Magnetization plateau in a two-dimensional multiple-spin exchange model

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(Received 28 September 1998)

We study a multiple-spin exchange model on a triangular lattice, which is a possible model for low-density solid ^{3}He films. Due to strong competitions between ferromagnetic three-spin exchange and the antiferromagnetic four-spin one, the ground states are highly degenerate in the classical limit. At least $2^{L/2}$-fold degeneracy exists on the $L\times L$ triangular lattice except for the SO(3) symmetry. In the magnetization process, we found a plateau at $m/m_{\mathrm{sat}}=\frac{1}{2}$, in which the ground state is *uuud* state (a collinear state with four sublattices). The $\frac{1}{2}$ plateau appears due to the strong four-spin exchange interaction. This plateau survives against both quantum and thermal fluctuations. Under a magnetic field which realizes the *uuud* ordered state, a phase transition occurs at a finite temperature due to the breakdown of translational symmetry. We predict that low-density solid ^{3}He thin films may show the $\frac{1}{2}$ plateau in the magnetization process. Experimental observation of the plateau will verify the strength of the four-spin exchange. It is also discussed that this magnetization plateau can be understood as an insulating-conducting transition in a particle picture. [S0163-1829(99)01514-3]

I. INTRODUCTION

In localized fermion systems, the magnetic interaction comes from permutations of particles.[1,2] Multiple-spin exchanges, e.g., cyclic exchanges of three or four spins, have been revealed to be strong in nuclear magnetism of two-dimensional (2D) solid ^{3}He films. Many experimental[3–5] and theoretical[6–10] studies suggested that exchange interactions of more than two spins are dominant in this system. A ferromagnetic behavior changes to the antiferromagnetic one when the coverage of ^{3}He decreases.[11–15] This tendency can be understood in terms of multiple-spin exchange (MSE): in fully packed systems, the three-spin exchange is dominant[6] and it is ferromagnetic (as shown by Thouless[2]) and, in loosely packed systems, the four- and six-spin exchanges become strong and favor antiferromagnetism. Effects of multiple-spin exchanges are not yet fully understood especially for the low-density region.[16,5] For example, recent specific-heat data at low densities show a peculiar behavior, which have a double-peak structure, and they also show that the ground state seems to be spin liquid (disordered) and the spin excitation gap is vanishing (or quite small).[5]

The multiple-spin exchange model has been studied to describe the nuclear magnetism of three-dimensional solid ^{3}He.[17] A general form of the spin Hamiltonian of quantum solid is[1,2] $\mathcal{H}=-\Sigma_n(-1)^nJ_n\Sigma_{P_n}P_n$, where P_n and $J_n(\leqslant 0)$ denote cyclic permutation of n spins and its exchange constant, respectively. For the 2D system, recent theoretical calculations[8–10] and experimental measurements[4] found that the exchange frequencies satisfy $|J_3|>|J_2|>|J_4|\gtrsim|J_6|\gtrsim|J_5|$ on the triangular lattice. In this paper, we consider a spin model with the two-, three-, and four-spin exchanges on the triangular lattice, which is the simplest 2D MSE model. Since the three-spin exchange can be transformed to the two-spin ones, the Hamiltonian can be written with two parameters J and K as

$$\mathcal{H}=J\sum_{\langle i,j\rangle}\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_j+K\sum_p h_p-\mu B\sum_i \sigma_i^z, \tag{1}$$

where $\boldsymbol{\sigma}_i$ denote Pauli matrices. The last term means the Zeeman energy, where μ denotes the nuclear magnetic moment of ^{3}He and B the magnetic field. The parameter $J(=J_3-J_2/2)$ is negative for most of densities (but it can change the sign) and $K(=-J_4/4)$ is always positive ($K\geqslant 0$). The first and the second summations run over all pairs of nearest neighbors and all minimum diamond clusters, respectively. The explicit form of h_p for four sites (1,2,3,4) is

$$\begin{aligned}h_p&=4(P_4+P_4^{-1})-1\\&=\sum_{1\leqslant i<j\leqslant 4}\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_j+(\boldsymbol{\sigma}_1\cdot\boldsymbol{\sigma}_2)(\boldsymbol{\sigma}_3\cdot\boldsymbol{\sigma}_4)+(\boldsymbol{\sigma}_1\cdot\boldsymbol{\sigma}_4)(\boldsymbol{\sigma}_2\cdot\boldsymbol{\sigma}_3)\\&\quad-(\boldsymbol{\sigma}_1\cdot\boldsymbol{\sigma}_3)(\boldsymbol{\sigma}_2\cdot\boldsymbol{\sigma}_4),\end{aligned} \tag{2}$$

where (1,3) and (2,4) are diagonal bonds of the diamond. The WKB approximations[7,9] show that exchange parameters vary depending on the particle density: At high densities, the exchange $J(\leqslant 0)$ is dominant, which mainly originates from the three spin exchange. As the density is lowered, the ratio $|K/J|$ increases rapidly and hence the four spin exchange $K(\geqslant 0)$ becomes important. This density dependence is consistent with experimental results of susceptibility.[11–15] Since multiple-spin exchanges produce frustration by themselves and strong competitions between exchanges also introduce frustration,[18] this model is expected to show various complex magnetic behaviors.

