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Ising Spin System with Biquadratic Exchange Interaction and Single-Ion Anisotropy

By

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Abstract

The magnetic properties of the spin S = 2 Ising system with the bilinear exchange interaction $J_1S_{iz}S_{jz}$, the biquadratic exchange interaction $J_2S_{iz}^2S_{jz}^2$ and the single-ion anisotropy DS_{iz}^2 are discussed by making use of the Monte Carlo (MC) simulation for the magnetization $<S_z>$, sub-lattice magnetizations $<S_z(A)>$ and $<S_z(B)>$, the magnetic specific heat C_M and spin structures. The phase diagram of this Ising spin system on a two-dimensional square lattice has been obtained for exchange parameter J_2/J_1 and anisotropy parameter D/J_1. The changes of sub-lattice magnetizations $<S_z(A)>$ and $<S_z(B)>$ are related with abnormal behavior of temperature dependence of $<S_z>$ at low temperatures and affected significantly by the single-ion anisotropy D. The staggered quadrupolar (SQ) ordering turns out to be different largely between Ising systems with the single-ion anisotropy ($D \neq 0$) and without the one (D =0).

Key words: biquadratic interaction, single-ion anisotropy, Ising model, 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 $(\boldsymbol{S}_i \cdot \mathrm{S}_j)(\boldsymbol{S}_k \cdot \boldsymbol{S}_l)$ are 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].

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]. The Ising system of S=1with a 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 [5] 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[6].

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Therefore, we have developed the Monte Carlo (MC) simulation to the Ising spin system with large spin of S=2, and investigated more precisely the growth of the spin ordering and the ground state (GS) spin structures. In the present study, the effects of the biquadratic interaction $J_2S_{iz}^2S_{jz}^2$ and the single-ion anisotropy DS_{iz}^2 on the magnetization $<S_z>$, sub-lattice magnetizations $<S_z(A)>$ and $<S_z(B)>$, the magnetic specific heat C_M of Ising spin system of S=2 on a two-dimensional square lattice are investigated by making use of the MC simulation. The obtained characteristic behaviors of $<S_z>$, C_M are discussed in conjunction with the GS spin structures determined by energy evaluations. The temperature dependences of spin structure are also studied for various values of parameters J_2/J_1 and D/J_1, and the phase diagram is obtained for these parameters (J_2/J_1 <0 and D/J_1>0).

2. Spin Hamiltonian and Methods of Simulation

The spin Hamiltonian for the present Ising spin system with S=2 can be written as follows:

$$H = -J_1\sum_{<ij>} S_{iz}S_{jz} - J_2\sum_{<ij>} S^2_{iz}S^2_{jz} - D\sum_{i} S^2_{iz} \qquad (1)$$

The GS spin structures are determined for the Ising spin system with both interactions J_1 and J_2 and without single-ion anisotropy term (D=0) by comparing the energies of various spin structures with each other (see e.g. [7]). The GS spin structures obtained for the spin system of S=2 with positive interaction J_1 and negative interaction J_2 are shown in Fig.1.

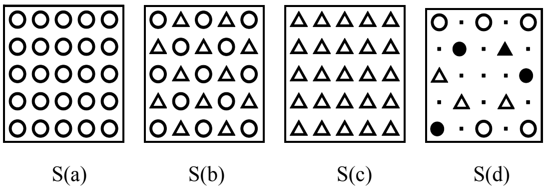


Fig.1. The GS spin structures S(a), S(b), S(c), S(d) for Ising spin system with S=2. Open and closed circles denote S_Z =2 and S_Z =-2, respectively, and open and closed triangles and dot denote S_Z =1 and S_Z =-1, S_Z =0, respectively.

The energies per one spin for the spin structures S(a)～S(d) are given as E_a=-8J_1 -32J_2 , E_b= -4J_1 -8J_2 , E_c=-2J_1 -2J_2 and E_d= 0, respectively. Therefore, by comparing these energies, phase transitions turn out to occur at the conditions of J_2/J_1 =-1/6, -1/3 and -1. Furthermore, the spin structures S(a)～S(d) may be the GS spin structures in the interaction range of -1/6<J_2/J_1, -1/3<J_2/J_1< -1/6, -1<J_2/J_1< -1/3, J_2/J_1< -1, respectively.

