Specific heat of the quantum Heisenberg antiferromagnet on the *kagomé* lattice

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We propose a numerical technique, the forced-oscillator method, to investigate finite-temperature properties of quantum spin systems. The space and time complexity of this method are linear in the matrix dimension, which should be compared with the square or quadratic dependence of conventional methods. We apply this method to calculations of the specific heat of the spin-$\frac{1}{2}$ quantum Heisenberg antiferromagnet on the *kagomé* lattice. We find only a single-peak in contrast to a double-peak structure claimed in the literature.

I. INTRODUCTION

Frustrated quantum spin systems are of active current interest. Mainly the ground-state properties are discussed in the literature, but finite-temperature behavior also has unresolved aspects. We discuss the latter finite-temperature problem in the present paper with our principal focus on numerical techniques.

Numerical investigation of frustrated quantum spin systems faces serious difficulties when one tries to study systems with large numbers of spins. Direct numerical diagonalization requires enormous amount of computer memory if the number of spins exceeds about 16 for $S = 1/2$ since all eigenvalues of the Hamiltonian must be evaluated to know finite-temperature properties. The quantum Monte Carlo method by Trotter decompositions has its own serious problem, the negative-sign problem.[1,2] The decoupled-cell Monte Carlo simulation[3] tends to reflect quite strongly finite-size effects of decoupled cells.

In this paper we present the forced-oscillator method, which enables us to calculate finite-temperature properties with relatively small computational effort and thus is supplementary to other numerical methods mentioned above. This method was originally developed for calculations of the density of states of classical oscillators on various lattices.[4,5] We show that the forced-oscillator method is useful in quantum spin systems, frustrated or not, if appropriate correspondence is assigned between classical oscillators and spin states. We apply the method to the $S = 1/2$ quantum Heisenberg antiferromagnet (QHAF) on the *kagomé* lattice with up to 18 spins. Our result shows that there is only one peak in the specific heat, which is in contrast to the conclusion of Elser who found two peaks from direct diagonalization of the 12-spin system and a decoupled-cell Monte Carlo simulation[3] but is in agreement with Wang[6] who used a variant of mean-field theory. We also investigate the QHAF on the triangular lattice with parts of interactions being weaker than the others. This model has been a target of research in relation with the low-temperature properties of ^{3}He adsorbed on graphite.[7,8] We find a small shoulder in the low-temperature side of the specific heat. Further calculations must be carried out to confirm the existence of the latter shoulder in the thermodynamic limit.

The forced-oscillator method is explained in the next section. Applications to the frustrated QHAF are given in Sec. III. Discussions on physical and technical aspects of the problem are also found there.

II. ALGORITHM

The forced-oscillator method is based on the algorithm developed in the study of fractons in percolation clusters.[4,5] The essential idea is that eigenmodes satisfying the resonance condition are excited in applying a periodic external force. By counting the number of excited eigenmodes, we have found that we can obtain the density of states around the frequency of applied field. The number of excited modes is directly related with the energy expectation value as explained below.

Consider the Hamiltonian (energy) H of a classical M-oscillator system with unit mass,

$$H = \sum_{l}^{M} \frac{1}{2}\dot{u}_l\dot{u}_l + \sum_{ll'} \frac{1}{2}\Phi_{ll'}u_l u_{l'} - \sum_{l} F_l u_l \cos(\Omega t), \quad (1)$$

where u_l is the amplitude of the lth oscillator and $\Phi_{ll'}$ is the spring constant between the lth and l'th oscillators. The coefficient F_l is the external force driving the lth oscillator and Ω is the driving frequency. The first and second terms in (1) represent ordinary harmonic oscillators, and the third one is the external driving force. Varying this frequency Ω, we have found that we can obtain the density of states in an arbitrary range as follows. When the matrix $\Phi_{ll'}$ is positive definite and symmetric, its eigenvectors always exist. Using the equation of motion and orthogonality of eigenvectors, we find

$$\omega_\lambda^2 e_l(\lambda) = \sum_{l'} \Phi_{ll'} e_{l'}(\lambda), \quad (2)$$

where $e_l(\lambda)$ is the amplitude of the lth oscillator in the mode λ. If we regard ω_λ^2 as E_λ, this equation represents the eigenvalue problem. This fact makes it possible to evaluate the density of states of quantum spin systems by the present method which deals only with classical numbers. After driving the system for a sufficiently long time following the classical equation of motion of the oscillator

