat. Phys.* **13** 1079

[118] Sandilands L J, Tian Y, Plumb K W, Kim Y J and Burch K S 2015 *Phys. Rev. Lett.* **114** 147201
[119] Nasu J, Knolle J, Kovrizhin D L, Motome Y and Moessner R 2016 *Nat. Phys.* **12** 912
[120] Wang W, Dong Z Y, Yu S L and Li J X 2017 *Phys. Rev. B* **96** 115103
[121] Majumder M, Schmidt M, Rosner H, Tsirlin A A, Yasuoka H and Baenitz M 2015 *Phys. Rev. B* **91** 180401
[122] Johnson R D, Williams S C, Haghighirad A A, Singleton J, Zapf V, Manuel P, Mazin I I, Li Y, Jeschke H O, Valentí R and Coldea R 2015 *Phys. Rev. B* **92** 235119
[123] Cui Y, Zheng J, Ran K, Wen J, Liu Z X, Liu B, Guo W and Yu W 2017 *Phys. Rev. B* **96** 205147
[124] Zheng J, Ran K, Li T, Wang J, Wang P, Liu B, Liu Z X, Normand B, Wen J and Yu W 2017 *Phys. Rev. Lett.* **119** 227208
[125] Wolter A U B, Corredor L T, Janssen L, Nenkov K, Schönecker S, Do S H, Choi K Y, Albrecht R, Hunger J, Doert T, Vojta M and Büchner B 2017 *Phys. Rev. B* **96** 041405
[126] Sears J A, Zhao Y, Xu Z, Lynn J W and Kim Y J 2017 *Phys. Rev. B* **95** 180411
[127] Leahy I A, Pocs C A, Siegfried P E, Graf D, Do S H, Choi K Y, Normand B and Lee M 2017 *Phys. Rev. Lett.* **118** 187203
[128] Aoyama T, Hasegawa Y, Kimura S, Kimura T and Ohgushi K 2017 *Phys. Rev. B* **95** 245104
[129] Little A, Wu L, Lampen-Kelley P, Banerjee A, Patankar S, Rees D, Bridges C A, Yan J Q, Mandrus D, Nagler S E and Orenstein J 2017 *Phys. Rev. Lett.* **119** 227201
[130] Wang Z, Reschke S, Hüvonen D, Do S H, Choi K Y, Gensch M, Nagel U, Room T and Loidl A 2017 *Phys. Rev. Lett.* **119** 227202
[131] Ponomaryov A N, Schulze E, Wosnitza J, Lampen-Kelley P, Banerjee A, Yan J Q, Bridges C A, Mandrus D G, Nagler S E, Kolezhuk A K and Zvyagin S A 2017 *Phys. Rev. B* **96** 241107
[132] Banerjee A, Lampen-Kelley P, Knolle J, Balz C, Aczel A A, Winn B, Liu Y, Pajerowski D, Yan J, Bridges C A, Savici A T, Chakoumakos B C, Lumsden M D, Tennant D A, Moessner R, Mandrus D G and Nagler S E 2018 *npj Quantum Materials* **3** 8
[133] Hentrich R, Wolter A U B, Zotos X, Brenig W, Nowak D, Isaeva A, Doert T, Banerjee A, Lampen-Kelley P, Mandrus D G, Nagler S E, Sears J, Kim Y J, Büchner B and Hess C 2018 *Phys. Rev. Lett.* **120** 117204
[134] Baek S H, Do S H, Choi K Y, Kwon Y S, Wolter A U B, Nishimoto S, van den Brink J and Büchner B 2017 *Phys. Rev. Lett.* **119** 037201
[135] Yu Y J, Xu Y, Ran K J, Ni J M, Huang Y Y, Wang J H, Wen J S and Li S Y 2018 *Phys. Rev. Lett.* **120** 067202
[136] Binder K and Young A P 1986 *Rev. Mod. Phys.* **58** 801
[137] Mydosh J A 1993 *Spin glasses: an experimental introduction* (London: Taylor & Francis)
