# Blume-Emery-Griffths Model of Mixed Ising Spin System of S=1 and R=2

**Takashi Iwashita[*], Hisanori Arai[*], Yui Ito[**], Yoshinori Muraoka[***]**

(Received October 31, 2011)

**Abstract.**

The phase diagram and magnetic properties such as the magnetizations $<S_z>$ and $<R_z>$, thermal averages $<S_z^2>$ and $<R_z^2>$, the Curie temperature $T_c$ and the ground state (GS) spin structures of the mixed Ising spin system ($S$=1and $R$=2) on two-dimensional square lattice with the bilinear exchange interaction $J_1S_{iz}R_{jz}$, the biquadratic exchange interaction $J_2S_{iz}^2R_{jz}^2$ and the single-ion anisotropies $DS_{iz}^2$ and $DR_{jz}^2$ have been discussed by making use of the Monte Carlo simulation. In this Ising spin system, we have 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 dependence of the magnetizations $<S_z>$ and $<R_z>$, thermal averages $<S_z^2>$ and $<R_z^2>$ and the ground state (GS) spin structures are investigated for various values of parameters of $J_2/J_1$ and $D/J_1$.

**Keywords:** Ising model; biquadratic interaction; mixed spin system; 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 ground state (GS) spin structure of the Ising ferromagnet [12,13] with $S$=1 by making use of

[*]General Education Center, Tokai University
[**] Department of Environment Conservation Sciences, School of Industrial Engineering, Tokai University
[***]Department of General Education, Ariake National College of Technology

the Monte Carlo (MC) simulation. Furthermore, we have applied this MC simulation to the Ising spin system of large spin $S = 2$ with interaction $J_2$, and investigated more precisely the growth of spin ordering and the GS spin structures [14]. We have also developed this MC simulation to the mixed Ising spin system with spins of $S = 1$ and $R=3/2$, and investigated precisely the growth of spin ordering and the ground state (GS) spin structures.

Therefore, in the present study, the effects of the biquadratic exchange interaction $J_2S_{iz}^2R_{jz}^2$ and the single-ion anisotropies $DS_{iz}^2$ and $DR_{jz}^2$ on the magnetizations $<S_z>$ and $<R_z>$, the thermal averages $<S_z^2>$ and $<R_z^2>$ and the ground state (GS) spin structure of the mixed Ising spin system of $S = 1$ and $R=2$ on two-dimensional square lattice are investigated by making use of the MC simulation. Here, spins $S$ and $R$ are situated on each two interpenetrating sub-lattices. The obtained phase diagram is discussed in conjunction with the GS spin structures determined by energy evaluations. The temperature dependences of the magnetizations $<S_z>$ and $<R_z>$, the thermal averages $<S_z^2>$ and $<R_z^2>$ and the spin structure are also studied for various values of parameters $J_2/J_1$ and $D/J_1$.

In Section 2, the spin Hamiltonian is given for present mixed Ising system with $S=1$ and $R=2$. 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 parameters $J_2/J_1$ and $D/J_1$ by the MC simulation of this Ising system. In the latter part of Section 3, the magnetic properties and the spin ordering are investigated for various temperatures. In the last Section 4, new interesting results obtained here are summarized.

## 2. Spin Hamiltonian, Methods of Simulation and Energy Estimation

The spin Hamiltonian for the present mixed Ising spin system with $S=1$ and $R=2$ on two-dimensional square lattice can be written as follows:

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

Here, $<ij>$ denotes the sum on the nearest neighboring spin pairs on two-dimensional square lattice. Furthermore, $S_z$ and $R_z$ in above expression represent $S_z = \pm 1,\ 0$ and $R_z = \pm 2, \pm 1,\ 0$, respectively. From a consideration of the Hamiltonian (1), magnetic properties and spin arrangements of this mixed Ising spin system of $S=1$ and $R=2$ 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 $J_1$, $J_2$ 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 magnetizations $<S_z>$ and $<R_z>$ and the thermal averages $<S_z^2>$ and $<R_z^2>$ estimated from the energy fluctuation are calculated using $2\times10^5$ MC steps per spin (MCS/s) after discarding first $3\times10^5$ MCS/s.

