Ising Spin System with Three-Site Four-Spin and Four-Site Four-Spin Interactions

by

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Abstract.

The phase diagram and magnetic properties such as the magnetization $<S_z>$, four-spin thermal average $<S_{iz}S_{jz}S_{kz}S_{lz}>$, the magnetic specific heat C_M and spin structures of spin-one (S=1) Ising spin system on two-dimensional square lattice with the bilinear exchange interaction $J_1S_{iz}S_{jz}$, the three-site four-spin interaction $J_3S_{iz}S_{jz}^2S_{kz}$, the four-site four-spin interaction $J_4S_{iz}S_{jz}S_{kz}S_{lz}$ and the single-ion anisotropy DS_{iz}^2 have been discussed by making use of the Monte Carlo simulation. In this Ising spin system, we have found new magnetic phases and determined the conditions of phase transitions and phase diagram. 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 dependences of the magnetization $<S_z>$, four-spin thermal average $<S_{iz}S_{jz}S_{kz}S_{lz}>$ and spin structures are investigated for various values of interaction parameters of J_3/J_1 and J_4/J_1.

Keywords: Ising model; four-spin interaction; phase diagram; 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 $J_2(\boldsymbol{S}_i\cdot\boldsymbol{S}_j)^2$, the three-site four-spin interaction $J_3(\boldsymbol{S}_i\cdot\boldsymbol{S}_j)(\boldsymbol{S}_j\cdot\boldsymbol{S}_k)$, the four-site four-spin interaction $J_4(\boldsymbol{S}_i\cdot\boldsymbol{S}_j)(\boldsymbol{S}_k\cdot\boldsymbol{S}_l)$ have been discussed extensively by many investigators [1-4]. 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 [4].

It was pointed out that the higher-order exchange interactions are smaller than the bilinear ones for the *3d* group ions [4], and comparable with the bilinear ones in the rare-earth compounds [5,6]. 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 [7]. Furthermore, the four-site four-spin interaction has been pointed out to be important to explain the magnetic properties of the solid helium [8,9] and the magnetic materials such as NiS_2 and C_6Eu [10].

The Ising system of S=1 with the bilinear interaction $J_1S_{iz}S_{jz}$ and the biquadratic exchange interaction $J_2S_{iz}^2S_{jz}^2$ and the single-ion anisotropy DS_{iz}^2 is quite famous as so-called Blume-Emery-Griffiths (BEG) model [1] 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 arrangements 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 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 $J_1S_{iz}S_{jz}$, the three-site four-spin

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interaction $J_3S_{iz}S_{jz}^{\ 2}S_{kz}$, the four-site four-spin interaction $J_4S_{iz}S_{jz}S_{kz}S_{lz}$ and with the single-ion anisotropy $DS_{iz}^{\ 2}$. 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. The obtained phase diagram is discussed in conjunction with the GS spin structures determined by energy evaluations. The temperature dependences of the magnetization $<S_z>$, the magnetic specific heat C_M and four spin thermal average $<S_{iz}S_{jz}S_{kz}S_{lz}>$ were also studied for various values of parameters J_2/J_1, J_4/J_1 and D/J_1.

In Section 2, the spin Hamiltonian is given for present Ising system with S=1. Furthermore, the method of the MC simulation is explained briefly. The energies per one spin of spin structures with lower energy are also obtained from this spin Hamiltonian. In Section 3, phase diagram is obtained for exchange parameters J_3/J_1 and J_4/J_1 by the MC simulation of this Ising system. In the Section 4, the magnetic properties and the spin ordering are investigated for new magnetic phases. In the last Section 5, new interesting results obtained here are summarized.

2. Spin Hamiltonian , Methods of Simulation and Energy Estimation

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

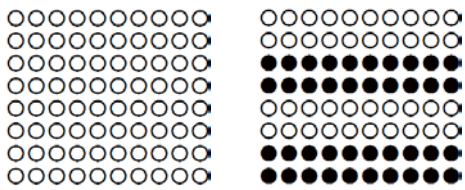
$$H = -J_1\sum_{<ij>} S_{iz}S_{jz} - 2J_3\sum_{<ijk>} S_{iz}S^2{}_{jz}S_{kz} - 2J_4\sum_{<ijkl>} S_{iz}S_{jz}S_{kz}S_{lz} - D\sum_i S^2{}_{iz} \quad (1)$$

Here, $<ij>$, $<ijk>$ and $<ijkl>$ denote the sum on the nearest neighboring spin pairs, and on the three spin sites and the square spin sites of two-dimensional square lattice. The coefficient 2 of the second and the third terms in this Hamiltonian is obtained by considering the sum of two terms $(S_{iz}\cdot S_{jz})(S_{jz}\cdot S_{kz})$ and $(S_{jz}\cdot S_{kz})(S_{iz}\cdot S_{jz})$, and $(S_{iz}\cdot S_{jz})(S_{kz}\cdot S_{lz})$ 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.

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_3, 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 $<S_z>$, the four-spin thermal average $<S_{iz}S_{jz}S_{kz}S_{lz}>$ 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 at whole temperature range.

