

Ising Spin System with Biquadratic Exchange Interaction and Four-Site Four-Spin Interaction

by

Takashi IWASHITA*, Ai NAGAKI**, Kakuko URAGAMI*

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Abstract

The phase diagram and magnetic properties such as the magnetization $\langle S_z \rangle$, the four-spin thermal average $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$, the specific heat C_M , the Curie temperature T_c , and spin structures of spin-one ($S=1$) Ising spin system with the bilinear exchange interaction $J_1 S_{iz} S_{jz}$, the biquadratic exchange interaction $J_2 S_{iz}^2 S_{jz}^2$ and the four-site four-spin interaction $J_4 S_{iz} S_{jz} S_{kz} S_{lz}$ have been discussed by making use of the Monte Carlo simulation on two-dimensional square lattice. In this Ising spin system with interactions J_1 , J_2 and J_4 , we have found new magnetic phases and determined the conditions of phase transitions between lots of magnetic phases with different ground state (GS) spin structures. Furthermore, it is confirmed that these conditions of phase transition agree well with those obtained from comparison of energies per one spin for various spin structures with low energy. The characteristic temperature dependence of the magnetization $\langle S_z \rangle$, the four-spin thermal average $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ and the interesting changes of spin structures are investigated for various values of interaction parameters of J_2/J_1 and J_4/J_1 .

Key words: biquadratic interaction, four-spin interaction, Ising model, Monte Carlo simulation

1. Introduction

In Heisenberg and Ising spin systems, the existence and the importance of such higher-order exchange interactions as the biquadratic exchange interaction $(\mathbf{S}_i \cdot \mathbf{S}_j)^2$, the three-site four-spin interaction $(\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_j \cdot \mathbf{S}_k)$, the four-site four-spin interaction $(\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_k \cdot \mathbf{S}_l)$ have been discussed extensively by many investigators [1, 2].

Theoretical explanations of the origin of these interactions have been given in the theory of the superexchange interaction, the magnetoelastic effect, the permutation operator, the perturbation expansion, the higher harmonics of oscillatory exchange coupling and the spin-phonon coupling [3].

It was pointed out that the higher-order exchange interactions are smaller than the bilinear ones for the $3d$ group ions [3], and comparable with the bilinear ones in the rare-earth compounds [4, 5]. On the other hand, in solid helium and in such phenomena as the quadrupolar ordering of molecules in solid hydrogen, in liquid crystals, or the cooperative Jahn-Teller phase transitions,

* Center of Kumamoto General Education, Tokai University, Kumamoto 862-8652, Japan

** School of Industrial Engineering, Tokai University, Kumamoto 862-8652, Japan

the higher-order exchange interactions turned out to be the main ones [6]. Furthermore, the four-site four-spin interaction has been pointed out to be important to explain the magnetic properties of the solid helium [7, 8] and the magnetic materials such as NiS_2 and C_6Eu [9].

In the Ising ferromagnet with a spin of $S \geq 1$, the dependences of the magnetization and the Curie temperature on the biquadratic exchange interaction [10, 11] and the three-site four-spin interaction [12] were investigated and the ground state (GS) spin structures were determined by pair-spin and three-spin models approximation. Recently present authors have investigated the effects of the three-site and the four-site four-spin interactions and biquadratic interaction on magnetic properties and the GS spin structure of the Ising ferromagnet [13, 14, 15] with $S=1$ by making use of the Monte Carlo (MC) simulation.

In the present paper, we extend this MC calculation to spin-one Ising spin system on the two-dimensional square lattice with three interactions such as the bilinear exchange $J_1 S_{iz} S_{jz}$ and the biquadratic exchange $J_2 S_{iz}^2 S_{jz}^2$, and the four-site four-spin interactions $J_4 S_{iz} S_{jz} S_{kz} S_{lz}$. The model in which the four-site four-spin interactions $J_4 S_{iz} S_{jz} S_{kz} S_{lz}$ for this $S=1$ Ising system is replaced with single-ion anisotropy term D is quite famous as so-called Blume-Emery-Griffiths (BEG) model [16] and has been applied for many problems, e.g. super-liquid helium, magnetic material, semiconductor, alloy, lattice gas and so on. In the BEG model, there appear various characteristic spin orders depending on the combinations of parameters J_1 , J_2 , D and on the lattice dimensionality [17, 18, 19, 20, 21].

In the present study, we have investigated the phase diagram and ground state (GS) spin structures for each phase on two-dimensional square lattice with interaction parameters J_1 , J_2 and J_4 . Furthermore, the magnetization $\langle S_z \rangle$, the four-spin thermal average

$\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$, the specific heat C_M and the Curie temperature T_c in the spin-one Ising spin system have been calculated for each phase and their phase boundary.

In Section 2, the spin Hamiltonian are given for present Ising system, and the energies per one spin of the spin structures with lower energy are calculated from this spin Hamiltonian. Furthermore, the method of the MC simulation is explained briefly. In Section 3, phase diagram are obtained for exchange parameters J_2/J_1 and J_4/J_1 by the MC simulation. In the latter part of this section, this result of MC simulation are confirmed by the one obtained from the comparisons of the energies per one spin of the spin structures with lower energy. In the Section 4, the magnetic properties and spin structures are investigated for each magnetic phase. In the last Section 5, new interesting results obtained here are summarized.

2. Spin Hamiltonian and Methods of Simulation

Let us consider the spin-one Ising model described by the following Hamiltonian

$$H = -J_1 \sum_{\langle ij \rangle} S_{iz} S_{jz} - J_2 \sum_{\langle ij \rangle} S_{iz}^2 S_{jz}^2 - 2J_4 \sum_{\langle ijkl \rangle} S_{iz} S_{jz} S_{kz} S_{lz}, \quad (1)$$

Here, $\langle ijkl \rangle$ denotes the sum on the square spin sites of the two-dimensional square lattice. The coefficient 2 of the second term in this Hamiltonian is obtained by considering the sum of two terms $(S_{iz} \cdot S_{jz})(S_{kz} \cdot S_{lz})$, and $(S_{iz} \cdot S_{kz})(S_{jz} \cdot S_{lz})$. Furthermore, S_z represents $S_z = \pm 1, 0$. From a consideration of the Hamiltonian (1), magnetic properties and spin arrangements of Ising spin system with two-dimensional square lattice are calculated by the MC simulation. Furthermore, the energies per one spin

are obtained for various spin structures with low energy (see e.g. [11]).

For Ising spin system with three kinds of interactions such as positive bilinear interaction J_1 , and four-spin interactions J_2 and J_4 , the energies of various spin structures with low energy have been calculated for positive and negative J_2 and J_4 . It is evident that the ferromagnetic spin structure (a) (S(a)) shown in Fig.1 becomes the GS spin structure for all positive interactions J_1 , J_2 and J_4 . The energy $E(a)=E_d/J_1$ per one spin for this ferromagnetic spin structure (S(a)) with $S=1$ is given by $E(a)=-2-2x-2y$. Here, these parameters x and y are defined as J_2/J_1 and J_4/J_1 , respectively.