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system, we find that the average value of the Hamiltonian (1) asymptotically takes the form which is obtained from orthogonality of random numbers [$F_l = F_0 \cos(\phi_l)$, where F_0 is a constant, ϕ_l is a uniform random number]:[5]

$$H = 2\sum_{\lambda}(\varrho_{\lambda}^{0})^2\Omega^2 \sin^2\left(\frac{\omega_\lambda - \Omega}{2}t\right) = \frac{F_0^2\pi t}{8}\sum_{\lambda}\delta(\omega_\lambda - \Omega), \tag{3}$$

where ϱ_λ^0 is defined as $\sum_l F_l e_l(\lambda)/(\Omega^2 - \omega_\lambda^2)$. The last expression is nothing but the density of states at Ω. Thus, by calculating the energy of an oscillator system, we can evaluate the density of states.

It should be remarked here that the asymptotic form (3) of the Hamiltonian is correct when $\Omega t \gg 1$ and $4\pi N/(\omega_{\max} t) \gg 1$. The former constraint comes from the fact that we should drive the system until resonance sets in. The latter inequality implies that too small a number of modes will be excited if one drives the forced oscillation too long as compared to the largest eigenfrequency $\omega_{\max}$, which prevents us from calculating the density of states as a smooth function of Ω.

This algorithm may be applied to calculate the density of states of any symmetric matrix including quantum spin systems. The idea is to regard a spin configuration as a state of an oscillator system. These oscillators are of course virtual ones. For instance, consider a two-spin system with $S = 1/2$; the first state 0 of an oscillator system is identified with a spin configuration 00(binary), where 0 represents a down spin. The amplitude of the spin state 00 is just the amplitude u_0 of the first oscillator. The matrix element between 0(00) and 1(01) is identified with $\Phi_{0,1}$.

Once the density of states $D(E_\lambda)$ is known, it is simple to evaluate the specific heat by calculating fluctuations of the energy eigenvalues, $\int E_\lambda^2 D(E_\lambda)dE_\lambda - \{\int E_\lambda D(E_\lambda)dE_\lambda\}^2$. We obtain more stable values by this numerical integration than the numerical differentiation of $\int E_\lambda D(E_\lambda)dE_\lambda$, which is equivalent to the formula above. For an arbitrary physical quantity A, the following formula is useful:

$$\sum_{\lambda}\delta(\omega_\lambda - \Omega)A_{\lambda\lambda} = \frac{4}{{F_0}^2\pi t}\sum_{ll'}(\dot{u}_l\dot{u}_{l'} + \Omega^2 u_l u_{l'})A_{ll'}, \tag{4}$$

where $A_{\lambda\lambda}$ is the eigenvalue of the arbitrary operator and $A_{ll'}$ is the matrix element in the u_l representation. Using (4), the expectation value of the given operator can be calculated.

For a sufficiently large system it will be unnecessary to take the average over all possible $\{\phi_l\}$ explicitly because of the self-averaging property. It suffices to use one random choice of $\{\phi_l\}$. The system size $M \sim 10^5$ turns out to be sufficient in the examples given below. Numerical integration of the differential equations to describe time development of the oscillator system also becomes stable as the system size grows, because statistical errors play less significant roles.

III. RESULTS AND DISCUSSION

We have applied our method to the QHAF on the *kagomé* lattice. The Hamiltonian is

$$H = J\sum \mathbf{S}_i \cdot \mathbf{S}_j, \tag{5}$$

where the $\mathbf{S}_j$ is a spin-1/2 operator. The sum is over nearest neighbors of the *kagomé* lattice. We followed Refs. 9 and 10 to cut periodic finite cells out of an infinite lattice.

We first tested our method in the 12-spin system. This system has $2^{12} = 4096$ states, which turns out slightly insufficient to assure stable results with respect to the self-averaging property mentioned above. The value of the specific heat depends upon the random choice of $\{\phi_l\}$. Thus we had to perform the sample average explicitly. After averaging over several samples, we have obtained the final values shown in Fig. 1 (points with diamond symbols). The exact result by direct numerical diagonalization is also shown for comparison. Qualitative agreement is satisfactory including the existence of a peak at $k_BT/J \sim 0.10$. The peak value is seen to be somewhat underestimated. This may come from the smallness of the number of states of the given matrix. When the distribution of states is not dense, the states excited by the external force do not distribute smoothly. Correspondingly the asymptotic value of H/t after sufficiently long time depends upon the initial condition. This lack of smoothness leads to relatively large statistical uncertainties at the lower temperature peak. This uncertainty is expected to decrease rapidly with the system size since the number of states increases exponentially with the number of spins.