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_2/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 $\times 10^5$ MC steps per spin (MCS/s) after discarding the first 3 $\times$ 10^5 MCS/s. The values of $<S_z>$, $<S_z(A)>$, $<S_z(B)>$, C_M and spin structures are calculated for the spin system with fixed positive interaction J_1 and various negative interaction J_2 and various positive anisotropy term D.

3. Results of Calculation and Discussion

3.1 Magnetization , Specific Heat and Sub-lattice Magnetization

The temperature dependences of $<S_z>$ and C_M have been calculated for the spin system both with interactions J_1 and J_2 (J_1>0 and J_2<0), and the results of $<S_z>/S$ for the spin system of S=2 on the two-dimensional square lattice are shown in Fig.2 and (a) in Fig.3, respectively.

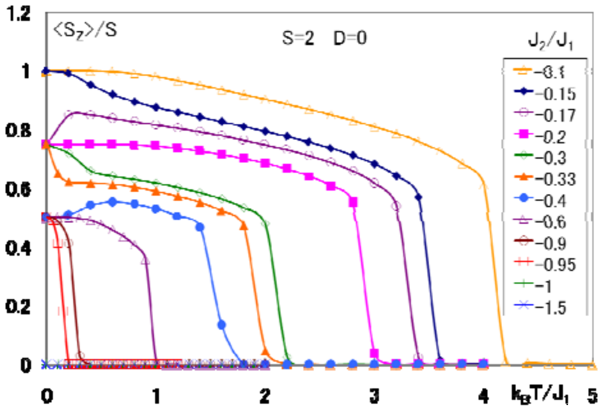
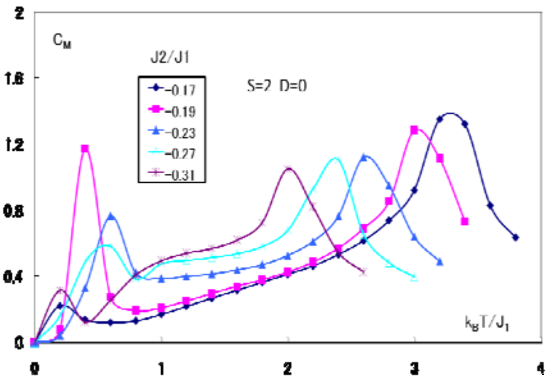


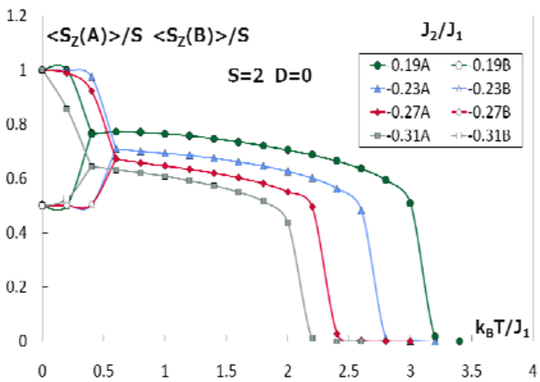
Fig.2. Temperature dependence of $<S_Z>/S$ for spin system of S=2 with both interactions of fixed positive J_1 and various negative values of J_2 (D=0).

The values of $<S_z>/S$ at T=0 have been turned out to be 1, 0.75, 0.5 and 0 for interaction parameter in the range of -1/6<J_2/J_1, -1/3<J_2/J_1< -1/6, -1<J_2/J_1< -1/3 and J_2/J_1< -1, respectively. These values of $<S_z>/S$ at T=0 are confirmed to correspond to those obtained for the spin structures S(a)～S(d) in Fig.1, respectively. Judging from the behaviors at low temperatures, the phase transitions are pointed out to occur at the conditions of J_2/J_1 =-1/6, -1/3 and -1. These conditions agree quite well with those obtained by above mentioned energy comparisons. As known from spin structure S(b) in Fig.1, the GS spin structure is constructed with two sub-lattices of S_z=2 and S_z=1 in the range of -1/3<J_2/J_1< -1/6. On the other hand, as seen from (a) in Fig.3 the temperature dependence curves of the magnetic specific heat C_M have two peaks in the same range of J_2/J_1. These facts may suggest that different abrupt spin orderings occur at two different temperatures. Therefore, let us investigate the temperature dependence of the sub-lattice magnetization. The calculated results of sub-lattice magnetizations $<S_z(A)>$ and $<S_z(B)>$ ($<S_z(A)> \geqq <S_z(B)>$) are shown by (b) in Fig.3 for above mentioned interaction range. Here, A and B represent the two-interpenetrating lattices.