For the negative interaction J_4 , the spin structures S(b) and S(c) with $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle = -1$ shown by (b) and (c) in Fig.1 has turned out to have a low energy [13,14] and the energies $E(b)=E_b/J_1$ and $E(c)=E_c/J_1$ per one spin for these S(b) and S(c) with $S=1$ are take the same value and given by $E(b)=E(c)=-2x+2y$. On the other hand, the values of magnetization $\langle S_z \rangle$ for S(b) and S(c) are different and takes 0 and 0.5, respectively. The spin structure (S(d)) shown by (d) in Fig.1 in which all spins $S_z=1$ and $S_z=-1$ are surrounded by spins of $S_z=0$ on the nearest neighbor spin site has been pointed out to become the spin structure with low energy for the negative interaction J_2 [11]. The energy $E(d)=E_d/J_1$ per one spin for this S(d) with $S=1$ is given by $E(d)=0$. For both negative interactions J_2 and J_4 , the spin structure (S(e)) shown by (e) in Fig.1 with the 1/3- ratio of the number of a spin $S_z=0$ to a spin $S_z=1$ is expected to have a low energy, and the energy $E(e)=E_e/J_1$ per one spin for this S(e) with $S_z=1$ is given by $E(e)=-x-y$.

The MC simulations based on the Metropolis method are carried out assuming periodic boundary condition for two dimensional square lattice with linear lattice size up to $L=160$. For fixed values of various parameters J_1 , J_2 and J_4 , we start the simulation at high temperatures adopting a random, a ferromagnetic, and an

antiferromagnetic initial configurations, respectively, and gradually advance this simulation to lower temperature.

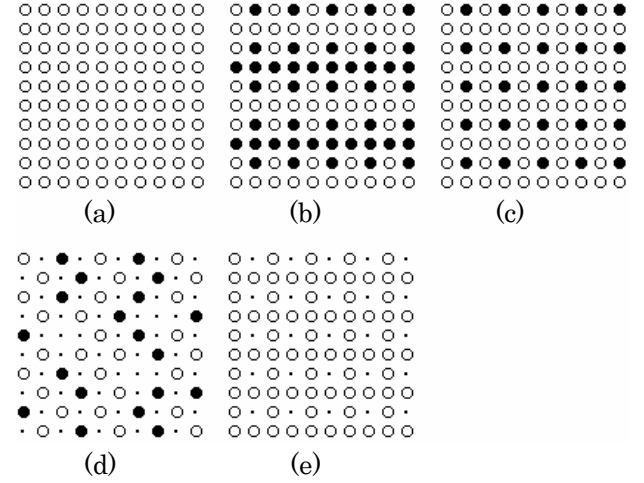


Fig. 1 Spin structures S(a), S(b), S(c), S(d), S(e) are defined by (a),(b),(c),(d),(e), respectively. Open and closed circles, and dot denote $S_z=1$, $S_z=-1$ and $S_z=0$, respectively.

We use the last spin configuration as input for the calculation at the next point. The magnetization $\langle S_z \rangle$, the four-spin thermal average $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$, the Curie temperature T_c and magnetic specific heat C_M estimated from the energy fluctuation are calculated using 2×10^5 MC steps per spin (MCS/s) after discarding first 3×10^5 MCS/s.

In order to check the reliability of these obtained average values, the thermal averages are also calculated separately for each interval of 0.5×10^5 MCS/s in the above mentioned total interval of 2×10^5 MCS/s. In the following section, results in the largest system of $L=160$ are given without showing error bars which were found to be negligibly small in our calculation.

3. Results of Calculation and Discussions

3.1 Phase Diagram of the Ground State (GS) with Interaction Parameters of $x(J_2/J_1)$ and $y(J_4/J_1)$

Let us calculate the magnetization $\langle S_z \rangle$, the four-spin thermal average $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$, the Curie temperature T_c and spin structures by the MC simulation, and investigate the conditions of phase transition of the Ising spin system with the biquadratic exchange interaction J_2 in the range of $-1.6 \leq x(J_2/J_1) \leq -0.8$ and the four-site four-spin interaction J_4 in the range of $-1.6 \leq y(J_4/J_1) \leq -0.8$. Furthermore, we have determined the GS spin structures for each spin phases of this Ising spin system. The phase diagram of the ground state is obtained for this spin system and is shown in Fig.2 for both interaction parameters x and y . The magnetic phases of PS(a)~PS(e) shown in Fig.2 represent the ones with spin structures (a)~(e) defined in the previous section.

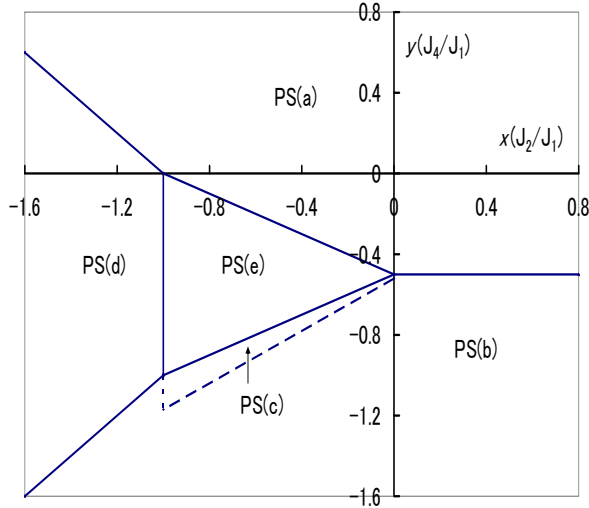


Fig. 2 Phase diagram of the ground state with exchange parameters $x(J_2/J_1)$ in the range of $-1.6 \leq x \leq 0.8$ and $y(J_4/J_1)$ in the range of $-1.6 \leq y \leq 0.8$.

As can be seen from this figure, the new phase of PS (e) is found in the negative ranges of both interaction parameters of x and y . This new phase PS(e) is also confirmed to exist in the range surrounded by three phases PS(a), PS(b) and PS(d).

By substituting these some pair values ($x=0, y=0$), ($x=0, y=-1$), ($x=-3/5, y=-9/10$), ($x=-2, y=0$), ($x=-1/2, y=-1/2$) for parameters x and y of energies ($E(a) \sim E(e)$) and comparing these energies, we can confirm that the spin structures S(a)~S(e) take the lowest energy in the

phases PS(a)~PS(e), respectively.