In order to check the reliability of these obtained average values, the thermal averages are also calculated separately for each interval of $0.5\times10^5$ MCS/s in the above mentioned total interval of $2\times10^5$ MCS/s. In the following section, results in the largest system of $L=240$ 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. [15]). The GS spin structures are determined for this mixed Ising spin system with interactions $J_1$ and $J_2$, and with single-ion anisotropy $D$ by comparing these energies per one spin with each other. The GS spin structures with low energy obtained for this spin system of $S = 1$ and $R=2$ with positive interaction $J_1$ are shown in Fig. 1. The spin structures S(a) and S(b) are consisted of two kinds of spins $S_z=1$

and $R_z$=2, $S_z$=1and $R_z$=1. Furthermore, the spin structures S(c) ～ S(e) are consisted of two kinds of spins $S_z$=0 and $R_z$=±2, ±1, 0, and spins $S_z$=0 and $R_z$=±2, and spins $S_z$=0 and $R_z$=0, respectively. Let us define parameters $x$ and $y$ as $J_2/J_1$ and $D/J_1$, respectively. The energies per one spin for the spin structures S(a) ～ S(e) of this mixed Ising spin system with $S$=1 and $R$=2 are given as $E(a) = E_a/NJ_1 = -8x -5y/2-4$, $E(b) = E_b/NJ_1 = -2x-y-2$ , $E(c) = E_c/NJ_1=0$, $E(d) = E_d/NJ_1= -2y$, $E(e) = E_e/NJ_1=0$, respectively.

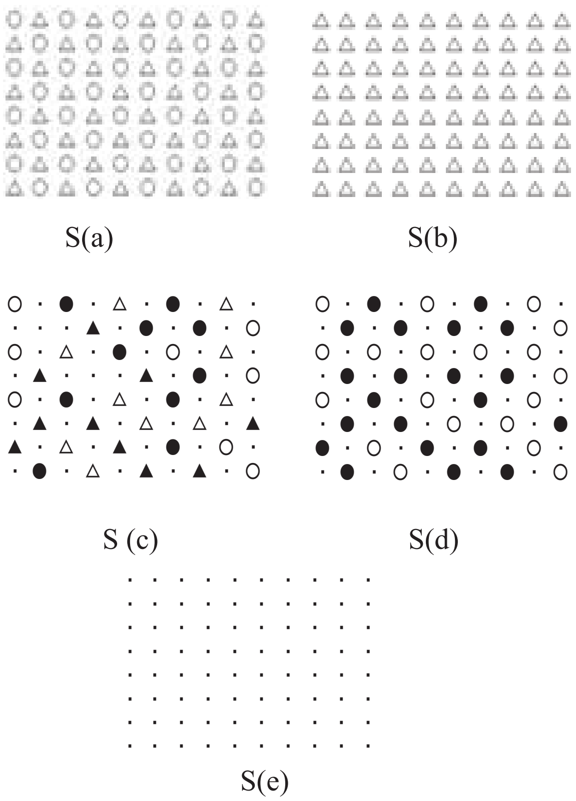



**Fig. 1 The GS spin structures S(a), S(b), S(c), S(d) and S(e) for the mixed Ising spin system of *S*=1 and *R*=2 with interactions $J_1$, $J_2$ and anisotropy *D*. Open and closed circles, and open and closed triangles denote $R_Z$ =±2 and $R_Z$= $S_Z$ =±1, and dot denote $R_Z$= $S_Z$ =0, respectively.**

## 3. Results of Simulation and Discussion

### 3.1 Phase Diagram of Mixed 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 biquadratic interaction $J_2$ in the range of -1.5≦$J_2/J_1$≦1.0 and the anisotropy D/$J_1$ in the range of -1.2≦$D/J_1$≦1.2. In this calculation, the interaction parameter $J_1$ was treated as a positive constant value. The phase diagram is obtained for this mixed Ising spin system on two-dimensional lattice and the result for both parameters $J_2/J_1$ in the range of -1.2≦$J_2/J_1$≦0.2 and $D/J_1$ in the range of -1.2≦$D/J_1$≦1.2 is shown in Fig.2.

The GS spin structures of magnetic phases (a) ～ (e) in Fig.2 determined by the MC simulation are confirmed to be the spin structures S(a) ～S(e) shown in Fig.1 obtained by the energy comparison, respectively. For the case of $D$=0, the conditions of phase transition are determined to be $J_2/J_1$=-1/3 and -1 by this MC simulation. These conditions of phase transition for $D$=0 are confirmed to agree well with those obtained from the equations of $E(a)= E(b)$ and $E(b)= E(c)$ given in the previous section under the condition of $D/J_1$ =0, respectively.

