Thermodynamic signature of SU(4) spin-orbital liquid and symmetry fractionalization from Lieb-Schultz-Mattis theorem

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The SU(4) Heisenberg model on a honeycomb lattice is expected to host a quantum spin-orbital liquid at low temperature with a surprising candidate material, α -ZrCl₃. We employed the canonical thermal pure quantum state method to investigate the finite-temperature phase of this model. Exploiting the full symmetry of SU(4), calculation up to a 24-site cluster, which is equivalent to 48 sites in the spin- $\frac{1}{2}$ language, is possible. This state-of-the-art computation with large-scale parallelization enables us to capture the thermodynamic properties of the SU(4) Heisenberg model on a honeycomb lattice. In particular, the specific heat shows a characteristic peak-and-shoulder structure, which should be related to the nature of the low-temperature quantum spin-orbital liquid phase. We also discuss what can be concluded from the assumption that the ground state is gapped and symmetric in view of the generalized Lieb-Schultz-Mattis theorem.

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Introduction. Quantum spin liquids are an unusual state of matter without long-range order beyond the Landau paradigm [1,2]. Specifically, Kitaev materials, such as d^5 iridates and α -RuCl₃, have attracted attention because of their explicit fractionalization [3–13]. As a consequence, the thermodynamic signatures including specific heat and thermal Hall conductivity show a characteristic behavior [14,15], which is difficult to explain without rewriting spin operators with Majorana fermions and is therefore regarded as a precursor of fractionalized topological excitations.

However, the Kitaev model does not have continuous symmetry, differently from usual SU(2)-symmetric spin liquids, and the signature of fractionalization must also be different. For example, the two-peak structure of specific heat was discussed in a kagome spin liquid [16], but later the low-temperature peak has been shown to disappear in large-scale thermal pure quantum (TPQ) state calculations [17–19], and now it is not regarded as an indication of fractionalization in the kagome spin liquid.

In spin liquids with a global symmetry G, a concept called symmetry fractionalization plays an important role. Low-energy excitations called anyons generically have a G charge which is described by a projective representation of G [3]. This means that the symmetry action gets nonlocal in space, which should be one of the most definite signatures of quantum spin liquids.

A completely different avenue of physics of symmetry fractionalization appears in d^1 systems with strong spin-orbit coupling [20–22], which seemingly have a lower symmetry in spin space. However, an emergent SU(4) symmetry appears in d^1 systems, which would lead to multicomponent frustration between spin and orbital degrees of freedom [23–29]. In this case, as is exemplified in honeycomb α -ZrCl₃, an emergent

SU(4) symmetry may lead to a rich symmetry fractionalization which is not expected in the usual SU(2) systems.

Then, how can we prove the existence of symmetry fractionalization? Is there any thermodynamic signature? To see this, it is important to define each thermodynamic quantity for each symmetry sector. For example, the calculation of the specific heat defined for each symmetry sector essentially requires a full treatment of SU(4) symmetry in finite-temperature simulations, which is challenging because the dimension of the Hilbert space gets larger.

Therefore, we developed a state-of-the-art canonical thermal pure quantum (cTPQ) state method [18] to solve this problem. Exploiting the full symmetry of SU(4) enables us to calculate a specific heat up to a 24-site cluster, which is equivalent to 48 sites in spin- $\frac{1}{2}$ language. This is comparable to the Hilbert space size achieved by the cutting-edge ground state exact diagonalization method [30]. We stress again that our calculation is done with finite temperature, which is enabled by a large-scale parallelization in supercomputers and a graphics processing unit (GPU).

Although the ground state of the SU(4) Heisenberg model on the honeycomb lattice was proposed to be a gapless Dirac spin liquid in a previous study [24], we modestly propose another scenario, the possibility of a gapped symmetric ground state. In this gapped spin-orbital liquid case, the fractionalization of PSU(4) symmetry can be proven from a refined version of the generalized Lieb-Schultz-Mattis theorem [20,22,31,32]. In this sense, symmetry fractionalization is more visible in gapped spin-orbital liquids, which may be detected by some thermodynamic signatures.

In this Letter, we theoretically present the finitetemperature specific heat of the SU(4) Heisenberg model on a honeycomb lattice. The specific heat shows a characteristic peak-and-shoulder structure, which would be useful to identify the realization of this model in real materials. The results are consistent with a gapped scenario, and we also

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discuss what can be concluded from the assumption that the ground state is gapped and symmetric.