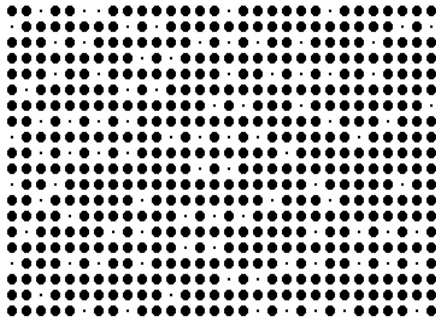
Next, let us determine the conditions of phase transition by making use of the energies per one spin obtained in the previous section. By comparing the energy $E(e)$ with each energy $E(a)$, $E(b)$ or $E(c)$, and $E(d)$, the conditions of phase transitions between PS (e) and PS (a), PS (e) and PS(b) or PS(c), PS(e) and PS(d) are obtained as $y = -x/2 - 1/2$, $y = x/2 - 1/2$ and $x = -1$, respectively. The spin structure for exchange parameters $x = -0.6$ and $y = -0.2$ on the boundary between PS(a) and PS(e) is shown by (a) in Fig.3. This spin structure mixed with S(a) and PS(e) may suggest the existence of the phase boundary between those of PS(a) and PS(e).

The condition of phase transition between PS (a) and PS(b) is also determined as $y = -1/2$ by comparing $E(a)$ with $E(b)$. Furthermore, the conditions of phase transitions between PS(d) and PS(a), PS(d) and PS(b) are also obtained as $y = -x - 1$, $y = x$ by comparing $E(d)$ with $E(a)$ and $E(b)$. These conditions of phase transition calculated by energies per one spin agree well with those obtained from the MC simulation shown in Fig.2.

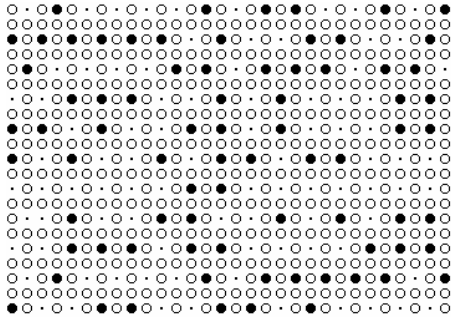
Though the energy per one spin of S(c) shown in Fig.1 with $\langle S_z \rangle = 0.5$ and $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle = -1$ has the same value with those of S(b) with $\langle S_z \rangle = 0$ and $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle = -1$, the phase of PS(c) is confirmed to exist between those of PS(b) and PS(e) only in the range of $-1 < x \leq 0$. This phase of PS(c) turns out to appear in more wide range of interaction parameter y for the smaller value of x in this range $-1 < x \leq 0$. The spin structure for exchange parameters $x = -0.6$ and $y = -0.8$ on the boundary between PS(c) and PS(e) is shown by (b) in Fig.3. This spin structure mixed with PS(c) and PS(e) may suggest the existence of the phase of PS(c) between those of PS(b) and PS(e).

The spin structures S(f) for exchange parameters $x = -0.6$ and $y = -1.0$ and S(g) for exchange parameters $x = -1.2$ and $y = -0.5$ obtained for $k_B T/J_1 = 0.1$ by the MC simulation which correspond to S(b) and S(d) in Fig.1

are shown (f) and (g) in Fig.4. This spin structure S(f) in Fig.4 as well as S(b) in Fig.1 satisfies the conditions of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle = -1$ and $\langle S_z \rangle = 0$. On the other hand, in the spin structure S(g) all spins of $S_z=1$ and $S_z=-1$ are surrounded by spins of $S_z=0$ on the nearest neighbor spin site. Furthermore, spin structures S(a), S(c), S(e) in Fig.1 agree completely with those obtained for GS spin structure by the MC simulation.

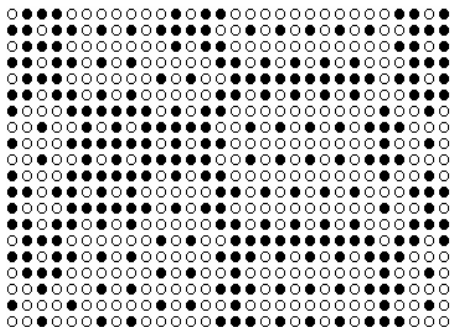


(a)

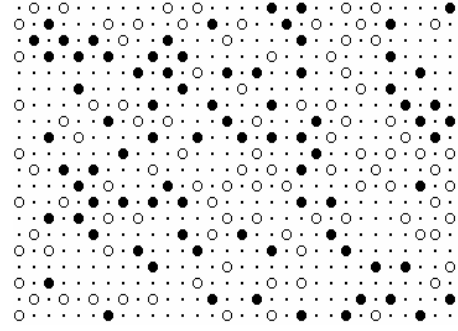


(b)

Fig. 3 Spin structures for exchange parameters (a) $J_2/J_1 = -0.6$ and $J_4/J_1 = -0.2$ (b) $J_2/J_1 = -0.6$ and $J_4/J_1 = -0.8$ on the boundaries between PS(a) and PS(e) and between PS(c) and PS(e), respectively. Open and closed circles, and dot denote $S_z = 1$, $S_z = -1$ and $S_z = 0$, respectively.



(f)



(g)

Fig. 4 Spin structures (f) and (g) obtained by MC simulation correspond to S (b) and S (d) shown in Fig.1, respectively. Open and closed circles and dot denote $S_z = 1$, $S_z = -1$ and $S_z = 0$, respectively.

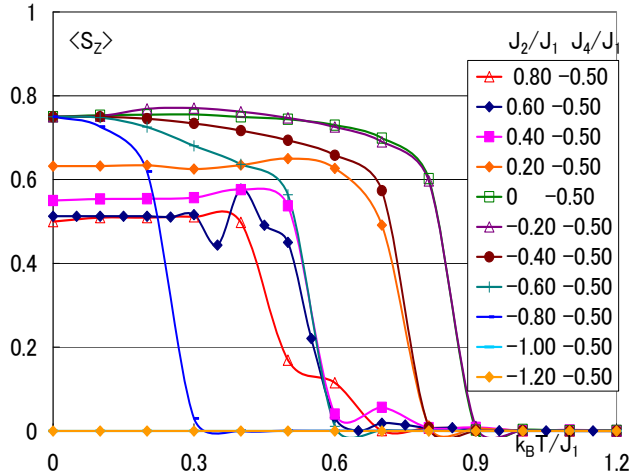
3.2 Magnetic Properties and Spin Structures of Ising Spin System with Interactions J_1 , J_2 and J_4

3.2.1 Effects of Interaction J_2 on the Magnetic Properties and Spin Structures of Ising Spin System with Interaction $J_4/J_1 = -0.5$

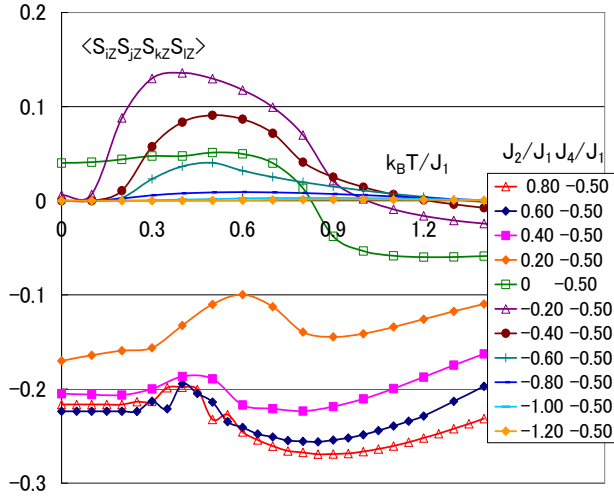
The temperature dependence of the magnetization $\langle S_z \rangle$ and four-spin thermal average $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ of the Ising spin system with $J_4/J_1 = -0.5$ was investigated for various values of interaction parameter J_2/J_1 in the range of $-1.2 \leq J_2/J_1 \leq 0.8$, and the results for $\langle S_z \rangle$ and $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ are shown by (a) and (b) in Fig.5, respectively.