In a previous paper, we studied the ground state of this model in the classical limit and found various phases:[19] (a) For $J<-8K$, the ground state shows the perfect ferromagnetism. (b) For $-8K<J<-8K/3$, ground states are highly degenerate. This degeneracy is a nontrivial one. (c) In $-8K/3<J<25K/3$, the ground state has a four-sublattice structure with zero magnetization, which we call as the tetrahedral structure.[20] (d) For $25K/3<J$, the ground state is the so-called 120° structure. Thus phases (b) and (c) appear due to the four-spin exchange interactions. In the region (c), we predicted chiral symmetry breaking at a finite temperature.[20]

The intermediate phase (b) seems to correspond to the parameter region of 2D low-density solid ^{3}He. The parameters J and K in the low density region are estimated as $J \simeq -1.5$ (mK) and $K \simeq 0.2$ (mK) from susceptibility and specific-heat data,[4] and $J/K = -4 \pm 2$ from path integral Monte Carlo simulations.[8–10] The five- and six-spin exchanges are also estimated to be comparable with four-spin one. This parameter region almost belongs to the phase (b) of our study in the classical limit. Our previous study[19] shows that competitions between the two- and four-spin exchanges are strong in this phase and many kinds of ground states exist due to frustration. Furthermore, we found that under the magnetic field a plateau appears at $m/m_{\mathrm{sat}} = \frac{1}{2}$ in the magnetization curve, where m_{sat} denotes the saturated magnetization. These various unusual phenomena occur due to frustration caused by the multispin exchanges. In this paper we study this phase further, especially considering quantum effects. Finite-temperature effects are also discussed.

In Sec. II we summarize the results for the phase (b) of the classical model. Among degenerate ground states, one collinear state, which we call *uuud* state, has the largest magnetization, $m/m_{\mathrm{sat}} = \frac{1}{2}$, and other states show $m/m_{\mathrm{sat}} < \frac{1}{2}$. In Sec. III we discuss the quantum model. Due to quantum effects, the *uuud* state disappears from the ground state at $B=0$ and the ground state belongs to the $S=0$ space. The magnetization process of the quantum model is studied in Sec. IV. Under the magnetic field, the *uuud* ordered state becomes stable, since it has the largest magnetization, and makes a plateau at $m/m_{\mathrm{sat}} = \frac{1}{2}$ in the magnetization curve. We discuss thermal effects in Sec. V. Under the magnetic field, which realizes the *uuud* ground state, the system shows a finite-temperature phase transition due to breakdown of translational symmetry. Section VI contains a summary and discussions.

II. CLASSICAL LIMIT

We studied the ground state of the model (1) in the classical limit,[19] where the Pauli matrices are replaced to unit vectors (u_i^x, u_i^y, u_i^z) with $|u_i^x|^2 + |u_i^y|^2 + |u_i^z|^2 = 1$. We searched the ground state using the mean-field theory and studied finite-size systems with the Monte Carlo method. We searched the ground state restricting ourselves to spin configurations with up to four-sublattice structures within mean-field theory. To take into account larger sublattice structures, we studied larger finite-size systems and searched the minimum energy state with the Monte Carlo method, gradually decreasing temperatures. Here we only discuss the phase (b), where the parameters are in $-8K < J < -8K/3$ and competitions of the two- and four-spin exchanges are strong.

If there is no magnetic field, ground states are highly degenerate for $-8K < J < -8K/3$. We list some ground states which we found.

(1) A collinear state with a four-sublattice structure. Up spins are on three sublattices and down spin on the other [see Fig. 1(a)]. We call this state *uuud* state.

(2) A coplanar state with nine sublattices, whose spin configuration is constructed from three kinds of spin vectors [see Fig. 1(b)].

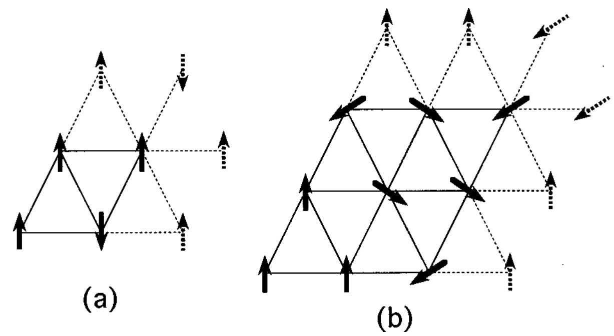

FIG. 1. Spin configuration of (a) *uuud* state with four sublattices and (b) a coplanar state with nine sublattices.

(3) Many other ground states can be made out of the *uuud* state by reversing all up spins to down on some parallel straight lines that consist of only up spins (see Fig. 2).

All these states have the minimum energy $E/N = -3K$. The *uuud* state has the largest magnetization $m/m_{\mathrm{sat}} = \frac{1}{2}$ among these states. The coplanar state shows $S^z = 0$ and the third series of ground states have a variety of magnetization between $-\frac{1}{2} < m/m_{\mathrm{sat}} < \frac{1}{2}$. The number of degeneracy except for the SO(3) symmetry is at least of order $2^{L/2}$ on the $L \times L$ triangular lattice, which comes from line degrees of freedom for spin flips. The number of the states in the $S^z = ML$ sector, where M is an integer in $-L/2 \leq M \leq L/2$, is at least ${}_{L/2}C_{(L-2M)/4}$ and thus the degeneracy is largest in the $S^z = 0$ sector. This degeneracy does not originate from the symmetry of the Hamiltonian and instead comes from frustration effects. We suspect that strong frustration makes density of states large near the lowest energy and hence nontrivial degeneracy appears in the ground states.

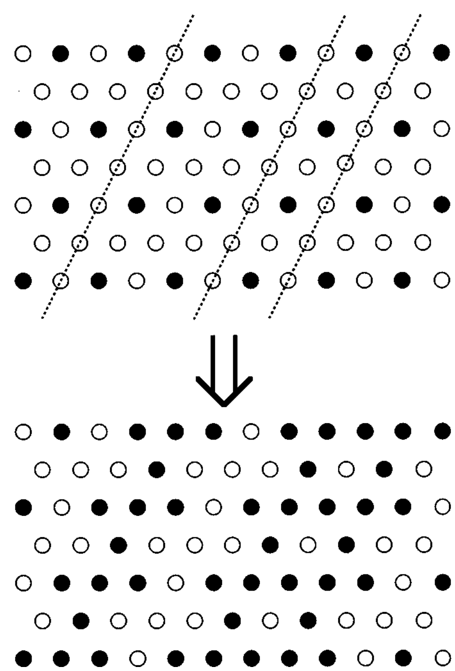

FIG. 2. Spin configuration of two kinds of ground states. Up (down) spins are denoted by white (black) circles. The upper configuration is that of the *uuud* state. The lower one is obtained by reversing the up spins to down on the dashed lines of the upper one.

