

Ising Spin System with Higher-Order Spin Interactions and Single-Ion Anisotropy

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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 on two-dimensional square lattice with the bilinear exchange interaction $J_1 S_{iz} S_{jz}$, the biquadratic exchange interaction $J_2 S_{iz}^2 S_{jz}^2$, the four-site four-spin interaction $J_4 S_{iz} S_{jz} S_{kz} S_{lz}$ and a single-ion anisotropy D have been discussed by making use of the Monte Carlo simulation. In this Ising spin system with the interactions J_1 , J_2 and J_4 and anisotropy term D 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 a 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 $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$, the thermal average $\langle S_z^2 \rangle$ 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 .

Keywords: Ising model; higher-order spin interaction; single-ion anisotropy; Monte Carlo simulation

1. Introduction

In Heisenberg and Ising ferromagnets, the existence and the importance of such higher-order exchange interactions as the biquadratic exchange interaction $J_2(\mathbf{S}_i \cdot \mathbf{S}_j)^2$, the three-site four-spin interaction $J_3(\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_j \cdot \mathbf{S}_k)$, the four-site four-spin interaction $J_4(\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_k \cdot \mathbf{S}_l)$ have been discussed extensively by many investigators [1-3]. Theoretical explanations of the origin of these interactions have been given in the theory of the super exchange interaction, the magnetoelastic effect, the perturbation expansion 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 some other materials showing such phenomena as quadrupolar ordering of molecules (solid hydrogen, liquid crystal) 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 NiS_2 and C_6Eu [9].

The Ising system of $S=1$ with the bilinear interaction $J_1 S_{iz} S_{jz}$ and the biquadratic exchange interaction $J_2 S_{iz}^2 S_{jz}^2$ and the single-ion anisotropy D is quite famous as so-called Blume-Emery-Griffiths

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(BEG) model [10] and applied for many problems, e.g. super-liquid helium, magnetic material, semiconductor, alloy, lattice gas and so on. This interaction J_2 is expected to have significant effects on magnetic properties and spin arrangement in the low-temperature region for the case of J_2 not negligible compared to J_1/S^2 [11]. 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 [12,13] 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 two-dimensional square lattice with three kinds of interactions such as the bilinear exchange $J_1 S_{iz} S_{jz}$, the biquadratic exchange $J_2 S_{iz}^2 S_{jz}^2$, the four-site four-spin interactions $J_4 S_{iz} S_{jz} S_{kz} S_{lz}$ and the single-ion anisotropy $D S_{iz}^2$. By making use of this simulation, we have investigated more precisely the growth of the spin ordering, conditions of phase transition and the ground state (GS) spin structures of the Ising spin system with $S=1$. In the present study, we determined the phase diagrams of the Ising system with anisotropy term D and without the one. The obtained phase diagram is discussed in conjunction with the GS spin structures determined by energy evaluations. The temperature dependences of the magnetization $\langle S_z \rangle$ and four spin thermal average $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ were also studied for various values of parameters J_2/J_1 and D/J_1 .

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 shown to be calculated from this spin Hamiltonian. Furthermore, the method of the MC simulation is explained briefly. In Section 3, phase diagram is obtained for exchange parameters J_2/J_1 and J_4/J_1 by the MC simulation of the Ising system without anisotropy term D . In the latter part of this section, this calculation of MC simulation are extended to the Ising system with anisotropy term D , and phase diagram is determined. Furthermore, these diagrams are confirmed by the one obtained from 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 a new magnetic phase. In the last Section 5, new interesting results obtained here are summarized.

2. Spin Hamiltonian and Methods of MC Simulation

The spin Hamiltonian for the present Ising spin system with $S=1$ on two-dimensional square lattice can be written as follows:

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

Here, $\langle ij \rangle$ and $\langle ijkl \rangle$ denote the sum on the nearest neighboring spin pairs and on the square spin sites of two-dimensional square lattice. The coefficient 2 of the third term in this Hamiltonian is obtained by considering the sum of two terms $(S_{iz} \cdot S_{jz})(S_{kz} \cdot S_{lz})$, and $(S_{iz} \cdot S_{lz})(S_{jz} \cdot S_{kz})$. Furthermore, S_z in above expression represents $S_z = \pm 1, 0$. From a consideration of the Hamiltonian (1), magnetic properties and spin arrangements of Ising spin system of $S=1$ on 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. [14]).

