Rapid Communications

Residual entropy and spin fractionalizations in the mixed-spin Kitaev model

Akihisa Koga[1] and Joji Nasu[2]

[1]*Department of Physics, Tokyo Institute of Technology, Meguro, Tokyo 152- 8551, Japan*
[2]*Department of Physics, Yokohama National University, Hodogaya, Yokohama 240-8501, Japan*

(Received 10 June 2019; revised manuscript received 6 August 2019; published 9 September 2019)

We investigate the ground-state and finite-temperature properties of the mixed-spin (s, S) Kitaev model. When one of the spins is a half integer and the other is an integer, we introduce two kinds of local symmetries, which results in a macroscopic degeneracy in each energy level. Applying the exact diagonalization to several clusters with $(s, S) = (1/2, 1)$, we confirm the presence of this large degeneracy in the ground states, in contrast to conventional Kitaev models. By means of the thermal pure quantum state technique, we calculate the specific heat, entropy, and spin-spin correlations in the system. We find that in the mixed-spin Kitaev model with $(s, S) = (1/2, 1)$, at least, the double-peak structure appears in the specific heat and the plateau in the entropy at intermediate temperatures, indicating the existence of spin fractionalization. Deducing the entropy in the mixed-spin system with $s, S \leqslant 2$ systematically, we clarify that the smaller spin s is responsible for the thermodynamic properties at higher temperatures.

DOI: 10.1103/PhysRevB.100.100404

The Kitaev model [1] and its related models have attracted much interest in condensed matter physics since the possibility of direction-dependent Ising interactions has been proposed in realistic materials [2]. Among them, the low-temperature properties in candidate materials such as A_2IrO_3 $(A = \mathrm{Na, K})$ [3–9] and α-$RuCl_3$ [10–14] have been examined extensively. To clarify the experimental results, the roles of the Heisenberg interactions [15–17], off-diagonal interactions [18,19], interlayer coupling [20–22], and the spin-orbit couplings [23] have been theoretically investigated for both ground-state and finite-temperature properties. One of the important issues characteristic of the Kitaev models is the fractionalization of the spin degree of freedom. In the Kitaev model with $S = 1/2$ spins, the spins are exactly shown to be fractionalized into itinerant Majorana fermions and localized fluxes, which manifest themselves in the ground-state and thermodynamic properties [24,25]. It has been observed as half-quantized thermal quantum Hall effects, which is clear evidence of Majorana quasiparticles fractionalized from quantum spins [14]. Recently, the Kitaev model with larger spins has theoretically been examined [26–31]. In the spin-S Kitaev model, the specific heat exhibits a double-peak structure, and a plateau appears in the temperature dependence of the entropy [28]. This suggests the existence of fractionalization even in this generalized Kitaev model. However, it is still hard to explain how the spin degree of freedom is divided in generalized Kitaev models beyond the exactly solvable $S = 1/2$ case [1,24,25].

The key to understanding "fractionalization" in the spin-S Kitaev model should be the multiple entropy release phenomenon. Half of the spin entropy $\sim\frac{1}{2}\ln(2S+1)$ in higher temperatures emerges with a broad peak in the specific heat. Then, a question arises as to how the plateau structure appears in the entropy in the Kitaev model composed of multiple kinds of spins (the mixed-spin Kitaev model). In other words, how is the many-body state realized in a system with decreasing temperatures? The extension to mixed-spin models should be a potential to exhibit an intriguing nature of the ground states. In fact, a mixed-spin quantum Heisenberg model has been examined [32–39], and the topological nature of the spins and lattice plays an important role in stabilizing the nonmagnetic ground states. Moreover, a mixed-spin Kitaev model can be realized by replacing transition-metal ions with other ions in the Kitaev candidate materials. Therefore, it is desired to study this model to discuss the nature of spin fractionalization in the Kitaev system.