Model. The SU(4) Heisenberg model on the honeycomb lattice is defined as follows,

$$H = \sum_{\langle ij\rangle} \left(2\mathbf{S}_i \cdot \mathbf{S}_j + \frac{1}{2} \right) \left(2\mathbf{T}_i \cdot \mathbf{T}_j + \frac{1}{2} \right), \qquad (1)$$

where S_i are spin- $\frac{1}{2}$ operators for the spin sector, and T_i are spin- $\frac{1}{2}$ operators for the orbital sector. $\langle ij \rangle$ runs over every nearest-neighbor bond of the honeycomb lattice. We put a fundamental representation of SU(4) per site, which consists of two spin and two orbital degrees of freedom. It is better to rewrite the Hamiltonian by the swapping operator,

$$H = \sum_{\langle ij \rangle} P_{ij},\tag{2}$$

where P_{ij} is a swapping operator for two fundamental representations on the *i*th and *j*th sites of the honeycomb lattice. In this way, the SU(4) symmetry is made explicit. This is a natural generalization of the SU(2) Heisenberg model to SU(4) and can be realized, for example, as a low-energy effective model of α -ZrCl₃ [20]. From now on we consider an *N*-site cluster of the honeycomb lattice with a periodic boundary condition.

Method. In order to compute finite-temperature quantities for the above model, we employ the cTPQ method (the Hamsde Raedt method [33]). In a finite-size system, the cTPQ method is regarded as a stochastic approximation of a trace of a large matrix. To compute a physical observable *A*, we use

$$\operatorname{Tr} A e^{-\beta H} \sim \langle 0|e^{-\beta H/2} A e^{-\beta H/2}|0\rangle, \qquad (3)$$

where $|0\rangle$ is a Haar random vector in the Hilbert space. In practice, it is better to expand the $e^{-\beta H/2}$ by a power of *H*, and we define

$$|k\rangle = (l - H)^k |0\rangle, \qquad (4)$$

where *l* is a real value larger than the maximal eigenvalue of *H*. $e^{-\beta H/2}$ is expanded up to the Λ th power of l - H. With a large *N* limit, a single random vector $|0\rangle$ is enough to compute any physical quantities, but in practice it is better to sample N_{sample} vectors and take an average.

For simplicity, we set $l = \max_i \{E_i\} + 1$, $\Lambda = 2000$, where E_i is each eigenvalue of H. For calculations with $N \leq 20$, we also set $N_{\text{sample}} = 100$. Errors are estimated by the jackknife method. For N = 24, it is difficult to take an average, so we show raw data for two samples (shown as No. 1 and No. 2).

In reality, we use the above cTPQ method for each symmetry sector of SU(4) and sum up the results afterwards because the Hamiltonian is block-diagonal. This direct-sum decomposition is achieved by the method developed for exact diagonalization by Nataf and Mila [34,35]. However, their original method is not suitable for a large-scale parallelization, so we modify their idea a little to improve the computational efficiency.

As is usually the case, difficulty in the exact diagonalization or the cTPQ method arises when we make a lookup dictionary for the subspace basis. In the case of Nataf and Mila's method [34], such a problem is reduced to the problem to index and retrieve standard Young tableaus (SYTs) because



FIG. 1. A graph associated with a Young tableau (3, 2, 1) in the Wilf-Rao-Shanker method. Arrows are connected when a reduced Young tableau is obtained by eliminating a corner box of the original Young tableau. Each path starting from (3, 2, 1) to (1) corresponds to a single SYT by assigning a number from 1 to 6 to an added box when going backwards from (1) to (3, 2, 1).

each basis of a specific symmetry sector associated with a Young tableau γ (with *N* boxes) is labeled by an SYT with the same shape as γ . We note that SYTs are prepared by filling γ with numbers, 1, 2, ..., *N*, with a constraint that they are aligning in ascending order both in the row and the column. The problem of indexing and retrieving such SYTs for a fixed γ is solved by Wilf, Rao, and Shanker [36,37]. We note that a similar idea was discussed in Ref. [38]. In their method, a graph is associated with each γ by eliminating a corner box recursively from a Young tableau γ (see Fig. 1). Then, we can identify each SYT with each path starting from γ to a single box. These paths are indexed and retrieved by Wilf's method [36] for any such graphs very efficiently, and by this method we can parallelize the exact diagonalization or the cTPQ method for any SU(N_c) Heisenberg models.

For calculations with $N \leq 20$, we use a GPU machine with a CUDA implementation. For N = 24, we use a largescale supercomputer with a message-passing-based cTPQ implementation. A flat message passing interface (MPI) parallelization up to 18 432 processes is used. The multiplication of off-diagonal components is possible through MPI ALLTOALLV. All codes are written in the JULIA language [39–41].

Specific heat. The specific heat shows a peak-and-shoulder structure for all the system sizes calculated (see Fig. 2). This typical shape becomes clearer in the larger system size. Physically the low-temperature peak is associated with a color gap, i.e., an energy scale of excitations with an SU(4) color charge, and the high-temperature shoulder is associated with the interaction energy scale.

