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Magnetic Properties and Spin Structures of Ising Spin System with Two-Spin and Four-Spin Interaction

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Abstract

The magnetic properties of the spin S (=3/2 and 2) Ising systems with the bilinear exchange interaction $J_1S_{iz}S_{jz}$ and the four-site four-spin interaction $J_4S_{iz}S_{jz}S_{kz}S_{lz}$ are discussed by making use of the Monte Carlo (MC) simulation for the magnetization $<S_z>$, the magnetic specific heat C_M and spin structures. The ferromagnetic spin arrangement ordered by the positive interaction J_1 is destroyed and changed into lots of complicated spin structures with low energy by introducing negative four-spin interaction J_4. Near the phase transitions of J_4/J_1=-2/9 and -1/8 for S=3/2 and 2, respectively, the thermal averages $<S_z>$, $<S_{iz}S_{jz}S_{kz}S_{lz}>$ and the specific heat C_M show characteristic temperature dependences in the low temperature region. The change of spin structures of the system with the variation of the values of J_1 and J_4 are also investigated by using the result of the MC simulation and by the comparison of energies of the possible ground state spin structures.

Keywords: Ising model; Four-site four-spin interaction; Monte Carlo simulation

1. Introduction

In Heisenberg and Ising ferromagnets, the existence and the importance of such higher-order exchange interactions as J_2 $(\boldsymbol{S}_i\cdot\boldsymbol{S}_j)^2$, J_3 $(\boldsymbol{S}_i\cdot\boldsymbol{S}_j)(\boldsymbol{S}_j\cdot\boldsymbol{S}_k)$, J_4 $(\boldsymbol{S}_i\cdot\boldsymbol{S}_j)(\boldsymbol{S}_k\cdot\boldsymbol{S}_l)$ are discussed extensively by many investigators [1-2]. 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].

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 [4]. In a spin system with a bilinear exchange interaction $J_1(\boldsymbol{S}_i\cdot\boldsymbol{S}_j)$ and a four-site four-spin interaction $J_4(\boldsymbol{S}_i\cdot\boldsymbol{S}_j)(\boldsymbol{S}_j\cdot\boldsymbol{S}_k)$, the interaction J_4 is expected to have significant effects on magnetic properties in the low-temperature region for the case of J_4 not negligible compared to J_1/S^2[5,6]. Furthermore, there exist many complicated spin structures with zero and non-zero magnetizations as the ground state (GS) in the Ising spin system only with negative interaction J_4.

In the previous studies, the magnetic properties of spin systems with S=1/2 and 1 with both interactions J_1

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and J_4 were investigated [5,6]. We cannot, however, discuss sufficiently the changes of magnetic properties and spin structures. Therefore, we have developed this calculation to the spin systems with S=3/2 and 2 and investigated more precisely the dependences on the temperature T and interaction parameter J_4 / J_1 of the magnetic properties and spin structures.

In the present study, the effects of the four-site four-spin interaction $J_4S_{iz}S_{jz}S_{kz}S_{lz}$ on the magnetization $<S_z>$, the magnetic specific heat C_M and the thermal average $<S_{iz}S_{jz}S_{kz}S_{lz}>$ of Ising spin systems of S=3/2 and 2 on a two-dimensional square lattice are investigated by making use of the Monte Carlo (MC) simulation. The temperature dependence of spin structures is also studied for various values of interaction parameter (J_4/J_1). The obtained characteristic behaviors of $<S_z>$, C_M and $<S_{iz}S_{jz}S_{kz}S_{lz}>$ are discussed in conjunction with the ground state (GS) spin structures determined by energy evaluations.

2. Ground State Spin Structure and Methods of MC Simulation

The spin Hamiltonian for the present Ising spin system can be written as follows:

$$H = -J_1 \sum_{<ij>} S_{iz} S_{jz} - 2J_4 \sum_{<ijkl>} S_{iz} S_{jz} S_{kz} S_{lz} \quad (1)$$

Here, $<ijkl>$ denotes the sum on the square spin sites. 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 ($S_{iz}\cdot S_{lz}$)($S_{jz}\cdot S_{kz}$).

The GS spin structures are determined by comparing the energies of various spin structures with each other (see e.g. [7]). Some GS spin structures with S_z =S and S_z =-S (S=3/2, 2) for the spin system only with positive J_4 and only with negative J_4 are shown by (a) and (b)～（f） in Fig.1, respectively. Non-zero magnetization ($<S_z>\neq 0$) and zero magnetization ($<S_z>$=0) are obtained for the spin structures (a)～(d) and (e), (f) , respectively.