In this Rapid Communication, we investigate the mixed-spin Kitaev model, where two distinct spins (s, S) $(s < S)$ are periodically arranged on a honeycomb lattice (see Fig. 1). First, we show the existence of Z_2 symmetry in each plaquette in the system. In addition, by considering another local symmetry, we show that macroscopic degeneracy exists in each energy level when one of the spins is a half integer and the other an integer. The exact diagonalization (ED) in the system with $(s, S) = (\frac{1}{2}, 1)$ reveals that the ground state has a macroscopic degeneracy, which is consistent with the presence of the two kinds of local symmetries. Using thermal pure quantum (TPQ) state methods [40,41], we find that, at least, the double-peak structure appears in the specific heat and the plateau appears at intermediate temperatures in the entropy, which are similar to those in the spin-S Kitaev models [28]. From systematic calculations of mixed-spin systems with $s, S \leqslant 2$, we clarify that the smaller spin s is responsible for the high-temperature properties. The deconfinement picture to explain the "spin fractionalization" in the Kitaev model is addressed.

We consider the Kitaev model on a honeycomb lattice, which is given by the following Hamiltonian as

$$\mathcal{H} = -J\sum_{\langle i,j\rangle_x} s_i^x S_j^x - J\sum_{\langle i,j\rangle_y} s_i^y S_j^y - J\sum_{\langle i,j\rangle_z} s_i^z S_j^z, \qquad (1)$$

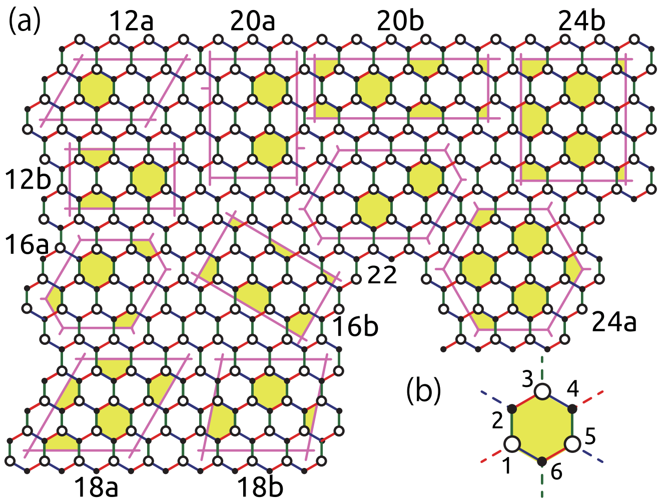

FIG. 1. (a) Mixed-spin Kitaev model on a honeycomb lattice. Solid (open) circles represent spin s (S). Red, blue, and green lines denote x, y, and z bonds between nearest-neighbor sites, respectively. (b) Plaquette with sites marked 1–6 is shown for the corresponding operator W_p defined in Eq. (2).

where s_i^α (S_i^α) is the α (=x, y, z) component of a spin-s(S) operator at the ith site. J is the exchange constant between the nearest-neighbor spin pairs $\langle i, j\rangle_\gamma$. The model is schematically shown in Fig. 1(a). We consider here the following local Hermite operator defined on each plaquette p as

$$W_p = \exp\left[i\pi\left(S_1^x + s_2^y + S_3^z + s_4^x + S_5^y + s_6^z\right) - i\pi\eta\right], \qquad (2)$$

where $\eta = [3(s+S)]$ is a phase factor. By using the following relation for the spin operators, $e^{i\pi S^\alpha} S^\beta e^{-i\pi S^\alpha} = (2\delta_{\alpha\beta} - 1)S^\beta$, we find $[\mathcal{H}, W_p] = 0$ for each plaquette and $W_p^2 = 1$. Therefore, the mixed-spin Kitaev system has Z_2 local symmetry.