This is in contrast to what has been observed for the kagome spin liquid. In the kagome case, the low-temperature physics below the spin gap is dominated by neutral excitations [30], which is consistent with a recent expectation that the low-temperature effective theory consists of neutral Dirac cones [42]. On the other hand, in the present case, the low-temperature peak structure is not dominated by singlet excitations, which is demonstrated by the comparison with the specific heat restricted to the singlet subspace (see Fig. 3). This is also consistent with the exact diagonalization observation that the color gap 0.8882 is much smaller than the singlet gap 1.1702 for N = 24.

We believe that our results are consistent with a gapped spin-liquid ground state, most probably a Z_4 spin liquid,



FIG. 2. (a) Shape of the 16*a* cluster. (b) Shape of the 16*b* cluster. (c) Shape of the 20-site cluster. (d) Shape of the 24-site cluster. (e) Temperature dependence of the specific heat for the above clusters calculated by the cTPQ method. For $N \leq 20$ -site clusters we take an average over $N_{\text{sample}} = 100$ samples, and errors are shown in ribbons, while for the 24-site cluster we show raw data for the two samples (shown as No. 1 and No. 2).

rather than a gapless one proposed previously [24]. This is because the low-temperature peak associated with the O(1)color gap seems to be converged to the thermodynamic limit with the peak temperature almost unchanged. Indeed, the peak temperature is almost constant for the 16*b*-, 20-, and 24-site calculations, and we cannot expect that this peak disappears or moves drastically in the thermodynamic limit. Of course, we cannot rule out the possibility that singlet excitations begin to dominate below the color gap for N > 24, but we believe that this is an unlikely scenario. In order to explain everything observed in our calculations, the gapped spin-liquid scenario



FIG. 3. Comparison of specific heats between the whole symmetry sectors and the singlet sector. Solid lines are the same as those in Fig. 2(e) with N = 24. Dashed lines are corresponding specific heats calculated by confining the symmetry sector to the singlet sector. It is clear that the low-temperature peak does not originate from singlet excitations.

is more likely. We note that a previous gapless Dirac spinliquid scenario [24] has the problem of a relevant monopole perturbation [43] and also cannot explain the existence of a color gap. Based on this observation, we will discuss what can be said from the assumption that the ground state is gapped and symmetric from now on.

Rigorous proof of the Lieb-Schultz-Mattis-Affleck-Yamada-Oshikawa-Jackeli theorem. Here, we briefly give a rigorous version of the two-dimensional case of the Lieb-Schultz-Mattis-Affleck-Yamada-Oshikawa-Jackeli (LSMAYOJ) theorem [20,22,31,32], which was first "proven" physically by the flux-insertion argument by Yamada, Oshikawa, and Jackeli [20,22]. This is a G = PSU(N) generalization of the Lieb-Schultz-Mattis theorem, which was proven rigorously for one-dimensional systems by Ogata, Tachikawa, and Tasaki [44]. Its generalization to two dimensions is indeed straightforward. We here give a very simple proof, utilizing the fact that the Ogata-Tachikawa-Tasaki proof [44] is directly treating the infinite system.

Let us consider an infinite cylinder geometry with a spiral boundary condition (sometimes called a tilted or twisted boundary condition) [45–47]. The spiral boundary condition is defined as follows. First, let us begin with \mathbb{Z}^2 . We glue it into an infinite cylinder by identifying $(i, j) \in \mathbb{Z}^2$ with $(i + 1, j + L_y) \in \mathbb{Z}^2$, where L_y is a circumference. The point is that this infinite cylinder has a one-unit-cell translation symmetry as a one-dimensional system, which we call a spiral translation symmetry. Thus, for any L_y we can use the Ogata-Tachikawa-Tasaki theorem [44] to prove that the ground state is degenerate [48] when the unit cell contains a projective representation of *G* and the interaction is short ranged. The degeneracy is maintained for any finite L_y with a spiral boundary condition, so the degeneracy will be preserved in the thermodynamic limit.

The advantage of this proof is that it is applicable to the discrete group G case. However, one caveat is that this kind of proof cannot give information on the number of degeneracies. The information on the degeneracy can be deduced from the argument of the next section.

Physical proof of the existence of symmetry fractionalization. The extension of the Lieb-Schultz-Mattis theorem not only can prove the ground state degeneracy, but also can prove the existence of symmetry fractionalization of *G*. Here, we assume that *G* is continuous and connected. This was physically proven in Refs. [49,50] in the G = SO(3) case. For simplicity, we also give our refined proof of this claim for G = SO(3)in the following. This is actually achieved by proving the degenerate ground states guaranteed by the LSMAYOJ theorem include different one-dimensional symmetry-protected topological (SPT) states.