The different temperature dependence of $<S_z(A)>$ and $<S_z(B)>$ at low temperatures may explain the abnormal behavior of $<S_z>$(=($<S_z(A)>$ + $<S_z(B)>$)/2). Next, we investigate the temperature dependences of $<S_z(A)>$ and $<S_z(B)>$ of the Ising spin system with single-ion anisotropy term D. The results for the Ising spin systems with D/J_1=1 and 1.5 are shown by (a) and (b) in Fig.4. It is worth noting that the temperature at which <$S_z(A)$> and <$S_z(B)$> begin taking different values becomes higher and close to the Curie temperature T_c with increasing anisotropy term D.

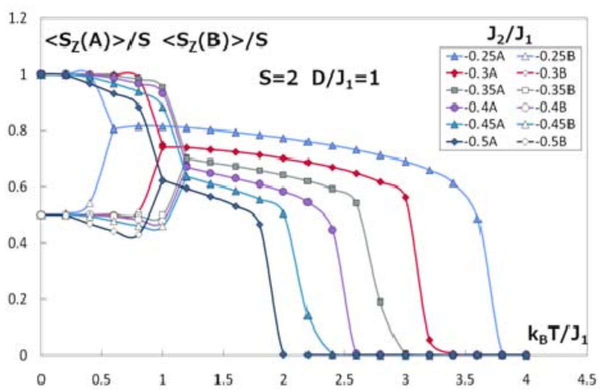


(a)

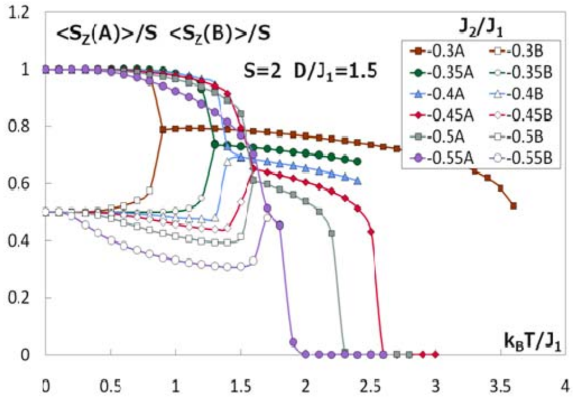


(b)

Fig. 3. The temperature dependence of (a) C_M and (b) <$S_Z(A)$>/S and <$S_Z(B)$>/S of spin system with S=2 for both interactions of fixed positive J_1 and various negative values of J_2 in the range of $-1/3<J_2/J_1<-1/6$.



(a)

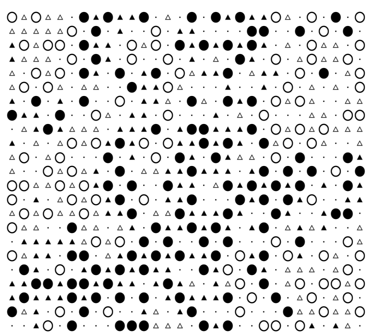


(b)

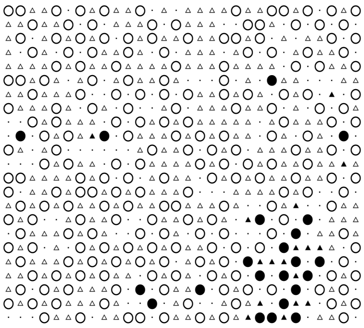
Fig. 4. The sub-lattice magnetizations <$S_Z(A)$>/S and <$S_Z(B)$>/S of spin system with S=2 for (a) D/J_1 =1 and (b) D/J_1 =1.5 .

The shapes of the temperature dependence of <$S_z(A)$> and <$S_z(B)$> also change largely with increasing anisotropy term D in the temperature range with different values of <$S_z(A)$> and <$S_z(B)$>. From the behaviors of <$S_z(A)$> and <$S_z(B)$> in Fig.3 and Fig.4, the range of spin structure S(b) as a GS spin arrangement also turns out to change with increasing anisotropy term D.

