Numerical-Diagonalization Study of Spin Gap Issue of the Kagome Lattice Heisenberg Antiferromagnet

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We study the system size dependence of the singlet-triplet excitation gap in the $S = 1/2$ kagome-lattice Heisenberg antiferromagnet by numerical diagonalization. We successfully obtain a new result of a cluster of 42 sites. The two sequences of gaps of systems with even-number sites and that with odd-number sites are separately analyzed. Careful examination clarifies that there is no contradiction when we consider the system to be gapless.

KEYWORDS: antiferromagnetic Heisenberg spin model, kagome lattice, frustration, numerical-diagonalization method, Lanczos method

The kagome-lattice antiferromagnet attracts the attention of many researchers because it is one typical case in which magnetic frustration occurs in systems. Recently, the discovery of candidate materials, i.e., herbersmithite, volborthite, and vesignieite, has promoted further investigation into clarifying their properties. However, the kagome-lattice antiferromagnet is still an unresolved system, some properties of which remain unclear.

One of them is the spin gap issue of the $S = 1/2$ kagome-lattice Heisenberg antiferromagnet, namely, whether or not a nonzero energy gap exists between the ground-state singlet level and the lowest triplet one. The reason why this issue is difficult to resolve is that some of the numerical methods are not adequate at present. The density matrix renormalization group (DMRG) method is not good because of the two-dimensionality of the system although much effort using the DMRG method has recently been carried out, which will be mentioned later. Under such circumstances, numerical diagonalization calculation can provide us with precise data; however, available system sizes are limited to being very small. In studies of the kagome-lattice antiferromagnet, only sizes up to 36 sites were treated before Sakai and Nakano treated a system with 39 sites for the first time. Although Sindzingre and Lhuillier attempted to resolve the spin gap issue in the argument on the basis of the data up to 36 sites, the authors finally concluded “it is impossible to distinguish between a gapless system and a system with a very small gap.” Therefore, numerical results of larger system sizes are required in order to tackle this issue; thus, carrying out large-scale numerical-diagonalization calculations is an urgent task.

The purpose of this study is to investigate the spin gap issue from the argument based on new numerical-diagonalization data of larger systems that have not yet been reported. We successfully obtain some new results in the case of up to 42 sites.

Here, we examine the excitation gap of the $S = 1/2$ kagome-lattice Heisenberg antiferromagnet. The Hamiltonian is given by

$$\mathcal{H} = \sum_{\langle i,j \rangle} J S_i \cdot S_j,$$

where $S_i$ denotes the $S = 1/2$ spin operator. The sum runs over all the nearest-neighbor pairs. Energies are measured in units of $J$; hereafter, we set $J = 1$. The number of spin sites is denoted by $N_s$, where $N_s/3$ is an integer. We impose the periodic boundary condition for clusters with site $N_s$. We calculate the lowest energy of $E$ in the subspace divided by $\sum_j S_j^z = M$. The energy is denoted by $E(N_s, M)$.

Using the numerical exact diagonalization based on the Lanczos algorithm, we have calculated the values of $E(N_s, M)$ of clusters up to $N_s = 42$, to obtain the finite-size energy differences. The convergence of our Lanczos calculations has been checked so that the relative changes of energies of the lowest and first-excited states become less than $10^{-10}$. The shapes of the finite-size clusters of system sizes from $N_s = 30$ to $N_s = 42$ are shown in Fig. 1. For $N_s = 36$ and 39, a rhombic cluster having an interior angle $\pi/3$ is available, which two-dimensionality may be captured well. For other system sizes of $N_s$, on the other hand, no rhombic cluster is available. For $N_s = 30$ and 33 in such cases, we examine more shapes than one specific case; we here present different results due to the difference between cluster shapes as many as we have obtained. Since the difference between cluster shapes is regarded as a kind of boundary conditions, the effect on our results for the gaps appears as a finite-size effect due to such various boundaries. In our analysis, it is desirable to extract information that is not largely affected from a specific cluster shape. To realize such an analysis, we treat various results from different nonrhombic shapes equivalently instead of rejecting the results in nonrhombic cases. The case of $N_s = 42$ shows the largest size in this study; thus, we treat only a single case of shape owing to the limitation of available computational time that we have prepared. For $N_s = 42$, the largest dimension is 538 257 874 440 for the subspace of $M = 0$. To treat such huge matrices in com-
The Hamiltonian is given by
\[ \mathcal{H} = \sum_{(i,j)} J \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{(i,j)} J' \mathbf{S}_i \cdot \mathbf{S}_j. \] (4)

Here, the ratio of \( J'/J \) denoted by \( \alpha \) is a controllable parameter. The case of \( \alpha = 1 \) is that of a simple square lattice in which the ground state is Néel-ordered. On the other hand, the case of \( \alpha = 0 \) is reduced to an assembly of isolated dimers; the gapped ground state is realized near \( \alpha = 0 \). Matsumoto et al. clarified the boundary between these phases to be \( \alpha = 0.52337(3) \). The examination of this model makes it possible to confirm an appropriate means of analyzing \( \Delta_{N_s} \) of diagonalization data in two-dimensional systems; results are shown in Fig. 2. In Fig. 2(a), analysis based on the case when a nonzero gap exists is demonstrated; finite-size data are fitted by
\[ \Delta_{N_s} = \Delta_\infty + C \exp(-DN_s^{1/2}), \] (5)
which was also used in ref. 20. One finds that the gapped case of \( \alpha = 0.4 \) gives us the result of \( \Delta_\infty \) that agrees well with the precise estimate from the QMC simulation. However, the case of \( \alpha = 0.52337 \) in which the system is gapless (and critical) incorrectly gives a nonzero \( \Delta_\infty \).