It is known that this local Z_2 symmetry is important to understand the ground-state properties in the Kitaev model. We wish to note that the local operator W_p on a plaquette p commutes with those on all other plaquettes in the spin-S Kitaev models, while this commutation relation is not always satisfied in the present mixed-spin Kitaev model. In fact, we obtain $[W_p, W_q] \propto [e^{i\pi(s_i^x + S_j^y)}, e^{i\pi(s_i^y + S_j^x)}] \propto \sin[\pi(s_i^y - S_j^x)]$ when the plaquettes p and q share the same z bond $\langle ij\rangle_z$. This means that the local operator does not commute with the adjacent ones in the mixed-spin Kitaev model with one of the spins being a half integer and the other an integer. Instead, we introduce another local symmetry specific in this case. When either s or S is a half integer and the other is an integer, the Hilbert space is divided into subspaces specified by the set of the eigenvalues w_p ($=\pm 1$) of the N_p ($\leqslant N/6$) local operators W_p defined on the plaquettes $p \in \mathcal{P}$, where $\mathcal{P}$ is a set of plaquettes whose corners are not shared with each other. Now, we assume the presence of the local operator R_p on a plaquette p ($\in\mathcal{P}$) so as to satisfy the conditions $R_p^2 = 1$ and the following commutation relations $[\mathcal{H}, R_p] = 0$, $[W_p, R_q] = 0$ ($p \neq q$), and $\{W_p, R_p\} = 0$. In the case that half-integer and integer spins are mixed, such an operator can be introduced so that the spins located on its corners are inverted as $\mathbf{S}_{2i-1} \to -\mathbf{S}_{2i-1}$, $\mathbf{s}_{2i} \to -\mathbf{s}_{2i}$ ($i = 1, 2, 3$) and the signs of the six exchange constants are inverted on the bonds connecting between the plaquette p and its outside sites, shown as the dashed lines in Fig. 1(b). When a wave function for the energy level E is given by the set of $\{w_p\}$ as $|\psi\rangle = |\psi; \{w_1, w_2, \ldots, w_p, \ldots\}\rangle$, we obtain $\mathcal{H}|\psi'\rangle = E|\psi'\rangle$ with the wave function $|\psi'\rangle = R_p|\psi\rangle = |\psi; \{w_1, w_2, \ldots, -w_p, \ldots\}\rangle$. Since the operators R_p for arbitrary plaquettes in $\mathcal{P}$ generate degenerate states, the presence of R_p results in, at least, $2^{N/6}$-fold degenerate ground states.

TABLE I. Ground-state energy E_g and its degeneracy N_d in the mixed-spin (s, S) Kitaev models with the 12-site clusters.

		$N = 12a$		$N = 12b$	
s	S	E_g/JN	N_d	E_g/JN	N_d
1/2	1/2	−0.20417	4	−0.21102	1
1/2	1	−0.33533	8	−0.34235	8
1/2	3/2	−0.47208	4	−0.47389	1
1/2	2	−0.60214	8	−0.60260	8
1	1	−0.66487	1	−0.67421	1
1	3/2	−0.92855	8	−0.93437	8
1	2	−1.19271	1	−1.19567	1
3/2	3/2	−1.37169	4	−1.38840	1
3/2	2	−1.76901	8	−1.77691	8
2	2	−2.33449	1	−2.35306	1

This qualitative difference in the spin magnitudes s and S can be confirmed in the small clusters. By using the ED method, we obtain ground-state properties in the 12-site systems, as shown in Table I. As for the ground-state degeneracy, the mixed-spin systems are naively expected to be divided into three groups. When both spins s and S are integers, the ground state is always a singlet. In the half-integer case, the fourfold degenerate ground state is realized in the $N = 12a$ system, while the singlet ground state is realized in the $N = 12b$ system. This feature is essentially the same as the ground-state properties in the $S = 1/2$ Kitaev model, where the ground-state degeneracy depends on the topology in the boundary condition. By contrast, the eightfold degenerate state is realized in the system with one of the spins being a half integer and the other an integer, which suggests macroscopic degeneracy in the thermodynamic limit.

To confirm this, we focus on the mixed-spin system with $(s, S) = (1/2, 1)$. By using the ED method, we obtain the ground-state energies for several clusters up to 24 sites [see Fig. 1(a)]. The obtained results are shown in Table II. It is clarified that a finite-size effect slightly appears in the

TABLE II. Ground-state profile for several clusters in the Kitaev model with $(s, S) = (1/2, 1)$. N_p is the number of plaquettes, where the local operator W_p is diagonal in the basis set. N_d is the degeneracy in the ground state.