By taking Hamiltonian (1) into consideration, the energies per one spin are obtained for various spin structures with low energy (see e.g. [14]). The GS spin structures are determined for the Ising spin system with interactions J_1, J_3 and J_4, and anisotropy term D by comparing these energies per one spin with each other. The GS spin structures with low energy obtained for the spin system of S = 1 with positive interaction J_1 are shown in Fig. 1. Let us define parameters x, y and d as J_3/J_1 , J_4/J_1 and D/J_1, respectively. The energies per one spin for the spin structures S(a) ～ S(e) of Ising spin system with S=1 are given as $E(a)=E_a/NJ_1$=-2x-12y-d-2, $E(b)=E_b/NJ_1$=-2x-d-1, $E(c)=E_c/NJ_1$=-2x+4y-d, $E(d)=E_d/NJ_1$=2x +4y-d and $E(e)=E_e/NJ_1$=2x-4y-d, respectively.



S(a) S(b)

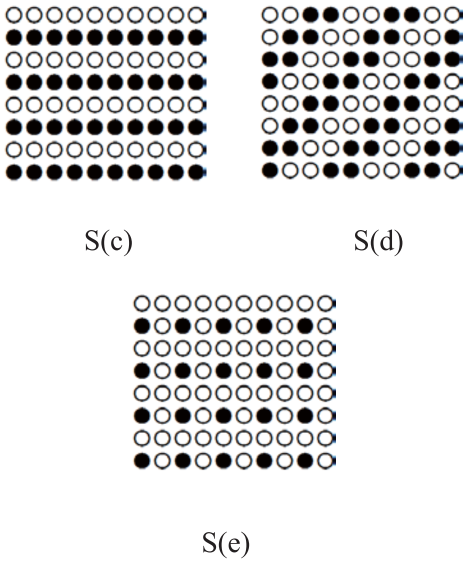


Fig. 1 The GS spin structures S(a), S(b), S(c), S(d) and S(e) for the Ising spin system of *S*=1 with interactions J_1, J_3, J_4 and anisotropy *D*. Open and closed circles denote S_Z =1 and S_Z =-1, respectively.

3. Results of Simulation and Discussion

3.1 Phase Diagram of Ising Spin System

Let us calculate magnetic properties and spin structures by making use of the MC simulation and investigate the condition of phase transitions, and determine the GS spin structures of the Ising spin system with the three-site four-spin interaction J_3 in the range of $-0.5 \leqq J_3/J_1 \leqq 0.3$ and the four-site four-spin interaction J_4 in the range of $-1.0 \leqq J_4/J_1 \leqq 0.2$. In this calculation, the interaction parameter J_1 was treated as a positive constant value. Here, we define these parameters J_4/J_1 and J_3/J_1 as x and y, respectively. The phase diagram is obtained for this Ising spin system on two-dimensional lattice without anisotropy (D = 0), and the result for both interaction parameters x and y is shown in Fig.2.

The GS spin structures of magnetic phases P(a)～P(e) determined by the MC simulation are confirmed to be the spin structures S(a) ～S(e) obtained by the energy comparison shown in Fig.1, respectively. The

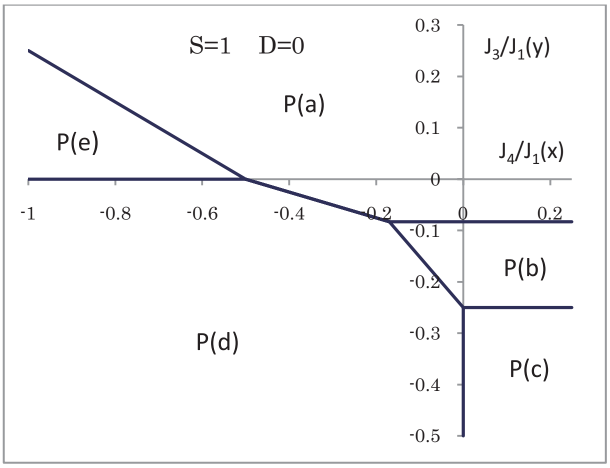


Fig. 2 Phase diagram of Ising spin system on two-dimensional square lattice with exchange parameters $x(J_4/J_1)$ in the range of $-1.0 \leqq x \leqq 0.2$ and $y(J_3/J_1)$ in the range of $-0.5 \leqq y \leqq 0.3$.

GS spin structure S(a) of phase P(a) is a ferromagnetic spin arrangement. The boundary conditions between P(a) and P(b), and between P(b) and P(c) are given by J_3/J_1 =-1/12and -1/4, respectively. As can be seen from Fig.2, the magnetic phases P(b) and P(e) disappear under the condition of $J_4/J_1 < -1/6$ and $J_4/J_1 > -1/2$, respectively.