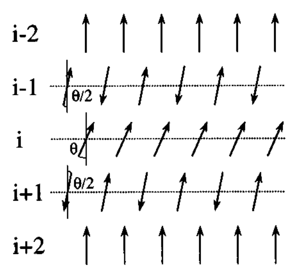

FIG. 3. A stripe excitation of the *uuud* state. Spins on the ith line are rotated with angle θ, and on the $(i-1)$th and $(i+1)$th lines with angle $\theta/2$.

Another characteristic property also appears in excitations. For the ground states in group (3), a huge number of excitations have extremely low energy, which is very close to the ground state energy. Consider a stripe excitation which is shown on Fig. 3. On the ith line, rotate spins with angle θ and, on the $(i-1)$th and $(i+1)$th lines, rotate spins with angle $\theta/2$, where ith line contains only up spins. For small θ, expanding the excitation energy with θ, one can find that $\mathcal{O}(\theta^2)$ terms of the excitation energy vanish and the leading term starts from the order $\mathcal{O}(\theta^4)$, if all spins on both $(i-2)$th and $(i+2)$th lines direct upward. We note that line excitation energy usually depends on the angle θ in the quadratic form. The condition for the low excitations of this kind to appear is that all spins on three lines in next neighbors, i.e., on $(i-2)$th, ith, and $(i+2)$th lines, are in the same direction. We hence find that these low-energy excitations exist for most of ground states in group (3) and there are a huge number of low-lying excitations of this kind.

By applying a weak magnetic field, this degeneracy quickly disappears. Since the *uuud* state has the largest magnetization among the degenerate states, the *uuud* state is stable under the magnetic field and it has the lowest energy. Figure 4 shows the parameter region where the *uuud* state becomes the ground state. The *uuud* state remains to be the ground state up to finite magnitude of the magnetic field,

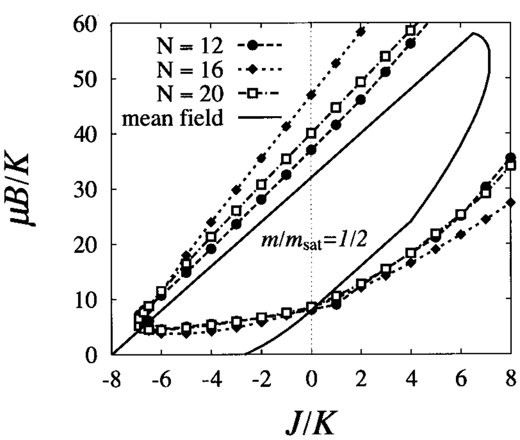

FIG. 4. Parameter dependence of phase boundaries of the $m/m_{\mathrm{sat}}=\frac{1}{2}$ state in the magnetization process at $T=0$, where the $\frac{1}{2}$ plateau appears in the region surrounded by data. The solid line denotes the result from the mean-field theory in the classical limit and other data with lines denote the results of the quantum model $(S=\frac{1}{2})$ on finite-size systems.

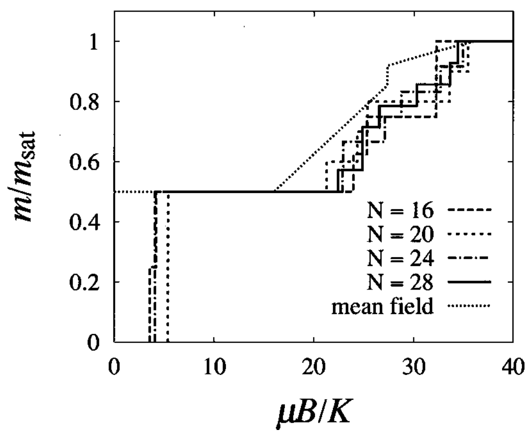

FIG. 5. Magnetization process at $T=0$ for $J=-4$ and $K=1$. The dotted line denotes the result from the mean-field theory in the classical limit (Ref. 19) and other lines denote the results of the quantum model $(S=\frac{1}{2})$ on finite-size systems.

which means the magnetic susceptibility to be vanishing in this phase. This behavior occurs due to singularity of collinear states. Since all spin vectors in the *uuud* state are parallel to the magnetic field, spins can be rigid against the field. The *uuud* state hence makes a plateau at the half magnetization $m/m_{\mathrm{sat}}=\frac{1}{2}$ in the magnetization process (see Fig. 5). These results are obtained with the mean-field theory and also confirmed with Monte Carlo method. The *uuud* state can stably exist even at low but finite temperatures and it also shows a finite-temperature phase transition due to the breakdown of the translational symmetry. We will further discuss finite-temperature properties in Sec. V.

III. QUANTUM $(S=\frac{1}{2})$ MODEL WITH $B=0$

When there are various degenerate ground states in the classical limit, quantum effects play an essential role in forming the ground state of the quantum model. One possibility is that a new quantum ground state appears due to tunneling between the classical states. This mixture of states can occur between the degenerate ground states in the same S^z sector. This possibility was discussed as an origin of the disordered state which is observed in low-density solid ^{3}He films.[19,5] Another possibility is that one of the degenerate ground states may be selected due to quantum effects.

