Thermodynamic properties of the quantum antiferromagnet on the triangular lattice in a magnetic field

N. Suzuki and F. Matsubara
Department of Applied Physics, Faculty of Engineering, Tohoku University, Sendai 980-8579, Japan
(Received 7 January 1998; revised manuscript received 7 April 1998)

We study the thermodynamic properties of the $S=\frac{1}{2}$ quantum antiferromagnetic Heisenberg model on finite triangular lattices with N ($\leqslant 27$) spins in a magnetic field using a quantum transfer Monte Carlo method. The size dependence of the order parameter suggests that a threshold ($H_1 \sim 2.8J$) exists, below which the long-range order is broken by the quantum fluctuation. For $H>H_1$, only the longitudinal component of the order parameter has a finite nonzero value at low temperatures. The transition temperature is estimated at different magnetic fields and the phase diagram is predicted. [S0163-1829(98)06334-6]

Recently, the spin ordering of the low-dimensional frustrated spin system has attracted much interest. The antiferromagnet on the triangular lattice is a typical frustrated spin system. In the classical case, the ground state has a 120° structure and is discretely degenerated with chirality. The 120° structure disappears at finite temperatures, but if an easy plane anisotropy exists, i.e., $J_z/J_{xy}<1$, the ground state degeneracy of chirality causes a phase transition.[1] In the quantum case, it is known that the *XY* model, i.e., $J_z=0$, has the long-ranged chiral order phase at finite temperatures.[2–4] On the other hand, in the Heisenberg model, it is still controversial whether or not the classical 120° ground state is stable against the quantum fluctuation.[4–7] Recently, the authors studied an anisotropic Heisenberg model at finite temperatures[8] by a quantum transfer Monte Carlo (QTMC) method.[9] They found a threshold of $J_z/J_{xy}\sim 0.4$ below which the chiral phase transition occurs at a finite temperature. The disappearance of long-range order for $0.4 \lesssim J_z/J_{xy} \leqslant 1$ has been attributed to the quantum fluctuation of the z component of the spins.[7] That is, in contrast with the classical case the spin structure may not depend only on the symmetry of the model but also on the nature of the quantum fluctuation.

In this paper, we consider the effect of a magnetic field which breaks the spin-reversal symmetry. In the classical Heisenberg antiferromagnet on a triangular lattice, the 120° structure with $f^{\parallel}\neq 0$ and $f^{\perp}\neq 0$ appears in an infinitesimal magnetic field, where $f^{\parallel}$ and $f^{\perp}$ are the longitudinal and transverse components of the order parameter.[10] As the magnetic field increases, it changes successively into a ferrimagnetic phase with $f^{\parallel}\neq 0$ and $f^{\perp}=0$ and a spin flop phase with $f^{\parallel}\neq 0$ and $f^{\perp}\neq 0$. We ask the following questions in the $S=1/2$ Heisenberg model. (1) Do both $f^{\parallel}$ and $f^{\perp}$ have nonvanishing values in the magnetic field? (2) If they do, is it true even in an infinitesimal magnetic field? (3) Does any phase transition occur as the magnetic field increases? We calculate the specific heat, the magnetization, and the order parameter at finite temperatures for lattices with $N\leqslant 27$ spins by using the QTMC method. The results suggest the following. (1) The magnetization curve has a 1/3 plateau[11,12] for $H_1<H<H_2$, which indicates the occurrence of a ferrimagnetic phase. (2) For $H<H_1$, the long-range order is broken by the quantum fluctuation. (3) For $H>H_1$, only $f^{\parallel}$ has a nonzero value at low temperatures. We note that $f^{\perp}$ disappears at any magnetic field, because the spin rotational symmetry around the magnetic field is not broken. That is, a collinear phase occurs for $H>H_1$. The transition temperature is estimated from the size dependence of the order parameter and the phase diagram is predicted.