The conditions of phase transition are obtained by comparing the energies per one spin expressed in the previous section. From the comparisons of $E(a)$ with $E(b)$, $E(d)$ and $E(e)$, the conditions of phase transition are obtained as y=-1/12, y=-x/4-1/8, y=-x/2-1/4, respectively. Furthermore, the conditions of phase transition are obtained as y=-1/4, y=-x-1/4 from the comparisons of $E(b)$ with $E(c)$, $E(d)$ and as x=0, y=0 from the comparisons of $E(d)$ with $E(c)$, $E(e)$, respectively. All these conditions calculated from energy comparison agree well with the ones obtained from the MC simulation shown in Fig.2. It is worth noting that all these conditions are not depended on the anisotropy parameter D (d).

3.2 Magnetic Properties of New Magnetic Phase P(e) on Ising Spin System

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(e) by making use of the MC simulation. The temperature dependence of $\langle S_z \rangle$, $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ and C_M in the Ising system with the fixed interaction J_4 (J_4/J_1=-0.8) and various values of interaction J_3 are shown in Fig.3, Fig.4 and Fig.5, respectively. These calculations by the MC simulation are performed on the Ising spin system with interaction J_3 in the range of $0 \leqq J_3/J_1 \leqq 0.2$ for $\langle S_z \rangle$ and $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$, and in the range of $0.06 \leqq J_3/J_1 \leqq 0.14$ for C_M.

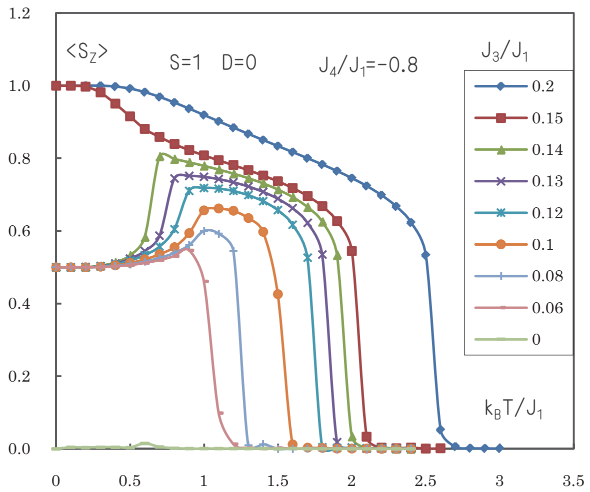


Fig. 3 Temperature dependence of $\langle S_z \rangle$ of the magnetic phase P(e) calculated by the MC simulation for fixed value of J_4 (J_4/J_1=-0.8) and various values J_3 of in the range of $0 \leqq J_3/J_1 \leqq 0.2$

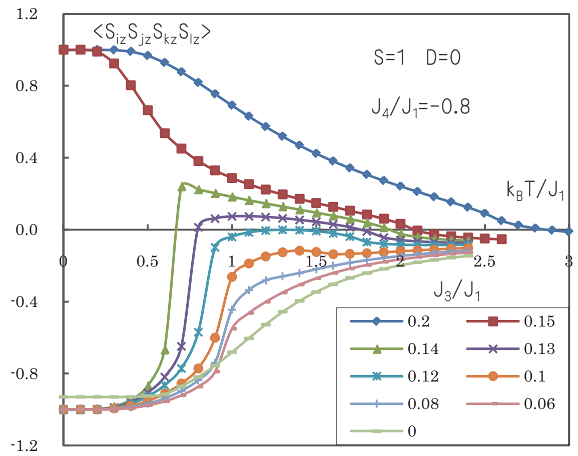


Fig. 4 Temperature dependence of $\langle S_{iz}\ S_{jz}\ S_{kz}\ S_{lz} \rangle$ of the magnetic phase P(e) calculated by the MC simulation for fixed value of J_4 (J_4/J_1=-0.8) and various values J_3 of in the range of $0 \leqq J_3/J_1 \leqq 0.2$

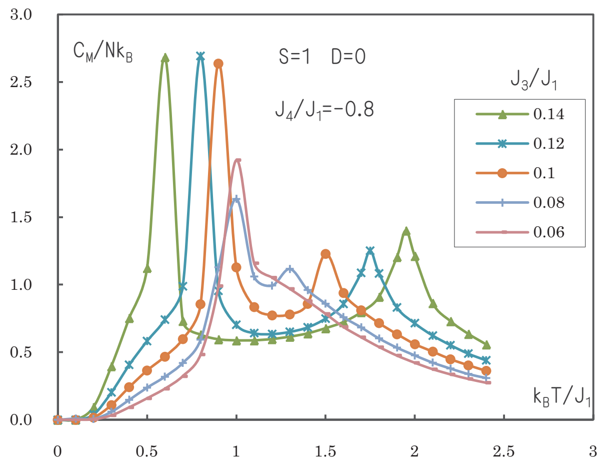


Fig. 5 Temperature dependence of C_M of the magnetic phase P(e) calculated by the MC simulation for fixed value of J_4 (J_4/J_1=-0.8) and various values J_3 of in the range of 0.6 $\leqq J_3/J_1 \leqq$0.14

It is noticeable that the ferromagnetic order $\langle S_z \rangle$ constructed at temperatures below T_c decreases abruptly in the middle temperature range for interaction J_3 in the range of $0.08 \leqq J_3/J_1 \leqq 0.14$. As can be seen from Fig.3, this abrupt change of $\langle S_z \rangle$ is large for J_3 such as J_3/J_1=0.12 ～0.14 just below the condition of phase transition(J_3/J_1=0.15). From the behavior of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ shown in Fig.4, the spin order of S(e) turns out to begin forming below this middle temperature at which abrupt decrease of $\langle S_z \rangle$ occurs.