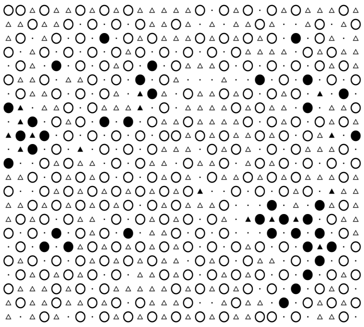
Next, we investigate the change of spin structures with two sub-lattice structure as a GS structure by gradually decreasing temperature. The changes of spin structures of Ising system with parameters of J_2/J_1=-0.55 and D/J_1=1.5 are shown in Fig.5. The simulation was done in the temperature range of 0< k_BT/J_1<2.2.



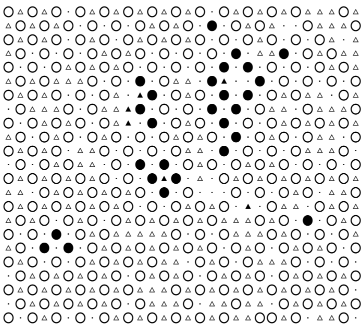
(a)



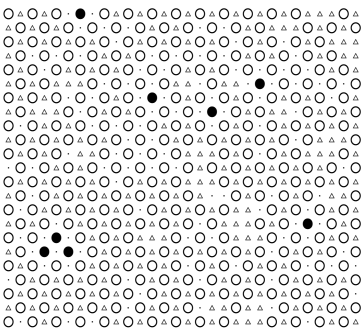
(b)



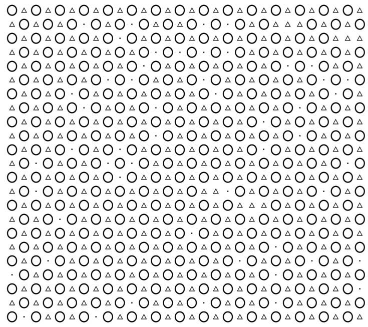
(c)



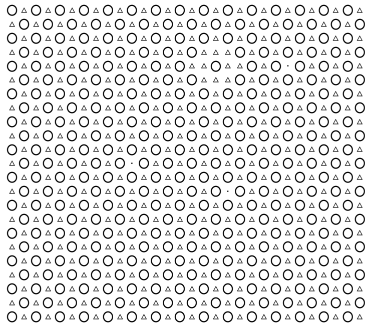
(d)



(e)



(f)

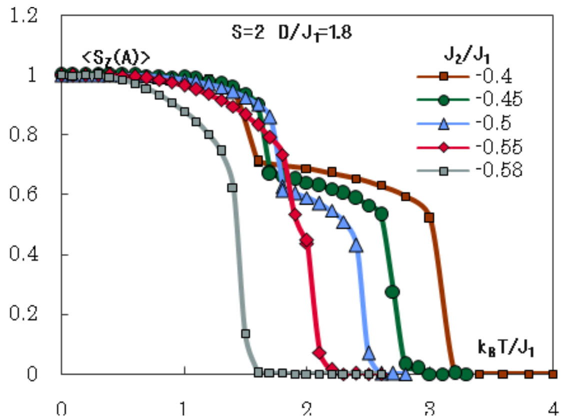


(g)

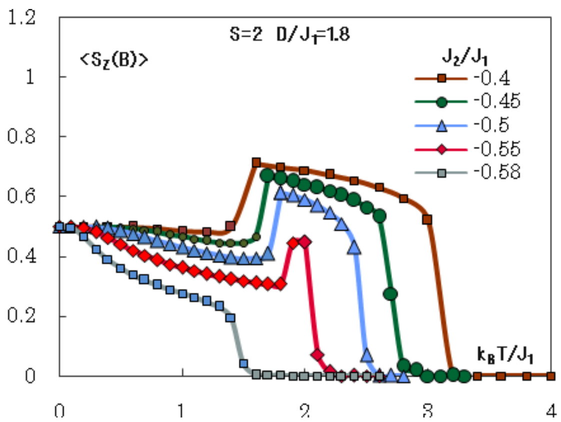
Fig.5. The change of spin structures for various values ((a)k_BT/J_1=2.2, (b)k_BT/J_1=1.8, (c)k_BT/J_1=1.6, (d)k_BT/J_1=1.2, (e)k_BT/J_1=0.8, (f)k_BT/J_1=0.4, (g)k_BT/J_1=0.1) of temperature in Ising spin system with J_2/J_1=-0.55 and D/J_1=1.5. Open and closed circles denote S_Z =2 and S_Z =-2, respectively, and open and closed triangles and dot denote S_Z =1 and S_Z =-1, S_Z =0, respectively.