For various values of J_2/J_1 in the range of $-1 < J_2/J_1 < 0$, the value of $\langle S_z \rangle$ at $T=0$ becomes constant and is 0.75 as the S(e) exists for a GS spin structure in this range. At the condition of $J_2=0$, $\langle S_z \rangle$ at $T=0$ takes becomes also 0.75 as the structure mixed with S(a) and S(c) may becomes the GS spin structure. The Curie temperature T_c turns out to increase with increasing interaction parameter J_2/J_1 in the range of $-1 < J_2/J_1 < 0$. The value of $\langle S_z \rangle$ is zero at all temperatures for interaction parameter of $J_2/J_1 \leq -1$.

For J_2/J_1 in positive range of $0.4 < J_2/J_1$, the value of $\langle S_z \rangle$ at $T=0$ becomes almost constant value in the range



(a)



(b)

Fig. 5 Temperature dependences of (a) $\langle S_z \rangle$ and (b) $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ of Ising spin system with both interactions of fixed J_4 ($J_4/J_1 = -0.5$) and various values of J_2 in the range of $-1.2 \leq J_2/J_1 \leq 0.8$.

of $0.5 < J_2/J_1 < 0.55$, and the behavior of $\langle S_z \rangle$ is almost the same for $0.4 \leq J_2/J_1$. The spin structure at $k_B T/J_1 = 0.1$ are shown in Fig.6 for interaction parameters $J_2/J_1 = 0.8$ and $J_4/J_1 = -0.5$.

Just below the Curie temperature T_c , the increases of $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ are observed for the interaction J_2 in the range of $-0.6 \leq J_2/J_1$. These abrupt rises are outstanding for the interaction $J_2/J_1 = -0.2$. The value of $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ at $T=0$ turns out to become zero in the range of $-1.0 \leq J_2/J_1 < 0$. This fact suggests that spin structure S(e) is the GS spin structure in this range.

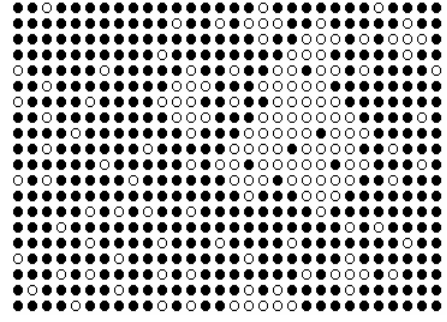


Fig. 6 Spin structure for exchange parameters $J_2/J_1 = 0.8$ and $J_4/J_1 = -0.5$ on the boundary between PS(a) and PS(b).

3.2.2 Effects of Interactions J_2 and J_4 on the Magnetic Properties of Ising Spin System on the Phase Boundaries between PS(a) and PS(e) Phases, between PS(c) and SS(e) Phases, and between PS(d) and SS(e) Phases

Let us investigate the temperature dependences of the magnetization $\langle S_z \rangle$ and four-spin thermal average $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ of the Ising spin system on the phase boundary between PS(a) and PS(e) phases. The results of $\langle S_z \rangle$ and $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ for J_2/J_1 in range of $-1 \leq J_2/J_1 \leq 0$ and J_4/J_1 in range of $-0.5 \leq J_4/J_1 \leq 0$ are shown by (a) and (b) in Fig.7, respectively. It is worth noting that the Curie temperature T_c takes the highest value for exchange parameters $J_2/J_1 = -0.2$ and $J_4/J_1 = -0.4$. As can be seen from this figure, the Curie temperature T_c becomes smaller gradually with decreasing interaction J_2 from the value of $J_2/J_1 = -0.2$. For the parameter J_2 in the range of $J_2/J_1 \leq -1$, T_c vanishes and non-zero magnetization can not appear at all temperatures.

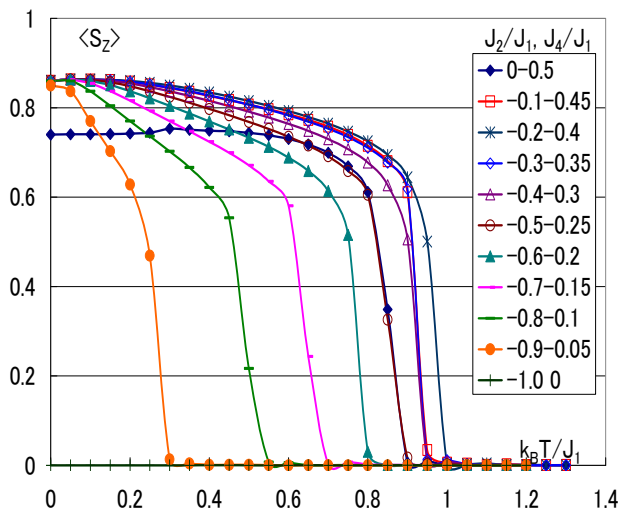
It is pointed out that the values of $\langle S_z \rangle$ at $T=0$ become about 0.863 for all pair interaction parameters on this boundary in the range of $-1 < J_2/J_1 < 0$ and $-0.5 < J_4/J_1 < 0$. This value of $\langle S_z \rangle$ is just smaller than 0.875 which correspond to the average value of $\langle S_z \rangle = 1$ for PS(a) phase and $\langle S_z \rangle = 0.75$ for PS(e) phase. This fact may suggest that on the phase boundary, the number of spin with $S_z = 0$ is slightly larger than one-seventh of that with

$S_z=1$. Therefore, on this phase boundary, the number of spin structure of S(e) turns out to be slightly larger than the one of S(a). On the other hand, the value of $\langle S_z \rangle$ at $T=0$ for parameters $J_2/J_1=0$ and $J_4/J_1=-0.5$ was confirmed to be 0.75 which correspond exactly to the average value of $\langle S_z \rangle=1$ for PS(a) phase and $\langle S_z \rangle=0.5$ for PS(b) phase.