It is interesting that $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ for J_3/J_1=0.12 ～ 0.14 takes positive value in the higher temperature range than the abrupt decrease of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$. From Fig.5, double peaks of C_M turn out to appear for interaction J_3 in the range of $0.08 \leqq J_3/J_1 \leqq 0.14$. It is worth noting that the peaks of lower temperature are large than those of higher temperature for each interaction parameter J_3, respectively. This fact suggests that the large change of spin structures occurs for the formation of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$. Therefore, let us investigate the change of spin ordering with decreasing temperature by the MC simulation. The spin structures on the Ising system with the interactions J_3/J_1=0.14 and J_4/J_1=-0.8 are shown in Fig.6 for various temperatures. It is confirmed that the spin structure of S(e) with these interactions J_3 and J_4 begin forming from ferromagnetic order under condition of $k_BT/J_1 \leqq 0.6$.

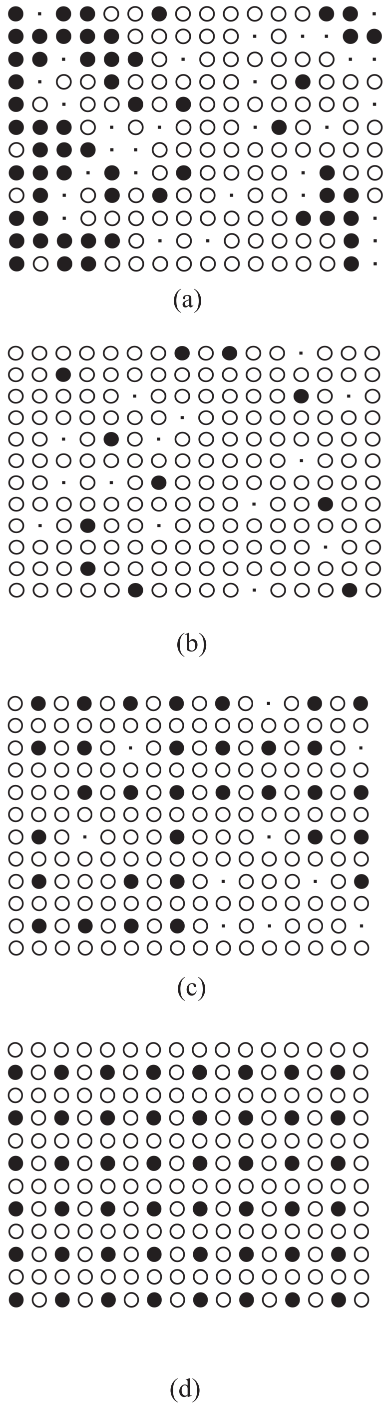


Fig.6 The formation of spin structure S(e) calculated by the MC simulation for the interactions J_3/J_1=0.14 and J_4/J_1=-0.8 at (a) k_BT/J_1=2.2, (b) k_BT/J_1=1.0, (c) k_BT/J_1=0.6 and (d) k_BT/J_1=0.1. Open and closed circles, and dot denote S_Z =1, S_Z =-1 and S_Z =0, respectively.

3.3 Magnetic Properties of Magnetic Phases P(b), P(c) and P(d) on Ising Spin System

Next, let us investigate the magnetic ordering of the magnetic phases P(b), P(c) and P(d) without non-zero magnetization ($<S_z>=0$) as GS spin structure. The magnetic specific heat C_M and the four-spin thermal average $<S_{iz}S_{jz}S_{kz}S_{lz}>$ of these magnetic phases have been calculated by making use of the MC simulation. The calculated results of C_M and $<S_{iz}S_{jz}S_{kz}S_{lz}>$ of the magnetic phases P(b) and P(d) with the fixed interaction J_3/J_1=-0.15 and various values of the interaction J_4 in the range $-0.4 \leqq J_4/J_1 \leqq 0.2$ are shown in Fig.7 and Fig.8, respectively. It is confirmed that the phase change between P(b) and P(d) occurs at the condition of J_3/J_1=-0.15 and J_4/J_1=-0.1 from behaviors of C_M and $<S_{iz}S_{jz}S_{kz}S_{lz}>$. The positions of peaks of C_M for magnetic phases P(b) and P(d) turn out to be almost symmetric against the condition of phase transition (J_3/J_1=-0.15 and J_4/J_1=-0.1). The peaks of C_M on the phase P(d) are, however, higher than those on the phase (b). It is remarkable that $<S_{iz}S_{jz}S_{kz}S_{lz}>$ takes positive value at the condition of the phase transition (J_3/J_1=-0.15 and J_4/J_1=-0.1). From Fig.8, the change of the temperature dependence of $<S_{iz}S_{jz}S_{kz}S_{lz}>$ on the phase P(b) turns out to be larger than that on the phase P(d). This behavior of $<S_{iz}S_{jz}S_{kz}S_{lz}>$ is consistent with that of C_M.