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 $L=240$. For fixed values of various parameters J_1 , J_2 , J_4 and D , 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. We use the last spin configuration as an 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 the 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 $L=240$ are given without showing error bars which were found to be negligibly small in our calculation.

3. Results of Simulation and Discussion

3.1 Phase Diagram of Ising System without Single-Ion Anisotropy ($D=0$)

Let us calculate magnetic properties and spin structures by the MC simulation and investigate the conditions of a phase transition, and determine the GS spin structures of the Ising spin system with the biquadratic exchange interaction J_2 in the range of $-1.6 \leq J_2/J_1 \leq -0.8$ and the four-site four-spin interaction J_4 in the range of $-1.6 \leq J_4/J_1 \leq -0.8$. In this calculation, the interaction parameter J_1 was treated as a constant value and an anisotropy parameter D was set as zero, a constant value. Here, we define these parameters J_2/J_1 and J_4/J_1 as x and y , respectively. The phase diagram of the ground state is obtained for this Ising spin system on two-dimensional lattice, and the result is shown in Fig.1 for both interaction parameters x and y .

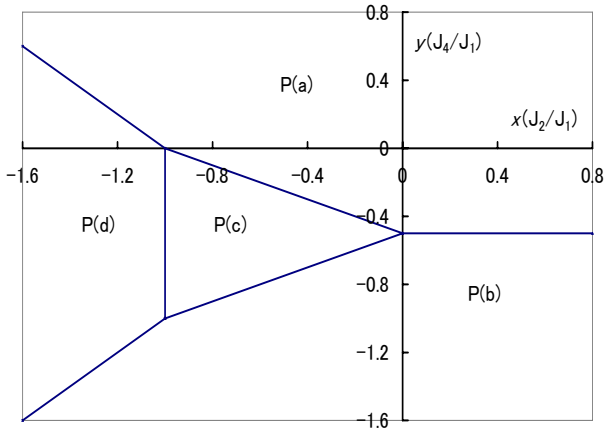


Fig. 1 Phase diagram of Ising spin system with exchange parameters $x(J_2/J_1)$ in the range of $-1.6 \leq x \leq 0.8$ and $y(J_4/J_1)$ in the range of $-1.6 \leq y \leq 0.8$ ($D=0$).

The GS spin structures of magnetic phases P(a)~P(d) are defined by the spin structures S(a)~S(d) shown in Fig.2, respectively. The GS spin structure S(a) of phase P(a) is a ferromagnetic spin arrangement characterized by magnetic parameters as $\langle S_z \rangle = 1$, $\langle S_{iz}S_{jz} \rangle = 1$ and $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle = 1$. The GS spin structure of phase P(b) calculated by the MC simulation are confirmed to be the spin structure mixed with S(b₁) and S(b₂) of the same energy shown in Fig.2. The example of the GS spin structure of P(b) obtained by the MC simulation is shown by

(a) in Fig.3. This GS spin structure of P(b) are characterized by magnetic parameters as $\langle S_z \rangle = 0$, $\langle S_{iz}S_{jz} \rangle = 0$ and $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle = -1$.

As can be seen from Fig.1, the new phase of P(c) is found in the negative ranges of both interaction parameters of x and y . This new phase P(c) is also confirmed to exist in the range surrounded by three phases P(a), P(b) and phase P(d). The GS spin structure S(c) of phase P(c) turns out to be characterized by magnetic parameters as $\langle S_z \rangle = 0.75$, $\langle S_{iz}S_{jz} \rangle = 0.5$ and $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle = 0$. Furthermore, the example of the GS spin structure of P(d) obtained by the MC simulation is shown by (b) in Fig.3. The GS spin structure of phase P(d) are characterized by magnetic parameters as $\langle S_z \rangle = 0$, $\langle S_{iz}S_{jz} \rangle = 0$ and $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle = 0$.

Furthermore, the magnetizations $\langle S_z(A) \rangle$ and $\langle S_z(B) \rangle$, and thermal averages $\langle S_z^2(A) \rangle$ and $\langle S_z^2(B) \rangle$ on the sub-lattice have been calculated for this phase P(d). Here, A and B represent the two-interpenetrating lattices. The conditions of $\langle S_z(A) \rangle = 0$, $\langle S_z(B) \rangle = 0$ and $\langle S_z^2(A) \rangle = \langle S_z^2(B) \rangle \neq 0$ are obtained by this simulation. Therefore, the GS spin structure of P(d) is confirmed not to be a staggered quadrupolar (SQ) ordering. In the Ising spin system with $J_2/J_1 = -1.5$ and $J_4/J_1 = -0.5$, thermal averages $\langle S_z^2(A) \rangle$ and $\langle S_z^2(B) \rangle$ turn out to take the same value as $\langle S_z^2(A) \rangle = \langle S_z^2(B) \rangle = 0.287$ at low temperatures. Therefore, these facts may suggest that the number of a spin with $S_z = 0$ is about 5 times larger than that with $S_z = 1$ or $S_z = -1$ in this region.