N	N_p	E_g/JN	Δ/J	N_d	N	N_p	E_g/JN	Δ/J	N_d
12a	1	−0.33981	0.0071	8	20a	2	−0.33550	0.0013	20
12b	2	−0.34235	0.0024	8	20b	3	−0.34210	0.0041	32
16a	2	−0.33543	0.0002	20	22	2	−0.33531	0.0016	20
16b	2	−0.33895	0.0019	16	24a	4	−0.33525	0.0031	64
18a	3	−0.33533	0.0018	8	24b	4	−0.33511	0.0010	64
18b	2	−0.33537	0.0015	40					

ground-state energy, and its value is deduced as $E_g/JN = -0.335$. We also find that the ground state is $N_d/2^{N_p}$-fold degenerate in each subspace and its energy is identical in all subspaces except for the $N = 18a$ system [42]. The large ground-state degeneracy $N_d \geqslant 2^{N/6}$ is consistent with the above conclusion. We also find that the first excitation energy Δ is much smaller than the exchange constant J, as shown in Table II. These imply the existence of multiple low-energy states in the system.

Next, we consider thermodynamic properties in the Kitaev model. It is known that there exist two energy scales in the $S = 1/2$ Kitaev model [1], which clearly appear as a double-peak structure in the specific heat and a plateau in the entropy [24,25]. Similar behavior has been reported in the spin-S Kitaev model [28]. These suggest the existence of fractionalization in the generalized spin-S Kitaev model. An important point is that the degrees of freedom for the high-energy part depend on the magnitude of spins $\sim(2S+1)^{N/2}$. On the other hand, in the mixed-spin case, it is unclear which spin is responsible for the high-temperature properties.

Here, we calculate thermodynamic quantities for 12-site clusters, by diagonalizing the corresponding Hamiltonian. Furthermore, we apply the TPQ state method [40,41] to larger clusters. In this calculation, the thermodynamic quantities are deduced by the statistical average of the results obtained from at least 25 independent TPQ states. Here, we calculate the specific heat $C(T) = dE(T)/dT$, entropy $\mathcal{S}(T) = \mathcal{S}_\infty - \int_T^\infty C(T')/T'dT'$, and the nearest-neighbor spin-spin correlation on the α bond $C_S(T) = \langle s_i^\alpha S_j^\alpha \rangle = -2E(T)/(3J)$, where $\mathcal{S}_\infty = \frac{1}{2}\ln(2s+1)(2S+1)$ and $E(T)$ is the internal energy per site.

The results for the mixed-spin systems with $(s, S) = (1/2, 1)$ are shown in Fig. 2. We clearly find the multiple-peak structure in the specific heat. Note that finite-size effects appear only at low temperatures. Therefore, our TPQ results for the 24 sites appropriately capture the high-temperature properties ($T \gtrsim 0.01J$) in the thermodynamic limit. Then, we find the broad peak around $T_H \sim 0.6J$, which is clearly separated by the structure at low temperatures ($T < 0.01J$). Now, we focus on the corresponding entropy, which is shown in Fig. 2(b). This indicates that, by decreasing temperature, the entropy monotonically decreases and the plateau structure is found around $T/J \sim 0.1$. The released entropy is $\sim\frac{1}{2}\ln 2$, which is related to the smaller spin ($s = 1/2$). Therefore, multiple temperature scales do not appear at high temperatures although the system is composed of two kinds of spins (s, S). However, it does not imply that only smaller spins are frozen and larger spins remain paramagnetic at the temperature since the spin-spin correlations develop around $T \sim T_H$ and a quantum many-body spin state is formed, as shown in Fig. 2(c). We have also confirmed that local magnetic moments do not appear even in a wave function constructed by the superposition of the ground states with different configurations of $\{w_p\}$.