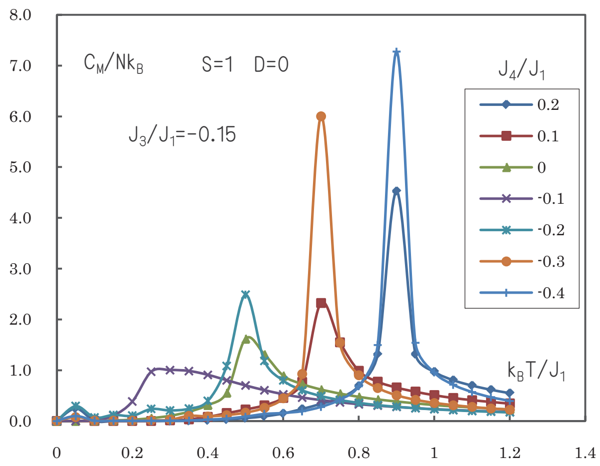


Fig. 7 Temperature dependence of C_M of the magnetic phases P(b) and P(d) calculated by the MC simulation for fixed value of J_3 (J_3/J_1=-0.15) and various values J_4 of in the range of $-0.4 \leqq J_4/J_1 \leqq 0.2$

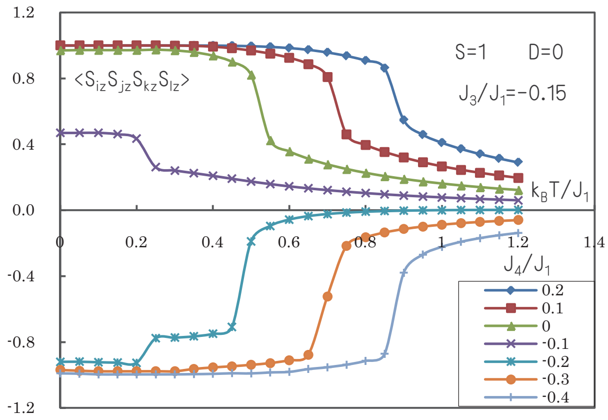
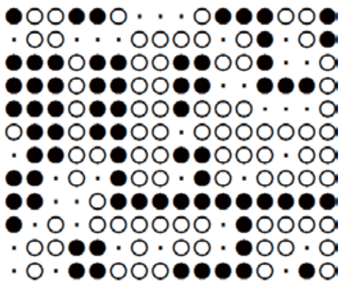
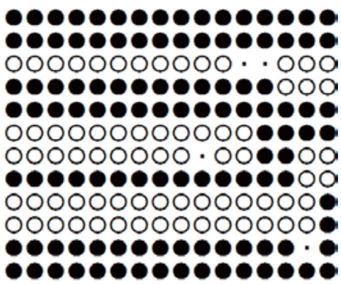


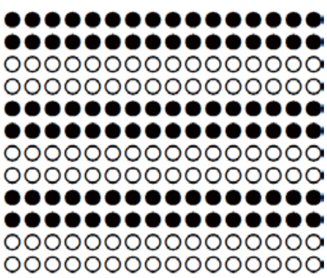
Fig. 8 Temperature dependence of <S_{iz} S_{jz} S_{kz} S_{lz}> of the magnetic phase P(b) and P(d) calculated by the MC simulation for fixed value of J_3 (J_3/J_1=-0.15) and various values J_4 of in the range of -0.4≦J_4/J_1≦0.2



(a)

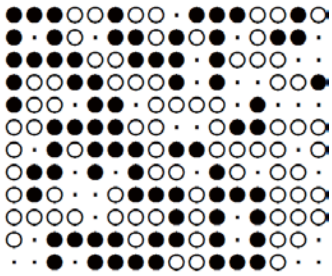


(b)

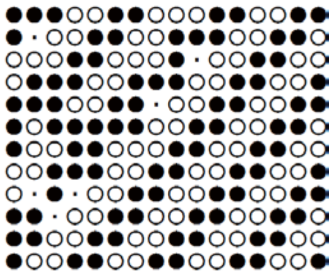


(c)

Fig. 9 The formation of spin structure S(b) calculated by the MC simulation for the interactions J_3/J_1=-0.15 and J_3/J_1=-0.1 at (a) k_BT/J_1=1.0, (b) k_BT/J_1=0.5, (c) k_BT/J_1=0.1. Open and closed circles, and dot denote S_Z =1, S_Z =-1 and S_Z =0, respectively.



