Ising Spin System with Biquadratic Exchange Interaction and Four-Site Four-Spin Interaction
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(Received OCTOBER 20, 2008)

Abstract

The phase diagram and magnetic properties such as the magnetization \(<S_z>\), the four-spin thermal average \(<S_{iz}S_{jz}S_{kz}S_{lz}>\), 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 \(J_1S_{iz}S_{jz}\), the biquadratic exchange interaction \(J_2S_{iz}^2S_{jz}^2\) and the four-site four-spin interaction \(J_4S_{iz}S_{jz}S_{kz}S_{lz}\) 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 \(<S_z>\), the four-spin thermal average \(<S_{iz}S_{jz}S_{kz}S_{lz}>\) 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 \((S_i \cdot S_j)^2\), the three-site four-spin interaction \((S_i \cdot S_j)(S_j \cdot S_k)\), the four-site four-spin interaction \((S_i \cdot S_j)(S_k \cdot S_l)\) 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,
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 NiS₂ and C₆Eu [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 \( J S_i S_j \) and the biquadratic exchange \( J S_i S_j^2 \), and the four-site four-spin interactions \( J S_i S_j S_k S_l \). The model in which the four-site four-spin interactions \( J S_i S_j S_k S_l \) 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. 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 \( <S_z> \), the four-spin thermal average \( <S_i S_j S_k S_l> \), 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

\[
H = -J_1 \sum_{ij} S_i^z S_j^z - J_2 \sum_{ij} S_i^z S_j^z S_k^z S_l^z - 2J_4 \sum_{ijkl} S_i^z S_j^z S_k^z S_l^z, \tag{1}
\]

Here, \( ijkl \) 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 \( (S_i^z \cdot S_j^z)(S_k^z \cdot S_l^z) \) and \( (S_i^z \cdot S_j^z)(S_k^z \cdot S_l^z) \). Furthermore, \( S \) 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 $E(a)=E_a/J_1$ 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 $<S_zS_xS_yS_z>=-1$ 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 the same value and given by $E(b)=E(c)=2x+2y$. On the other hand, the values of magnetization $<S_z>$ 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.

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![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.](image)

We use the last spin configuration as input for the calculation at the next point. The magnetization $<S_z>$, the four-spin thermal average $<S_zS_xS_yS_z>$, the Curie temperature $T_c$ and magnetic specific heat $C_M$ estimated from the energy fluctuation are calculated using $2\times10^5$ MC steps per spin (MCS/s) after discarding first $3\times10^5$ MCS/s.

In order to check the reliability of these obtained average values, the thermal averages are also calculated separately for each interval of $0.5\times10^5$ MCS/s in the above mentioned total interval of $2\times10^5$ 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 $y(J_4/J_1)$

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Let us calculate the magnetization $\langle S_i \rangle$, the four-spin thermal average $\langle S_i S_j S_k S_l \rangle$, 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.

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 ($E(a)$ ~ $E(e)$) 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 $y = -x/2$, $y = x/2$ 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 S(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 $y = -x$, $y = x$ 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 $<S_i>=0.5$ and $<S_i S_j S_k S_l>=-1$ has the same value with those of S(b) with $<S_i>=0$ and $<S_i S_j S_k S_l>=-1$, the phase of PS(c) is confirmed to exist between those of PS(b) and PS(e) only in the range of $-1 < x < 0$. 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 $k_BT/J_1=0.1$ 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 $<S_z S_y S_x S_z S_x> = -1$ and $<S_z> = 0$. 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.

3.2 Magnetic Properties and Spin Structures of Ising Spin System with Interactions $J_p$, $J_2$ 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 $<S_z>$ and four-spin thermal average $<S_z S_y S_x S_z S_x>$ 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 $<S_z>$ and $<S_z S_y S_x S_z S_x>$ 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 $<S_z>$ 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 $J_2 = 0$, $<S_z>$ 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 $-1 < J_2/J_1$. The value of $<S_z>$ 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 $<S_z>$ at $T=0$ becomes almost constant value in the range...
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 $\langle S_i S_j S_k S_l \rangle$ of the Ising spin system on the phase boundary between PS(a) and PS(e) phases. The results of $\langle S_z \rangle$ and $\langle S_i S_j S_k S_l \rangle$ 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 $J_4/J_1=-0.5$. 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.

Just below the Curie temperature $T_c$, the increases of $\langle S_i S_j S_k S_l \rangle$ 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 $\langle S_i S_j S_k S_l \rangle$ at $T=0$ turns out to become zero in the range of $-1.0 \leq J_2/J_1 \leq 0$. This fact suggests that spin structure S(e) is the GS spin structure in this range.
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 $<S_z>$ 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 $<S_z>=1$ for PS(a) phase and $<S_z>=0.5$ for PS(b) phase.

The four-spin thermal average $<S_{i\alpha}S_{j\beta}S_{k\gamma}S_{l\delta}>$ 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 $<S_{i\alpha}S_{j\beta}S_{k\gamma}S_{l\delta}>$ can be said to be consistent to the one of $<S_z>$. For the interaction range of $-1<J_2/J_1\leq-0.1$ and $-0.45\leq<J_4/J_1<0$, $<S_{i\alpha}S_{j\beta}S_{k\gamma}S_{l\delta}>$ takes positive values at all temperatures.