To test these possibilities, we first examine the *uuud* state, which is one of the classical ground states, with the spin-wave approximation. Using the Holstein-Primakoff transformation, we expand the Hamiltonian up to the quadratic form of bosons

$$\mathcal{H}=-3NK+\sum_{k}\mathbf{A}_k^{\dagger}\mathcal{D}_k\mathbf{A}_k \tag{3}$$

with $A_k^{\dagger}=(a_k^{\dagger},b_k^{\dagger},c_k^{\dagger},d_{-k})$ and

$$\mathcal{D}_k=\begin{pmatrix} -4J & B_{\mathbf{e}_2,2\mathbf{e}_1} & B_{\mathbf{e}_2-\mathbf{e}_1,2\mathbf{e}_1} & C_{\mathbf{e}_1} \\ B_{\mathbf{e}_2,2\mathbf{e}_1} & -4J & B_{\mathbf{e}_1,2\mathbf{e}_2} & C_{\mathbf{e}_1-\mathbf{e}_2} \\ B_{\mathbf{e}_2-\mathbf{e}_1,2\mathbf{e}_1} & B_{\mathbf{e}_1,2\mathbf{e}_2} & -4J & C_{\mathbf{e}_2} \\ C_{\mathbf{e}_1} & C_{\mathbf{e}_1-\mathbf{e}_2} & C_{\mathbf{e}_2} & 12(J+8K) \end{pmatrix}, \tag{4}$$

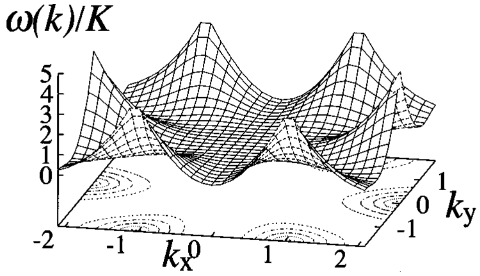

FIG. 6. The lowest mode of spin-wave spectrum on the *uuud* ordered state. The contours of constant energy are written on the bottom plane. Zero modes appear on the three lines, $k_x=0$ and $k_y=\pm\sqrt{3}k_x/2$.

where *a, b, c,* and *d* denote bosons for four sublattices and

$$B_{\mathbf{r},\mathbf{r}'}=4(J+2K)\cos\mathbf{k}\cdot\mathbf{r}+8K\cos\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}'),$$

$$C_{\mathbf{r}}=4(J+8K)\cos\mathbf{k}\cdot\mathbf{r}. \tag{5}$$

Numerically diagonalizing this Hamiltonian with Copla's method,[21] we evaluate the excitation spectrum of spin waves.[22] The spectrum has four branches of excitation modes in the first Brillouin zone of the four-sublattice structure. The lowest mode shows an ill behavior (see Fig. 6); it has flat modes along three lines in momentum space, $k_x=0$ and $k_y=\pm\sqrt{3}k_x/2$, and furthermore the dispersion curves except on the three lines behave as $|\mathbf{k}-\mathbf{k}_0|^6$. These modes correspond to the line excitations that we have shown for the classical model in Sec. II. This flat mode suggests that the *uuud* state does not have spin stiffness. Higher-order terms of spin-wave expansions may destroy the *uuud* state due to nonlinear effects.

We also studied the ground state of finite-size systems with the exact-diagonalization method. The systems with the size $N=12$, 16, 20, 24, and 28 are treated. (See Appendix A for shapes of the finite-size clusters.) The results reveal that the ground state belongs to the $S=0$ space and hence it is not the *uuud* state for $-7K\leqslant J$, which almost covers the phase (b). A recent numerical study by Misguich *et al.*[23] also suggests that the ground state in the same parameter region is spin liquid with $S=0$.

All the above results are consistent with each other and give a unique picture. The ground state belongs to the $S=0$ space and the *uuud* state has a little higher energy than the ground state due to quantum effects. The *uuud* state thus disappears from the ground state in the quantum model. But it again becomes stable under the magnetic field. We will discuss this point in the next section.

IV. QUANTUM ($S=\frac{1}{2}$) MODEL UNDER THE MAGNETIC FIELD ($B>0$)

To test the appearance of magnetization plateau at $m/m_{\rm sat}=\frac{1}{2}$ which we found in the classical limit, we investigate ground states of the quantum model (1) in finite-size systems under the magnetic field. We study finite-size ($N\leqslant 28$) systems with periodic-boundary conditions. Figure 5 shows the magnetization process of finite-size systems with $J=-4$ and $K=1$. Though the magnetization increases stepwisely due to finite-size effects, there clearly exists a broad plateau at $m/m_{\rm sat}=\frac{1}{2}$ in every-size data. Width between the lower and upper critical fields of the plateau does not vanish, as the system size increases, and remains significantly large (see Fig. 7). This result strongly suggests that this magnetization plateau survives in the thermodynamic limit. To examine whether this $m/m_{\rm sat}=\frac{1}{2}$ state has *uuud* long-range order, we consider the following *uuud* order parameter:

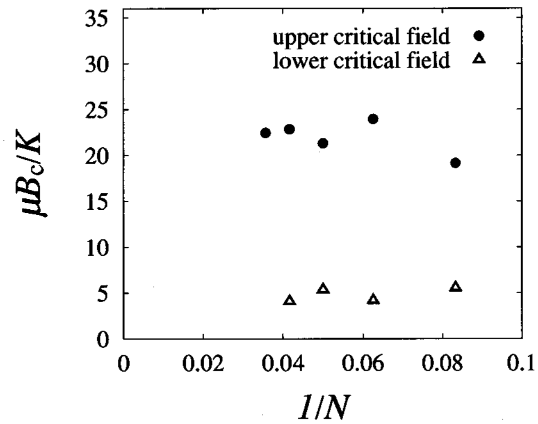

FIG. 7. Size dependence of the lower- and upper-critical fields of the magnetization plateau at $S^z_{\rm total}=N/4$ for the model with $J=-4$ and $K=1$.

$$\mathcal{O}=\frac{1}{2}\left(\sum_{i\in A}\sigma_i^z+\sum_{i\in B}\sigma_i^z+\sum_{i\in C}\sigma_i^z-\sum_{i\in D}\sigma_i^z\right) \tag{6}$$

and calculate long-range order

$$(\langle\mathcal{O}^2\rangle-\langle\mathcal{O}\rangle^2)/N^2 \tag{7}$$

in the ground state of the $S^z_{\rm total}=N/4$ space. Results are shown in Fig. 8, which clearly suggest that data are extrapolated to a finite value in the $N\to\infty$ limit for $J=-4K$. The extrapolated value is estimated as about 0.03 in the $N\to\infty$ limit. Long-range order of the *uuud* structure brings breakdown of translational symmetry in the thermodynamic limit.