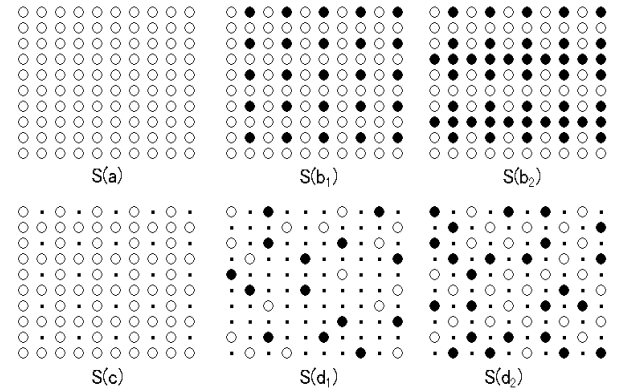
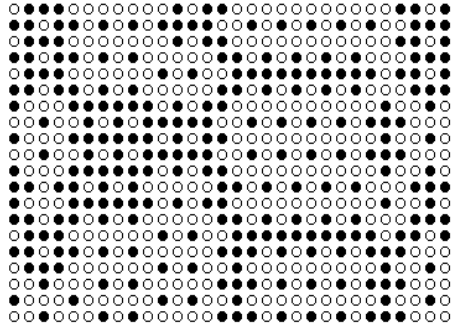
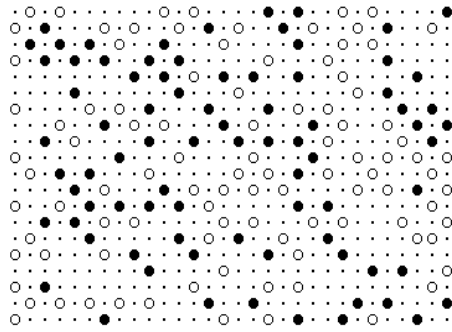


Fig. 2 The GS spin structures S(a), S(b₁), S(b₂), S(c), S(d₁) and S(d₂) of magnetic phases P(a), P(b), P(c) and P(d), respectively ($D=0$). Open and closed circles, and dot denote $S_z = 1$, $S_z = -1$ and $S_z = 0$, respectively. The GS spin structures S(d₂) appears only for spin system with single-ion anisotropy D ($D \neq 0$)



(a)



(b)

Fig. 3 The GS spin structures of P(b) and P(d) calculated by MC simulation. Open and closed circles, and dot denote $S_z = 1$, $S_z = -1$ and $S_z = 0$, respectively.

Next, let us calculate the energies $E(a) \sim E(d)$ per one spin for spin structures S(a) $\sim$ S(d) shown in Fig.2 by taking spin Hamiltonian (1) into consideration (see e.g.[14]). The energies per one spin for S(a) $\sim$ S(d) obtained from this calculation are given as $E(a) = -2x - 2y - 2$, $E(b_1) = E(b_2) = -2x + 2y$, $E(c) = -x - 1$ and $E(d) = 0$. Furthermore, we have determined the conditions of phase transition by making use of the energies per one spin. By comparing the energy $E(c)$ with each energy $E(a)$, $E(b)$ and $E(d)$, the phase transitions turn out to occur at $y = -x/2 - 1/2$, $y = x/2 - 1/2$ and $x = -1$, respectively. The conditions of phase transition are obtained from same procedures as $y = -1/2$ by comparing $E(a)$ with $E(b)$, and as $y = -x - 1$, $y = x$ by comparing $E(d)$ with $E(a)$ and $E(b)$, respectively. It is worth noting that these conditions obtained from comparisons of energies per one spin agree quite well with the results calculated from the MC simulation shown in Fig.1.