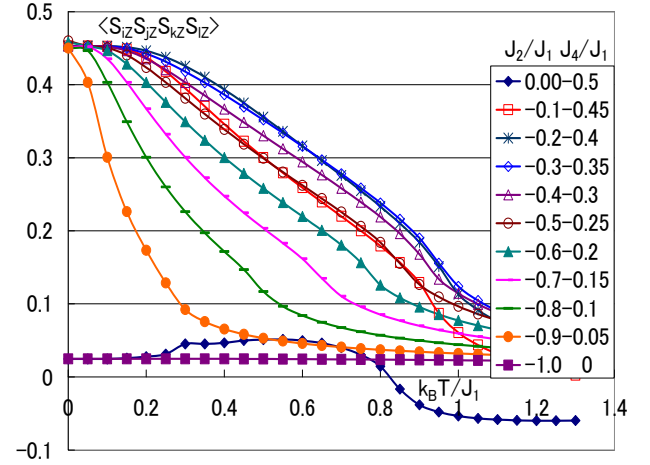
The four-spin thermal average $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ takes high values at all temperatures for the cases of interaction pairs of $J_2/J_1=-0.2$ and $J_4/J_1=-0.4$, and pairs of $J_2/J_1=-0.3$ and $J_4/J_1=-0.35$. Therefore, the behavior of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ can be said to be consistent to the one of $\langle S_z \rangle$. For the interaction range of $-1 < J_2/J_1 \leq -0.1$ and $-0.45 \leq J_4/J_1 < 0$, $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ takes positive values at all temperatures.

It is worth noting that the value of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ at $T=0$ is 0.453 for all pair interaction parameters on the boundary in the range of $-1 < J_2/J_1 < 0$ and $-0.5 < J_4/J_1 < 0$. This value of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ is just smaller than 0.5 which correspond to the average value of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle = 1$ for PS(a) phase and $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle = 0$ for PS(e) phase. This fact may also suggest that on the phase boundary, the number of the spin structure of S(e) is slightly larger than the one of S(a).

The value of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ for interaction parameters of $J_2/J_1=0$ and $J_4/J_1=-0.5$ shows no temperature



(a)



(b)

Fig. 7 Temperature dependences of (a) $\langle S_z \rangle$ and (b) $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ of Ising spin system on the boundary between PS(a) and PS(e) with both interactions J_2 in the range of $-1.0 \leq J_2/J_1 \leq 0$ and J_4 in the range of $-0.5 \leq J_4/J_1 \leq 0$.

dependence for all temperatures. On the other hand, the value of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ for interaction parameters $J_2/J_1=-1.0$ and $J_4/J_1=0$ is positive for low temperature range of $k_B T / J_1 \leq 0.82$, and negative for $0.82 < k_B T / J_1$.

Next, we investigate the dependence of the magnetization $\langle S_z \rangle$ and the four-spin thermal average $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ on the the Ising spin system on the boundary between PS(c) and PS(e). The results of $\langle S_z \rangle$ and $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ for pair interaction parameters J_2/J_1 in the range of $-1 \leq J_2/J_1 \leq 0$ and J_4/J_1 in the range of $-0.5 \leq J_4/J_1 \leq 0$ are shown by (a) and (b) in Fig. 8, respectively.

As can be seen from (a) of this figure, the Curie temperature T_c becomes small with decreasing interaction J_2 in the range of $-1 < J_2/J_1 \leq 0$ and vanishes at $J_2/J_1=-1$. It is very interesting that the curves of $\langle S_z \rangle$ have small bump at high temperatures below T_c for pair interactions J_2 and J_4 in the range of $-0.4 \leq J_2/J_1 < 0$ and $-0.7 \leq J_4/J_1 < -0.5$. These behaviors may suggest that the effect of ferromagnetic spin order appears even on the boundary in this range. The value of $\langle S_z \rangle$ at $T=0$ is 0.625 for the interactions on this boundary in the range $-1 < J_2/J_1 < 0$ and $-1.0 < J_4/J_1 \leq -0.5$, and this value agrees well with the average value of the $\langle S_z \rangle=0.5$ for spin structure S(c) and $\langle S_z \rangle=0.75$ for spin structure S(e).

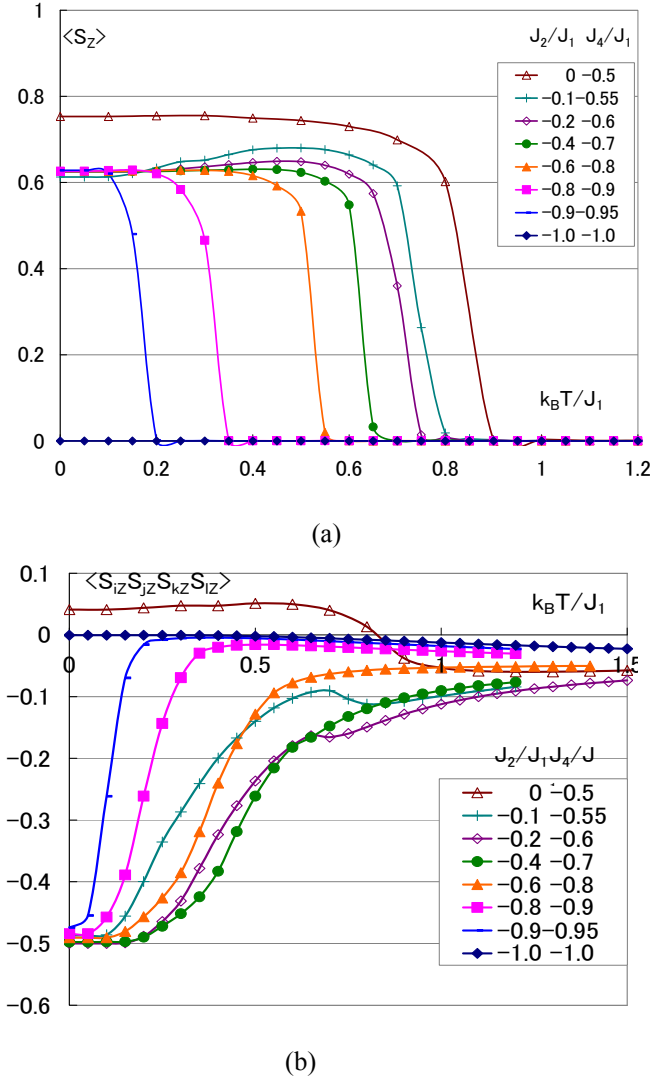


Fig. 8 Temperature dependences of (a) $\langle S_z \rangle$ and (b) $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ of Ising spin system on the boundary between PS(c) and PS(e) with both interactions of J_2 in the range of $-1 \leq J_2/J_1 \leq 0$ and J_4 in the range of $-1 \leq J_4/J_1 \leq -0.5$.