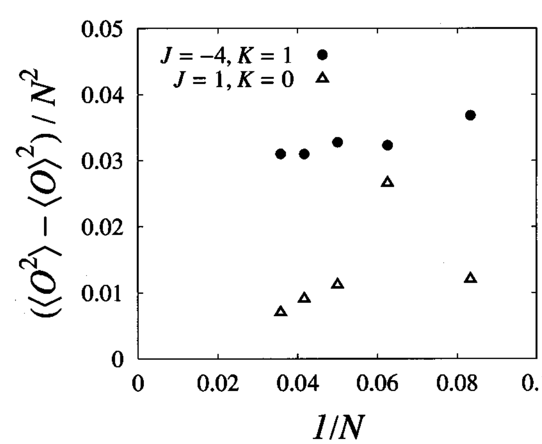

FIG. 8. Size dependence of long-range order of the *uuud* structure $(\langle\mathcal{O}^2\rangle-\langle\mathcal{O}\rangle^2)/N^2$ in the ground state of the $S^z_{\rm total}=N/4$ space. Black circles denote data for the model with $J=-4$ and $K=1$. Data for the Heisenberg antiferromagnet ($J=1$, $K=0$) are also shown with triangle symbols for a comparison.

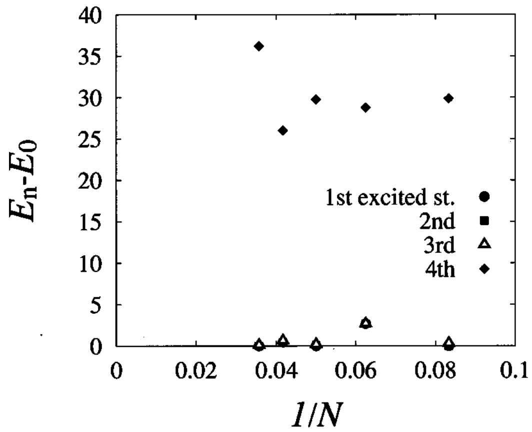

FIG. 9. Size dependence of excitation energy of four low-lying excited states in the $S^z_{\rm total}=N/4$ space for the model with $J=-4$ and $K=1$. The symbol for the third excited state almost overlaps those for the first and second states.

A careful estimation (see Appendix B) leads, when the translational symmetry is spontaneously broken, to the expectation value of spin on each sublattice $\langle \sigma^z_i/2\rangle = 0.45$ for $i \in A$, B, or C sublattice, and $\langle \sigma^z_i/2\rangle = -0.35$ for $i \in D$ sublattice, respectively. The ground state in the $S^z_{\rm total}=N/4$ space thus has a rigid *uuud* long-range order and deviation of the sublattice magnetization from the classical value is small. By the way, the total magnetization in the *uuud* ordered state does not change from the classical value $m/m_{\rm sat}=\frac{1}{2}$ by quantum effects. Note that the expectation values of spin satisfy $\Sigma_i\langle\sigma^z_i/2\rangle/N=(3\times 0.45-0.35)/4=1/4$. The spin-wave analysis also does not give any quantum correction to the total magnetization in the *uuud* state.

Magnetization plateaus of two-dimensional systems have been observed both theoretically and experimentally. On the triangular lattice, a magnetization plateau was observed at $m/m_{\rm sat}=\frac{1}{3}$ in the measurements of C_6Eu (Ref. 24) and $CsCuCl_3$,[25] and in theoretical studies of antiferromagnets (with two-spin exchange).[26–28] The plateau comes from the three-sublattice *uud* ordered state. The quantum correction to the magnetization $m/m_{\rm sat}=\frac{1}{3}$ is also vanishing.[27,28] For MSE models, a similar magnetic plateau was also observed at $m/m_{\rm sat}=\frac{1}{2}$ by finite-size studies. Roger and Hetherington first discovered the magnetic plateau in a MSE model with four-spin exchange on the square lattice.[29,30] [In the mean-field approximation, the ground state of this plateau is also the *uuud* state (see Ref. 30).] Recently Misguich *et al.* also observed a plateau in a model with two-, four-, five-, and six-spin exchanges on the triangular lattice. We believe that these plateaus in MSE models come from the *uuud* ordered state and that the four-spin exchange is the main cause for the plateaus as well.

Next, we study the excitations in the $S^z_{\rm total}=N/4$ space. Figure 9 shows excitation energy of up to fourth excited state. Three low-lying excited states have energy very close to the ground-state one and they converge to the ground state as the system size is enlarged. Above them there is a large gap, which seems not to vanish in the $N\to\infty$ limit. Thus four states exist around the lowest level and are clearly separated from other excited states. The number of low-lying levels, four, is equal to the degeneracy of *uuud* states, which comes from the spatial translation of sublattices. Furthermore, the four low-lying states have the same translation group as the following schematic states, respectively,

$$
\begin{aligned}
|1\rangle &= (1+\mathcal{R}_1+\mathcal{R}_2+\mathcal{R}_1\mathcal{R}_2)|\uparrow\uparrow\uparrow\downarrow\rangle,\\
|2\rangle &= (1+\mathcal{R}_1-\mathcal{R}_2-\mathcal{R}_1\mathcal{R}_2)|\uparrow\uparrow\uparrow\downarrow\rangle,\\
|3\rangle &= (1-\mathcal{R}_1+\mathcal{R}_2-\mathcal{R}_1\mathcal{R}_2)|\uparrow\uparrow\uparrow\downarrow\rangle,\\
|4\rangle &= (1-\mathcal{R}_1-\mathcal{R}_2+\mathcal{R}_1\mathcal{R}_2)|\uparrow\uparrow\uparrow\downarrow\rangle,
\end{aligned}
\tag{8}
$$

where $|\uparrow\uparrow\uparrow\downarrow\rangle$ denotes one of the *uuud* states, $\otimes_{i\in A,B,C}|\uparrow\rangle_i \otimes_{j\in D}|\downarrow\rangle_j$, and $\mathcal{R}_i$ $(i=1,2)$ mean the translation of sites by unit vectors $\mathbf{e}_i$. We hence believe that these four levels form four ground states which have *uuud* order in the thermodynamic limit and translational invariance is spontaneously broken in the ground states. This argument leads to a conclusion that the fifth low-lying state corresponds to the lowest excitation in the thermodynamic limit and hence the spin excitation spectrum has a finite gap in the same $S^z_{\rm total}$ sector.