(a)



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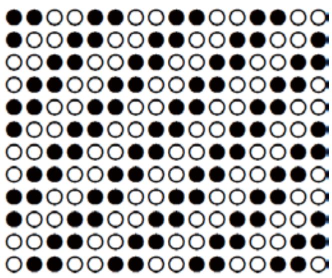


(c)

Fig.10 The formation of spin structure S(d) calculated by the MC simulation for the interactions J_3/J_1=-0.15 and J_3/J_1=-0.3 at (a) k_BT/J_1=1.0, (b) k_BT/J_1=0.5, (c) k_BT/J_1=0.1. Open and closed circles, and dot denote S_Z =1, S_Z =-1 and S_Z =0, respectively.

The formation of the GS spin structure S(b) has investigated for the spin system with interactions J_3/J_1=-0.15 and J_3/J_1=-0.1. The spin structures at k_BT/J_1=1.0, 0.5 and 0.1 are shown in Fig.9. Furthermore, the formation of the GS spin structure S(d) has investigated for the spin system with interactions J_3/J_1=-0.15 and J_3/J_1=-0.3. The spin structures at k_BT/J_1=1.0, 0.5 and 0.1 are shown in Fig.10. Both GS spin structures S(b) and S(d) turns out to be almost random at the temperature of k_BT/J_1= 1.0. They are, however, formed over large area at the temperature of k_BT/J_1= 0.5.

Furthermore, the calculated results of C_M and $<S_{iz}S_{jz}S_{kz}S_{lz}>$ of the magnetic phases P(c) and P(d) with the fixed interaction J_3/J_1=-0.4 and the various values of interaction J_4 in the range $-0.2 \leqq J_4/J_1 \leqq 0.2$ are shown in Fig.11 and Fig.12, respectively. It is confirmed that the phase change between P(c) and P(d) occurs at J_3/J_1=-0.4 and J_4/J_1=0 from behaviors of C_M and $<S_{iz}S_{jz}S_{kz}S_{lz}>$. The magnetic ordering of P(d) turns out to start at higher temperature than that of P(c) from the behaviors of the temperature dependence of C_M and $<S_{iz}S_{jz}S_{kz}S_{lz}>$. The C_M on the phase P(d) has higher peak than that on the phase P(c) as shown in Fig.11, and this result reflects the abrupt larger change of $<S_{iz}S_{jz}S_{kz}S_{lz}>$ on the phase P(d) than on the phase P(c) as shown in Fig.12.

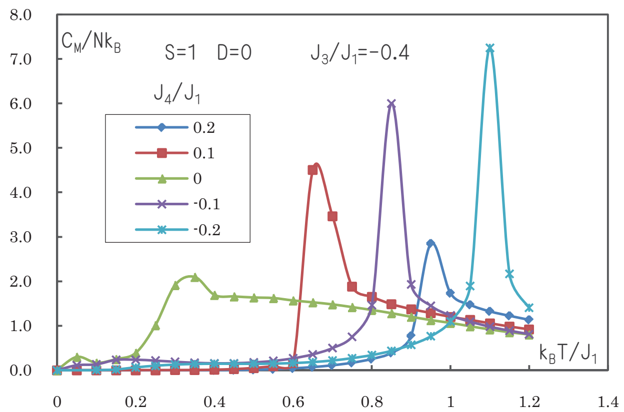


Fig. 11 Temperature dependence of C_M of the magnetic phases P(c) and P(d) calculated by the MC simulation for fixed value of J_3 (J_3/J_1=-0.4) and various values J_4 of in the range of $-0.2 \leqq J_4/J_1 \leqq 0.2$

The formation of the GS spin structure S(c) has investigated for the spin system with interactions J_3/J_1=-0.4 and J_4/J_1=0.2. The spin structures at k_BT/J_1=1.2, 1.0, 0.8 and 0.1 are shown in Fig.13. Furthermore, the formation of the GS spin structure S(d) has investigated for the spin system with interactions J_3/J_1=-0.4 and J_4/J_1=-0.2. The spin structures at k_BT/J_1=1.2, 1.0, 0.8 and 0.1 are shown in Fig.14. Both GS spin structures S(b) and S(d) turns out to be almost random at the temperature of k_BT/J_1= 1.2. The spin structures S(c) and S(d) are, however, start forming at the temperature of k_BT/J_1= 0.8 and 1.0, respectively.

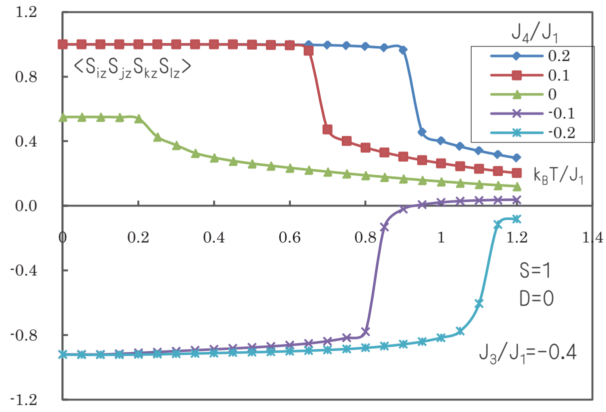
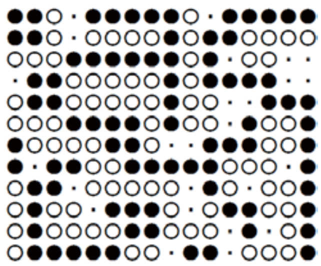
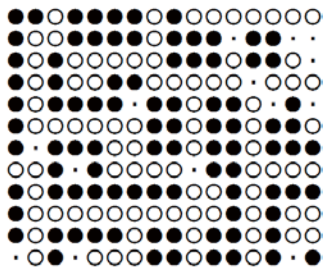