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 $-1<J_2/J_1\leq0$ and vanishes at $J_2/J_1=-1$. It is very interesting that the curves of $<S_z>$ 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 $<S_z>$ at $T=0$ is 0.625 for interaction parameters $J_2/J_1=0$ and $J_4/J_1=-0.5$, which agrees well with the average value of the $<S_z>=0.5$ for spin structure S(c) and $<S_z>=0.75$ for spin structure S(e).
The curves of $\langle S_z \rangle$ and $\langle S_z S_x S_y S_t \rangle$ 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 $\langle S_z S_x S_y S_t \rangle$ 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 $\langle S_z S_x S_y S_t \rangle$ in the range of $-0.2<J_2/J_1<0$ and $-0.6<J_2/J_1<0.5$. The values of $\langle S_z S_x S_y S_t \rangle$ 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 $\langle S_z S_x S_y S_t \rangle=-1$ and S(e) with $\langle S_z S_x S_y S_t \rangle=0$. It is noticeable that the value of $\langle S_z S_x S_y S_t \rangle$ for pair interactions $J_2=-1.0$ and $J_4=-1.0$ is almost zero at all temperatures.

Let us investigate the temperature dependences of the magnetization $\langle S_z \rangle$ and four-spin thermal average $\langle S_z S_x S_y S_t \rangle$ of the Ising spin system on the phase boundary between PS(d) and PS(e) phases. The results of $\langle S_z \rangle$ and $\langle S_z S_x S_y S_t \rangle$ for $J_2/J_1$ with $-1.0$ and $J_4/J_1$ in range of $-0.5<J_2/J_1<0$ 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 $<S_{iz}S_{jz}S_{kz}S_{lz}>$ 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 $<S_z>$ and the four-spin thermal average $<S_{iz}S_{jz}S_{kz}S_{lz}>$ 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 $<S_z>$ and $<S_{iz}S_{jz}S_{kz}S_{lz}>$ 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 $<S_z>$ 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 $<S_z>$ 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 $<S_{iz}S_{jz}S_{kz}S_{lz}>$ 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 $<S_{iz}S_{jz}S_{kz}S_{lz}>$ 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 the is remarkable that in the range of $-1.0 \leq J_4/J_1<-0.5$, the values of $<S_{iz}S_{jz}S_{kz}S_{lz}>$ 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 $S(b)$.

Next, we investigate the dependence of the magnetization $<S_z>$ and the four-spin thermal average $<S_{iz}S_{jz}S_{kz}S_{lz}>$ on the interaction parameter $J_4/J_1$ of the Ising spin system with $J_2/J_1=-0.6$. The results of $<S_z>$ and $<S_{iz}S_{jz}S_{kz}S_{lz}>$ 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, $<S_z>$ at $T=0$ takes different values for each phase and each phase boundary. The value of $<S_z>$ at $T=0$ for phase boundary
It is worth noting that the curves of $<S_Z>$ 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 $<S_{\alpha}\alpha S_{\beta}S_{\gamma}S_{\delta}>$ 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 $<S_{\alpha}\alpha S_{\beta}S_{\gamma}S_{\delta}>$ are positive for interaction $J_4$ in the range of $-0.5 \leq J_4/J_1 \leq 0$ and gradual increase with decreasing temperature. On the other hand, the values of $<S_{\alpha}\alpha S_{\beta}S_{\gamma}S_{\delta}>$ 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 $<S_{\alpha}\alpha S_{\beta}S_{\gamma}S_{\delta}>$ at $T=0$ are 1, 0, -1 for the interaction $J_2$ in the range of $-0.2 < J_2/J_1 < -0.8$, 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 spin system with fixed interaction $J_2$ of $J_2=-0.6$ and various values of interaction $J_4$. The results for

![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 $-1.0 \leq J_4/J_1 \leq 0$.](image)
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 $-0.8 < J_4/J_1 < -0.2$. These peaks correspond to the constructions of non-zero magnetization and spin structure $S(e)$.

![Graph](a)

**Fig. 13** Temperature dependences of (a) $<S_z>$ and (b) $<S_{iz}S_{jz}S_{kz}S_{lz}>$ of Ising spin system with fixed interaction $J_2/J_1$ of $J_2/J_1= -1.2$ 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 $<S_z>$ and the four-spin thermal average $<S_{iz}S_{jz}S_{kz}S_{lz}>$ on the interaction parameter $J_2/J_1$. On the other hand, in the negative range of $-1.3 \leq J_4/J_1 < -1.2$, the construction of negative thermal average $<S_{iz}S_{jz}S_{kz}S_{lz}>$ 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 $<S_{iz}S_{jz}S_{kz}S_{lz}>$ 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 $J_1S_{iz}S_{jz}$, the biquadratic exchange interaction $J_2S_{iz}S_{jz}^2$ and the four-site four-spin interaction $J_4S_{iz}S_{jz}S_{kz}S_{lz}$, the magnetization $<S_z>$, the four-spin thermal average $<S_{iz}S_{jz}S_{kz}S_{lz}>$, the Curie temperature $T_c$, the specific heat $C_V$ 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 $J_4$. 

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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 $<S_z>$ 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 $J_2/J_1=-0.2$.

References