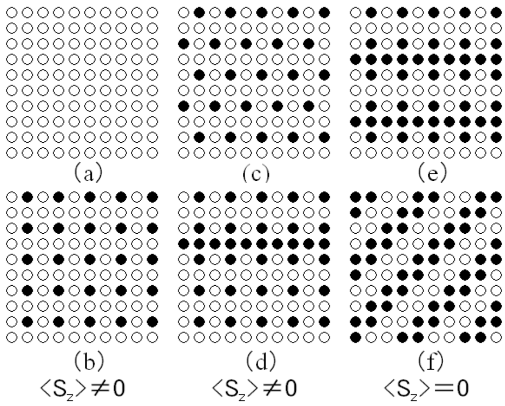


Fig. 1. The GS spin structures with $<S_Z>\neq 0$ and $<S_Z>$=0. Open and closed circles denote S_Z =S and S_Z =-S (S=3/2, 2), respectively.

For spin system with S=3/2, the energies per one spin are given as E_a(3/2)=-9/2J_1-81/8J_4 and E_b(3/2) ～ E_f(3/2)= 81/8J_4. Therefore, by comparing these energy, phase change turns out to occur at the condition of J_4/J_1 =-2/9. Furthermore, the spin structures (b)～（f）may be the GS spin structure in the range of J_4 / J_1< -2/9.

On the other hand, the energies per one spin are given as E_a(2)=-8J_1-32J_4 and E_b(2) ～ E_f(2)=32J_4 for spin system with S=2. Therefore, phase change turns out to occur at the condition of J_4 / J_1 =-1/8. For the interaction J_4 in the range of J_4 / J_1< -1/8, the spin structures (b)～（f）may be the GS spin structure.

MC simulations based on the Metropolis method are carried out assuming a periodic boundary condition for a two-dimensional square lattice with linear lattice size L=240. For fixed values of J_4/J_1, we start the simulation at high temperatures adopting random initial configurations, and advance gradually this simulation to lower temperature. We use the last spin configuration as an input for the calculation at the next point. Thermal averages of $<S_z>$ are calculated using 2×10^5 MC steps per spin (MCS/s) after discarding the first 3×10^5 MCS/s. The values of $<S_z>$, C_M and $<S_{iz}S_{jz}S_{kz}S_{lz}>$ and spin structures are calculated for the spin system both with fixed positive interaction J_1 and various negative interaction J_4.

3. Results of Calculation and Discussions

(A) Effect of the Negative Interaction J_4 on the Spin System with Positive Interaction J_1

The temperature dependence of $<S_z>$ have been calculated for the spin system both with interactions J_1 and J_4 (J_1>0, J_4<0), and the results of $<S_z>/S$ for the spin systems of S=3/2 and 2 are shown by (a) and (b) in Fig.2, respectively. Judging from the behaviors at low temperatures, the phase transitions are pointed out to occur at the conditions of J_4/J_1 =-2/9 for S= 3/2 and J_4/J_1 =-1/8 for S=2. These conditions agree quite well with those obtained by above mentioned energy comparisons.

Under the conditions of phase transition, the values of $<S_z>/S$ at low temperature are approximately 0.77 for both spin systems with S=3/2 and 2. The spin arrangement at various temperatures is shown in Fig.3 for spin system with S=2. At the condition of phase transition, the spin structures like the ones (b) and (c) may appear mainly at low temperatures because these spin structures can change easily from ferromagent-like order shown by (d) in this figure right after the appearance of non–zero magnetization with decreasing temperature.

Let us consider many spin structures in which closed circle in the spin structure (b) or (c) of Fig.1 represent the spin with S_z =±3/2, ±1/2 for S=3/2 and with S_z = ±2, ±1, 0 for S=2. The spin structures with S_z = ±1/2 for spin system of S=3/2 are shown by (g) and (h) in Fig.4, respectively, and the energies per one spin are given as $E_g(3/2)=-3J_1-27J_4/8$ and $E_h(3/2)=-3J_1/2+27J_4/8$. The J_4/J_1 -dependence of these energies are compared with $E_a(3/2)$ and $E_b(3/2)$～$E_f(3/2)$ in Fig.5. As can be seen from this figure, all these spin structures (a) ～(h) have the same energies at the condition of phase transition (J_4/J_1 =-2/9).