We start with the Hamiltonian

$$\mathcal{H}=2J\sum_{\langle i,j\rangle}\mathbf{S}_i\cdot\mathbf{S}_j-H\sum_i S_i^z, \tag{1}$$

where J (>0) is the exchange integral, H is an external magnetic field along the z direction, and $\langle i,j\rangle$ runs all the nearest neighbor pairs. We consider properties of the model *at finite temperatures* using the QTMC method.[9] In the QTMC method, we calculate the following quantity:

$$\widetilde{\langle A\rangle}=\sum_k^M \langle\psi_k|A\exp(-\beta\mathcal{H})|\psi_k\rangle \Big/ \sum_k^M \langle\psi_k|\exp(-\beta\mathcal{H})|\psi_k\rangle, \tag{2}$$

where A is some physical operator and the sum runs over M states, each of which is given by $|\psi_k\rangle=\sqrt{6/M}\Sigma_i^{2^N}C_{ik}|i\rangle$; here C_{ik} is a uniform random number of $-1\leqslant C_{ik}\leqslant 1$. We can easily show $\widetilde{\langle A\rangle}=\langle A\rangle+O(1/\sqrt{M})$, where $\langle A\rangle$ is the thermal average calculated by using all the 2^N Ising states. In this calculation, the lattice size is limited by the memory size of the computer. Here, we divide the lattice into two parts, so that we can treat the model on the lattice $N\leqslant 27$.[13] We can get results at *arbitrary* field *from those of* $H=0$. For example, the specific heat at $H=h$ is calculated as follows. First, we calculate

$$Z_m=\sum_k \langle \psi_k^{(m)}|\exp(-\beta\mathcal{H}_0)|\psi_k^{(m)}\rangle,$$

$$E_m=\sum_k \langle \psi_k^{(m)}|\mathcal{H}_0\exp(-\beta\mathcal{H}_0)|\psi_k^{(m)}\rangle, \tag{3}$$

$$C_m=\sum_k \langle \psi_k^{(m)}|\mathcal{H}_0^2\exp(-\beta\mathcal{H}_0)|\psi_k^{(m)}\rangle.$$

Here $|\psi_k^{(m)}\rangle$ is a similar state composed of Ising states with the total spin m, and $\mathcal{H}_0$ is the Hamiltonian (1) with $H=0$. Then, we obtain

$$C=[C_h/Z_h-(E_h/Z_h)^2]/T^2, \tag{4}$$

where

$$Z_h=\sum_m \exp(\beta mh)Z_m,$$

$$E_h=\sum_m \exp(\beta mh)[E_m-mhZ_m], \tag{5}$$

$$C_h=\sum_m \exp(\beta mh)[C_m-2mhE_m+m^2H^2Z_m].$$

The numbers of the states M are as follows: $M=50$ for $N\leqslant 21$, $M=10$ for $N=24$, and $M=2$ for $N=27$. For every size of the lattice except for $N=27$, the set of M states is divided into five subsets and quantities of interest are calculated in every subset. Error bars presented in the figures given below mean deviations of the values obtained for different subsets. Since the results depend markedly on whether N is even or odd,[14] we use the data only for odd N to consider the size dependence.

First we consider the magnetization M^z and the specific heat C given by

$$M^z=(1/N)\left\langle \sum_i S_i^z \right\rangle, \tag{6}$$

$$C=(1/NT^2)(\langle\mathcal{H}^2\rangle-\langle\mathcal{H}\rangle^2). \tag{7}$$

In Figs. 1(a) and 1(b), we show the magnetizations at $T=0.20J$ and $0.33J$ together with that in the ground state for $N=27$. The data for different size N lie almost on the same line. The 1/3 plateau appears and the positions of the edges depend little on the temperature. Therefore we believe that the 1/3 plateau occurs in the thermodynamic limit and its range in the magnetic field is almost independent of the temperature. Hereafter, we denote the edges of the plateau as $H_1(\sim 2.8J)$ and $H_2(\sim 4.2J)$. In Figs. 2(a)–2(d), we present results of the specific heat. In the plateau range, the peak height becomes higher with increasing N, suggesting the occurrence of a phase transition. This is not unexpected, because in the Ising model the ferrimagnetic phase with $M_z=1/3$ occurs at finite temperatures.