The curves of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ on this boundary have more sharp temperature dependence for small value of J_2 like $J_2/J_1 = -0.8$ and $J_4/J_1 = -0.9$ in the range of $-1 < J_2/J_1 < 0$. It is worth noting that the sign of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ for $J_2/J_1 = 0$ and $J_4/J_1 = -0.5$ changes from negative to positive with decreasing temperature, and the effects of this change appear a little even on the temperature dependence of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ in the range of $-0.2 < J_2/J_1 < 0$ and $-0.6 < J_4/J_1 < -0.5$. The values of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ at $T=0$ turns out to take -0.5 roughly for pair interactions J_2 on this boundary in the range of $-1 < J_2/J_1 < 0$ and J_4 in the range $-1 < J_2/J_1 < 0.5$. This fact may suggest that the spin

structure on this boundary is the one equally mixed with both spin structures S(c) with $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle = -1$ and S(e) with $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle = 0$. It is noticeable that the value of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ for pair interactions $J_2 = -1.0$ and $J_4 = -1.0$ is almost zero at all temperatures.

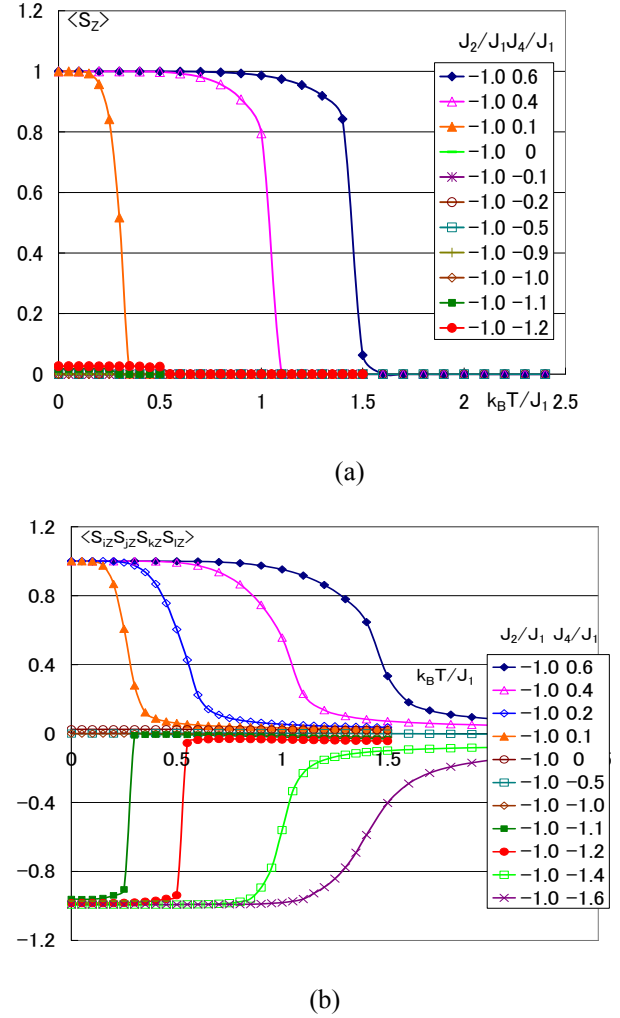


Fig. 9 Temperature dependences of (a) $\langle S_z \rangle$ and (b) $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ of Ising spin system on the boundary PS(d) and PS(e) with fixed interaction J_2 of $J_2/J_1 = -1.0$ and various values of interaction J_4 in the range of $-1.6 \leq J_4/J_1 \leq 0.6$.

Let us investigate the temperature dependences of the magnetization $\langle S_z \rangle$ and four-spin thermal average $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ of the Ising spin system on the phase boundary between PS(d) and PS(e) phases. The results of $\langle S_z \rangle$ and $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ for J_2/J_1 with -1.0 and J_4/J_1 in range of $-0.5 \leq J_4/J_1 \leq 0$ are shown by (a) and (b) in Fig.7, respectively.

As seen from (a) in Fig.9, ferromagnetic order cannot

appear for interactions J_2 of $J_2/J_1 = -1.0$ and J_4 in the range $J_4/J_1 \leq 0$. It is worth noting that the construction of four-spin order with positive value for interaction in the range of $0 < J_4/J_1$ and with negative value for interaction in the range of $J_4/J_1 < -1.0$ appears almost symmetrically against the parameter $J_4/J_1 = -0.5$ at which phase transition occurs on the Ising spin system only with interaction J_4 . The temperature dependence of $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ is confirmed to be more sharp for the interaction parameter in the range of $-1.4 \leq J_2/J_1 < -1.0$.

3.2.3 Effects of Interaction J_4 on the Magnetic Properties and Spin Structures of Ising Spin System with Interaction $J_2/J_1 = 0.6, -0.6$ and -1.2

Let us investigate the dependence of the magnetization $\langle S_z \rangle$ and the four-spin thermal average $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ on the interaction parameter J_4/J_1 of the Ising spin system with fixed interaction of $J_2/J_1 = 0.6$. The result of $\langle S_z \rangle$ and $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ for J_4/J_1 in range of $-1 \leq J_4/J_1 \leq 0$ is shown by (a) and (b) in Fig.10, respectively. The temperature dependence of the magnetization $\langle S_z \rangle$ turns out to show a large change with decreasing the interaction parameter J_4/J_1 from -0.45 to -0.55 . This change of $\langle S_z \rangle$ suggest the existence of a phase transition at $J_2/J_1 = 0.6$ and $J_4/J_1 = -0.5$. As can be seen from this figure, the values of $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ for phase boundary with $J_2/J_1 = 0.6$ and $J_4/J_1 = -0.5$ are negative and take almost constant value in the wide temperature range.

The temperature dependence of $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ for J_4 in the range of $J_4/J_1 < -0.5$ turns out to be more gradual than that for J_4 in the range of $-0.5 < J_4/J_1$. It is remarkable that in the range of $-1.0 \leq J_4/J_1 < -0.5$, the values of $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ at $T=0$ can not become -1 even for $J_4/J_1 = -1.0$. From this fact, we can realize that the spin structure at $T=0$ even for $J_4/J_1 = -1.0$ is not the complete

spin structure S(b). The interaction J_2 with positive value may disturb a creation of spin structure S(b).