By contrast, the value $\frac{1}{2}\ln 2$ reminds us of the high-temperature feature for itinerant Majorana fermions in the spin-1/2 Kitaev model [24,25]. Then, one expects that, in the mixed-spin (s, S) Kitaev model, higher-temperature properties are described by the smaller spin-s Kitaev model, where

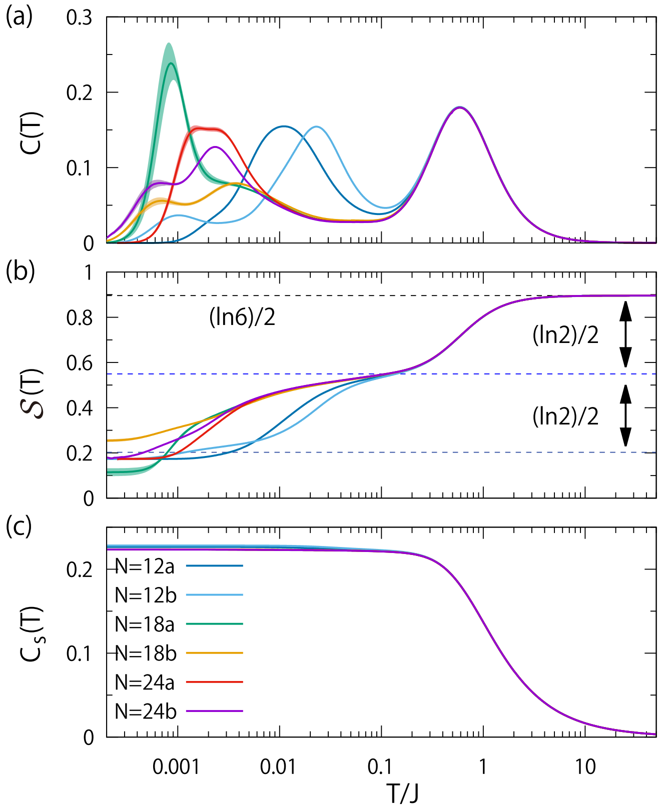

FIG. 2. (a) Specific heat, (b) entropy, and (c) spin-spin correlation as a function of temperatures. Shaded areas stand for the standard deviation of the results obtained from the TPQ states.

degrees of freedom $\sim(2s+1)^{1/2}$ are frozen at each site [28]. In this case, a peak structure appears in the specific heat and the plateau structure at $\sim\mathcal{S}_\infty - \ln(2s+1)/2$ in the entropy. These interesting properties at higher temperatures will be examined systematically. A further decrease of temperatures decreases the entropy and finally $\mathcal{S} \sim \mathcal{S}_\infty - \ln 2$ at lower temperatures, as shown in Fig. 2(b). This may suggest that thermodynamic properties in this mixed-spin Kitaev model with $(s, S) = (1/2, 1)$ are dominated by the degrees of freedom $(2s+1)^N$ corresponding to the smaller spin s for all sites including the larger spins, which are fractionalized into two kinds of quasiparticles. In this case, the existence of the remaining entropy $\mathcal{S} \sim \mathcal{S}_\infty - \ln 2$ should be consistent with macroscopic degeneracy in the ground state as discussed before. However, our TPQ data have a large system size dependence at low temperatures, and conclusive results could not be obtained. Therefore, a systematic analysis is desired to clarify the nature of the low-temperature properties.

To clarify the role of smaller spins in the mixed-spin Kitaev models, we calculate the entropy in systems with $s, S \leqslant 2$ and $N = 12a$ by means of the TPQ state methods. The results are shown in Fig. 3. The plateau structure is clearly observed in the curve of the entropy in the mixed-spin Kitaev models. In addition, we find that the plateau is located around $\mathcal{S} = \mathcal{S}_\infty - \frac{1}{2}\ln(2s+1)$, as expected above.

This may be explained by the deconfined-spin picture in the Kitaev model. In this picture, each spin S is divided into two kinds of quasiparticles with distinct energy scales: $2S$

