Possible chiral phase transition in two-dimensional solid $^{3}$He

Tsutomu Momoi, Kenn Kubo and Koji Niki

Institute of Physics, University of Tsukuba, Tsukuba, Ibaraki 305, Japan

(Received 19 May 1997)

We study a spin system with two- and four-spin exchange interactions on the triangular lattice, which is a possible model for the nuclear magnetism of solid $^{3}$He layers. It is found that a novel spin structure with scalar chiral order appears if the four-spin interaction is dominant. Ground-state properties are studied using the spin-wave approximation. A phase transition concerning the scalar chirality occurs at a finite temperature, even though the dimensionality of the system is two and the interaction has isotropic spin symmetry. Critical properties of this transition are studied with Monte Carlo simulations in the classical limit.

PACS numbers: 67.80.Jd, 64.70.Kb, 67.70.+n, 75.40.Cx

Solid $^{3}$He films adsorbed on the surface of graphite are purely two-dimensional (2D) magnetic systems with isotropic interactions [1]. Recently the solid phase of the adsorbed $^{3}$He layer at a low density has been attracting extensive interest, since it is a typical example of frustrated quantum spin systems; nuclei of $^{3}$He form a spin-1/2 quantum antiferromagnet on the triangular lattice.

In a double-layer solid $^{3}$He film, only the second layer is responsible for its magnetism at mK region. Specific-heat measurement of the second layer showed a peculiar behavior [2] and the effective exchange coupling obtained from the susceptibility was revealed to be antiferromagnetic at a coverage where the layer just solidifies. It is believed that the second layer forms a triangular lattice. Elser proposed that the second layer may decompose into two sublattices, and atoms on only one sublattice which form a kagomé net are responsible for the magnetism [3]. Recent specific-heat data, however, showed that all spins on the triangular lattice contribute to the magnetism and they cannot be fully separated to the kagomé lattice and the other [4]. It is not yet clear whether the inequivalence of the two sublattices gives relevant effects on the translational symmetry of the magnetic interactions. Another example of 2D solid $^{3}$He is realized in a monolayer of solid $^{3}$He adsorbed on graphite preplated with $^{4}$He [5] or HD [6].

It is well established that the multi-spin exchange interactions are important in the nuclear magnetism of solid $^{3}$He [7]. These multi-spin interactions originate from particle-ring-exchange of $^{3}$He atoms. It was shown by Thouless that the whole Hamiltonian has the form $H = \sum_n (-1)^n \sum J_n P_n$, where $P_n$ denotes cyclic permutation of $n$ spins and the exchange constant $J_n$ is always negative [1]. According to numerical calculations for the triangular lattice [3,4], two- and three-spin exchanges are large and, furthermore, four- and six-spin exchanges are not small. As pointed out in Ref. [5], the multi-spin interactions can create frustration, and hence they should be properly considered in a theoretical treatment. In our previous study [6], we found that the four-spin interaction can produce novel phases with four-sublattice structures.

In this Letter, we reveal effects of the four-spin interaction and predict a chiral phase transition in a certain parameter region where four-spin interactions are dominant. It is found that the ground state of a spin model for the solid $^{3}$He layer has a scalar chiral order. The spin-wave approximation shows that quantum fluctuations are not strong enough to destroy the chiral order at $T = 0$. We hence expect that the real $^{3}$He layer shows a chiral phase transition at a finite temperature. Critical properties of this transition are also studied in the classical limit. The critical exponent $\alpha$ of the specific heat clearly deviates from that of the 2D Ising model. To our knowledge, the present model gives the first example of a chiral phase transition in a 2D realistic spin system with $SO(3)$ symmetry.

We consider a spin model with two-, three- and four-spin exchange interactions on the triangular lattice, and assume that the interactions have the same translational symmetry as that of the triangular lattice. Since the three-spin permutations can be transformed into two-spin exchanges, the Hamiltonian can be written as

$$H = J \sum_{\langle i,j \rangle} \sigma_i \cdot \sigma_j + K \sum_p h_p, \quad (1)$$

where $J = J_3 - J_2/2$, $K = -J_4/4 (\geq 0)$, and $\sigma_i$ denote Pauli matrices. 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