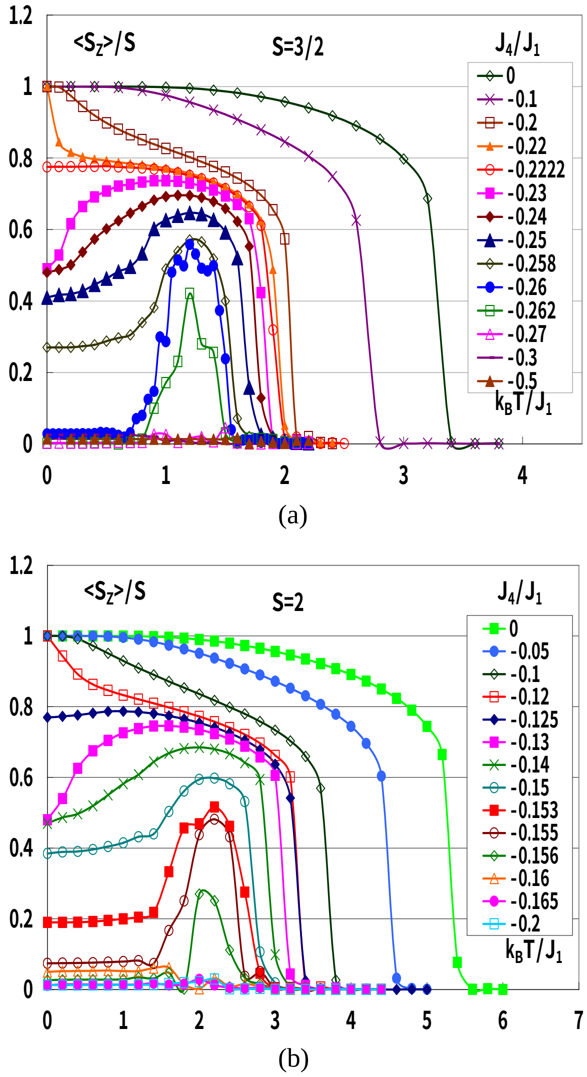
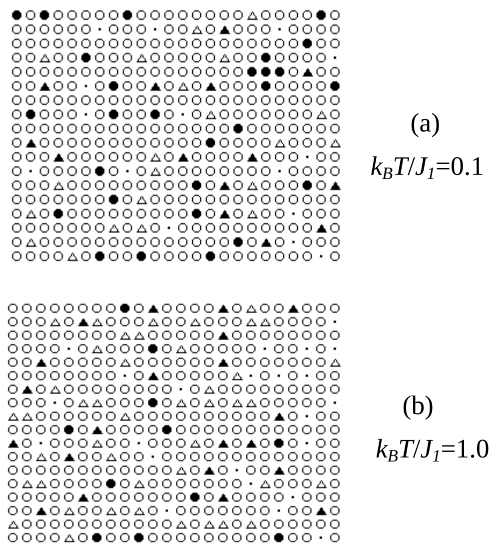


Fig. 2. Temperature dependence of $<S_z>/S$ for spin systems of (a) S=3/2 and (b) S=2 with both interactions of fixed positive J_1 and various negative values of J_4.



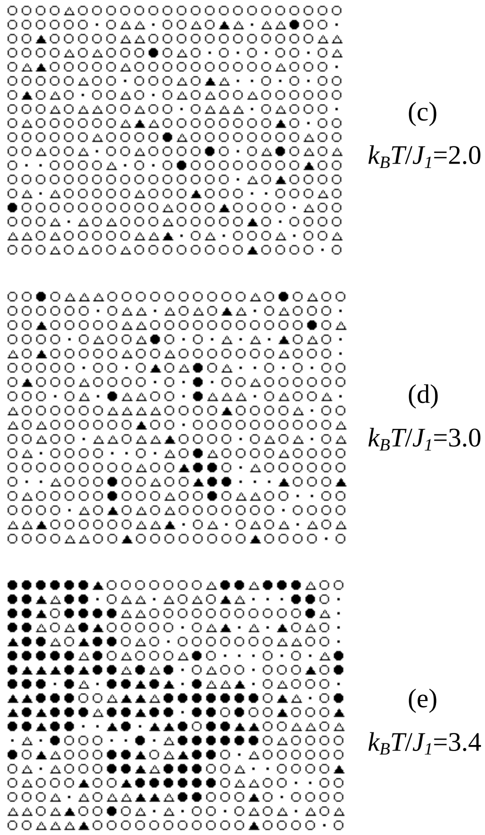


Fig. 3. The spin arrangement at the temperature of (a) k_BT/J_1=0.1, (b) k_BT/J_1=1, (c) k_BT/J_1=2, (d) k_BT/J_1=3, (e) k_BT/J_1=3.4 for the spin system of S=2 with J_4/J_1=-1/8. Open and closed circles denote S_Z =2 and S_Z =-2, respectively. Open and closed triangles, and dot denote S_Z =1 and S_Z =-1,and S_Z =0, respectively.

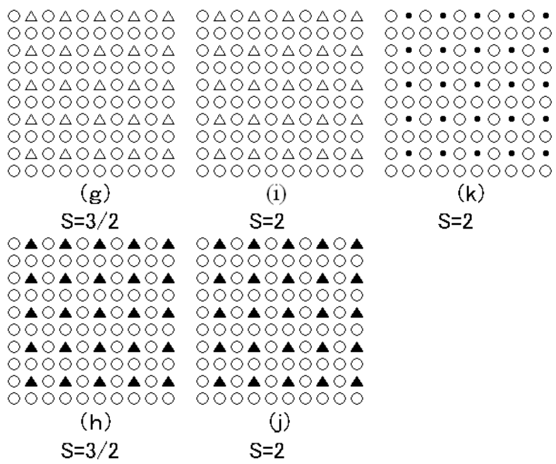


Fig. 4. The GS spin structures with at J_4/J_1=-2/9,-1/8 for the spin system of S=3/2,2, respectively. Open and closed circles, and open and closed triangles denote S_Z =3/2,-3/2,1/2,-1/2 for S=3/2 and S_Z =2,-2,1,-1 for S=2, respectively. Dot denotes S_Z =0 for S=2.