3.2 Phase Diagram of Ising System with Single-Ion Anisotropy ($D \neq 0$)

Next, let us investigate magnetic properties and spin structures by the MC simulation and investigate the conditions of a phase transition, and determine the GS spin structures of the Ising spin system with an anisotropy term D and the biquadratic exchange interaction J_2 in the range of $-1.6 \leq J_2/J_1 \leq -0.8$ and the four-site four-spin interaction J_4 in the range of $-1.6 \leq J_4/J_1 \leq -0.8$. In this calculation, the interaction parameter J_1 was treated as a constant value, and the MC simulation was performed for the Ising spin system with single-ion anisotropic parameter D in the range of $0 < D/J_1 \leq 1.5$. The conditions of phase transition and GS spin structures for Ising system with an anisotropy D on two-dimensional square lattice are calculated, and the phase diagram of the ground state for Ising system with $D/J_1 = 1$ is shown in Fig. 4.

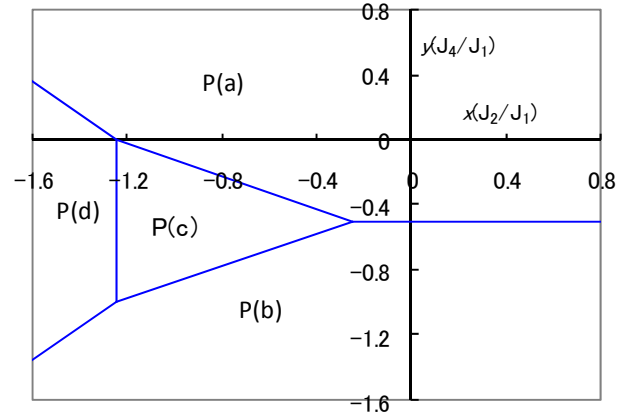


Fig. 4 Phase diagram of Ising spin system with exchange parameters $x(J_2/J_1)$ in the range of $-1.6 \leq x \leq 0.8$ and $y(J_4/J_1)$ in the range of $-1.6 \leq y \leq 0.8$ and an anisotropy parameter of $D/J_1 = 1$.

As can be seen from Fig.4, all phases P(a) $\sim$ P(d) for $D/J_1 = 1$ are given by parallel translation along $x(J_2/J_1)$ axis of those for $D/J_1 = 0$. These facts suggest that the effects of interaction J_2 with negative value which has a tendency to decrease the value of S_z may deny by operation of a single-ion anisotropy term D with a tendency to increase the value of S_z . On the other hand, as a single-ion anisotropy term D and the interaction J_4 with negative value have almost the same operation to increase the value of S_z , this single-ion anisotropy term D cannot give any change

to the effect by interaction J_4 . The GS spin structures for magnetic phases P(a) $\sim$ P(c) in Fig.4 are the same with those in Fig.1. It is worth noting that the GS spin structures for magnetic phase P(d) is $S(d_2)$ shown in Fig.2. This spin structure $S(d_2)$ is characterized by the conditions of $\langle S_z(A) \rangle = 0$, $\langle S_z(B) \rangle = 0$ and $\langle S_z^2(A) \rangle = 1$, $\langle S_z^2(B) \rangle = 0$. Therefore, the GS spin structure of P(d) is confirmed to be the staggered quadrupolar (SQ) ordering. The example of the GS spin structure of P(d) obtained by the MC simulation is shown in Fig.5.

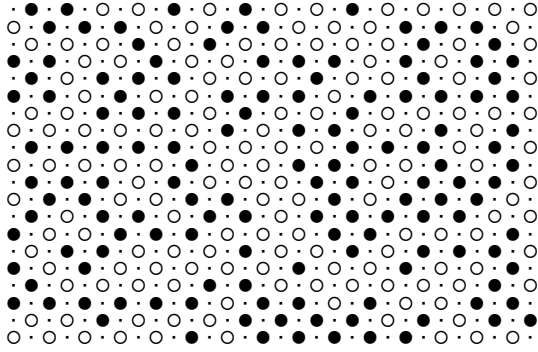


Fig. 5 The GS spin structure of P(d) calculated by MC simulation. Open and closed circles, and dot denote $S_z = 1$, $S_z = -1$ and $S_z = 0$, respectively.