The spin arrangement is random and paramagnetic state for spin structure (a) at k_BT/J_1 =2.2. The ordered spin state ($\langle S_z \rangle \neq 0$) appears in the spin structure (b) at k_BT/J_1 =1.8. The spin structure becomes the one with two sub-lattices in the spin structure (c) at k_BT/J_1 =1.6. In the spin structure (d) at k_BT/J_1 =1.2, two sub-lattices with $\langle S_z(A) \rangle$ and $\langle S_z(B) \rangle$ are constructed almost with spins of S_z=±2, ±1 and S_z=±1, 0, respectively. On the other hand, in the spin structure (e) at k_BT/J_1 =0.8, two sub-lattices with $\langle S_z(A) \rangle$ and $\langle S_z(B) \rangle$ are constructed almost with spins of S_z=±2 and S_z=1, 0, respectively. The spin arrangement for the spin structure (f) at k_BT/J_1 =0.4 is constructed with two sub-lattices of $\langle S_z(A) \rangle$ with S_z=2 and $\langle S_z(B) \rangle$ with S_z=1, 0. The spin arrangement for the spin structure (g) at k_BT/J_1 =0.1 is constructed with two sub-lattices of $\langle S_z(A) \rangle$ with S_z=2 and $\langle S_z(B) \rangle$ with S_z=1 and becomes completely the spin structure S(b) in Fig.1.

Furthermore, the temperature dependence of sub-lattice magnetizations $<S_z(A)>$ and $<S_z(B)>$ was studied for Ising system with D/J_1=1.8. The results for various values of J_2 are shown by (a) for $<S_z(A)>$ and by (b) for $<S_z(B)>$ in Fig. 6. As can be seen from these figures, the abrupt changes of $<S_z(A)>$ and $<S_z(B)>$ occur in ordered state. These changes may be caused by the movement of spins S_z=1, 0 between two sub-lattices. As the split of two sub-lattices occurs at the Curie temperature T_c for J_2/J_1 =-0.58, the abrupt changes of $<S_z(A)>$ and $<S_z(B)>$ caused by the movement of spins S_z=1, 0 cannot appear.



(a)



(b)

Fig.6. The sub-lattice magnetizations (a) $<S_Z(A)>/S$ and (b) $<S_Z(B)>/S$ of spin system with S=2 and D/J_1 =1.8.

3.2 Thase Diagram of Ising Spin System with Interaction J_2 and Anisotropy D

Next, we have investigated the phase diagram of the Ising spin system of S=2. The phase diagram obtained from the MC simulation is shown in Fig.7 for parameters J_2/J_1 (J_2/J_1<0) and D/J_1 (D/J_1>0). For the spin system without single-ion anisotropy, the GS spin structure S(c) appears in the wide interaction range of -1/3< J_2/J_1 < -1/6. As can be seen from this figure, the spin structure S(c) as a GS spin structure, however, varnishes for the anisotropy term D in the range of D/J_1>0.9. On the other hand, the ranges of spin structures S(a), S(b) and S(d) as GS spin structures expand with increasing the anisotropy term D. It is worth noting that the GS spin structure S(d) becomes to appear for more large interaction J_2 with increasing anisotropy D in the range of 0< D/J_1 < 0.9.