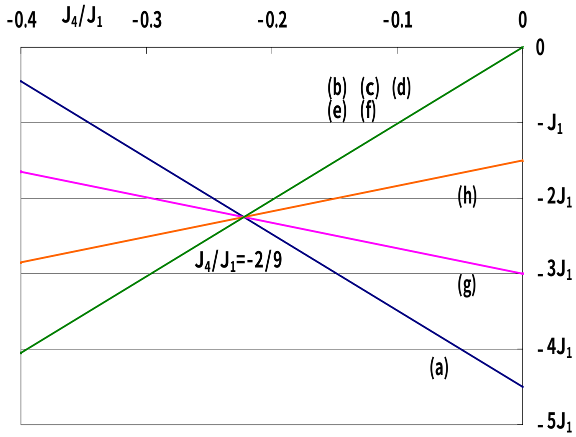


Fig. 5. The temperature dependence of energies per one spin for various spin structures of S=3/2 with low energy.

On the other hand, the spin structures with S_z = ±1 and 0 for spin system of S=2 are shown by (i), (j) and (k) in Fig.4, respectively, and the energies per one spin are given as $E_i(2)$=-6J_1-16J_4, $E_j(2)$=-2J_1+16J_4 and $E_k(2)$=-4J_1/2. The J_4/ J_1 -dependence of these energies are compared with $E_a(2)$ and $E_b(2)$～$E_f(2)$ in Fig.6. We can confirm that all these spin structures (a) ～(f) and (i) ～ (k) have the same energies at the condition of phase transition (J_4 /J_1 =-1/8).

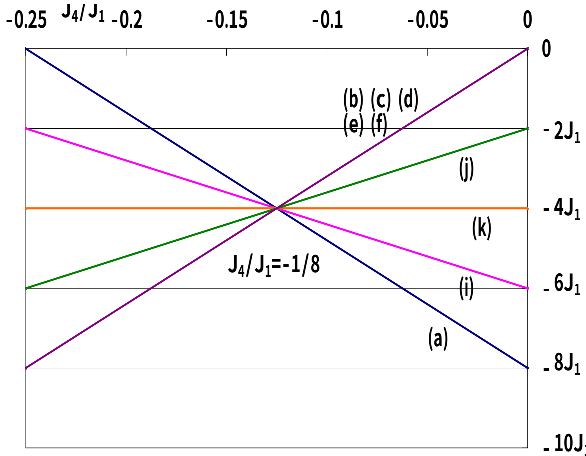


Fig. 6. The temperature dependence of energies per one spin for various spin structures of S=2 with low energy.

The values of <S_z>/S are estimated as 1.0, 0.5, 0.833, 0.667 for spin structures (a), (b), (g), (h) in the spin system with S=3/2, respectively. On the other hand, in the spin system with S=2, the values of <S_z>/S are obtained as 1.0, 0.5, 0.875, 0.625, 0.75 for spin structures (a), (b), (i), (j), (k), respectively. We assume that all these spin structures (a),(b),(g) ～(k) appear at the same probability for each spin system. Then, the average value of $<S_z>/S$ becomes 0.75 for both spin systems with S=3/2 and with S=2. This estimation may agree roughly with the one (0.77) obtained for the spin arrangement in Fig.3.

The temperature dependence of the magnetic specific heat C_M were also calculated for the spin system with fixed positive interaction J_1 and various negative interaction J_4, and the results for S=2 are shown in Fig. 7. As can be seen from this figure, specific heat curves shows complicated behaviors in the interaction range of -0.16< J_4 / J_1<-0.125. These behaviors suggest the large

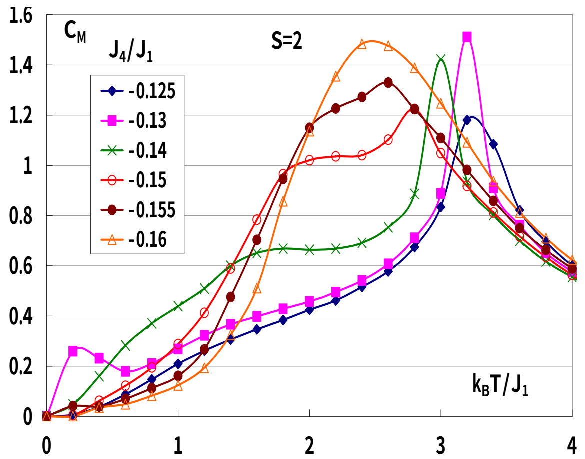
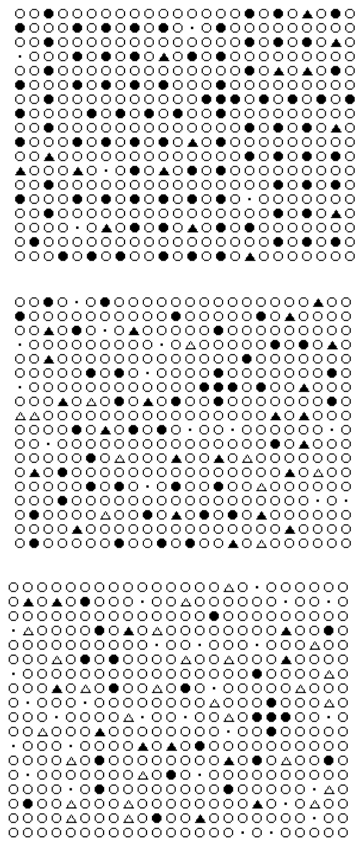


