Thermodynamic properties of the quantum Heisenberg antiferromagnet on the *kagomé* lattice

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We have performed Monte Carlo simulations on the quantum Heisenberg antiferromagnet on the *kagomé* lattice with up to 72 spins. We have used the transfer-matrix Monte Carlo method, which enabled us to do efficient samplings at rather low temperatures where other methods suffer from serious sign problem. Specific-heat data exhibit the double peak. Results of the chirality and short-range spin correlations show no tendency of any magnetic ordering even at the lowest temperature we observed.

Recent interest in the *kagomé* antiferromagnet originates in the experiment of a ^{3}He layer adsorbed on graphite in the mK temperature region.[1–3] A peak of the specific heat was observed at the coverage $\rho \simeq 0.18$ atoms/Å^2, where the second layer is just filled. An unexpected finding is a lack of entropy. Namely, if we assume $C \propto T$ in the low temperature region where the experiment could not be achieved, the entropy calculated by the integration C/T is just half the expected value.[2] Therefore they speculated that some structure, or the second peak, might be present in the lower temperature region not yet observed experimentally.

Elser proposed that the atoms on the second layer form the $\sqrt{7} \times \sqrt{7}$ triangular lattice and that a quarter of the atoms become free from the exchange interactions so that the remainder form a *kagomé* lattice.[1] The $\sqrt{7} \times \sqrt{7}$ structure with respect to the first layer atoms is supported by path integral simulations.[4] In this proposal, half the missing entropy is to come from these free spins. The other half is to be supplied by the additional low temperature peak of the specific heat. He showed the double peak by the numerical diagonalization of a small cluster[1] and by the decoupled-cell Monte Carlo simulation.[5] It should be noted that there is another explanation of the specific heat; the multispin interactions may play an important role in this triangular system as well as the two-body interactions.[6]

Apart from the experiment, the *kagomé* system provides attracting problems concerning both the thermodynamic properties and the ground state, and thus numerous efforts have been done both analytically and numerically.[5,7–12] Classically, the *kagomé* antiferromagnet possesses macroscopic local degeneracy in the ground states. In the quantum system, two typical structures have been considered as candidates for the ground state selected among the classical degenerate ground states by quantum fluctuations. One is a $\sqrt{3} \times \sqrt{3}$ structure and the other is a $q = 0$ structure. Within the linear spin-wave theory, both states have an identical dispersionless zero-energy mode throughout the Brillouin zone.[5,7] It is reported that higher order corrections to the spin-wave calculation lift the degeneracy and the two states are stabilized for the systems with large S value.[8] A large-N expansion gives a similar result but a disordered ground state at small S.[9] The disordered ground state is predicted by the series expansion from the Ising limit[10] and the numerical diagonalization of finite systems.[5,11,12] Results of the high temperature expansion depend on the orders of expansion.[7,12] The presence of the magnetic order in the ground state is not clarified yet and therefore left for a future problem.

On the thermodynamic properties, the specific heat exhibits a broad plateau at low temperatures in the classical system.[13] Contrarily, in the quantum system, numerical diagonalization up to size $N = 18$ displays the double peak, but the size dependence is still strong; the data of $N = 15$ do not show the second peak but a shoulder.[12] The existence of the third peak was also observed at a lower temperature.[12] The specific heat for the system with 24 spins is not so definite to determine the existence of the second peak.[14] An elementary polygon of the *kagomé* lattice is a hexagon with 12 spins. Thus the small lattices studied so far include only one or two hexagons, where they suffer from nontrivial boundary conditions. The minimum size which is free from the problem of the boundary condition is 36. The system contains four hexagons. In this sense, we consider that we have to study systems with $N \geq 36$ in order to study intrinsic properties of the *kagomé* lattice.

In this paper, we report that the second peak stably exists at $T \sim 0.1$ by comparing the data of $N = 18$ with those of $N = 36$. Here, the temperature is scaled by the coupling constant J in the notation

$$H = J \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j, \qquad |\boldsymbol{S}| = 1/2. \tag{1}$$

The staggered and the uniform chirality as well as the short-range spin correlations were also calculated to investigate whether the $q = 0$ structure or the $\sqrt{3} \times \sqrt{3}$ structure grows when the temperature decreases.