Next, let us calculate the energies $E(a) \sim E(d)$ per one spin for spin structures S(a) $\sim$ S(d) for the Ising spin system with a single-ion anisotropy D . The energies per one spin for S(a) $\sim$ S(d) for $D/J_1 \neq 0$ obtained from the same calculation with the one for $D = 0$ are given as $E(a) = -2x - 2y - d - 2$, $E(b_1) = E(b_2) = -2x + 2y - d$, $E(c) = -x - 3d/4 - 1$ and $E(d) = -d/2$. Here, the parameter d is defined as D/J_1 . Furthermore, we have determined the conditions of phase transition by making use of these energies per one spin. These conditions are given as $y = -x/2 - d/8 - 1/2$, $y = x/2 + d/8 - 1/2$, $x = -d/4 - 1$, $y = -1/2$, $y = -x - d/4 - 1$ and $y = x + d/4$. By substituting $d = 1$ in above mentioned equations, the conditions of phase transition for the Ising spin system with $D/J_1 = 1$ are obtained as $y = -x/2 - 5/8$, $y = x/2 - 3/8$, $x = -5/4$, $y = -1/2$, $y = -x - 5/4$ and $y = x + 1/4$. It is quite remarkable that the results by these straight lines obtained from energy comparison agree well with those shown in Fig.4 determined from the MC simulation.

3.3 Magnetic Properties of New Phase P(c) on Ising System with Interactions J_2 and J_4 ($D = 0$)

Let us investigate the magnetic properties such as the magnetization $\langle S_z \rangle$ and the four-spin thermal average $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$, the magnetic specific heat C_M of a new magnetic phase P(c) by making use of the MC simulation. The changes of the temperature dependence of $\langle S_z \rangle$ for Ising system with the fixed interaction J_4 ($J_4/J_1 = -0.5$) and various values of J_2 are shown in Fig.6. Furthermore, the temperature dependences of $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ and C_M for Ising system with the fixed interaction J_4 ($J_4/J_1 = -0.5$) and various values of J_2 are shown by (a) and (b) in Fig.7, respectively. These calculations by the MC simulation are performed for the Ising spin system with no anisotropy ($D = 0$).

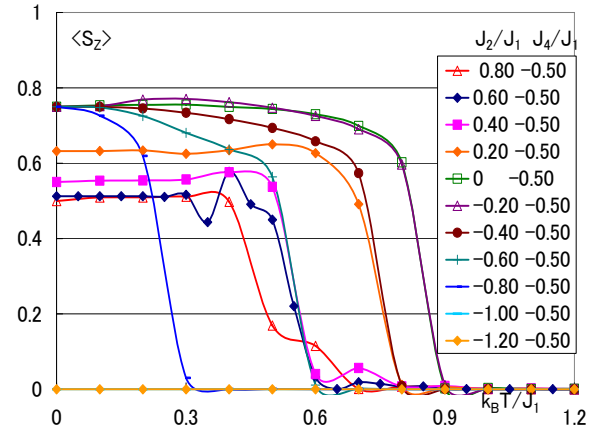
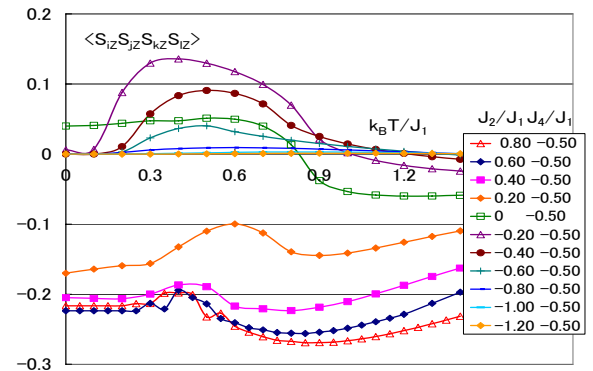


Fig. 6 Temperature dependence of $\langle S_z \rangle$ of the magnetic phase P(c) calculated by MC simulation for fixed value of J_4 ($J_4/J_1 = -0.5$) and various values J_2 of in the range of $-1.2 \leq J_2/J_1 \leq 0.8$.



(a)

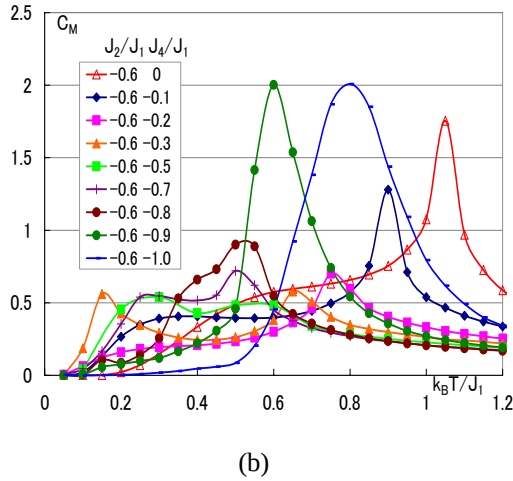


Fig. 7 Temperature dependence of (a) $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ and (b) C_M of the magnetic phase P(c) calculated by MC simulation for fixed value of (a) J_4 ($J_4/J_1 = -0.5$) and fixed value of (b) J_2 ($J_2/J_1 = -0.6$).