Fig. 7. The temperature dependence of C_M of spin system with S=2 for both interactions of fixed positive J_1 and various negative values of J_4.

change of spin arrangement in this interaction range. The behaviors of temperature dependence of $<S_z>/S$ for spin system with S=2 shown by (b) in Fig.2 consistent well with above suggestion obtained from C_M curve.

It is quite remarkable that in the interaction ranges of -0.155< J_4 / J_1< -1/8 for S=2 and -0.26< J_4 / J_1< -2/9 for S=3/2, the values of $<S_z>/S$ decrease with decreasing temperature, and take finite values at low temperature, even at T=0. The temperature dependence of spin arrangements for spin system of S=2 with J_4 / J_1 =-0.13 is shown in Fig.5. At high temperature like k_BT/J_1=0.6, the numbers of spins with S_z =±1,0 are considerably large. These numbers, however, decrease with decreasing temperature, and in inverse, the spin S_z =-2 increases and begins to make the spin arrangement like the one shown by (b) or (c) in Fig.1. By the speculation from the spin arrangement at k_BT/J_1=0.2, the spin structure mixed with the ones (b) and (c) in Fig.1 may appear as the GS spin structure at T=0. Therefore, these changes of spin structure lead the decrease of $<S_z>/S$ with decreasing temperature.

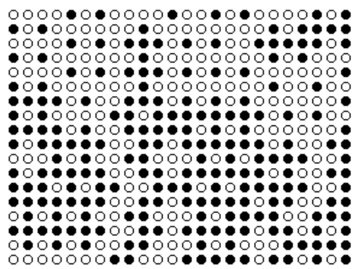


(a) k_BT/J_1=0.2

(b) k_BT/J_1=0.6

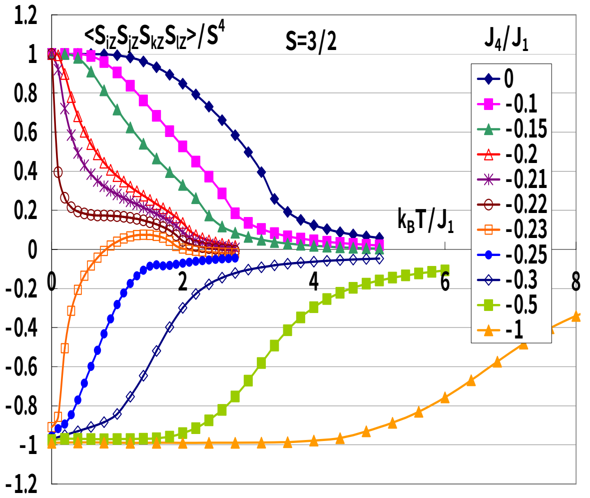
(c) k_BT/J_1=1.2

Fig. 8. The spin arrangements at (a) k_BT/J_1=0.2 and (b) k_BT/J_1=0.6 for the spin system of S=2 with J_4 /J_1=-0.13.

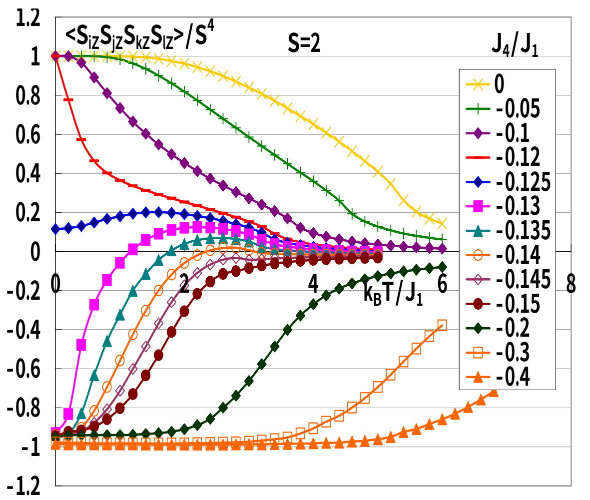


k_BT/J_1=0.2

Fig. 9. The spin arrangement at k_BT/J_1=0.2 for the spin system of S=2 with J_4 /J_1=-0.155.



(a)



(b)

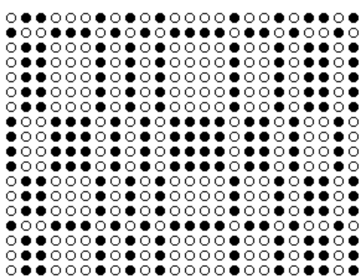
Fig. 10. Temperature dependence of $<S_{iz}S_{jz}S_{kz}S_{lz}>/S^4$ for spin systems with (a) S=3/2 and (b) S=2 with both interactions of fixed positive J_1 and various negative values of J_4.