So far, unfortunately, the quantum Monte Carlo

(QMC) method is not a good tool to study this system because of the negative-sign problem.[15] We employed the transfer-matrix Monte Carlo method proposed recently.[16] In this method, we decompose all the spins in the real space into two parts; one is composed of the spins (•) which are traced out beforehand and the other is composed of the spins (○) which we sample by MC as depicted in Fig. 1. For such local traceout, the • spins should be enclosed by the ○ spins. In the present simulations, two-thirds of total spins are already traced out. This causes an improvement in the sign problem. In the case of the size $N = 36$, the sign ratio stays almost unity for $T \geq 0.2$ and it decreases for $T < 0.125$. Inverse temperature dependence of the ratio is roughly $r \sim 51.7 \exp[-0.74\beta]$ for $T < 0.2$. This should be compared with a case of an ordinary world line simulation on the Δ-chain system, in which the ratio is 10^{-7} at the same temperature for $N = 16$,[17] although we have not yet performed the ordinary world line QMC for the *kagomé* lattice.

Simulations were carried out at $1.5 \geq T \geq 0.15$ for $N = 18$, at $3.5 \geq T \geq 0.10$ for $N = 36$, and at $T = 0.2, 0.25, 0.35, 0.5$ for $N = 72$. We have used the reweighting method[18] to improve the sampling efficiency. We obtained data at three to five different temperatures from a single temperature simulation, and consequently a local tendency of the data with respect to the temperature was observed. It should be noted that this method also works to improve the sign problem: error bar of a quantity observed at $T_{\rm obs}$ by the reweighting from the simulation at $T_{\rm sim}$ becomes smaller than that of a quantity observed at $T_{\rm obs}$ from the simulation at $T_{\rm obs}$ for $T_{\rm obs} > T_{\rm sim}$. We performed simulation of several values of the Trotter number in the range $0.32 < \beta/m < 0.80$, and extrapolated the data to $m \to \infty$, which gives good linearity. The autocorrelation time is also short because the simulated spins are all isolated alone. Besides these merits of the method, there is a disadvantage; it takes a rather long time for one MC step because of the local traceout. For example, a simulation with 2000 steps for the Trotter number $m = 18$ at $T = 0.2$ with $N = 36$ takes about 106 *minutes* by the FACOM VPP500 parallel supercomputer with a single processor. Typical numbers of steps we sampled are about 20 000 for most of the temperatures except for the low temperatures, $T < 0.2$, where we have done eight independent samplings with different initial conditions, each of which has more than 40 000 steps. Error bars are estimated from the deviation of

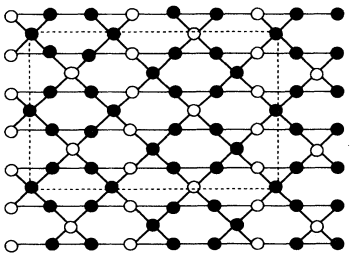

FIG. 1. The *kagomé* lattice. Spins that are traced out are indicated by •, while spins that are sampled are indicated by ○. The broken line is a periodic boundary for $N = 36$.

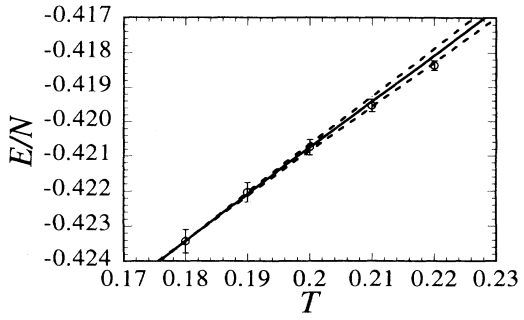

FIG. 2. Energies obtained by the reweighting method and the slope obtained as the specific heat.

data sets among these eight samplings. We have used the local three-spin flip along the Trotter axis, the world line flip, and a local winding number flip around a hexagon. Generally, the last type of the flip is necessary around each elementary polygon of the lattice. We have done the data analysis proposed by Hatano[19] at the temperatures where the sign problem is serious. We adopt the algorithm of Kawashima, Hatano, and Suzuki[20] to calculate correlation functions along the Trotter axis. This algorithm enables us to do MC sampling of the specific heat as the second derivative of the free energy. We also estimate the specific heat by the temperature derivative of the values of the energy which are obtained by the reweighting analysis. Both data agree with each other as shown in Fig 2.