As can be seen from Fig.6, the shape of temperature dependence of $\langle S_z \rangle$ for $J_2/J_1 = -0.2$ is almost the same with the one for $J_2/J_1 = 0$. Therefore, ferromagnetic property of this Ising system with $J_4/J_1 = -0.5$ turns out to keep in the interaction range of $-0.4 < J_2/J_1 \leq 0$. The effect of negative interaction J_2 to decrease the value of S_z at $J_2/J_1 = -0.2$ may be canceled by the one of negative interaction J_4 with $J_4/J_1 = -0.5$ to increase the value of S_z .

As can be seen from (a) in Fig.7, four-spin thermal average $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ with large positive value appears in the range of $-0.4 < J_2/J_1 < 0$. This result supports the existence of rather strong ferromagnetic property in this interaction range. It is interesting that the sign of $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ of the Ising system with $J_4/J_1 = -0.5$ and $J_2/J_1 = 0$ is negative for temperature range in the paramagnetic state ($T_c < T$) and positive for temperature range in ordered state ($T < T_c$). Furthermore, two peaks are observed on the specific heat C_M curve (b) in Fig.7 in the interaction range of $-0.8 < J_4/J_1 < 0$. These two peaks may correspond to the appearance of ordered state (T_c) and the creation of the GM spin structure of magnetic phase P(c).

The changes of the temperature dependence of $\langle S_z \rangle$ and $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ for the Ising system with the fixed interaction J_2 ($J_2/J_1 = -0.6$) and various values of J_4 are shown by (a) and (b) in Fig. 8. The values of $\langle S_z \rangle$ at $T=0$ are 1, 0.75 and 0 for the interaction ranges of $-0.2 < J_4/J_1$, $-0.8 < J_4/J_1 < -0.2$ and $J_4/J_1 < -0.9$, respectively. Furthermore, $\langle S_z \rangle$ at $T=0$ for the phase boundaries of $J_4/J_1 = -0.2$ and $J_4/J_1 = -0.8$ are 0.875 and 0.625, respectively.

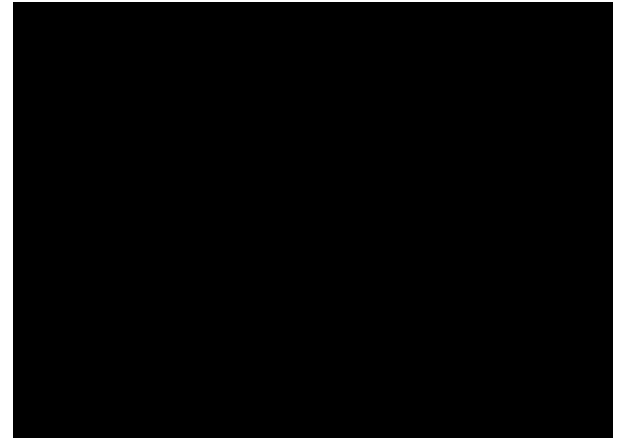
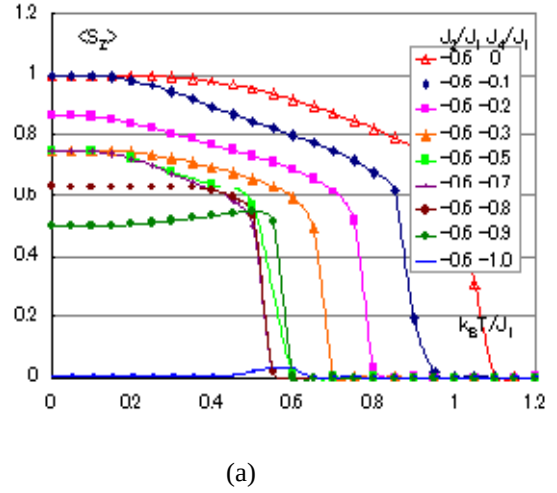


Fig. 8 Temperature dependence of (a) $\langle S_z \rangle$ and (b) $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ of the magnetic phase P(c) calculated by MC simulation for fixed value of J_2 ($J_2/J_1 = -0.6$) and various values of in the range of $-1 \leq J_4/J_1 \leq 0$.