It is worth noting that in the interaction ranges of -0.16< J_4 / J_1 ≤ -0.155 for S=2 and -0.264< J_4 / J_1≤ -0.26 for S=3/2, the values of $<S_z>/S$ take finite values only at finite temperature and become almost zero at T=0. For spin system of S=2 with J_4 /J_1 =-0.155, the spin arrangement at k_BT/J_1=0.2 are shown in Fig. 6. As can be seen from this figure, the spin structure at k_BT/J_1=0.2 is the one mixed with spin structures (a)～(d) with non-zero magnetization ($<S_z>\neq0$), and with the ones with reversed spin of S_z = ±2 in these spin structures (a)～(d), and with the spin structures (e), (f) with zero magnetization ($<S_z>$=0). Therefore, the total magnetization becomes almost zero at low temperatures.

The temperature dependence of the thermal average $<S_{iz}S_{jz}S_{kz}S_{lz}>$ has also been calculated for the spin system with fixed positive interaction J_1 and various negative interactions J_4, and the results for S=3/2 and 2 are shown by (a) and (b) in Fig.7. As can be seen from these figures, abrupt increases of $<S_{iz}S_{jz}S_{kz}S_{lz}>$ are observed just after the appearance of non-zero magnetization with lowering temperature in the range of -2/9< J_4 / J_1< 0 for S=3/2 and -1/8< J_4 / J_1< 0 for S=2. Moreover, these abrupt increases appear in the small range of J_4 / J_1 even right after these phase transition. These facts may support the appearance of non-zero magnetization for J_4 / J_1right after these phase transition. The value of $<S_{iz}S_{jz}S_{kz}S_{lz}>/S$ at T=0 can become almost -1 only in the range of J_4 / J_1<-0.5 for S=3/2 and J_4 / J_1<-0.3 for S=2. These facts may suggest that the complete GS spin arrangement by negative interaction J_4 can be realized at low temperature only by starting the construction of GS spin structure at sufficient high temperature.

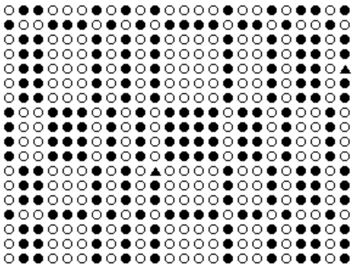
(B) Effect of the Positive Interaction J_1 on the Spin System with Positive Interaction J_4

Let us investigate the temperature dependence of spin structure of the spin system only with positive four-site four-spin interaction J_4 and obtain the ground state (GS) spin structure, and moreover determine the temperature range in which this GS spin structure is fixed. For the spin system of S=2 only with positive J_4, the dependence of spin structures on the temperature is shown in Fig.11 in the range of $0.5\leqq k_BT/J_4\leqq 19.0$.

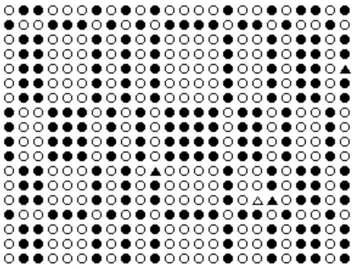


(a)

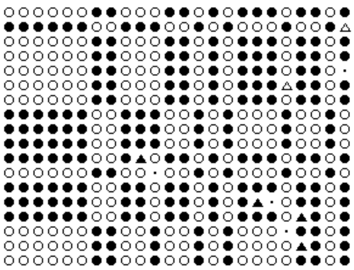
k_BT/J_1=0.5



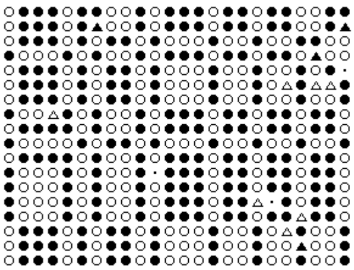
(b)
k_BT/J_1=6



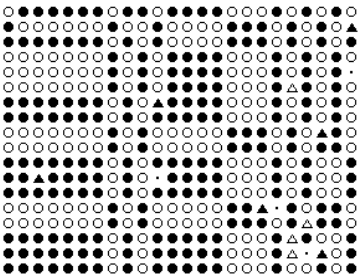
(c)
k_BT/J_1=10



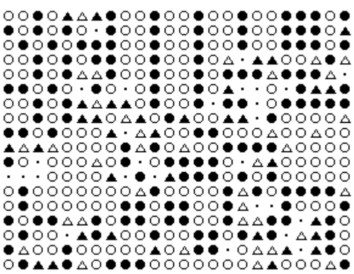
(d)
k_BT/J_1=13



(e)
k_BT/J_1=14



(f)
k_BT/J_1=15



(g)
k_BT/J_1=19

Fig. 11. The spin arrangement at (a) k_BT/J_4=0.5, (b) k_BT/J_4=6, (c) k_BT/J_4=10, (d) k_BT/J_4=13, (e) k_BT/J_4=14, (f) k_BT/J_4=15, (g) k_BT/J_4=19 for the spin system of S=2 only with positive J_4. Open and closed circles denote S_Z =2 and S_Z =-2, respectively. Open and closed triangles, and dot denote S_Z =1 and S_Z =-1,and S_Z =0, respectively.