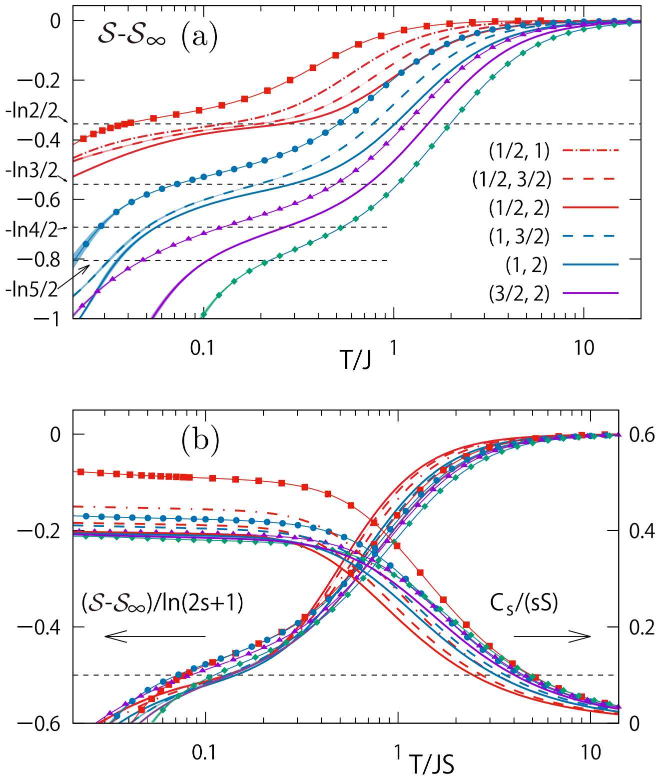

FIG. 3. (a) $\mathcal{S}-\mathcal{S}_\infty$ in the generalized (s,S) Kitaev model at higher temperatures. Squares with lines represent data for the $S=1/2$ Kitaev model obtained from the Monte Carlo simulations [24,25]. Circles, triangles, and diamonds with lines represent the TPQ data for the $S=1, 3/2$, and 2 cases [28]. (b) $(\mathcal{S}-\mathcal{S}_\infty)/\ln(2s+1)$ and $C_s/(sS)$ as a function of T/JS.

L-quasiparticles and $2S$ H-quasiparticles, which are dominant at lower and higher temperatures, respectively. In the exactly solvable $S=1/2$ Kitaev model, H- (L-)quasiparticles are identical to itinerant Majorana fermions (localized fluxes). In addition, this should explain the double-peak structure in the specific heat of the spin-S Kitaev model, each of which corresponds to the half-entropy release [28]. In our mixed-spin (s,S) system, the entropy release at higher temperatures can be interpreted as follows: The thermodynamic properties are governed by the smaller spin s and the portion of the larger spin S corresponding to the spin-s degree of freedom. Namely, the dominant degree of freedom is interpreted as a spin s on both smaller- and larger-spin sites. This "effective" spin s for each site is fractionalized into H- and L-quasiparticles, where the former exhibit energy scales of $\sim J$ with the two-dimensional network and the latter possess a smaller energy scale than J. On the other hand, the remaining degree of freedom at the site with larger spin S is localized and does not contribute to the thermodynamic properties. This is expected from Fig. 2(b), but the low-temperature entropy release is difficult to be discussed in the larger spin cases as shown in Fig. 3(a). Therefore, the low-temperature properties remain as a future issue.

Interestingly, the temperature T^* characteristic of the plateau in the entropy, which may be defined such that $\mathcal{S}(T^*)=\mathcal{S}_\infty-\frac{1}{2}\ln(2s+1)$, depends on the magnitude of the larger spin. In fact, we find that T^* should be scaled by the larger spin $T^*\sim JS$, which is shown in Fig. 3(b). Namely, the peak temperature in the specific heat $T_H\sim JS$ (not shown). This is in contrast to the conventional temperature scale $T^{**}\sim J\sqrt{s(s+1)S(S+1)}$, which is derived from the high-temperature expansion. This discrepancy is common to the spin-S Kitaev model [28], implying that quantum fluctuations are essential even in this temperature range in the mixed-spin Kitaev models. As for the spin-spin correlation, by decreasing temperatures, it develops around $T/JS\sim 1$ and is almost saturated around T^*, as shown in Fig. 3(b). This means that the many-body spin state is indeed realized at the temperature. We also find that at low temperatures, the normalized spin-spin correlation $C_s/(sS)\sim 0.4$ is less than unity when s and S are large. This suggests that the quantum spin-liquid state is, in general, realized in the generalized mixed-spin Kitaev model, which is consistent with the presence of magnetic fluctuations even in the classical limit [27].