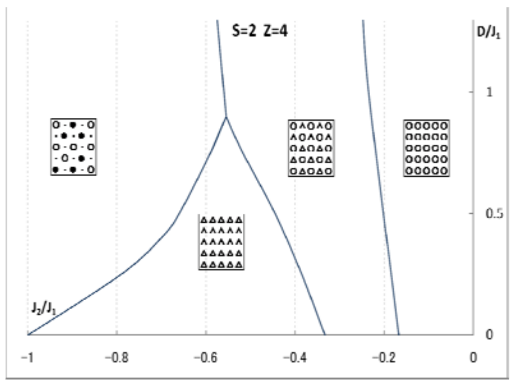


Fig. 7. The phase diagram of Ising system of S=2 on two-dimensional square lattice with negative interaction parameter J_2/J_1 and positive anisotropy parameter D/J

3.3 SQ Ordering of Ising Spin System in the GS spin structure S(d)

In the magnetic phase of spin structure S(d) as a GS spin structure, non-zero magnetization can not appears ($<S_z>$=0), the values of $<S_z^2(A)>$ and $<S_z^2(B)>$ are, however, pointed out to take non-zero value at low temperature. This spin arrangement is called as staggered quadrupolar (SQ) ordering [5]. Now, let us consider three kinds of SQ orderings defined as

$$\begin{aligned}
&\text{SQ I}: 0<<S_z^2(A)><1 \text{ and } 0<<S_z^2(B)><1,\\
&\text{SQ II}: <S_z^2(A)> \neq 1 \text{ and } <S_z^2(B)> = 0, \qquad (2)\\
&\text{SQ III}: <S_z^2(A)> =1 \text{ and } <S_z^2(B)> = 0.
\end{aligned}$$

The examples of spin arrangement of these three kinds of SQ states for Ising spin system with S=2 are shown in Fig.8.

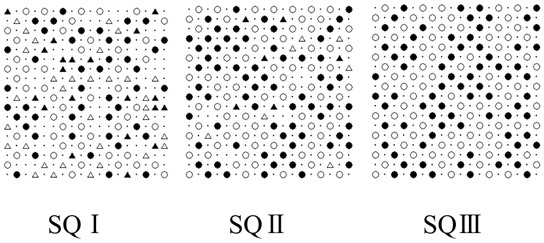


Fig. 8. Three kinds of SQ spin structure. Open and closed circles denote S_Z =2 and S_Z =-2, respectively, and open and closed triangles and dot denote S_Z =1 and S_Z =-1, S_Z =0, respectively.

First, let us investigate the spin arrangement, $<S_z^2(A)>$ and $<S_z^2(B)>$ of the Ising system without anisotropy term (D=0) in the range of spin structure S(d) as a GS spin structure. The temperature dependences of $<S_z^2(A)>/S^2$ and $<S_z^2(B)>/S^2$ calculated by MC simulation are shown in Fig.9 for various values of negative interaction J_2/J_1. As can be seen from this figure, the values of $<S_z^2(A)>/S^2$ and $<S_z^2(B)>/S^2$ at T=0 for interaction J_2/J_1 = -1.5 are about 0.423 and 0.035. These values of $<S_z^2(A)>/S^2$ and $<S_z^2(B)>/S^2$ cannot take 1 or 0 for decreasing interaction J_2 in the negative range. Therefore, we may conclude that the SQ ordering for Ising system without anisotropy (D=0) is only SQ I even at sufficiently low temperature. Next, we investigate the change of spin structure of Ising system with fixed interaction J_2/J_1 and without anisotropy term (D=0). The changes of spin structure for interaction J_2/J_1 =-1.5 are shown in Fig.10.

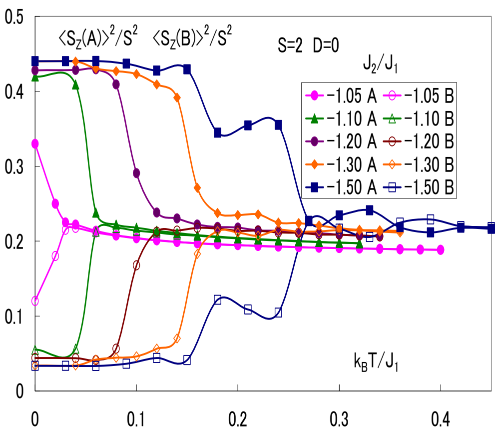


Fig.9. The temperature dependences of $<S_Z^2(A)>/S^2$ and $<S_Z^2(B)>/S^2$ of spin system of S=2 with various values of J_2/J_1 and without anisotropy D.