$$h_p = \sum_{1 \leq i < j \leq 4} \sigma_i \cdot \sigma_j + (\sigma_1 \cdot \sigma_2)(\sigma_3 \cdot \sigma_4)$$
$$+ (\sigma_1 \cdot \sigma_4)(\sigma_2 \cdot \sigma_3) - (\sigma_1 \cdot \sigma_3)(\sigma_2 \cdot \sigma_4), \quad (2)$$

where $(1, 3)$ and $(2, 4)$ are diagonal bonds of the diamond. Bernu et al. first studied this model discussing the temperature dependence of the specific heat and the...
magnetic susceptibility [11]. Roger estimated density dependence of interactions using the WKB approximation [10]. It shows that \( J_3 \) is dominant in the high-density region and \( |J_2| \) increases faster than \( |J_3| \) as the density is lowered. Hence the value of \( J \) changes rapidly depending on the density and \( J \) may vanish at a low-density region.

In the previous paper [13], we studied the ground state manifold has two-fold degeneracy. In this Letter, we employ the following order-parameter operator of the chirality [14] which indicates that quantum fluctuations are not very strong and the ground state has the sublattice magnetic order. The value of the chirality per unit triangle is \( \kappa/2N = 0.2858 \), whereas \( \kappa/2N = 4/3\sqrt{3} \) in the classical limit. The chirality has 37% of its classical value and hence quantum effects should be properly treated. We employ the spin-wave approximation to investigate the strength of quantum fluctuations. Using the Holstein-Primakoff transformation, we expand the Hamiltonian in terms of Bose operators up to bilinear terms from the tetrahedral structure. After the Bogoliubov transformation of four kinds of bosons, we obtain

\[
H = -\frac{49}{3}KN + \sum_\kappa \sum_{i=1}^4 \omega_\kappa(k)(a_\kappa^\dagger a_\kappa + \frac{1}{2}),
\]

where the first summation runs over the reduced Brillouin zone of the four-sublattice structure. The frequency \( \omega_\kappa(k) \) is given by

\[
\omega_\kappa(k) = \frac{8K}{9}(4A_k^2 - B_k^2 - 3C_k^2)^{1/2},
\]

where

\[
A_k = 12 - \cos\frac{k_1}{2} - \cos\left(\frac{k_1}{4} + \frac{\sqrt{3}k_2}{4}\right) - \cos\left(\frac{3k_1}{4} + \frac{\sqrt{3}k_2}{4}\right) - \cos\left(\frac{3k_1}{4} - \frac{\sqrt{3}k_2}{4}\right),
\]

\[
B_k = 5\left\{- \cos\frac{k_1}{2} + 2\cos\left(\frac{k_1}{4} - \frac{\sqrt{3}k_2}{4}\right) - \cos\frac{3k_1}{4} + \frac{\sqrt{3}k_2}{4}\right\} - \cos\frac{3k_1}{4} - \frac{\sqrt{3}k_2}{4},
\]

\[
C_k = 5\left\{ \cos\frac{k_1}{2} - \cos\left(\frac{k_1}{4} - \frac{\sqrt{3}k_2}{4}\right) \right\} - \cos\frac{3k_1}{4} + \frac{\sqrt{3}k_2}{4}.
\]

The others are \( \omega_2(k) = \omega_1(k + q_1) \), \( \omega_3(k) = \omega_1(k + q_2) \) and \( \omega_4(k) = \omega_1(k + q_1 + q_2) \), where \( q_1 \) and \( q_2 \) are reciprocal lattice vectors of the four-sublattice structure. Three frequencies \( \omega_i(k) \) with \( i = 2, 3, 4 \) are gapless at \( k = 0 \) and the other one, \( \omega_1(k) \), is massive. The ground-state energy per site is estimated as \( \varepsilon_g = -7.4615K \), whereas \( \varepsilon_g = -17K/3 \) in the classical limit. The sublattice magnetization and the chirality are evaluated in the same way. The estimate of the sublattice magnetization per spin is \( m_s/S = 0.5937 \), where \( S = 1/2 \). The deviation from the classical value, \( \Delta m_s/S = 0.4063 \), is small compared with the value \( \Delta m_s/S = 0.522 \) of the pure Heisenberg antiferromagnet on the triangular lattice [13], which indicates that quantum fluctuations are not very strong and the ground state has the sublattice magnetic order. The value of the chirality per unit triangle is \( \kappa/2N = 0.2858 \), whereas \( \kappa/2N = 4/3\sqrt{3} \) in the classical limit. The chirality has 37% of its classical value and this ratio is larger than the third power of the sublattice magnetization. This shows that the chiral order is more stable against quantum fluctuations than the sublattice magnetization. A similar tendency was observed in the vector chiral order of the Heisenberg and \( XY \) antiferromagnets on the triangular lattice [13]. The spin-wave
The analysis of the $uuud$ and $120^\circ$ structure ground states indicates that the stable region for the tetrahedral ground state will become narrower than the mean field result [17].