[138] Mydosh J A 1986 *Hyperfine Interactions* **31** 347
[139] Hüser D, Wenger L E, van Duyneveldt A J and Mydosh J A 1983 *Phys. Rev. B* **27** 3100
[140] Mulder C A M, van Duyneveldt A J and Mydosh J A 1981 *Phys. Rev. B* **23** 1384
[141] Eiselt G, Kötzler J, Maletta H, Stauffer D and Binder K 1979 *Phys. Rev. B* **19** 2664
[142] Mehlawat K, Sharma G and Singh Y 2015 *Phys. Rev. B* **92** 134412
[143] Sonier J E 2002 http://phys.columbia.edu/fcmp/files//FCMP2015_MuSR_Brochure.pdf
[144] Watanabe K, Kawamura H, Nakano H and Sakai T 2014 *J. Phys. Soc. Jpn.* **83** 034714
[145] Kawamura H, Watanabe K and Shimokawa T 2014 *J. Phys. Soc. Jpn.* **83** 103704
[146] Shimokawa T, Watanabe K and Kawamura H 2015 *Phys. Rev. B* **92** 134407
[147] Freedman D E, Han T H, Prodi A, Müller P, Huang Q Z, Chen Y S, Webb S M, Lee Y S, McQueen T M and Nocera D G 2010 *J. Am. Chem. Soc.* **132** 16185
[148] Han T, Chu S and Lee Y S 2012 *Phys. Rev. Lett.* **108** 157202
[149] Olariu A, Mendels P, Bert F, Duc F, Trombe J C, de Vries M A and Harrison A 2008 *Phys. Rev. Lett.* **100** 087202
[150] Chitra R and Rozenberg M J 2008 *Phys. Rev. B* **77** 052407
[151] Imai T, Nytko E A, Bartlett B M, Shores M P and Nocera D G 2008 *Phys. Rev. Lett.* **100** 077203
[152] de Vries M A, Kamenev K V, Kockelmann W A, Sanchez-Benitez J and Harrison A 2008 *Phys. Rev. Lett.* **100** 157205
[153] Bert F, Nakamae S, Ladieu F, L'Hôte D, Bonville P, Duc F, Trombe J C and Mendels P 2007 *Phys. Rev. B* **76** 132411
[154] Lee S H, Kikuchi H, Qiu Y, Lake B, Huang Q, Habicht K and Kiefer K 2007 *Nat. Mater.* **6** 853
[155] Rozenberg M J and Chitra R 2008 *Phys. Rev. B* **78** 132406
[156] Wu J, Wildeboer J S, Werner F, Seidel A, Nussinov Z and Solin S A 2011 *Europhys. Lett.* **93** 67001
[157] Kitagawa K, Takayama T, Matsumoto Y, Kato A, Takano R, Kishimoto Y, Bette S, Dinnebier R, Jackeli G and Takagi H 2018 *Nature* **554** 341
[158] Lefebvre S, Wzietek P, Brown S, Bourbonnais C, Jérome D, Méziére C, Fourmigué M and Batail P 2000 *Phys. Rev. Lett.* **85** 5420
[159] Kato R, Tajima A, Nakao A and Tamura M 2006 *J. Am. Chem. Soc.* **128** 10016
[160] Kelly Z A, Gallagher M J and McQueen T M 2016 *Phys. Rev. X* **6** 041007
[161] Ueno K, Shimotani H, Yuan H, Ye J, Kawasaki M and Iwasa Y 2014 *J. Phys. Soc. Jpn.* **83** 032001
[162] Lei B, Cui J H, Xiang Z J, Shang C, Wang N Z, Ye G J, Luo X G, Wu T, Sun Z and Chen X H 2016 *Phys. Rev. Lett.* **116** 077002
[163] Bollinger A T, Dubuis G, Yoon J, Pavuna D, Misewich J and Božović I 2011 *Nature* **472** 458
[164] Lu N, Zhang P, Zhang Q, Qiao R, He Q, Li H B, Wang Y, Guo J, Zhang D, Duan Z, Li Z, Wang M, Yang S, Yan M, Arenholz E, Zhou S, Yang W, Gu L, Nan C W, Wu J, Tokura Y and Yu P 2017 *Nature* **546** 124