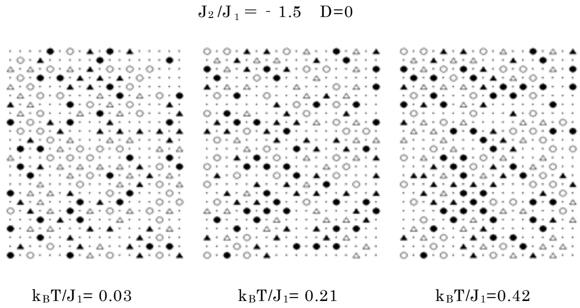


Fig.10. Temperature dependence of spin structures for spin system of S=2 with fixed interaction J_2/J_1=-1.5 and without anisotropy (D=0).

The spin structures for temperatures k_BT/J_1 =0.03 and 0.21 are SQ state, the one for temperature k_BT/J_1 =0.42 is not, however, SQ state and is paramagnetic spin state.

Furthermore, let us investigate the spin ordering for fixed interaction J_2/J_1 and various values of anisotropy term D in the positive range. The temperature dependences of $<S_z^2(A)>/S^2$ and $<S_z^2(B)>/S^2$ for fixed interaction J_2/J_1 =-1.5 are shown in Fig. 11.

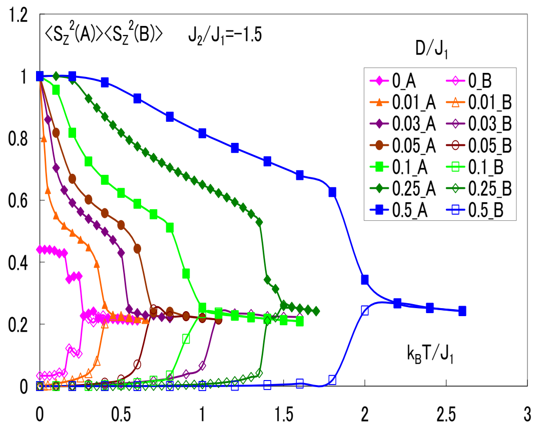


Fig.11. The temperature dependences of $<S_Z^2(A)>/S^2$ and $<S_Z^2(B)>/S^2$ of spin system of S=2 with fixed interaction J_2/J_1=-1.5 and with various values of anisotropy D.

As can be seen from this figure, the values of $<S_z^2(A)>/S^2$ and $<S_z^2(B)>/S^2$ at T=0 are completely one and zero, respectively for anisotropy parameter D>0. These facts suggest us that SQIII state can appear for Ising system with anisotropy parameter D. The temperature of at which the value of $<S_z^2(B)>/S^2$ becomes zero turns out to be higher than that at which the value of $<S_z^2(A)>/S^2$ becomes one. Therefore, the appearance of SQ II state is pointed out to be confirmed from these behavior of $<S_z^2(A)>/S^2$ and $<S_z^2(B)>/S^2$.

The ratio of increase of temperature range of $<S_z^2(B)>/S^2=0$ is much larger than that of $<S_z^2(A)>/S^2=1$ with increasing anisotropy term D.

We investigate the temperature range of existence of SQ state for various values anisotropy term D. The results for anisotropy parameter in the range of $0<D<0.5$ are shown in Fig.12.

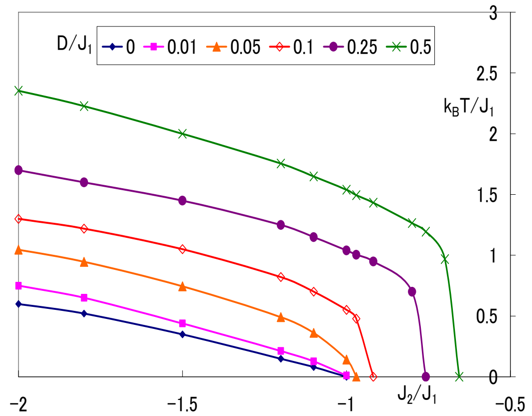
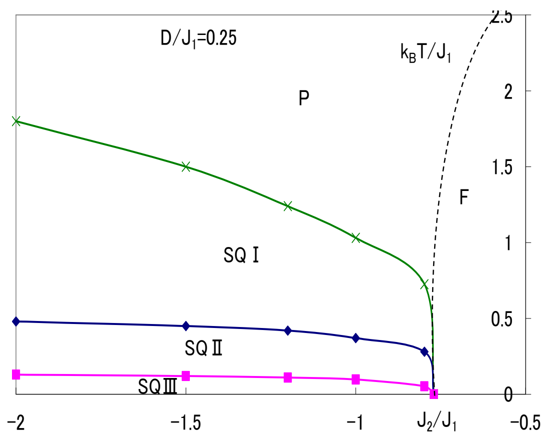
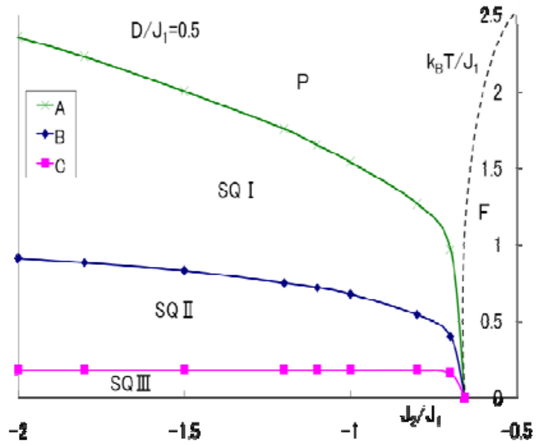