These behaviors of $\langle S_z \rangle$ at low temperature support the phase diagram shown in Fig.1 and the GS spin structure shown in Fig.2. It is quite remarkable that near the phase boundary of the interaction range of $-0.9 \leq J_2/J_1 < -0.8$, the spin structure with almost only structure S (b₁) in Fig.2 appears as the GS spin structure of phase P(b). In the interaction range of $J_2/J_1 < -0.9$, the spin structure mixed with structures S (b₁) and S (b₂) of the same energy turns out to be the GS spin structure of phase P(b). Therefore, the value of $\langle S_z \rangle$ at $T=0$ becomes zero. As can be seen from (b) in Fig.8, the temperature dependence curves of $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ for phase P(b) are more sharp than those for P(a) near the temperature of phase

transition from paramagnetic state to ordered state. It should be noted that the value of $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle$ for parameters $J_2/J_1 = -0.6$ and $J_4/J_1 = -0.7$ is almost zero at all temperatures.

3.4 Spin Arrangements of Phase P(d) on Ising System with Interactions J_2 and J_4

The temperature dependence of the spin arrangements of phase P(d) have been investigated for various values of an anisotropy parameters D/J_1 by the MC simulation. As the value of $\langle S_z \rangle$ is zero in all region of P(d), we have calculated the thermal averages of $\langle S_z^2(A) \rangle$ and $\langle S_z^2(B) \rangle$. Here, these values of $\langle S_z^2(A) \rangle$ and $\langle S_z^2(B) \rangle$ are defined as $\langle S_z^2(A) \rangle > \langle S_z^2(B) \rangle$, and A and B represent the two-interpenetrating lattices. The temperature dependences of $\langle S_z^2(A) \rangle$ and $\langle S_z^2(B) \rangle$ of the Ising spin system with $J_2/J_1 = -1.5$ and $J_4/J_1 = -0.5$ are shown in Fig. 9 for various values of anisotropy term D in the range of $0 \leq D/J_1 \leq 1.5$.

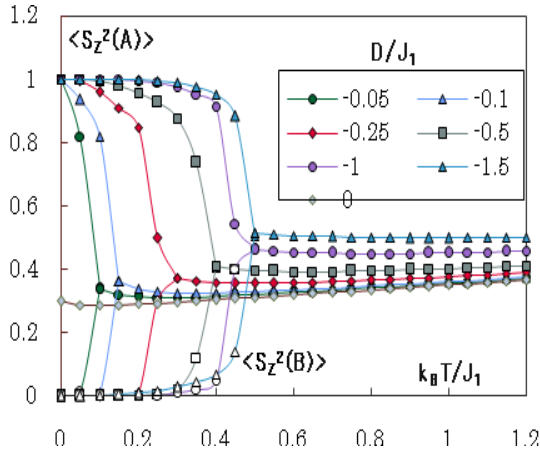
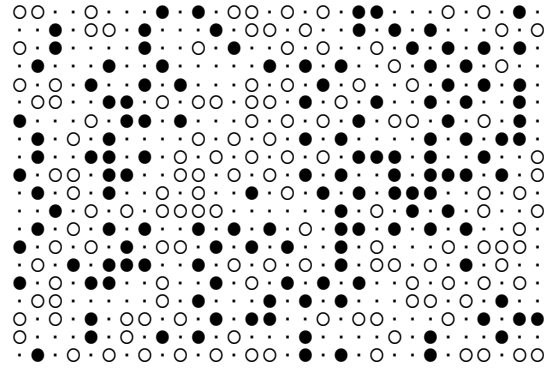


Fig. 9 Temperature dependence of $\langle S_z^2(A) \rangle$ and $\langle S_z^2(B) \rangle$ of the magnetic phase P(d) calculated by MC simulation for various values of D in the range of $0 \leq D/J_1 \leq 1.5$.