Increase of statistical stability as the system size grows has actually been observed in the time development of the classical oscillator system which we solved by numerical integration. For $N = 12$, we have observed from time to time that the value of H/t increases linearly in t after some critical time $t > t_c$, which is not allowed from (3) if the system satisfies the self-averaging condition. For $N = 15$ or larger, this type of instability was only rarely observed.

In the case of the system with $N = 15$ spins, we find the high-temperature peak almost at the same temperature as in the 12-spin case (Fig. 2). The lower-temperature

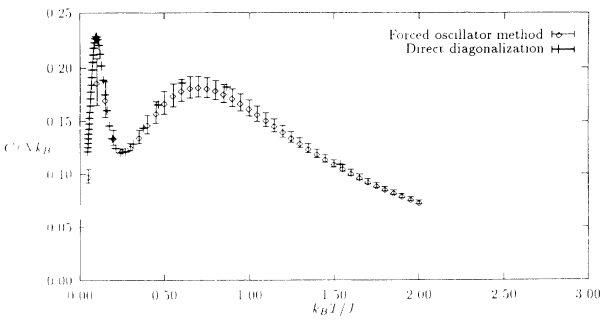

FIG. 1. The specific heat calculated by the forced-oscillator method and direct diagonalization for the QHAF on the *kagomé* lattice with $N = 12$.

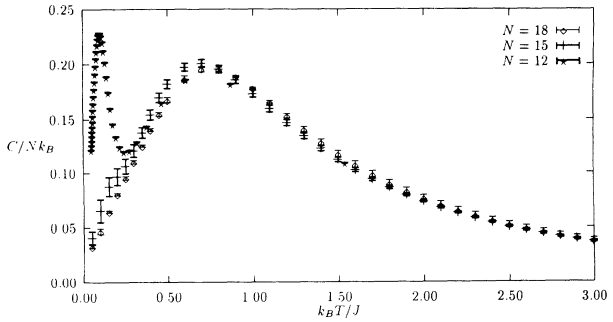

FIG. 2. The specific heat of the QHAF on the *kagomé* lattice with $N = 12$–18 with periodic boundaries. For $N = 12$ only, the data are from direct numerical diagonalization. Statistical errors are of the order of $1/\sqrt{M}$, where M is the number of spin configurations $M \sim 2^N$. Thus the larger-system data ($N = 18$) are much more reliable than smaller-system counterparts.

peak found in the $N = 12$ system is not clearly observed; only a small shoulder remains. The lower-temperature peak disappears completely in the 18-spin system. Decrease of this peak with system size is seen also in the density of states (Fig. 3).

Our result up to 18 spins contradicts the conjecture proposed in Refs. 9 and 3, and clearly shows disappearance of the lower-temperature peak for large systems. The lower-temperature peak may be simply a finite-size effect. Wang noticed that the specific heat of the present model calculated using the Wigner-Jordan transformation and a decoupling approximation has only a single peak.[6] Our result agrees with his prediction on the location of the high-temperature peak, but does not agree quantitatively in the peak value. As suggested by Wang,[6] the higher-temperature peak, which he assumed to correspond to that at 2.5 mK in experiments of ^{3}He adsorbed on graphite,[7,8] occurs from interactions between spins on the *kagomé* net. The assumption of Elser[9] that the exchange interactions between spins on the *kagomé* lattice and those on other sites are weak may be inappropriate

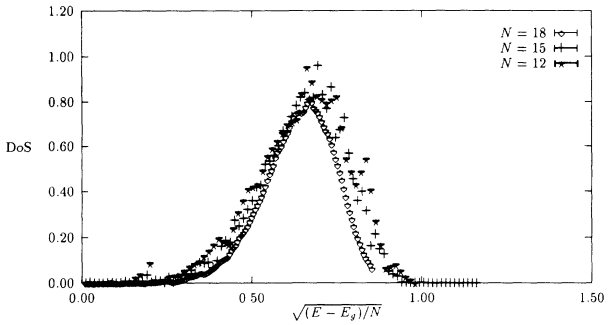

FIG. 3. The density of states of the QHAF on the *kagomé* lattice with $N = 12$–18 with periodic boundaries. If E_g denotes the ground energy, the range of the density of states, $\sqrt{E - E_g}$, becomes large in proportion to $\sqrt{N}$ as the system size grows. Thus the range of this density of states is normalized by a factor $\sqrt{N}$. The range around 0.2 for $N = 12$ causes the low-temperature peak. The value of the density of states in this range decreases with the system size. For $N = 12$ only, the data are from direct numerical diagonalization.