Fig. 12. Temperature range of SQ state for Ising spin system of S=2 in the range of $0\leqq D/J_1\leqq 0.5$. The lower part and upper part of each line represent the SQ state and paramagnetic spin states. .

Next, let us investigate more precisely the temperature dependence of spin arrangement for various values of D. The phase diagrams obtained for anisotropy parameter D= 0.25 and 0.5 are shown by (a) and (b) in Fig.13.



(a)



(b)

Fig.11. Phase diagram for Ising spin system of S=2 with (a) D=0.25 and (b) D=0.5. F and P represent the ferromagnetic and paramagnetic spin states. .

The existence of three kinds of SQ state is confirmed under the presence of anisotropy parameter D. Furthermore, the temperature ranges of these three SQ states become much wider with increasing anisotropy term D.

4. Conclusions

In the previous section, for the Ising spin system of S=2 with the bilinear exchange interaction $J_1S_{iz}S_{jz}$, the biquadratic exchange interaction $J_2S_{iz}^2S_{jz}^2$ and the single-ion anisotropy DS_{iz}^2, the magnetization $<S_z>$, sub-lattice magnetizations $<S_z(A)>$ and $<S_z(B)>$, magnetic specific heat C_M, the values of $<S_z^2(A)>$ and $<S_z^2(B)>$, 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 transitions occur under the conditions of J_2/J_1 =-1/6, -1/3 and -1. Furthermore, the spin structures S(a)～S(d) are the GS spin structures in the interaction range of $-1/6<J_2/J_1$, $-1/3<J_2/J_1<-1/6$, $-1<J_2/J_1<-1/3$, $J_2/J_1<-1$, respectively.

(2) The different temperature dependences of sub-lattice magnetizations $<S_z(A)>$ and $<S_z(B)>$ at low temperatures explain the abnormal behavior of temperature dependence of total magnetization $<S_z>(=(<S_z(A)>+<S_z(B)>)/2)$. The position of a split of sub-lattice magnetizations moves to higher

temperature with increasing anisotropy term *D*. Furthermore, the shape of sub-lattice magnetization also changes largely with increasing anisotropy term *D*.

(3) The abrupt changes of sub-lattice magnetizations $<S_z(A)>$ and $<S_z(B)>$ at ordered state are caused by the movement of spins S_z=1, 0 between two sub-lattices.

(4) The phase diagram of this Ising spin system of *S*=2 is obtained for parameters J_2/J_1 (J_2/J_1<0) and D/J_1 (D/J_1>0). The spin structure S(c) as a GS spin structure varnishes for the anisotropy term *D* in the range of D/J_1>0.9. On the other hand, the ranges of spin structures S(a), S(b) and S(d) as GS spin structures expand with increasing the anisotropy term *D*.

(5) The SQ spin ordering for Ising system without anisotropy term (*D*=0) is only SQⅠstate even at sufficiently low temperature. The three kinds of SQ states(SQⅠ,SQⅡ,SQⅢ) can appear for Ising system with anisotropy parameter *D*. The ranges of these three SQ states become more wide with increasing anisotropy parameter *D*.

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