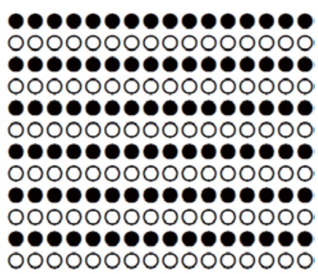
Fig. 12 Temperature dependence of $<S_{iz}\ S_{jz}\ S_{kz}\ S_{lz}>$ of the magnetic phase P(c) and P(d) calculated by the MC simulation for fixed value of J_3 (J_3/J_1=-0.4) and various values J_4 of in the range of $-0.2 \leqq J_4/J_1 \leqq 0.2$



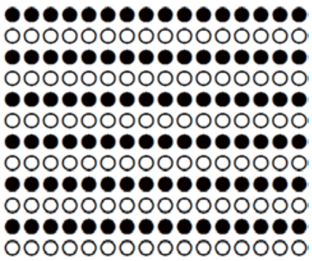
(a)



(b)

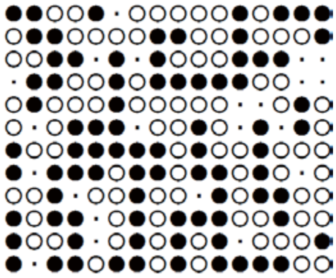


(c)

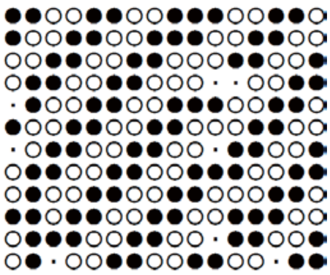


(d)

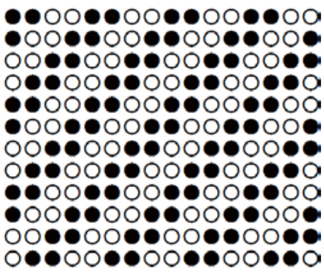
Fig. 13 The formation of spin structure of S(c) calculated by the MC simulation at (a) k_BT/J_1=1.2, (b) k_BT/J_1=1.0, (c) k_BT/J_1=0.8, (d) k_BT/J_1=0.1. Open and closed circles, and dot denote S_Z =1, S_Z =-1 and S_Z =0, respectively.



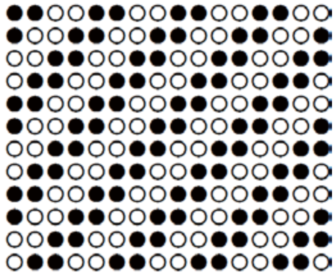
(a)



(b)



(c)

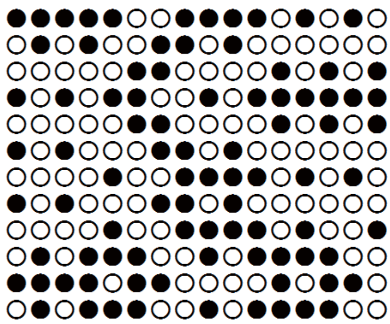


(d)

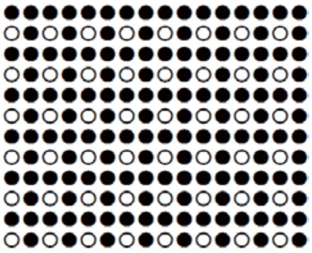
Fig. 14 The formation of spin structure of S(d) k_BT/J_1=1.0, (c) k_BT/J_1=0.8, (d) k_BT/J_1=0.1 Open and closed circles, and dot denote S_Z =1, S_Z =-1 and S_Z =0, respectively.

3.4 GS Spin Structures on *X*-axis (J_3=0) and *Y*-axis (J_4=0)

We have investigated the GS spin structure of the phase boundary (*x*-axis) between P(d) and P(e) by the MC simulation. This phase boundary between P(d) and P(e) appears on the *x*-axis under the condition of J_4/J_1< -1/2. The GS spin structure on this boundary is shown by (a) in Fig.11. This GS spin structure is constructed by the mixture of S(d) and S(e) in Fig.1, and S(f) shown by (b) in Fig.11 which is the spin structure with the reversed spin of S_z= ± 1 in S(e). These results agree with those obtained by the previous study [15].