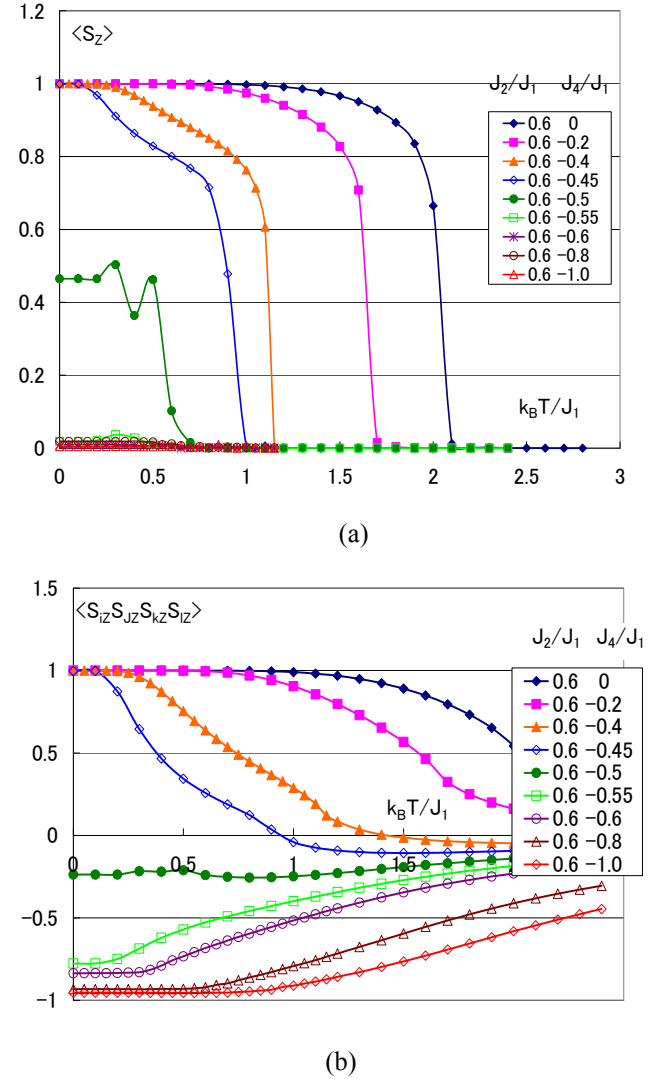


Fig. 10 Temperature dependences of (a) $\langle S_z \rangle$ and (b) $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ of Ising spin system with fixed interaction $J_2/J_1 = 0.6$ and various values of interaction J_4 in the range of $-1.0 \leq J_4/J_1 \leq 0$.

Next, we investigate the dependence of the magnetization $\langle S_z \rangle$ and the four-spin thermal average $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ on the interaction parameter J_4/J_1 of the Ising spin system with $J_2/J_1 = -0.6$. The results of $\langle S_z \rangle$ and $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ for J_4/J_1 in range of $-1 \leq J_4/J_1 \leq 0$ are shown by (a) and (b) in Fig.11, respectively.

As can be seen from (a) in this figure, $\langle S_z \rangle$ at $T=0$ takes different values for each phase and each phase boundary. The value of $\langle S_z \rangle$ at $T=0$ for phase boundary

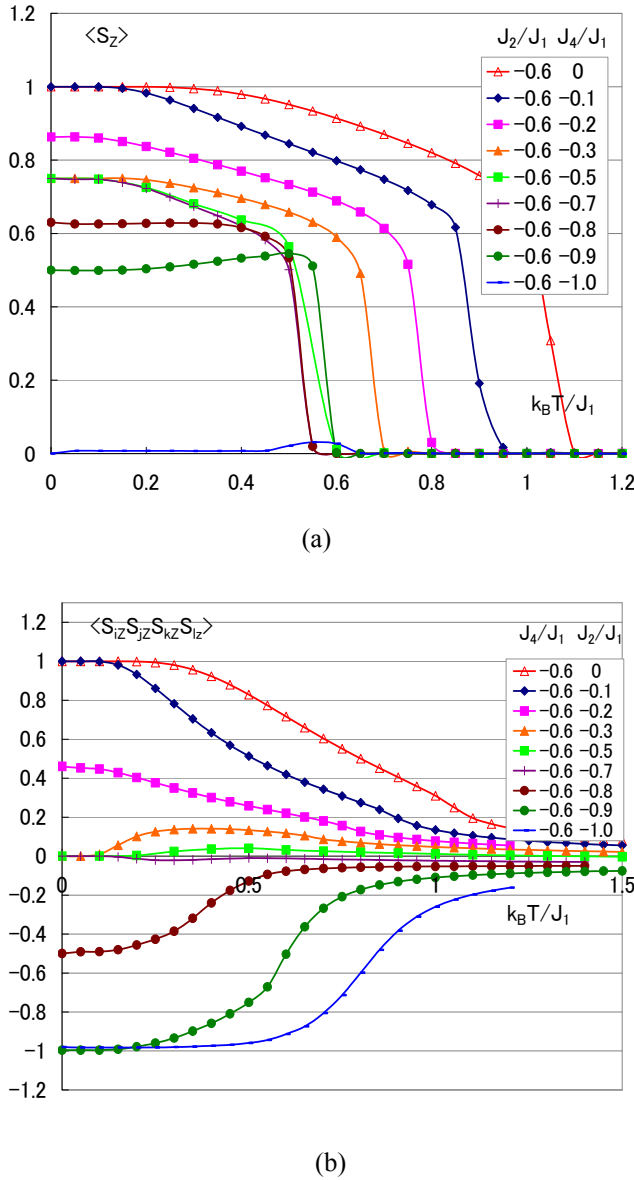


Fig. 11 Temperature dependences of (a) $\langle S_z \rangle$ and (b) $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ of Ising spin system with fixed interaction J_2 of $J_2 = -0.6$ and various values of interaction J_4 in the range of $-1.0 \leq J_4/J_1 \leq 0$.

with parameters $J_2/J_1 = -0.6$ and $J_4/J_1 = -0.2$ turns out to be 0.875 and agree well with the average value of $\langle S_z \rangle = 1$ for PS(a) phases and $\langle S_z \rangle = 0.75$ for PS(e) phases. Furthermore, the value of $\langle S_z \rangle$ at $T=0$ for phase boundary with parameters $J_2/J_1 = -0.6$ and $J_4/J_1 = -0.8$ turns out to be 0.625 and agree well with the average value of $\langle S_z \rangle = 0.75$ for PS(e) phases and $\langle S_z \rangle = 0.5$ for PS(c) phases. The existence of PS(c) may be confirmed by this value on the boundary and the behavior of $\langle S_z \rangle$ for interaction parameter of $J_4/J_1 = -0.9$.

It is worth noting that the curves of $\langle S_z \rangle$ for S(c) with interactions $J_2/J_1 = -0.6$ and $J_4/J_1 = -0.9$ and for phase boundary between S(c) and S(e) with $J_2/J_1 = -0.6$ and $J_4/J_1 = -0.8$ show abrupt rise just below T_c and keep almost constant value at wide low temperature range with decreasing temperature.

It turns out that the thermal average $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ of Ising spin system with interactions $J_2/J_1 = -0.6$ and $J_4/J_1 = -0.7$ shows almost no temperature dependence. The values of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ are positive for interaction J_4 in the range of $-0.5 \leq J_4/J_1$ and gradual increase with decreasing temperature. On the other hand, the values of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ are negative for interaction J_4 in the range of $J_2/J_1 \leq -0.8$ and shows rapid decrease with decreasing temperature. The values of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ at $T=0$ are 1, 0, -1 for the interaction J_4 in the range of $-0.2 < J_4/J_1$, $-0.8 < J_4/J_1 < -0.2$, $J_4/J_1 < -0.8$, respectively. These values may confirm the existence of PS(a), PS(e), PS(c) or PS(b).