In summary, we have studied the mixed-spin Kitaev model. First, we have clarified the existence of the local Z_2 symmetry at each plaquette. Using the TPQ state methods for several clusters, we have found a double-peak structure in the specific heat and plateau in the entropy, which suggests the existence of fractionalization in the mixed-spin system. Deducing the entropy in the mixed-spin system with $s,S\leqslant 2$ systematically, we have clarified that the smaller spin plays a crucial role in the thermodynamic properties at higher temperatures. We expect that the present mixed-spin Kitaev systems are realizable in real materials by substituting the magnetic ions in the Kitaev candidate materials with other magnetic ions with larger spins, and therefore the present work should stimulate material research for mixed-spin Kitaev systems.

Parts of the numerical calculations were performed in the supercomputing systems in ISSP, the University of Tokyo. This work was supported by Grant-in-Aid for Scientific Research from JSPS, KAKENHI Grants No. JP18K04678, No. JP17K05536 (A.K.), No. JP16K17747, No. JP16H02206, and No. JP18H04223 (J.N.).

[1] A. Kitaev, Ann. Phys. **321**, 2 (2006).
[2] G. Jackeli and G. Khaliullin, Phys. Rev. Lett. **102**, 017205 (2009).
[3] Y. Singh and P. Gegenwart, Phys. Rev. B **82**, 064412 (2010).
[4] Y. Singh, S. Manni, J. Reuther, T. Berlijn, R. Thomale, W. Ku, S. Trebst, and P. Gegenwart, Phys. Rev. Lett. **108**, 127203 (2012).
[5] S. K. Choi, R. Coldea, A. N. Kolmogorov, T. Lancaster, I. I. Mazin, S. J. Blundell, P. G. Radaelli, Y. Singh, P. Gegenwart, K. R. Choi, S.-W. Cheong, P. J. Baker, C. Stock, and J. Taylor, Phys. Rev. Lett. **108**, 127204 (2012).
[6] R. Comin, G. Levy, B. Ludbrook, Z.-H. Zhu, C. N. Veenstra, J. A. Rosen, Y. Singh, P. Gegenwart, D. Stricker, J. N. Hancock, D. van der Marel, I. S. Elfimov, and A. Damascelli, Phys. Rev. Lett. **109**, 266406 (2012).