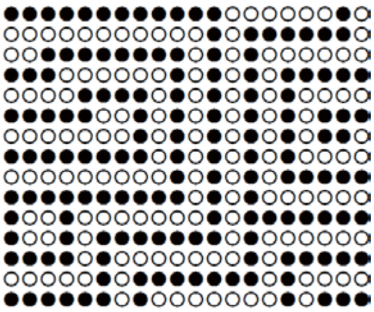
(a)



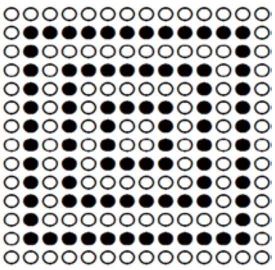
(b)

Fig. 15 (a) the GS spin structures on the boundary (x-axis) and (b) the GS spin structure S(f) with reversed spin of Sz= ±1 of the structure S(e). Open and closed circles denote S_Z =1and S_Z =-1, respectively.

Next, we have investigated the GS spin structure of the phase boundary (y-axis) between P(c) and P(d) by the MC simulation. This phase boundary between P(c) and P(d) appears on the y-axis under the condition of J_3/J_1< -1/4. The GS spin structure on this boundary is shown by (a) in Fig.12. This GS spin structure is constructed by the mixture of S(c) in Fig.1,and S(g) shown by (b) in Fig.12 which is the same energy with S(c) in the case of J_4=0. These results agree with those obtained by the previous study [13]. It is remarkable that this GS spin structure on the boundary between P(c) and P(d) is not the mixture of S(c) and S(d).



(a)



(b)

Fig. 16 (a) GS spin structures on the boundary (y-axis) and (b) the GS spin structure S(g) in the case of J_4=0. Open and closed circles denote S_Z =1and S_Z =-1, respectively.

4. Concluding Remarks

In the previous section, for the Ising spin system of S=1 with the bilinear exchange interaction $J_1S_{iz}S_{jz}$, the three-site four-spin interaction $J_3S_{iz}S_{jz}^2S_{kz}$, the four-site four-spin interaction $J_4S_{iz}S_{jz}S_{kz}S_{lz}$ and a single-ion anisotropy D, the magnetization $<S_z>$, the four-spin thermal average $<S_{iz}S_{jz}S_{kz}S_{lz}>$, 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 two-dimensional square lattice, we may conclude as follows:

(1) The phase diagram of the ground state of the Ising spin system of S=1 with interaction parameters J_3/J_1 and J_4/J_1 without a single-ion anisotropy D are obtained by the MC simulation. This phase diagram does not depend on the anisotropy 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 magnetic phases P(d) and P(e) with new GS spin structures S(d) and S(e) are found in the negative range of interaction J_4. The temperature dependences of $<S_z>$ and $<S_{iz}S_{jz}S_{kz}S_{lz}>$ show interesting abrupt change in the process of construction of the GS spin structure S(e).
(3) The behaviors of the temperature dependence of C_M and $<S_{iz}S_{jz}S_{kz}S_{lz}>$ may suggest that the spin ordering for spin structure S(d) is more rapid than those for spin structures S(b) and S(c).
(4) The GS spin structure on the x-axis (J_3=0) in the range of J_4/J_1 <-1/2 is constructed by the mixture of S(d) and S(e). On the other hand, the GS spin structure on the y-axis (J_4=0) in the range of J_3/J_1 <-1/4 is constructed by the mixture of S(c) and S(g). Therefore, this GS spin structure has no relation with structure S(d).

Refernces

1. M.Blume, V.J. Emerry and Robert B. Griffiths,*Phys. Rev.* A4, *1071* (1971).
2. H. A. Brown, *Phys. Rev. B* **4**, 115 (1971).
3. J. Adler and J. Oitmaa, *J. Phys. C* **12**, 575 (1979).

4. T. Iwashita and N. Uryu, *J. Phys. C* **17**, 855 (1984).
5. H.H.Chen and P.M.Levy, *J. Phys. Rev*. **7***, 4267* (1973).
6. J.M.Baker, *JRep.Prog. Phys*. 34*, 109* (1971).
7. M. Roger, J. M. Delrieu and J. H. Hetherington, *Phys. Rev. Lett*. **45**, 137 (1980).
8. A.K.McMahan and R.A.Guyer, *Phys. Rev.* **A7***, 1105* (1973).
9. J.H.Hetherington and F.D.C.Willard, *Phys. Rev*. Lett. **35***, 1442*(1975).
10. M.Roger, J.M.Delrieu and A. Landesman, *Phys. Lett.* **62A***, 449* (1977).
11. T. Iwashita, R. Satou, T. Imada and T. Idogaki, *Physica B* **284**, 1203 (2000).
12. T. Iwashita, K. Uragami, K. Goto, K.Kasama and T. Idogaki, *J.Magn.Magn.Mater.* **272**,672 (2004).
13. T. Iwashita, K. Uragami, A.Shimizu, A.Nagaki, T.Kasama and T. Idogaki, *J.Magn.Magn.Mater.* **310**,435 (2007).
14. T. Iwashita and N. Uryu, *J. Phys*. Condens. Matter **3**, 8257 (1991).
15. T. Iwashita, K. Uragami, K. Goto, A.Shimizu, K.Kasama and T. Idogaki, American Institute of Physics **850**,1077 (2006).