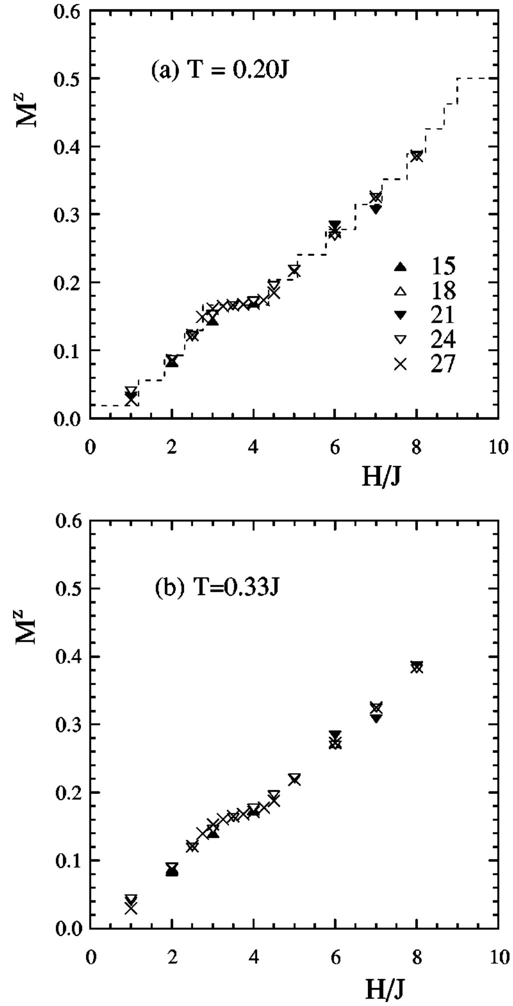

FIG. 1. The magnetization curves at (a) $T=0.20J$ and (b) $T=0.33J$. The dashed line in (a) is the result in the ground state for $N=27$.

Now we consider temperature dependences of the longitudinal and transverse components $f^{\parallel}$ and $f^{\perp}$ of the order parameter:

$$f^{\parallel}=f^z, \tag{8}$$

$$f^{\perp}=(f^x+f^y)/2, \tag{9}$$

with

$$f^{\alpha}=\frac{1}{2}\langle(M_A^{\alpha}-M_B^{\alpha})^2+(M_B^{\alpha}-M_C^{\alpha})^2+(M_C^{\alpha}-M_A^{\alpha})^2\rangle, \tag{10}$$

where M_{ζ}^{α} $[\equiv\Sigma_{i\in\zeta}S_i^{\alpha}/(N/3)]$ means the α component of the magnetization of the ζ sublattice. Here we divide the lattice into three sublattices A, B, and C, because the classical model has the 120° structure at $T=0$. If the classical 120° phase or the spin flop phase occurs, both $f^{\parallel}$ and $f^{\perp}$ have some nonzero value. In the ferrimagnet phase, only $f^{\parallel}$ has some nonzero value. In Figs. 3(a)–3(d), we plot $f^{\parallel}$ as a function of T. For all H, $f^{\parallel}$ increases with decreasing T. When $H\lesssim H_1$, the size dependence is large even at low temperatures. On the other hand, when $H\gtrsim H_1$, the size dependence is small at low temperatures, suggesting that $f^{\parallel}$ re-