[165] Cui Y, Zhang G, Li H, Lin H, Zhu X, Wen H H, Wang G, Sun J, Ma M, Li Y, Gong D, Xie T, Gu Y, Li S, Luo H, Yu P and Yu W 2018 *Sci. Bull.* **63** 11
[166] Hosono H 2018 *Sci. Bull.* **63** 5
[167] Okamoto Y, Nohara M, Aruga-Katori H and Takagi H 2007 *Phys. Rev. Lett.* **99** 137207
[168] Singh Y, Tokiwa Y, Dong J and Gegenwart P 2013 *Phys. Rev. B* **88** 220413
[169] Dally R, Hogan T, Amato A, Luetkens H, Baines C, Rodriguez-Rivera J, Graf M J and Wilson S D 2014 *Phys. Rev. Lett.* **113** 247601
[170] Rehn J, Sen A and Moessner R 2017 *Phys. Rev. Lett.* **118** 047201
[171] Shockley A C, Bert F, Orain J C, Okamoto Y and Mendels P 2015 *Phys. Rev. Lett.* **115** 047201
[172] Li Y, Pan B, Li S, Tong W, Ling L, Yang Z, Wang J, Chen Z, Wu Z and Zhang Q 2014 *New J. Phys.* **16** 093011
[173] Feng Z, Li Z, Meng X, Yi W, Wei Y, Zhang J, Wang Y C, Jiang W, Liu Z and Li S 2017 *Chin. Phys. Lett.* **34** 077502
[174] Wen X G 2017 *Chin. Phys. Lett.* **34** 090101
[175] Koteswararao B, Kumar R, Khuntia P, Bhowal S, Panda S K, Rahman M R, Mahajan A V, Dasgupta I, Baenitz M, Kim K H and Chou F C 2014 *Phys. Rev. B* **90** 035141
[176] Khuntia P, Bert F, Mendels P, Koteswararao B, Mahajan A V, Baenitz M, Chou F C, Baines C, Amato A and Furukawa Y 2016 *Phys. Rev. Lett.* **116** 107203
[177] Fåk B, Bieri S, Canévet E, Messio L, Payen C, Viaud M, Guillot-Deudon C, Darie C, Ollivier J and Mendels P 2017 *Phys. Rev. B* **95** 060402
[178] Balz C, Lake B, Reuther J, Luetkens H, Schonemann R, Herrmannsdorfer T, Singh Y, Nazmul Islam A T M, Wheeler E M, Rodriguez-Rivera J A, Guidi T, Simeoni G G, Baines C and Ryll H 2016 *Nature Phys.* **12** 942
[179] Balz C, Lake B, Nazmul Islam A T M, Singh Y, Rodriguez-Rivera J A, Guidi T, Wheeler E M, Simeoni G G and Ryll H 2017 *Phys. Rev. B* **95** 174414
[180] Balz C, Lake B, Reehuis M, Islam A T M N, Prokhnenko O, Singh Y, Pattison P and Tóth S 2017 *J. Phys.: Condens. Matter* **29** 225802
[181] Law K T and Lee P A 2017 *Proceedings of the National Academy of Sciences* **114** 6996
[182] Kratochvilova M, Hillier A D, Wildes A R, Wang L, Cheong S W and Park J G 2017 *npj Quantum Materials* **2** 42
[183] Klanjšek M, Zorko A, Žitko R, Mravlje J, Jagličić Z, Biswas P K, Prelovšek P, Mihailovic D and Arčon D 2017 *Nat. Phys.* **13** 1130
[184] Ribak A, Silber I, Baines C, Chashka K, Salman Z, Dagan Y and Kanigel A 2017 *Phys. Rev. B* **96** 195131
[185] Yu Y J, Xu Y, He L P, Kratochvilova M, Huang Y Y, Ni J M, Wang L, Cheong S W, Park J G and Li S Y 2017 *Phys. Rev. B* **96** 081111