Furthermore, the temperature dependence of the specific heat C_M has also been calculated for the Ising

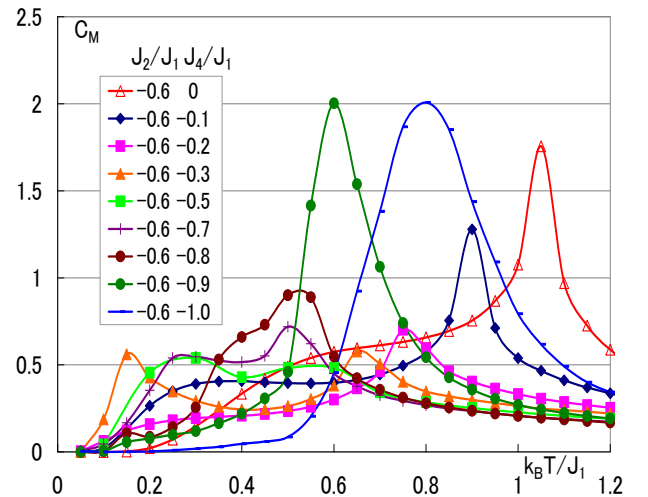
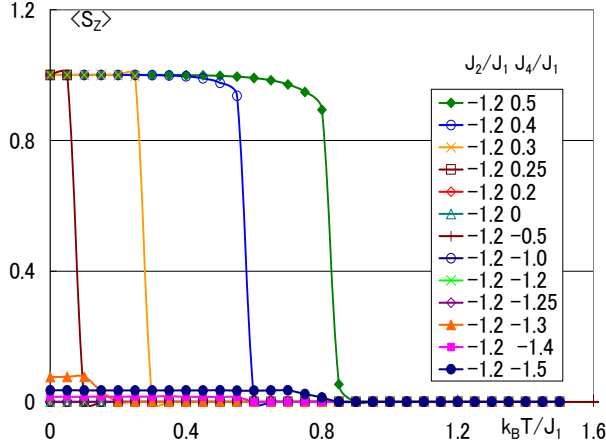


Fig. 12 Temperature dependences of C_M of Ising spin system with fixed interaction J_2 of $J_2 = -0.6$ and various values of interaction J_4 in the range of $-1.0 \leq J_4/J_1 \leq 0$.

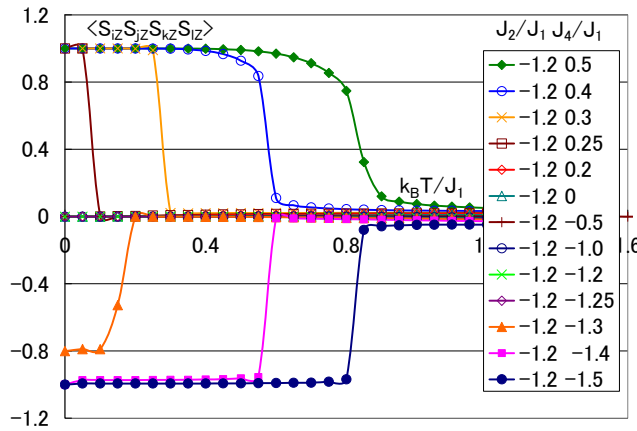
spin system with fixed interaction J_2 of $J_2 = -0.6$ and various values of interaction J_4 . The results for

interaction J_4 in the range of $-1 \leq J_4/J_1 \leq 0$ are shown in Fig.12.

It is worth noting that double peaks are observed for the phase PS(e) in the range $-0.8 < J_4/J_1 \leq -0.2$. These peaks correspond to the constructions of non-zero magnetization and spin structure S(e).



(a)



(b)

Fig. 13 Temperature dependences of (a) $\langle S_z \rangle$ and (b) $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ of Ising spin system with fixed interaction J_2 of $J_2 = -1.2$ and various values of interaction J_4 in the range of $-1.5 \leq J_4/J_1 \leq 0.5$.

Furthermore, let us investigate the dependence of the magnetization $\langle S_z \rangle$ and the four-spin thermal average $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ on the interaction parameter J_4/J_1 of the Ising spin system with $J_2/J_1 = -1.2$. The result of $\langle S_z \rangle$ and $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ for J_4/J_1 in range of $-1.5 \leq J_4/J_1 \leq 0.5$ is

shown by (a) and (b) in Fig.13, respectively. As can be seen from phase boundary between PS(a) and PS(d) in Fig.2, the temperature dependence of $\langle S_z \rangle$ suggests the existence of phase transition at $J_2/J_1 = -1.2$ and $J_4/J_1 = 0.2$. In the positive range of $0.2 < J_4/J_1$, the temperature dependence of $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ has more steep inclination with decreasing interaction parameter J_4/J_1 . On the other hand, in the negative range of $-1.3 \leq J_4/J_1 < -1.2$, the construction of negative thermal average $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ may be considerably unstable. Furthermore, for interaction J_4 in the negative range of $J_4/J_1 \leq -1.4$, the temperature dependence of negative thermal average $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ turns out to have very steep inclination.

4. Conclusions

In the previous section, for the Ising spin system of $S=1$ with the bilinear exchange interaction $J_1 S_{iz} S_{jz}$, the biquadratic exchange interaction $J_2 S_{iz}^2 S_{jz}^2$ and the four-site four-spin interaction $J_4 S_{iz} S_{jz} S_{kz} S_{lz}$, the magnetization $\langle S_z \rangle$, the four-spin thermal average $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$, the Curie temperature T_c , the specific heat C_M and the GS spin structures have been calculated by making use of the MC simulation.

Summarizing the present results on the two-dimensional square lattice, we may conclude as follows:

- (1) Phase diagram of the ground state of Ising spin system of $S=1$ with interaction parameters J_2/J_1 and J_4/J_1 are obtained by the MC simulation. The conditions of phase transition and the GS spin structures determined by this MC simulation show good agreements with those calculated from the comparison of energies per one spin for various spin structures with low energy.
- (2) The new phase PS(e) has been confirmed to exist in this Ising spin system with interactions J_1 , J_2 and J_4 .

Though the structure S(c) have the same energy with the structure S(b), the phase PS(c) is also confirmed to exist between phases PS(b) and PS(e).

- (3) The spin arrangement in the PS(d) is not the SQ state. Therefore, this arrangement can describe by single lattice model, not by two sub-lattice model.
- (4) At the phase PS(c) and on the phase boundary between PS(c) and PS(e), the magnetization $\langle S_z \rangle$ shows abrupt rise just below T_c and keep a constant value in the wide temperature range.
- (5) At the phase PS(e) and on the phase boundary between PS(a) and PS(e), the Curie temperature T_c has a maximum value for interaction parameter $J_2/J_1 = -0.2$. Therefore, ferromagnetic properties appear most strongly near or at this parameter $J_2/J_1 = -0.2$.

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