The appearance of the magnetization plateau and the excitation gap can be understood by introducing a particle picture into the spin model.[31] Let us regard the down spins as particles moving in background of up spins and the magnetic field μB as minus of the chemical potential of the particle. Then the present spin system is mapped to a hard-core boson system and we can recognize the *uuud* ordered state as a Mott insulating state with charge-density wave (CDW). Note that this density wave can be ordered by repulsion between particles on nearest- or next-nearest-neighbor pairs of sites, which originates from the four-spin exchange interaction. In incompressible CDW, particles are insulating and charge-density excitations have a finite gap. And the compressibility is vanishing. Through the mapping, these features correspond to the finite excitation gap and the magnetization plateau of the original spin model. Thus the magnetization plateau can be understood as an insulating-conducting transition in the particle picture. We hence have an explanation for the plateau from the particle limit $(S=\frac{1}{2})$.[32] As we mentioned in Sec. II, the appearance of the plateau in the $S\to\infty$ limit originates from the rigidity of the collinear state. If the scenario in the particle picture is valid, the mechanism for the appearance of plateau is understood both from the classical limit $(S\to\infty)$ and the particle limit $(S=\frac{1}{2})$. Note that, in both cases, the *uuud* order is the key property for the appearance of the magnetization plateau.

The $\frac{1}{2}$ plateau appears for a wide parameter region. In Fig. 4, we show the phase boundary of the parameter region of the plateau. Except for weak magnetic field cases, the region of the $m/m_{\rm sat}=\frac{1}{2}$ phase becomes wider in comparison with the classical model. Quantum effects thus stabilize the plateau and enhance its appearance.

V. THERMAL EFFECTS

It is also important to discuss thermal effects to the magnetization plateau for a comparison with experiments at finite temperatures. A favorable property of the *uuud* order is that it accompanies a phase transition at a finite temperature. Since the *uuud* order is a discrete symmetry breaking, i.e.,

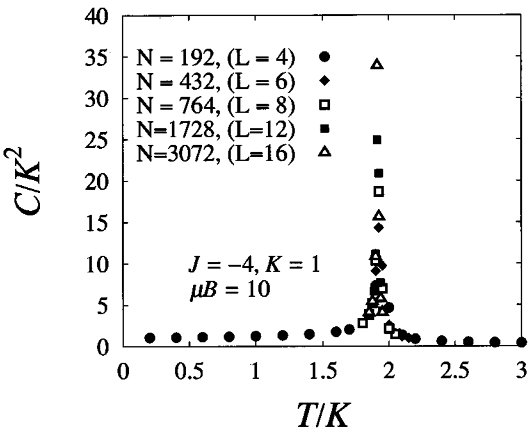

FIG. 10. Temperature dependence of the specific heat of the classical model with $J=-4$, $K=1$, and $\mu B=10$. Monte Carlo simulations were done on triangular lattices with finite size $N=12L^2$.

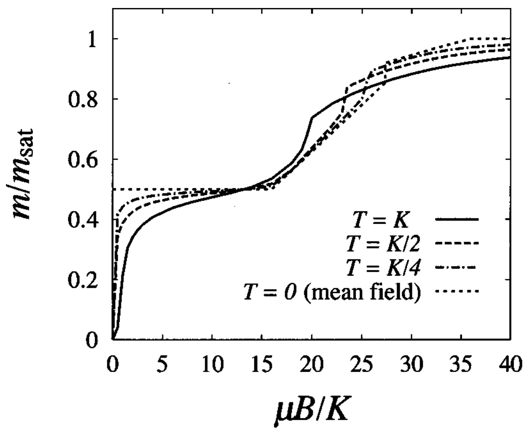

FIG. 11. Magnetization process of the classical model with $J=-4$ and $K=1$ at finite temperatures $T/K=0.25$, 0.5, and 1.0. The magnetization process at zero temperature, which we obtained with the mean-field theory (Ref. 19) is also shown for a comparison.

the number of ground states is four under the magnetic field, the symmetry breakdown occurs even at low but finite temperatures in two dimensions. The *uuud* order hence survives against thermal fluctuations and a phase transition occurs at a finite temperature. (Note that this argument does not depend on whether the system is quantum or classical.) Fortunately, the magnetization plateau survives even at low temperatures due to ordering of the *uuud* structure. Magnetization in the *uuud* ordered state is close to $m/m_{\mathrm{sat}}=\frac{1}{2}$ at low temperatures and it makes a plateau (or a shoulder) near the half of the saturated magnetization. This argument comes from only the degeneracy of the ground states, using the aspect of universality in phase transition.

To confirm the above argument, we study the finite-temperature properties in the classical limit using Monte Carlo simulations. We believe that critical properties of the phase transition at finite temperatures are governed by thermal fluctuations and quantum effects do not change its universality, though the value of the critical temperature or order parameter will deviate from the quantum one. Monte Carlo simulations were performed with the METROPOLIS algorithm. If a spin flip is rejected, we randomly rotate the spin about the local molecular field. We construct finite-size systems with a unit cluster which has 12 sites. The system sizes are $N=12L^2$ with $L=4$, 6, 8, 12, and 16 with periodic-boundary conditions. After discarding the initial 30 000–50 000 Monte Carlo steps per spin (MCS) for equilibration, subsequent $3\times10^5-5\times10^5$ MCS are used to calculate the average.