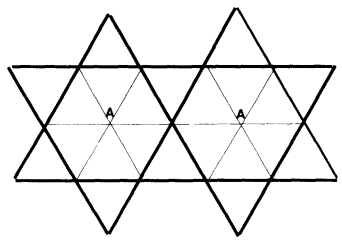

FIG. 4. The bold lines constitute the *kagomé* lattice. The thin lines between the points marked as A and *kagomé* lattice are the weak interactions with exchange value $0.4J$.

in the present system.

We therefore added spins at the center of the hexagon of the $N = 12$ *kagomé* lattice to form a triangular lattice with $N = 16$ spins (Fig. 4). The newly added interactions are assumed to be $0.4J$ following Wang.[6] Our result (Fig. 5) shows a small peak at the low-temperature side $(0.05 - 0.1)J$. This is in qualitative agreement with the calculations in Ref. 6. However, this $N = 16$ triangular lattice has been formed from the $N = 12$ pure *kagomé* lattice, and hence this result may reflect boundary effects like the $N = 12$ *kagomé* cluster in Fig. 2. Therefore the lower-temperature peak in Fig. 5 should be taken with special caution. It is not easy at present to treat larger triangular lattices.

It will be useful to comment on the efficiency of the forced-oscillator method. This algorithm may be applied to calculate the density of states of an arbitrary Hermitian matrix including quantum spin systems. The only constraint is that the matrix be positive definite and symmetric. Positive definiteness is easily achieved by adding a constant to diagonal elements. For matrix dimension of the order of $M \sim 10^5$ or larger, the asymptotic time complexity is proportional to $M \times$ (maximum of the number of nonzero elements at each matrix row). This complexity is one of the smallest among the well-known methods to calculate all eigenvalues.[11] The memory requirement is also small as compared with the conventional methods to calculate all eigenvalues:[11] The density of states can be calculated under the same memory requirement as that for calculating the ground-state energy. The weakness of the method is in calculating the density of states near

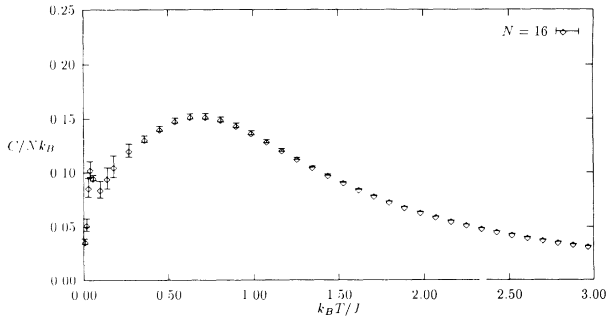

FIG. 5. The specific heat of the QHAF on the triangular lattice with two kinds of couplings derived from the *kagomé* lattice by adding weak ($0.4J$) interactions. This figure is for $N = 16$ with periodic boundaries which corresponds to $N = 12$ on the original *kagomé* lattice. We can see a small peak at $0.05J - 0.1J$.

the ground-state energy, which comes from the relative smallness of the number of states near the ground state and resulting large statistical errors.

Imada and Takahashi's quantum transfer Monte Carlo method is also effective for calculating finite-temperature properties.[12] The system size one can calculate using his method is the same as ours. The time complexity is comparable to our method. His method contains no Monte Carlo procedure in the matrix transfer; thus it has no difficulty such as the negative-sign problem.[1,2] However, in the quantum transfer Monte Carlo method, the initial vector is chosen as a random linear combination of the basis vectors, which is a rather arbitrary process. Our method does not contain such ambiguities.

In summary, we have shown that the forced-oscillator method is useful to calculate physical properties of quantum spin systems. Finite-temperature behavior can be treated with the same memory requirement as that for the ground-state problem. Application to the Heisenberg antiferromagnet on the *kagomé* lattice has shown that the specific heat has only a single peak.

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