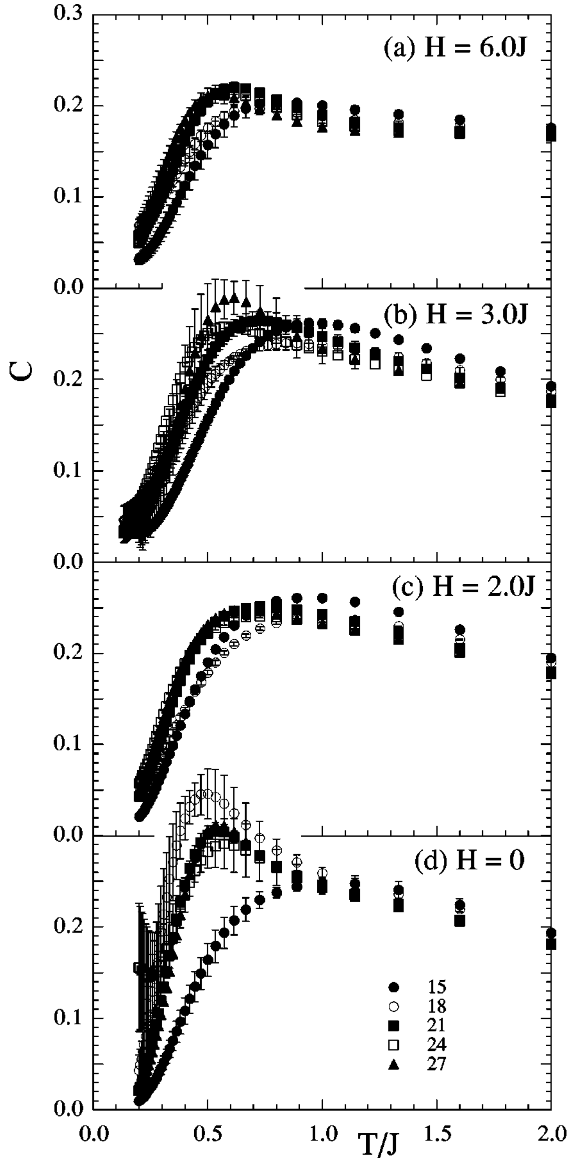

FIG. 2. The specific heat at different magnetic fields.

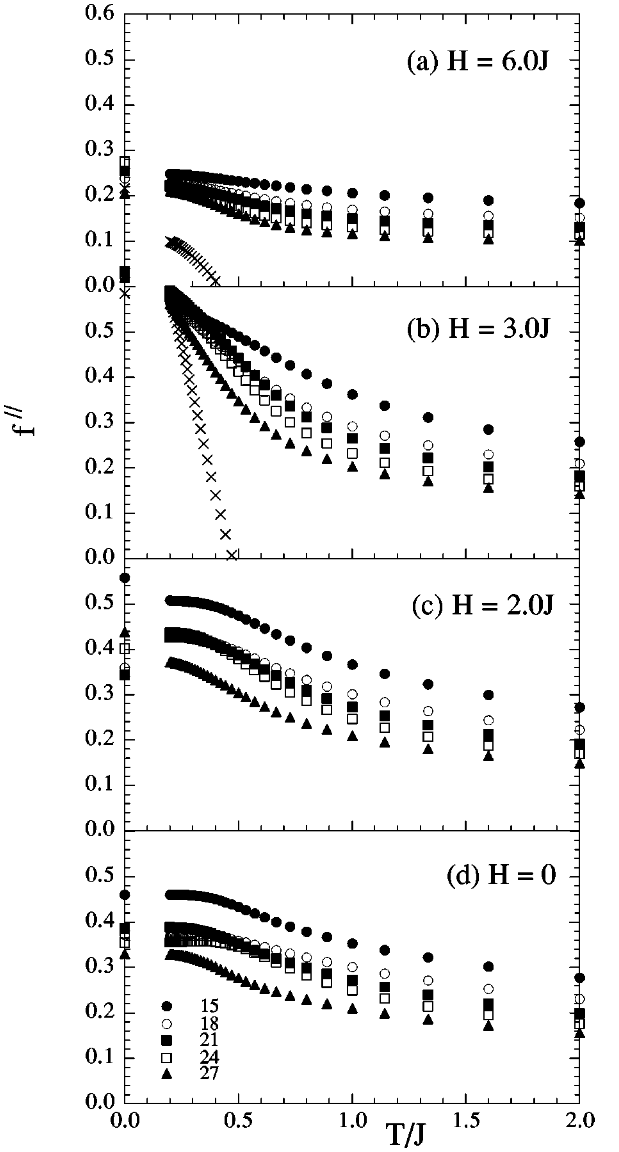

FIG. 3. The order parameter $f^{\parallel}$ at different magnetic fields. The symbol $\times$ denotes an extrapolated value described in the text.

mains nonzero as $N\rightarrow\infty$. We fit the data on a conventional $1/\sqrt{N}$ function and estimate values for $N\rightarrow\infty$,[15] which is shown in Fig. 4. The temperature dependence of the extrapolated values is also plotted in Figs. 3(a) and 3(b). When $H \gtrsim H_1$, $f^{\parallel}$ has a nonzero value at low temperatures, and becomes smaller and vanishes as the temperature increases. In Figs. 5(a)–5(c), we plot $f^{\perp}$ as a function of T. Although $f^{\perp}$ increases with lowering the temperature except for $H=4.0J$, its size dependence is not changed considerably at all temperatures, suggesting the disappearance of it for $N\rightarrow\infty$. In fact, we get a small negative value of $f^{\perp}$ by the same extrapolation.