Assuming the unit cell contains a projective representation of *G* and the same setting as that of the proof of the LSMAYOJ theorem, a unique and gapped ground state is forbidden. Let us assume that the ground states are degenerate with a gap. The point is that the SPT phase is distinguished by the Ogata index $\sigma_x^R \in H^2[G, U(1)] = Z_2$, where x = (i, j) is the origin of the unit cell [44,51–53], and $\sigma_x^R = 0$ and $\sigma_x^R = 1$ are essentially equivalent. This is because $\sigma_x^R = 0$ and $\sigma_x^R = 1$ are exchanged just by changing the definition of the origin *x* through a single spiral translation. This "democracy" of the Ogata index automatically guarantees that the same number of $\sigma_x^R = 0$ states and $\sigma_x^R = 1$ states should be included in the degenerate ground states. Such a distinction must be independent of the way of the dimensional reduction, which physically finishes the proof [54].

It is not clear that the degeneracy of different SPT states means symmetry fractionalization. Intuitively, this can be understood as follows. On a finite cylinder with an even number of sites, the distinction of SPT phases can be captured by edge states. A trivial SPT phase has no edge states, while a nontrivial SPT phase has spin- $\frac{1}{2}$ edge states. In the large-cylinder limit, these two correspond to a vacuum sector and a spinon sector of the topological phase, respectively, assuming that the vacuum sector does not have an edge state. This means that an original spin-1 excitation (magnon) is fractionalized into two spinons and separated into both edges with long-range entanglement. This is exactly the proof of the existence of fractionalization in the G = SO(3) case.

The generalization to the case with G = PSU(N) is straightforward. In the case of the present SU(4) model, G = PSU(4) and $H^2[G, U(1)] = Z_4$. As for the honeycomb lattice model, *i* and *i* + 2 (*i* = 0, 1) in $H^2[G, U(1)] = Z_4$ are democratic, so this proves the degeneracy of $\sigma_x^R = i$ and $\sigma_x^R = i + 2$ states as ground states.

In the thermodynamic limit, this not only means the existence of topological order, but also the fractionalization of the symmetry G, i.e., the existence of spinons and orbitalons which behave as a (six-dimensional) projective representation of G. On the finite cylinder, the vacuum sector without edge anyons, and the anyon sector with edge anyons must be almost degenerate.

Counterexamples. There exists a counterexample such as Wen's plaquette model [55] for the discrete group G case. This is because degenerate ground states guaranteed by the Lieb-Schultz-Mattis theorem are exchanged directly by G, not by translation. The Ogata index is defined only when all the ground states in the infinite system are G symmetric, and thus the above discussion fails in this case. However, this never happens when G is continuous and connected, so our conclusion is not affected by such a possibility. More detailed discussions can be found in Refs. [49,50].

Origin of the peak. So far we have identified the energy scale of the specific heat peak as the color gap, but the physical origin of an existing peak is still not clear. If we assume that there is no symmetry breaking, we have to come up with another mechanism beyond the Landau theory to describe the existence of a peak.

In contrast to the Landau theory, fermionic partons can acquire a gap without spontaneous symmetry breaking because a four-fermion condensate dynamically generates a gap without a bilinear term. This condensate can be described in terms of fermionic partons (spinons and orbitalons) f_{ia} , where *i* stands for a site index and a = 1, ..., 4 stands for a color index, as follows [43,56],

$$\Delta = \langle \epsilon_{abcd} f_{ia} f_{ib} f_{ic} f_{id} \rangle, \tag{5}$$

where ϵ_{abcd} is a completely antisymmetric tensor. Thus, we would see a crossover between the $\Delta \sim 0$ disordered phase to the $\Delta > 0$ ordered phase at some temperature T_c , which potentially produces a peaklike behavior in the specific heat.

The gap opening and symmetry fractionalization occurs simultaneously at T_c because we have proven that the symmetric gapped state inevitably leads to symmetry fractionalization. In this sense, we can regard the low-temperature peak as reminiscent of fractionalization, while it is not clear how the action of SU(4) is transformed in this crossover.

Discussion. The existence of a low-temperature peak implies that the ground state of the SU(4) Heisenberg model on the honeycomb lattice is a gapped spin-orbital liquid rather than a gapless liquid proposed previously [24]. If the ground state is really gapped and symmetric, the LSMAYOJ theorem guarantees ground state degeneracy, i.e., the existence of topological order. Even more, the extended version of this theorem proves the fractionalization of PSU(4) symmetry, and the existence of spinons and orbitalons which behave as a projective representation of PSU(4).

Finally, we conjecture a generalized version of the Lieb-Schultz-Mattis-type theorem. The claim is that if the representation of G per unit cell is projective, there must be multiple degenerate ground states which are distinguished as one-dimensional G-SPT phases after the dimensional reduction. This version is yet to be proven mathematically rigorously, and left for the future work.

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