**Fig. 1.** Shapes of finite-size clusters for (a) \( N_s = 42 \), (b) \( N_s = 39 \), (c) \( N_s = 36 \), (d) \( N_s = 33 \), (e) \( N_s = 33 \), (f) \( N_s = 30 \), (g) \( N_s = 30 \), and (h) \( N_s = 30 \). Note that clusters (b) and (c) are rhombic and the others are not.

**Fig. 2.** Analysis of the spin gap of the Heisenberg antiferromagnet on the dimerized square lattice. The \( 1/N_s \) and \( 1/N_s^{1/2} \) dependences of \( \Delta_{N_s}/J \) are plotted in panels (a) and (b), respectively. The inset in panel (a) illustrates the lattice structure, in which thick green bonds and thin black bonds denote the interactions of \( J \) and \( J' \), respectively. Green squares, blue diamonds, and black circles represent the cases of \( \alpha = 1, 0.52337, \) and 0.4, respectively. Closed symbols mean that the shapes of the clusters are regular squares; open symbols mean that they are not. A red double circle in each panel denotes an estimate from the QMC simulation. The explanations of fitting lines and curves are given in the text.
In the gapless case of $\alpha = 1$, the fitting is a failure; even the linear line obtained by fitting data of systems larger than $N_s = 16$ intersects at the intercept of a positive value with the line of $N_s \to \infty$.

In Fig. 2(b), on the other hand, analysis based on the gapless case is carried out; the fitting is based on the line

$$\Delta N_s = F + \frac{G}{N_s^{1/2}},$$

which is related to the assumption that the system has a dispersion relation proportional to the wave number having the unit of the inverse length. The linear fitting of data $N_s=36$, 32, and 28 in the case of $\alpha = 0.52337$ ($\alpha = 1$) gives a negative (almost vanishing) $F$, which suggests that the system is gapless. In the gapped case of $\alpha = 0.4$, a positive $F$ is very close to the precise estimate from the QMC simulation. Therefore, these examinations of Figs. 2(a) and (b) indicate that, in two-dimensional systems, the analysis based on Fig. 2(a) is good at capturing gapped cases and the analysis based on Fig. 2(b) is better for recognizing gaplessness.

Now, we present our new results of the kagome-lattice antiferromagnet of $N_s = 42$ accompanied by the results of smaller clusters, some of which have already been reported; these results are shown in Table I. Note that the present study is the first study, to the best of our knowledge, that presents the result of a singlet-triplet energy difference in the $S = 1/2$ system with $N_s = 42$, whereas the ground-state energy of the system with $N_s = 42$ in several cases of the star lattice, one-dimensional uniform chain, and kagome lattice has been reported. Note that our results of the ground-state energy per site are very close to the recent estimate $e_g/J = -0.4379(3)$ from the DMRG calculations. In this study, we focus our attention on the singlet-triplet energy differences and their system size dependence hereafter.

First, let us observe the results of $\Delta N_s/J$ for $N_s = 30$. The cases shown in Figs. 1(f)-1(h) depart from the ideal two-dimensional case in different degrees. When one examines the degree of strip, the departure becomes stronger in the order of clusters (f), (h), (g), and (f). However, $\Delta N_s/J$ becomes larger in the order of clusters (f), (g), and (h). The two orders disagree with each other. The disagreement indicates that, within the viewpoint of the results of $\Delta N_s/J$, determining whether the twodimensionality is better or worse is not very trivial. Thus, this suggests that there is considerable significance in examining various cluster shapes in nonrhombic cases, irrespective of the degree of strip.

Let us plot the $1/N_s$ dependence of singlet-triplet gaps of the kagome-lattice antiferromagnet in Fig. 3(a), where the data listed in Table I and some other results for clusters with a smaller $N_s$ are presented. One finds that the two sequences of even-$N_s$ and odd-$N_s$ data are different from each other. Thus, we analyze these two sequences separately on the basis of the assumption of the gapped case, as explained in Fig. 2(a). We obtain $\Delta_\infty = 0.088(69)$ from the data of even $N_s$ and $\Delta_\infty = 0.209(18)$ from the data of odd $N_s$. The two extrapolated results disagree with each other even though the fitting errors are taken into account. The disagreement suggests that the analysis under the assumption of the gapped case is invalid. There are two possibilities for this invalidity. One is that the system is not gapped. The above nonzero estimates of the spin gap are artificial consequences from the invalid analysis. (See the case of $\alpha = 0.52337$ in Fig. 2(a)). The other possibility is that although the system is intrinsically gapped, the system size dependence is completely