The greatest significance of the chiral order is that it can exist even at a finite temperature since it is stable against the spin-wave fluctuations with long wavelength. The spin-wave approximation indeed shows that the estimate of the chiral order is nonvanishing at sufficiently low temperatures. We thus expect a phase transition to occur at a finite temperature, which is accompanied by ordering of the scalar chirality. 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 hence the quantum effects do not change its universality class, though they can change values of the exponents. Our estimates of the exponents $(2-\alpha)/\nu$, $\beta/\nu$, and $\gamma/\nu$ appear to equal to 2, 1/8, and 7/4, respectively, and hence the present transition still belongs to the Ising universality-class in the sense of the weak universality [20].

Of course, there is no sublattice magnetic order at finite temperatures even in the chiral ordered phase. Thus the divergence of the specific heat corresponds to the second-order phase transition from the disordered phase to the chiral ordered one. This is the first example of the scalar-chiral phase transition in the 2D realistic spin model with SO(3) symmetry. We estimate the critical exponent $\alpha$ of the specific heat, $C(T) \sim |T - T_c|^{-\alpha}$. The maximum values of the specific heat are plotted in Fig. 4 for various sizes. The finite-size scaling reveals that the peak value behaves as $C(T_p(L)) \sim L^{\alpha/\nu}$. The plot fits well by setting $\alpha/\nu = 0.9(1)$. We hence obtain $\alpha = 0.62(5)$ and $\nu = 0.69(3)$ using the scaling relation $\gamma + 2\nu = d$. This is clearly different from the log-divergence, i.e. $\alpha = 0$, of the 2D Ising model. This result might be thought peculiar, since the chirality is an Ising-type variable and the chiral phase transition is expected to have the Ising universality. There is however a similar example; the four-spin interaction added to the Ising model changes the exponent $\alpha$ continuously from 0 [14]. The four-spin interaction may have a universal effect to change the critical exponent. Other values are estimated as $\beta/\nu = 0.13(2)$, $\gamma/\nu = 1.75(5)$ and $T_c = 0.9935(10)$ from the finite-size scaling of the order parameter and the response function [15]. The critical indices may depend on the parameter $J/K$ as they do in the 2D Ising model with four-spin interactions. Our estimates of the exponents $(2-\alpha)/\nu$, $\beta/\nu$, and $\gamma/\nu$ appear to equal to 2, 1/8, and 7/4, respectively, and hence the present transition still belongs to the Ising universality-class in the sense of the weak universality [20].

Even for the vector chirality in the fully frustrated 2D XY model, there remain controversies whether the chiral phase transition has the Ising universality or not. Since Miyashita and Shiba [21] estimated $\alpha = 0$ using Monte Carlo simulations, this problem has been studied repeat-
edly [24]. Though the discussions have not yet settled down, all estimates of $\alpha$ are less than 0.4. Our estimate is clearly different from these values.

We mention the possibility that the chiral phase transition may be observed experimentally in the solid $^3\text{He}$ films. The $^3\text{He}$ films behave as ferromagnets in a high-density region [5,24] and, by lowering the coverage, the exchange coupling $J_\chi$ derived from the susceptibility turns antiferromagnetic [25]. On the other hand, the high-temperature series expansion shows that $J_\chi = -2(J + 6K)$ [24]. Hence the negativity of $J_\chi$ indicates that $J > -6K$ and not necessary that $J$ is antiferromagnetic. For all densities, no magnetic phase transition has been observed at finite temperatures [24,25]. Comparing these results with the phase diagram of the present spin model [13], we find that the low-density $^3\text{He}$ layer seems to correspond to the intermediate phase ($-8K < J < -8K/3$) between the ferromagnetic phase and the tetrahedral-structure phase. According to the WKB approximation [10], the value of $|J| (= |J_2/2 - J_3|)$ decays fast whereas $K$ increases as the density is lowered. We hence expect that, in a certain lower-density (but well solidified) region, the four-spin exchange interaction is dominant and the chiral phase transition may occur. At this phase transition, one can observe sharp divergence of the specific heat, as we have shown in this Letter, and a cusp-like behavior in the magnetic susceptibility (see Ref. [18]). Recently, it was reported that, by preplating HD on graphite, the solid phase of a monolayer of $^3\text{He}$ can be stabilized and then the $^3\text{He}$ film can first solidify around $\rho = 0.0555 A^{-2}$ [24], which is the lowest density ever observed. (The second layer of double-layer $^3\text{He}$ solidifies around $\rho_2 = 0.064 A^{-2}$.) We expect that this material may be a plausible candidate for showing the chiral phase transition.