Monte Carlo simulations were done for the model under a magnetic field which realizes the *uuud* ground state. Figure 10 shows specific-heat data for the case $J=-4$, $K=1$, and $\mu B=10$. The data show a sharp divergence around $T=1.9$. We also found that the *uuud* long-range order exists below the critical temperature. Near the critical temperature, the magnetization curve is still rounded and smooth. As lowering the temperature, the slope of the curve decreases and becomes flat around $m/m_{\mathrm{sat}}=\frac{1}{2}$, as shown in Fig. 11. Misguich *et al.*[23] also demonstrated stability of the plateau at finite low temperatures in the quantum MSE model on a finite-size ($N=16$) lattice. Similarly, in the Heisenberg antiferromagnet on the triangular lattice, the magnetization plateau at $m/m_{\mathrm{sat}}=\frac{1}{3}$ was successfully observed at finite temperatures in the measurements of C_6Eu (Ref. 24) and $CsCuCl_3$,[25] and in Monte Carlo simulations.[26]

The critical temperature is estimated as $T_c=1.9K$ for the model with $J=-4K(<0)$ and $\mu B=10K$. For a weak magnetic field $\mu B=5K$, the estimate of T_c is about $1.7K$. These values can change due to quantum effects in the $S=\frac{1}{2}$ model. From the degeneracy of the ground states, the phase transition is expected to be of second order and to belong to the four-state Potts universality class. The finite-size scaling analysis, however, shows that critical exponent α is much larger than the expected value $\alpha=\frac{2}{3}$ for the four-state Potts model.[33] Moreover the distribution of energy histogram in Monte Carlo simulations has two peaks at the critical temperature, which suggests that the phase transition is of first order. A similar deviation of the universality was also found in the phase transition of chiral symmetry breaking in the model (1) for strong K.[20,34] These deviations may come from frustration effects or the singularity of four-body interactions. Critical properties of these phase transitions will be discussed further in a forthcoming paper.

The discrete symmetry of the *uuud* order in a magnetic field accompanies two favorable properties that a sharp phase transition occurs at a finite temperature and the magnetization plateau stably exists at low but finite temperatures. We expect that these properties make the experimental observation of the magnetization plateau possible.

VI. SUMMARY AND DISCUSSION

In this paper, we examined the appearance of the magnetization plateau at $m/m_{\mathrm{sat}}=\frac{1}{2}$ in a 2D MSE model on the triangular lattice, which we predicted in our previous work. This plateau appears when the two- and four-spin exchange interactions compete strongly, which may be realized in 2D low-density solid ^{3}He, and if the five and six exchanges are not too strong. The four-spin exchange is important and relevant to make the plateau at $m/m_{\mathrm{sat}}=\frac{1}{2}$. Since ordering of *uuud* structure is breakdown of discrete symmetry, it accompanies a phase transition at a finite temperature. Then the plateau would appear below the critical temperature. Experimental observation of this plateau in the magnetization process of the solid ^{3}He films will confirm that the four-spin exchange is strong and important.

Finally we discuss possibility of observing plateau in experiments. Comparing our results with measurements of susceptibility and specific heat, we estimate the effective exchanges J and K for low-density films. Susceptibility measurements at low densities show that the effective coupling J_χ is antiferromagnetic, $J_\chi > 0$, though it is close to vanishing.[4] In our model (1), the effective coupling behaves as $J_\chi = 2(J+6K)$ and hence we can estimate as $J \gtrsim -6K$. Specific heat measurements indicate that energy (spin) gap is small or vanishing,[5] where the system might be close to the critical point between the ferromagnetic phase and the liquid phase.[23] Since the critical point is around $J/K \simeq -8$ in the model (1), the exchange parameters can be estimated as $J \gtrsim -8K$ from the specific-heat data. The value J/K seems to be close to the boundary of the region where $\frac{1}{2}$ plateau appears (see Fig. 4), though two estimates are not consistent with each other. In real systems, more than four spin exchanges may move the boundary to right or left (in Fig. 4). For quantitative arguments, further studies will be needed on the effect of five- or six-spin exchanges. To make the plateau stable and wider, we would need lower-density solid films and enlarge the four-spin exchange effect. It was reported that, by preplating HD on grafoil, ^{3}He atoms solidify at lower density than double layer ^{3}He films.[13,15] Thus solid ^{3}He preplated HD may be a plausible candidate for observing the magnetization plateau. In our model with $J = -4K(<0)$, the lower critical field of the plateau is estimated about $4K/\mu$. Setting the parameter as $K = 1.0$(mK) and $\mu = 2.13\mu_N$, where $\mu_N = 0.366$(mK/T), we estimate the lower critical field as $B_c \simeq 5$[T]. (The value may be changed by other multiple-spin exchanges, i.e., six-spin exchange, in real systems.) Since this magnitude of the field is accessible with the present experimental equipments, we expect experimental verification of the magnetization process to be possible in 2D low-density solid ^{3}He.

ACKNOWLEDGMENTS

The authors would like to thank Hiroshi Fukuyama and Hikaru Kawamura for stimulating discussions and comments. One of the authors (T.M.) acknowledges Keisuke Totsuka for useful discussions and K.K. acknowledges J. Saunders, C. Lhuillier, and G. Misguich for useful discussions. This work was supported by Grant-in-Aid Nos. 09440130 and 10203202 from the Ministry of Education, Science and Culture of Japan. The numerical calculations were done on Facom VPP500 at the ISSP of the University of Tokyo and DEC Alpha 500 personal workstation at the Institute of Physics of the University of Tsukuba. K.K. also acknowledges the EPSRC Grant No. GR/L 90804 and the kind hospitality at the Department of Mathematics, Imperial College, London, where a part of this work was accomplished.