### Table I. List of numerical data of the ground state energy and the singlet-triplet energy difference.

| $N_s$ | shape | $-e_g/J$ | $\Delta N_s/J$ | first report |
|-------|-------|----------|----------------|-------------|
| 42    | Fig. 1(a) | 0.4381425934 | 0.14909214 | new         |
| 39    | Fig. 1(b) | 0.4361425934 | 0.12243392 | ref. 12     |
| 36    | Fig. 1(c) | 0.4383765311 | 0.16420678 | ref. 4      |
| 33    | Fig. 1(d) | 0.4366725772 | 0.22243392 | ref. 11     |
| 32    | Fig. 1(e) | 0.4430755530 | 0.25405132 | new         |
| 30    | Fig. 1(f) | 0.4452555475 | 0.4458946475 | new         |
| 33    | Fig. 1(g) | 0.4458946475 | 0.16823452 | new         |
| 30    | Fig. 1(h) | 0.44525556126 | 0.17565828 | ref. 1      |

Fig. 3. Analysis of the spin gap of the Heisenberg antiferromagnet on the kagome lattice. The $1/N_s$ and $1/N_s^{1/2}$ dependences of $\Delta N_s/J$ are plotted in panels (a) and (b), respectively. Red squares (black circles) represent the case of odd (even) $N_s$. Closed symbols mean that the clusters are rhombus with an interior angle $\pi/3$; open symbols mean that they are not. The explanations of the fitting lines and curves are given in the text.
different from eq. (5). The estimate $\Delta_{\infty}=0.088(69)$ from even $N_s$ coincides within the error with an estimate 0.05 of the singlet-singlet gap obtained from the recent DMRG calculations$^2$ as a lower bound for the singlet-triplet (spin) gap. However, $\Delta_{\infty}=0.209(18)$ from odd $N_s$ is not in agreement with 0.05 from the DMRG method. If the kagome-lattice antiferromagnet has a nonzero spin gap close to 0.05, this leads to the data sequence of finite-size gaps of odd $N_s$, revealing an unknown and/or peculiar system size dependence. Even though one does not use eq. (5), one finds that the data sequence of odd $N_s$ does not show the behavior of approaching a value close to 0.05. Thus, we think that this possibility is quite small.

Next, we carry out an analysis of the data in Fig. 3(b), under the assumption in the gapless case in the manner explained in Fig. 2(b). We obtain the black solid line from the data of $N_s = 42$ and 36 as representatives of the data of even $N_s$; the line indicates $F = -0.03955$. The black dotted line is also drawn so that the line runs through the origin and the data point of $N_s = 42$. Note that the solid and dotted black lines approach the data point of the largest $\Delta_{30}/J$ and the data point of the smaller $\Delta_{24}/J$. In the cases of odd $N_s$, we obtain the red solid line from the data point of $N_s = 39$ and the average of the two data of $N_s = 33$ as representatives of the data of odd $N_s$; the line indicates $F = 0.0007$. It is worth mentioning that the red line goes very near the data point of $N_s = 27$. This fact supports the notion that treating the average of the two data for $N_s = 33$ is reasonable. Since $F = 0.0007$ is quite small, if one compares it with the difference between the two values of $\Delta_{N_s}$ for $N_s = 33$, we can conclude that $F$ is vanishing in the case of odd $N_s$. Although the two values of $F$ from even $N_s$ and odd $N_s$ are different from each other, the difference comes from the effect that higher-order terms appear differently between the cases of even $N_s$ and odd $N_s$. The two results from even $N_s$ and odd $N_s$ commonly reveal the gapless feature of the two-dimensional system shown in Fig. 2(b). We emphasize here that no contradictory points are found in our numerical-diagonalization results in the viewpoint that the system is gapless.

In summary, we have studied the spin gap issue of the kagome-lattice Heisenberg antiferromagnet by the numerical-diagonalization method using the Lanczos algorithm. We have successfully obtained a finite-size gap of the 42-site cluster; we have examined the system-size dependence of gaps of all the available clusters up to 42 sites. From the results of the analyses from the viewpoints of the gapped and gapless cases, we here conclude that the system is gapless. This conclusion is in agreement with the fact that no spin gap is observed experimentally.$^1$ On the other hand, the present conclusion is not in agreement with the recent result from the DMRG analysis; the discrepancy should be resolved in future studies. The consequence of the gaplessness will be followed by the examination of whether or not the long-range order exists, although it is difficult to appropriately choose possible candidates from among the classical spin arrangements. To find a good candidate, the spin correlation functions of large systems would be a good help. To better understand the properties of the system, the wave numbers of the wave functions and their degeneracy should be examined. References 3 and 4 showed part of such results for $N_s = 27$ and $N_s = 36$, respectively; information on larger systems is desired in future studies.$^{25}$ Numerical-diagonalization calculations of systems as large as possible will provide us with a new and better understanding of condensed-matter physics.

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