Figure 3 shows low temperature behavior of the specific heat of systems of $N = 18, 36, 72$ together with the diagonalization result of $N = 18$ by Elstner and Young[12] for a check of our method. For $N = 18$, the results of both methods agree with each other for $T > 0.2$ and also the Monte Carlo data for these three sizes coincide. The data are proportional to T^{-2} for $T > 2.0$. This contradicts the experiment of Greywall and Busch, which showed the T^{-1} behavior.[2] For low temperatures, the existence of the second peak is confirmed. It is also seen that the data of $N = 36$ tend to be even larger than those of $N = 18$, although the differences are within the error bars. Therefore we conclude that the double peak is not a spurious finite size effect.

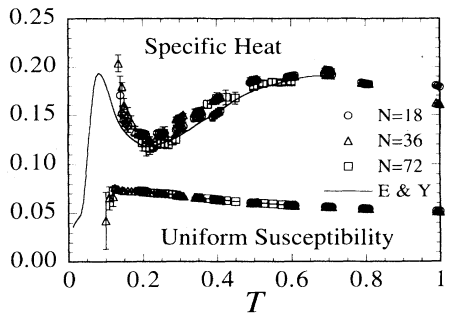

FIG. 3. Specific heat of $N = 18$ (○), $N = 36$ (△), and $N = 72$ (□) together with the diagonalization result of Elstner and Young of $N = 18$ (solid line). We also plotted the uniform susceptibility.

We have calculated the total entropy change from the specific heat data by

$$\Delta S = \int_0^\infty \frac{C}{T} dT, \tag{2}$$

assuming that $C \propto T^{-2}$ for the higher temperature region in which we did not simulate, and also that $C \propto T$ below the lowest temperature we observed. The integrated entropy changes are $(0.86 \pm 0.04) \ln 2$ for $N = 18$, and $(0.94 \pm 0.02) \ln 2$ for $N = 36$. Without the second peak taken into account, the integration gives $\sim 0.67 \ln 2$ for $N = 36$, which is consistent with the former calculations.[2,12] Uniform susceptibility is also shown in Fig. 3. The consistency of the Monte Carlo data of $N = 18$ with those of the diagonalization[12] was confirmed. The size dependence is very weak, and the susceptibility falls down to zero nearly at the temperature of the second peak of the specific heat. Below this temperature, singlet states are considered to be dominant because of the vanishing of the susceptibility. As has been obtained by the Lanczos diagonalization up to $N = 36$, the low-lying excited states are all singlet in the lattice with even numbers of spins, and there exists an energy gap between the lowest state of $S = 1$ and that of $S = 0$. The gap is about $0.25J$ as $N \to \infty$.[11] A sharp dump of the susceptibility is explained by the existence of the energy gap. Furthermore, we expect some structure in the density of states due to the energy gap, and that the structure would cause the second peak of the specific heat. The similar situation holds in the Δ-chain system where a finite energy gap exists above the ground state and the second peak of the specific heat stands at the temperature roughly equal to one-quarter of the gap.[17,21] Further simulations under the magnetic field may be necessary to check this speculation.

In Fig. 4, we have shown two types of chiral susceptibility; the staggered chirality corresponding to the $\sqrt{3} \times \sqrt{3}$ structure and the uniform chirality to the $q = 0$ structure. Here, chirality of three spins on a triangle is defined as

$$\kappa_v \equiv \frac{2}{\sqrt{3}} (\boldsymbol{S}_1 \times \boldsymbol{S}_2 + \boldsymbol{S}_2 \times \boldsymbol{S}_3 + \boldsymbol{S}_3 \times \boldsymbol{S}_1). \tag{3}$$

There is no clear difference between the two types within the temperature region simulated here. Moreover, there is no size enhancement. We could not observe evidence for the existence of any order in the ground state within the present calculations.