[7] K. Modic, T. E. Smidt, I. Kimchi, N. P. Breznay, A. Biffin, S. Choi, R. D. Johnson, R. Coldea, P. Watkins-Curry, G. T. McCandless *et al.*, Nat. Commun. **5**, 4203 (2014).
[8] T. Takayama, A. Kato, R. Dinnebier, J. Nuss, H. Kono, L. S. I. Veiga, G. Fabbris, D. Haskel, and H. Takagi, Phys. Rev. Lett. **114**, 077202 (2015).
[9] K. Kitagawa, T. Takayama, Y. Matsumoto, A. Kato, R. Takano, Y. Kishimoto, S. Bette, R. Dinnebier, G. Jackeli, and H. Takagi, Nature (London) **554**, 341 (2018).
[10] K. W. Plumb, J. P. Clancy, L. J. Sandilands, V. V. Shankar, Y. F. Hu, K. S. Burch, H.-Y. Kee, and Y.-J. Kim, Phys. Rev. B **90**, 041112(R) (2014).
[11] Y. Kubota, H. Tanaka, T. Ono, Y. Narumi, and K. Kindo, Phys. Rev. B **91**, 094422 (2015).
[12] J. A. Sears, M. Songvilay, K. W. Plumb, J. P. Clancy, Y. Qiu, Y. Zhao, D. Parshall, and Y.-J. Kim, Phys. Rev. B **91**, 144420 (2015).
[13] M. Majumder, M. Schmidt, H. Rosner, A. A. Tsirlin, H. Yasuoka, and M. Baenitz, Phys. Rev. B **91**, 180401(R) (2015).
[14] Y. Kasahara, T. Ohnishi, Y. Mizukami, O. Tanaka, S. Ma, K. Sugii, N. Kurita, H. Tanaka, J. Nasu, Y. Motome, T. Shibauchi, and Y. Matsuda, Nature (London) **559**, 227 (2018).
[15] J. Chaloupka, G. Jackeli, and G. Khaliullin, Phys. Rev. Lett. **105**, 027204 (2010).
[16] H.-C. Jiang, Z.-C. Gu, X.-L. Qi, and S. Trebst, Phys. Rev. B **83**, 245104 (2011).
[17] R. R. P. Singh and J. Oitmaa, Phys. Rev. B **96**, 144414 (2017).
[18] V. M. Katukuri, S. Nishimoto, V. Yushankhai, A. Stoyanova, H. Kandpal, S. Choi, R. Coldea, I. Rousochatzakis, L. Hozoi, and J. van den Brink, New J. Phys. **16**, 013056 (2014).
[19] T. Suzuki, T. Yamada, Y. Yamaji, and S.-i. Suga, Phys. Rev. B **92**, 184411 (2015).
[20] H. Tomishige, J. Nasu, and A. Koga, Phys. Rev. B **97**, 094403 (2018).
[21] U. F. P. Seifert, J. Gritsch, E. Wagner, D. G. Joshi, W. Brenig, M. Vojta, and K. P. Schmidt, Phys. Rev. B **98**, 155101 (2018).
[22] H. Tomishige, J. Nasu, and A. Koga, Phys. Rev. B **99**, 174424 (2019).
[23] A. Koga, S. Nakauchi, and J. Nasu, Phys. Rev. B **97**, 094427 (2018).
[24] J. Nasu, M. Udagawa, and Y. Motome, Phys. Rev. B **92**, 115122 (2015).
[25] J. Nasu, J. Yoshitake, and Y. Motome, Phys. Rev. Lett. **119**, 127204 (2017).
[26] G. Baskaran, D. Sen, and R. Shankar, Phys. Rev. B **78**, 115116 (2008).
[27] T. Suzuki and Y. Yamaji, Physica B: Condens. Matter **536**, 637 (2018).
[28] A. Koga, H. Tomishige, and J. Nasu, J. Phys. Soc. Jpn. **87**, 063703 (2018).
[29] J. Oitmaa, A. Koga, and R. R. P. Singh, Phys. Rev. B **98**, 214404 (2018).
[30] P. P. Stavropoulos, D. Pereira, and H.-Y. Kee, Phys. Rev. Lett. **123**, 037203 (2019).
[31] T. Minakawa, J. Nasu, and A. Koga, Phys. Rev. B **99**, 104408 (2019).
[32] M. Fujii, S. Fujimoto, and N. Kawakami, J. Phys. Soc. Jpn. **65**, 2381 (1996).
[33] S. K. Pati, S. Ramasesha, and D. Sen, Phys. Rev. B **55**, 8894 (1997).
[34] T. Fukui and N. Kawakami, Phys. Rev. B **55**, R14709 (1997).
[35] T. Tonegawa, T. Hikihara, M. Kaburagi, T. Nishino, S. Miyashita, and H.-J. Mikeska, J. Phys. Soc. Jpn. **67**, 1000 (1998).
[36] A. Koga, S. Kumada, N. Kawakami, and T. Fukui, J. Phys. Soc. Jpn. **67**, 622 (1998).
[37] A. Koga, S. Kumada, and N. Kawakami, J. Phys. Soc. Jpn. **68**, 2373 (1999).
[38] A. K. Kolezhuk, H.-J. Mikeska, and S. Yamamoto, Phys. Rev. B **55**, R3336 (1997).
[39] Y. Takushima, A. Koga, and N. Kawakami, Phys. Rev. B **61**, 15189 (2000).
[40] S. Sugiura and A. Shimizu, Phys. Rev. Lett. **108**, 240401 (2012).
[41] S. Sugiura and A. Shimizu, Phys. Rev. Lett. **111**, 010401 (2013).
[42] In the $N = 18a$ system, the ground state belongs to two subspaces with all eigenvalues of W_p being ± 1. Eigenstates with inharmonious distributions of w_p are not the ground states. This distribution of w_p is quite different from those in the other clusters. We expect that the local operators R_p cannot be defined as the $N = 18a$ system because of the periodic boundary condition.