From the results mentioned above, we predict the phase diagram which is shown in Fig. 6. We plot the critical magnetic field of H_1. Only when $H \gtrsim H_1$ does the long-range order appear which is characterized by $f^{\parallel}\neq 0$ and $f^{\perp}=0$. We estimate the transition temperature from $T_c=(T_1+T_2)/2$, where T_1 is the temperature at which the extrapolated value of $f^{\parallel}$ becomes the half maximum of it at $T=0$ and T_2 is the temperature at which the extrapolated

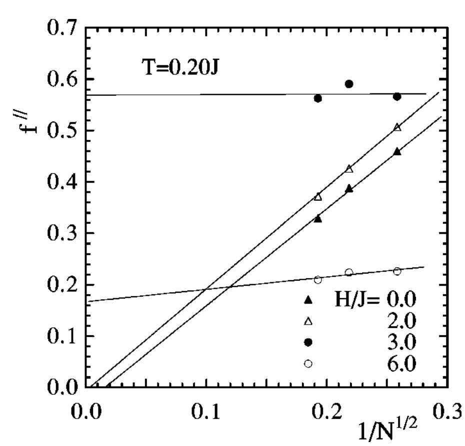

FIG. 4. An extrapolation of the order parameter $f^{\parallel}$ at different magnetic fields.

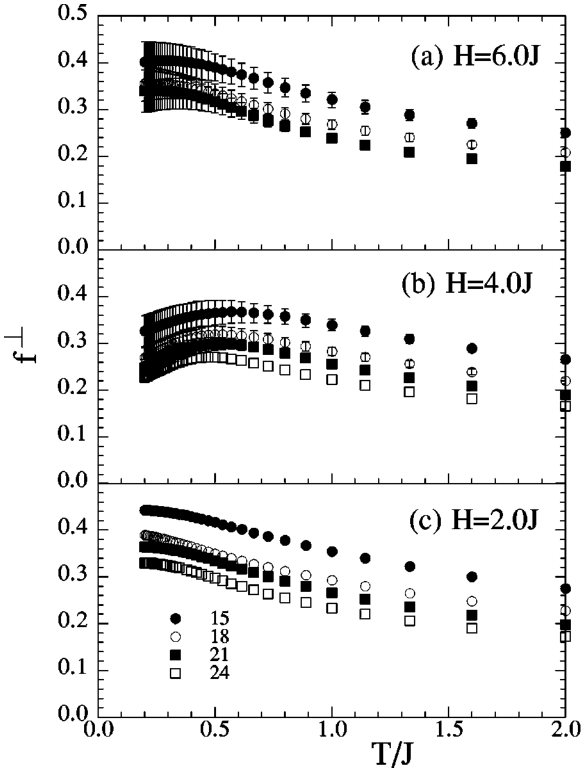

FIG. 5. The order parameter $f^{\perp}$ at different magnetic fields.

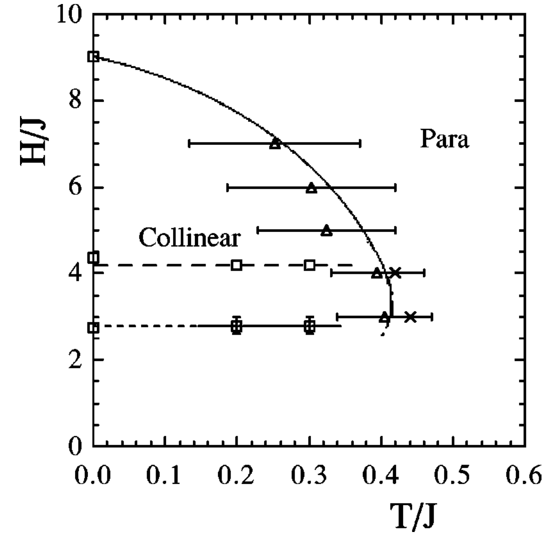

FIG. 6. The phase diagram in the temperature-magnetization plane. The symbols □, △, and × are estimated from the edges of the plateau of the magnetization curve, the order parameter, and the peak temperature of the specific heat, respectively. The lines are guides to the eye.