Short-range spin correlations $\langle S_i^z S_j^z \rangle$ are plotted in Fig. 5. We would expect at least a short-range magnetic order at a finite temperature if the ground state had some ordering structure. The nearest-neighbor correlation data are consistent with the energy and take about $3 \times 60\%$ of the classical value. For far neighbors the correlations rapidly decay with distance as was found previously by the diagonalization method.[5,11] There are two types of relative position for pairs connected by two steps, and three types for the three-step pairs. The correlations for the two-step pairs take small positive values. No clear difference between the two types was observed. For three-step pairs, the data take small negative values and the difference is also small. If the magnetic order, either the $\sqrt{3} \times \sqrt{3}$ structure or the $q = 0$ structure, was realized, the sign of the above correlations must depend on the relative position of the pair. We have calculated a dimer correlation defined by $(1/4 - \boldsymbol{S}_1 \cdot \boldsymbol{S}_2)(1/4 - \boldsymbol{S}_3 \cdot \boldsymbol{S}_4)(1/4 - \boldsymbol{S}_5 \cdot \boldsymbol{S}_6)$. Location of the spins is depicted in the inset of Fig. 5. We could not observe any anomalous behavior within the present calculations. The present results suggest the absence of order in the ground state.

At $T = 0.1$, the lowest temperature we observed for $N = 36$, the data fluctuate with large error bars especially for the quantities that are usually difficult to be measured, e.g., the specific heat and the chiral susceptibility. The staggered chirality decreases although the uniform chirality remains to increase at $T = 0.1$ in Fig. 4. This observation contradicts the results of the spin correlations discussed above, which do not show any sharp change at that temperature. Thus the data include a spurious error caused by the large fluctuation. Not only the sign problem but also the unsuitableness of the S^z representation for the complete set makes the sampling difficult,[22] and the important sampling cannot be achieved at low temperatures $T \leq 0.1$.

We confirmed that the second peak of the specific heat

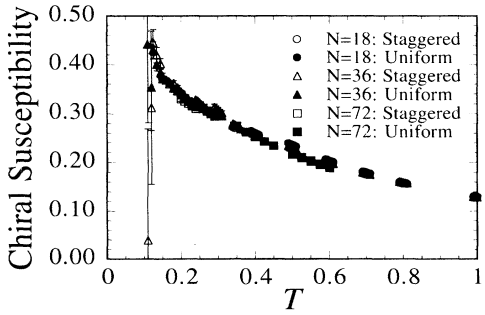

FIG. 4. Two types of chiral susceptibility; the staggered chirality (open symbols) and the uniform chirality (solid symbols) for $N = 18$ (circles), $N = 36$ (triangles), and $N = 72$ (squares). Neither size dependence nor the difference between the two types of chirality was observed for $T > 0.1$.

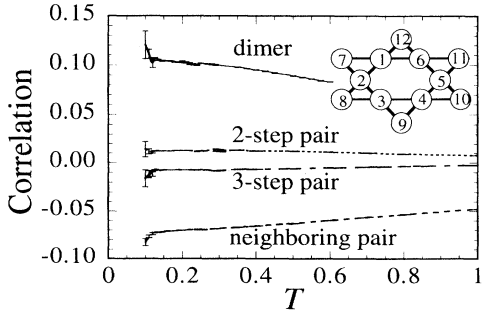

FIG. 5. The short-range spin correlations and the dimer correlation of the systems with $N = 36$ and $N = 72$. The inset shows the spatial configuration of spins. For two-step pairs, we calculated $\langle S_7^z S_8^z \rangle$ and $\langle S_7^z S_3^z \rangle$, and for three-step pairs we calculated $\langle S_7^z S_9^z \rangle$, $\langle S_7^z S_4^z \rangle$, and $\langle S_2^z S_5^z \rangle$.

exists in the system of $N = 36$, the smallest size for intrinsic properties for the *kagomé* lattice. For properties at lower temperatures, it is difficult to predict any conclusive nature. But it should be noted that in the temperature region where the specific heat shows relevant structure, namely, the double peak, we do not find any growth of short-range order. Actually the weak size dependences of various physical quantities suggest that the relevant excitations bear a local aspect. The nature of the ground state and the origin of the second peak should be investigated in future studies.

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