The authors wish to thank Hiroshi Fukuyama, Hikaru Kawamura and Harumi Sakamoto for stimulating discus-

![FIG. 4. Finite-size scaling plot of the peak values of the specific heat.](image)

[1] H. Franco, R. Rapp and H. Godfrin, Phys. Rev. Lett. 57, 1161 (1986); H. Godfrin, R. Ruel and D. Osheroff, Phys. Rev. Lett. 60, 305 (1988).
[2] D.S. Greywall and P.A. Busch, Phys. Rev. Lett. 62, 1868 (1989); D.S. Greywall, Phys. Rev. B 41, 1842 (1990).
[3] V. Elser, Phys. Rev. Lett. 62, 2405 (1989).
[4] M. Siqueira, J. Nyéki, B. Cowan and J. Saunders, Phys. Rev. Lett. 76, 1884 (1996).
[5] K. Ishida, M. Morishita, K. Yawata and H. Fukuyama, to be submitted to Phys. Rev. Lett..
[6] C.P. Lusher, J. Saunders and B.P. Cowan, Europhys. Lett. 14, 809 (1991).
[7] M. Siqueira, C.P. Lusher, B.P. Cowan and J. Saunders, Phys. Rev. Lett. 71, 1407 (1993).
[8] M. Roger, J.H. Hetherington and J.M. Delrieu, Rev. Mod. Phys. 55, 1 (1983).
[9] D.J. Thouless, Proc. Phys. Soc.(London) 86, 893 (1965).
[10] M. Roger, Phys. Rev. B 30, 6432 (1984).
[11] B. Bernu, D. Ceperley and C. Lhuillier, J. Low Temp. Phys. 89, 589 (1992).
[12] M. Roger, Phys. Rev. Lett. 64, 297 (1990).
[13] K. Kubo and T. Momoi, Z. Phys. B. 103, 485 (1997).
[14] X.G. Wen, F. Wilczek and A. Zee, Phys. Rev. B 39, 11413 (1989).
[15] Th. Jolicoeur and J. C. Le Guillou, Phys. Rev. B 40, 2727 (1989).
[16] T. Momoi and M. Suzuki, J. Phys. Soc. Jpn. 61, 3732 (1992).
[17] K. Kubo, K. Niki and T. Momoi, in preparation.
[18] T. Momoi and K. Kubo, in preparation.
[19] R.J. Baxter, “Exactly Solved Models in Statistical Mechanics” (Academic Press, London, 1982) p. 253.
[20] M. Suzuki, Prog. Theor. Phys. 51, 1992 (1974).
[21] S. Miyashita and H. Shiba, J. Phys. Soc. Jpn. 53, 1145 (1984).
[22] J. Lee, J.M. Kosterlitz and E. Granato, Phys. Rev. B 43, 11531 (1991); G. Ramirez-Santiago and J.V. José, Phys. Rev. B 49, 9567 (1994); P. Olsson, Phys. Rev. Lett. 75, 2758 (1995); and references therein.
[23] P. Schiffer, M.T. O’Keefe, D.D. Osheroff and H. Fukuyama, Phys. Rev. Lett. 71, 1403 (1993); J. Low Temp. Phys. 94, 489 (1994).
[24] M. Morishita, K. Ishida, K. Yawata and H. Fukuyama, Czech. J. Phys. Suppl. S1 46, 409 (1996).
[25] M. Siqueira, J. Nyéki, B. Cowan and J. Saunders, Czech. J. Phys. Suppl. S6 46, 3033 (1996).