value vanishes. The value of T_c estimated in this way is not incompatible with the peak temperature of the specific heat. Especially for $H_1 \lesssim H \lesssim H_2$, the extrapolation of the peak temperature by the $1/N^2$ function[8] gives a very close value. We also plot the other edge of the plateau, H_2. Of course, the phase for $H_1 \lesssim H \lesssim H_2$ is related to the ferrimagnetic phase in the classical case. The phase for $H \gtrsim H_2$ will correspond to the spin flop phase in the classical case, but the transverse component $f^{\perp}$ is absent.[16] Both phases above and below H_2 are the collinear phases, but some differences in the magnetizatic property appear between two phases, (1) the size dependence of the specific heat and (2) the temperature dependence of the order parameter $f^{\perp}$. Further studies are necessary to see whether H_2 is a phase boundary or nearly a crossover line. We note that the occurrence of the collinear phase was speculated in an Ising-like model.[17] The important point predicted in this study is that the same is true in the isotropic model and the other long-range order phase occurs at higher fields. Finally, it should be mentioned that the breaking of spin symmetry is the necessary condition to the occurrence of a long-range order, but there is a finite, nonzero threshold below which the long range-order is broken by quantum fluctuation.

We would like to thank Dr. T. Nakamura for valuable discussions. The diagonalization programs are based on the subroutine package KOBEPACK/S Ver. 1.1 by Professor T. Tonegawa, Professor M. Kaburagi, and Dr. T. Nishino. The computation in this work has partially been done using the facilities of the Supercomputer Center, Institute for Solid State Physics, University of Tokyo.

[1] S. Miyashita and H. Shiba, J. Phys. Soc. Jpn. **53**, 1145 (1984); S. Miyashita, Prog. Theor. Phys. Suppl. **87**, 112 (1986).
[2] F. Matsubara and S. Inawashiro, Solid State Commun. **67**, 229 (1988).
[3] T. Momoi and M. Suzuki, J. Phys. Soc. Jpn. **61**, 3277 (1992).
[4] P. W. Leung and K. J. Runge, Phys. Rev. B **47**, 5861 (1993).
[5] P. W. Anderson, Mater. Res. Bull. **8**, 153 (1973); P. Fazekas and P. W. Anderson, Philos. Mag. **30**, 432 (1974).
[6] B. Bernu, C. Lhuillier, and L. Pierre, Phys. Rev. Lett. **69**, 2590 (1992).
[7] N. Suzuki and F. Matsubara, J. Phys. Soc. Jpn. **65**, 1121 (1996).
[8] N. Suzuki and F. Matsubara, Phys. Rev. B **51**, 6402 (1995).
[9] M. Imada and M. Takahashi, J. Phys. Soc. Jpn. **55**, 3354 (1986).
[10] H. Kawamura and S. Miyashita, J. Phys. Soc. Jpn. **54**, 4530 (1985).
[11] H. Nishimori and S. Miyashita, J. Phys. Soc. Jpn. **55**, 4450 (1986).
[12] A. V. Chubukov and D. I. Golosov, J. Phys. C **3**, 69 (1991).
[13] We use an algorithm similar to that used in ''KOBEPACK/S.''
[14] The data for even N exhibit an unusual temperature dependence at low field as seen in Fig. 3. This behavior will come from a property of the ground state as discussed in Ref. 8.
[15] We also fit the data on the $1/N$ function, but the extrapolated value at $H=0$ becomes nonzero, which is of course unphysical.
[16] Though the peak height of the specific heat for $H>H_2$ does not increase with N, we think that the phase transition occurs at $T=T_c$, because in the classical case, the height of the specific heat does not increase although the spin flop phase exists as seen in Ref. 10.
[17] S. Miyashita, M. Takasu, and M. Suzuki, in *Quantum Monte Carlo Methods in Equilibrium and Non Equilibrium Systems*, edited by M. Suzuki (Springer-Verlag, Berlin, 1986), p. 104.