Thermal averages $\langle S_z \rangle$ and $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle$ have also been calculated for this spin system of S=2. Judging from the behavior of $\langle S_z \rangle$, non-zero magnetization can not appear for this spin system in all temperature range. The four-spin thermal average $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle/S^4$ is pointed out to be just one and spin arrangement is completely fixed in the temperature range of $k_BT/J_4 \leqq 4.5$. For the temperature in the range of $4.5 < k_BT/J_4 < 6.0$, the spin structure is constructed only by spins of $S_z = \pm 2$, but the value of $\langle S_{iz}S_{jz}S_{kz}S_{lz} \rangle/S^4$ cannot be one. Furthermore, the spin structure in the temperature range of $6.0 \leqq k_BT/J_4 < 13.0$ is constructed by spins of $S_z = \pm 2$ and ± 1, and in the temperature range of $13.0 \leqq k_BT/J_4$, the spin structure is constructed by spins $S_z = \pm 2$, ± 1, and 0, and change largely with increasing temperature.

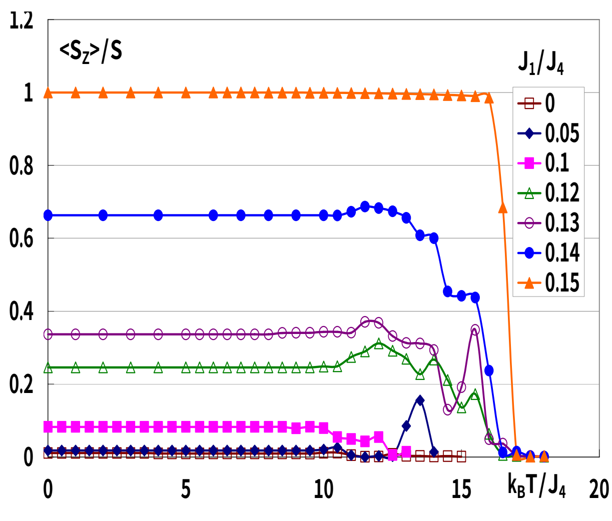


Fig. 12. Temperature dependence of $\langle S_Z \rangle/S$ for spin system of S=2 with both interactions of fixed positive J_4 and various positive values of J_1.

Let us investigate the effects of the bilinear exchange interaction J_1 on the magnetization $\langle S_z \rangle$, the magnetic specific heat C_M, the Curie temperature T_c, and the GS spin structures of the Ising spin system constructed with

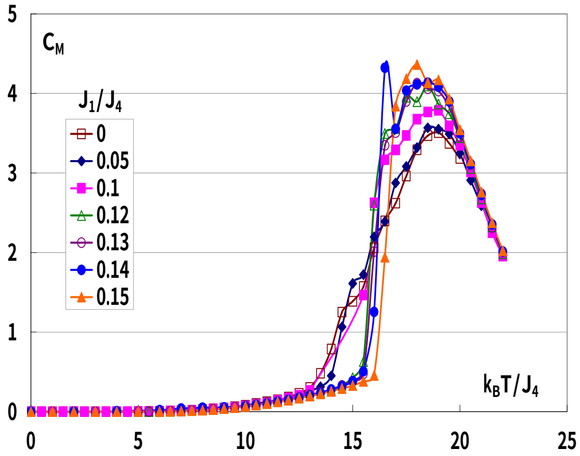


Fig. 13. The temperature dependence of C_M for spin system of S=2 for both interactions of fixed positive J_4 and various positive values of J_1.

positive four-site four-spin interaction J_4 by making use of the MC simulation. In the spin system of S=2 with positive interaction J_1 in the range of $0 \leqq J_1/J_4 \leqq 0.15$, the results of MC simulation for magnetization $\langle S_z \rangle /S$ and magnetic specific heat C_M are shown in Fig.12 and Fig.13, respectively.

As can be seen from Fig.12, the ferromagnetic order appears by a small positive interaction J_1. It is worth noting that for the interaction J_1 in the range of $0.10 \leqq J_1/J_4 \leqq 0.14$, the magnetizations $\langle S_z \rangle /S$ at T=0 become the values in the range of $0 < \langle S_z \rangle /S < 1$ and an imperfect ferromagnetic orders appear. Furthermore, it turns out that the value of $\langle S_z \rangle /S$ is constant at broad low temperature range for the interaction J_1 in the range of $0.10 \leqq J_1/J_4$. These facts are consistent with those that the magnetic specific heat C_M is almost zero at low temperatures for various values of J_1.