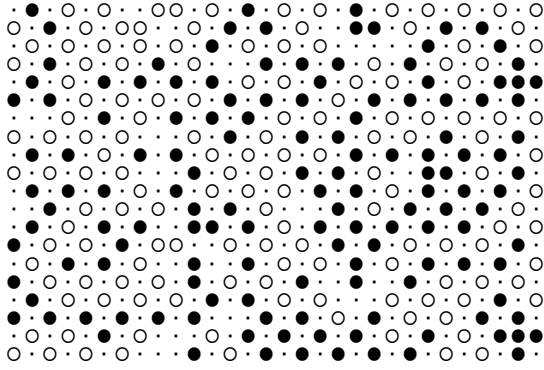
As can be seen from Fig.9, for Ising system without anisotropy ($D=0$), these thermal average $\langle S_z^2(A) \rangle$ and $\langle S_z^2(B) \rangle$ take the same value in all temperature range and have a small temperature dependence. Furthermore, these values $\langle S_z^2(A) \rangle$ and $\langle S_z^2(B) \rangle$ become 0.287 at $T=0$.

On the other hand, $\langle S_z^2(A) \rangle$ and $\langle S_z^2(B) \rangle$ turn out to take the different values at low temperatures for Ising system with anisotropy ($D \neq 0$). The values of $\langle S_z^2(A) \rangle$ and $\langle S_z^2(B) \rangle$ become 1 and 0 at $T=0$, respectively. Therefore, this spin state with anisotropy ($D \neq 0$) is confirmed to be a staggered quadrupolar (SQ) ordering state. It is worth noting that the value of $\langle S_z^2(B) \rangle$ become zero at higher temperature than the value of $\langle S_z^2(A) \rangle$ become one for anisotropy in the range of $0 < D/J_1 \leq 1.0$, and the value of $\langle S_z^2(A) \rangle$ become one at higher temperature than the value of $\langle S_z^2(B) \rangle$ become zero for anisotropy in the range of $1.0 < D/J_1$. These behaviors of $\langle S_z^2(A) \rangle$ and $\langle S_z^2(B) \rangle$ may depend the values of $\langle S_z^2(A) \rangle$ and $\langle S_z^2(B) \rangle$ at the temperature at which these values begin taking different values with decreasing temperature.

Next, let us investigate the change of a spin arrangement in this phase P(d). The temperature dependences of a spin arrangement for Ising spin system with $J_2/J_1 = -1.5$, $J_4/J_1 = -0.5$ and $D/J_1 = 1$ are shown in Fig.10. The spin arrangements at temperature of $k_B T / J_1 = 0.5$ and 0.4 are shown by (a) and (b) in this figure. The spin arrangements (a) and (b) correspond to paramagnetic state and staggered quadrupolar (SQ) state, respectively. As can be seen from Fig.10, the numbers of spins with $S_z = \pm 1$ and $S_z = 0$ are almost constant even by change of temperature, and an ordered state (SQ state) are made by changing the position of these spins with $S_z = \pm 1$ and $S_z = 0$.



(a)



(b)

Fig. 10 The spin structure of P(d) calculated by MC simulation at (a) $k_B T/J_1=0.5$ (paramagnetic state) and (b) $k_B T/J_1=0.4$ (SQ state). Open and closed circles, and dot denote $S_z=1$, $S_z=-1$ and $S_z=0$, respectively.

4. Concluding Remarks

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 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 diagrams of the ground state of Ising spin system of $S=1$ with interaction parameters J_2/J_1 and J_4/J_1 without a single-ion anisotropy D and with the one are obtained by the MC simulation. The conditions of phase transition for parameters J_2/J_1 and J_4/J_1 are symmetric against the axis of $J_4/J_1 = -1/2$. The Phase diagram with anisotropy parameter D is confirmed to be obtained by parallel translation along J_2/J_1 axis of the one without D . 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 condition of phase transitions and the GS spin structures on the $x(J_2/J_1)$ and $y(J_4/J_1)$ axes agree completely with those obtained in the previous studies [12,13]. The new phase P(c)

exists in this Ising spin system with interactions J_1 , J_2 and J_4 , and this new phase has been confirmed to appear also in the Ising spin system with interactions J_1 , J_2 and J_4 and anisotropy D . This phase PS(c) is characterized by $\langle S_z \rangle = 0.75$, $\langle S_{iz} S_{jz} \rangle = 0.5$ and $\langle S_{iz} S_{jz} S_{kz} S_{lz} \rangle = 0$.

- (3) The phase P(d) of the Ising spin system with interactions J_1 , J_2 and J_4 and anisotropy D is a SQ state, on the other hand, the one with interactions J_1 , J_2 and J_4 and without anisotropy D is not a SQ state. The speeds of spin ordering of $\langle S_z^2 \rangle$ (A) and $\langle S_z^2 \rangle$ (B) are different for the cases of $0 < D/J_1 \leq 1$ and $1 < D/J_1$.

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