APPENDIX A: CLUSTERS USED IN FINITE-SIZE STUDIES

In the numerical study of the $S = \frac{1}{2}$ model on finite-size systems, we used finite-size clusters with the periodic boundary condition which matches the four-sublattice structure. Clusters are set on the triangular lattice as shown in Fig. 12.

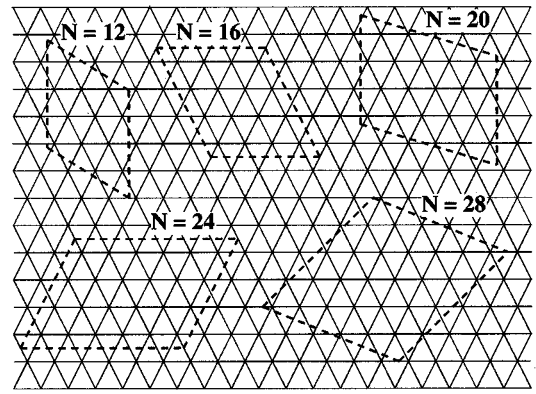

FIG. 12. Finite-size clusters which we used in the exact diagonalization study.

APPENDIX B: EVALUATION OF EXPECTATION VALUES OF THE SUBLATTICE MAGNETIZATION

The estimation of the sublattice magnetization from long-range order (7) requires that we carefully consider symmetry breaking. In the thermodynamic limit, translational symmetry is spontaneously broken in a pure natural ground state with *uuud* order. But the ground states of finite-size systems become mixed symmetric ones because of finite-size effects.

Here we write the mixed symmetric ground state in the $S^z_{\rm total} = N/4$ space as $|\phi\rangle$. We also consider a state $|\psi_1\rangle$ in which translational symmetry is broken and whose thermodynamic limit is the natural pure ground state. By the space translation of the state, $|\psi_1\rangle$ relates to other three *uuud* ordered states in the form

$$|\psi_2\rangle = \mathcal{R}_1|\psi_1\rangle, \quad |\psi_3\rangle = \mathcal{R}_2|\psi_1\rangle, \quad |\psi_4\rangle = \mathcal{R}_1\mathcal{R}_2|\psi_1\rangle, \tag{B1}$$

where $\mathcal{R}_1(\mathcal{R}_2)$ denotes spatial translation by $\mathbf{e}_1(\mathbf{e}_2)$. In the mixed state, there is translational invariance,

$$\langle\phi|S^z_i|\phi\rangle = \frac{1}{4} \quad \text{for any site } i. \tag{B2}$$

On the other hand, in the *uuud* ordered state $|\psi_1\rangle$, the translational symmetry is broken,

$$\langle\psi_1|S^z_i|\psi_1\rangle = m_1 \quad \text{for } i \in A,B, \text{ or } C, \tag{B3}$$

$$\langle\psi_1|S^z_i|\psi_1\rangle = m_2 \quad \text{for } i \in D, \tag{B4}$$

where $m_1 \neq m_2$. Since the total magnetization is $N/4$, the sublattice magnetization m_1 and m_2 satisfy

$$\frac{3}{4}m_1 + \frac{1}{4}m_2 = \frac{1}{4}. \tag{B5}$$

It is expected that the symmetric mixed state can be decomposed into the pure states in the thermodynamic limit in the form $|\phi\rangle=(|\psi_1\rangle+e^{i\theta}|\psi_2\rangle+e^{i\psi}|\psi_3\rangle+e^{i\phi}|\psi_4\rangle)/2$, where θ, ψ, and ϕ denote arbitrary real numbers. The following relation hence holds in the large N limit:

$$\frac{1}{N^2}\langle\Phi|\mathcal{O}^2|\phi\rangle=\frac{1}{4N^2}(\langle\psi_1|\mathcal{O}^2|\psi_1\rangle+\langle\psi_2|\mathcal{O}^2|\psi_2\rangle+\langle\psi_3|\mathcal{O}^2|\psi_3\rangle+\langle\psi_4|\mathcal{O}^2|\psi_4\rangle). \quad \text{(B6)}$$

From the numerical calculations in Sec. IV, we have

$$\lim_{N\to\infty}\frac{1}{N^2}\langle\phi|\mathcal{O}^2|\phi\rangle=0.03+\left(\frac{1}{8}\right)^2. \quad \text{(B7)}$$

For the pure state, the clustering property of the state leads to

$$\lim_{N\to\infty}\frac{1}{N^2}\langle\psi_1|\mathcal{O}^2|\psi_1\rangle=\left(\lim_{N\to\infty}\frac{1}{N}\langle\psi_1|\mathcal{O}|\psi_1\rangle\right)^2=\left\{\frac{1}{4}(3m_1-m_2)\right\}^2. \quad \text{(B8)}$$

and

$$\lim_{N\to\infty}\frac{1}{N^2}\langle\psi_2|\mathcal{O}^2|\psi_2\rangle=\left(\lim_{N\to\infty}\frac{1}{N}\langle\psi_2|\mathcal{O}|\psi_2\rangle\right)^2=\left\{\frac{1}{4}(m_1+m_2)\right\}^2. \quad \text{(B9)}$$

In the same way,

$$\lim_{N\to\infty}\frac{1}{N^2}\langle\psi_3|\mathcal{O}^2|\psi_3\rangle=\lim_{N\to\infty}\frac{1}{N^2}\langle\psi_4|\mathcal{O}^2|\psi_4\rangle=\left\{\frac{1}{4}(m_1+m_2)\right\}^2. \quad \text{(B10)}$$

Inserting these relations into Eq. (B6) and using Eq. (B5), we have

$$48m_1^2-24m_1+1.08=0 \quad \text{(B11)}$$

and then we obtain two solutions $(m_1,m_2)=(0.45,-0.35)$ and (0.005,0.985). From the constraints $|m_1|\leqslant 0.5$ and $|m_2|\leqslant 0.5$, we conclude that the former one is the physical solution.

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