The temperature dependence of spin structures has been calculated for the spin system with both interactions J_1 and J_4, and the results for interaction of J_1/J_4 =0.10 are shown in Fig.14. Large ferromagnetic spin groups exist in the temperature range of $k_BT/J_4 \leqq 16$ and these spin groups are fixed at low temperatures in the range of $k_BT/J_4 \leqq 5.5$.

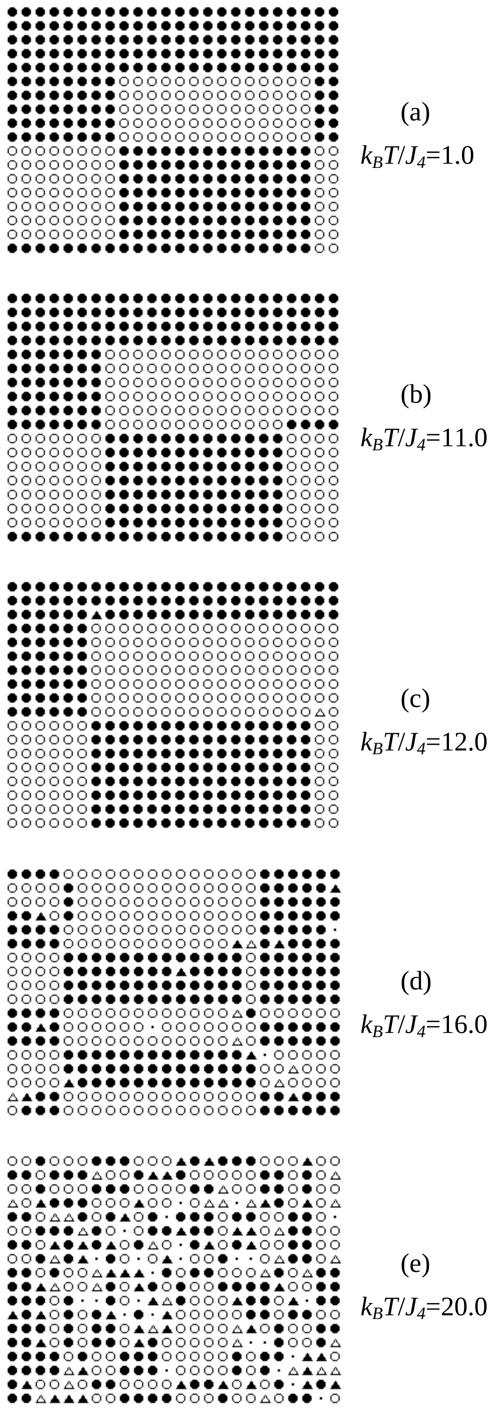


Fig. 14. The spin arrangement at (a) k_BT/J_4=0.5, (b) k_BT/J_4=6, (c) k_BT/J_4=10, (d) k_BT/J_4=13, (e) k_BT/J_4=14, (f) k_BT/J_4=15 for the spin system of S=2 with J_1/J_4=0.1. Open and closed circles denote S_Z =2 and S_Z =-2, respectively. Open and closed triangles, and dot denote S_Z =1 and S_Z =-1, and S_Z =0, respectively.

4. Concluding Remarks

In the previous section, for the Ising spin systems of S=3/2 and 2 on the two-dimensional square lattice with two-spin and interactions J_1 and J_4, the magnetization $<S_z>$, the magnetic specific heat $C_{M,}$, the four-spin thermal average $<S_{iz}S_{jz}S_{kz}S_{lz}>$ and the GS spin structures have been calculated by making use of the MC simulation. The conditions of phase transition are determined and the temperature dependences of spin structures have also been investigated for the Ising spin system with both interactions J_4 and $J_{1,}$ and with only interaction J_4.

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

(1) The conditions of phase transition obtained by the temperature dependence of magnetization $<S_z>$ agree well with the ones (J_4/J_1 =-2/9 for S=3/2 and J_4/J_1 =-1/8 for S=2) calculated by comparison of energy levels of many spin structures.

(2) The decreases of temperature dependence curves of $<S_z>$ with decreasing temperature are caused by the existence of two kinds of GS spin structures with zero and non-zero magnetizations.

(3) Abrupt increases of $<S_{iz}S_{jz}S_{kz}S_{lz}>$ are observed just after the appearance of non-zero magnetization with lowering temperature. These facts may support the appearance of non-zero magnetization for J_4/J_1 right after these phase transition.

(4) By introducing interaction J_1 in the range of $0.15 \leqq J_1/J_4$, the complete ferromagnetic order appears on the Isng spin system with only interaction $J_{4.}$ On the other hand, the magnetization $<S_z>/S$ becomes the value in the range of $0<<S_z>/S<1$ for the interaction of $0.10 \leqq J_1/J_4<0.15$ and an imperfect ferromagnetic spin arrangement appears in this rigion. Furthermore, these magnetizations turn out to be almost constant at low temperatures for the interaction in the range of $0.10 \leqq J_1/J_4$.

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