By comparing by the energies $E(a)$ and $E(b)$ given in the previous section, the condition of phase transition between magnetic phases (a) and (b) as $y= -4x-4/3$. This condition agrees well with that obtained by the MC simulation shown in Fig.2. The conditions of phase transition between magnetic phases (b) and (d), and magnetic phases (b) and (e) are given by the MC simulation as $y=10x/7+10/7$ and $y= -2x-2$. On the other hand, the conditions obtained by the energy comparison of $E(b)$ and $E(d)$, and $E(b)$ and $E(e)$ are $y= 2x+2$ and $y= -2x-2$. The condition of phase transition between magnetic phases (b) and (e) obtained by this energy comparison is completely agree with that by the MC simulation. The slight disagreement of the condition of phase transition between magnetic phases (b) and (d) may be confirmed in the later section to be occurred by spin arrangement on the S-site.

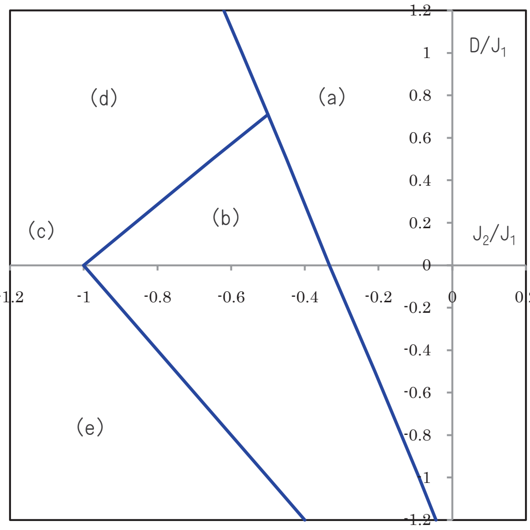



**Fig. 2 Phase diagram of mixed Ising spin system on two-dimensional square lattice with exchange parameter $x(J_2/J_1)$ in the range of $-1.2 \leqq x \leqq 0.2$ and anisotropy $y(D/J_1)$ in the range of $-1.2 \leqq y \leqq 1.2$.**

## 3.2 Magnetic Properties of Mixed Ising Spin System of $S$=1 and $R$=2

### (A) Magnetic Properties of Spin System without anisotropy ( $D/J_1$=0)

Let us investigate the magnetic properties such as the magnetizations $\langle S_z \rangle$ and $\langle R_z \rangle$, and the thermal averages $\langle S_z^2 \rangle$ and $\langle R_z^2 \rangle$ of the mixed Ising spin system of $S$=1 and $R$=2 without anisotropy $D$=0 by making use of the MC simulation. The temperature dependences of $\langle R_z \rangle / R$ and $\langle S_z \rangle / S$ (=$\langle S_z \rangle$) of the mixed spin system for various values of interaction $J_2$ in the range of $-1.0 \leqq J_2/J_1 \leqq -0.1$ are shown in Fig.3 and Fig.4, respectively.

As can be seen from Fig.3, the values of $\langle R_z \rangle / R$ at $T$=0 are 1.0, 0.5 and 0 for the interaction $J_2$ in the range of $-1/3 < J_2/J_1$, $-1 < J_2/J_1 < -1/3$ and $J_2/J_1 \leqq -1$, respectively. These facts suggest that the phase change occurs at $J_2/J_1$=-1/3 and -1. It is remarkable that the temperature dependence curves of $\langle R_z \rangle / R$ show characteristic behavior for the interaction $J_2$ near the condition of phase change ($J_2/J_1$=-1/3). It should be noticeable that for the interaction in the range of $-1 < J_2/J_1 < -1/2$, the curve of $\langle R_z \rangle / R$ show linear dependence in the middle temperature range below $T_c$.

It can be seen from Fig.4 that the temperature dependence curves of $\langle S_z \rangle / S$ show rapid increase just below $T_c$ for the interaction $J_2$ near the condition of phase change ($J_2/J_1$=-1/3). As can be seen from Fig.3 and Fig.4, not only magnetizations $\langle S_z \rangle$ and $\langle R_z \rangle$ but also $T_c$ varnish at the same time under the condition of phase transition ($J_2/J_1$=-1).

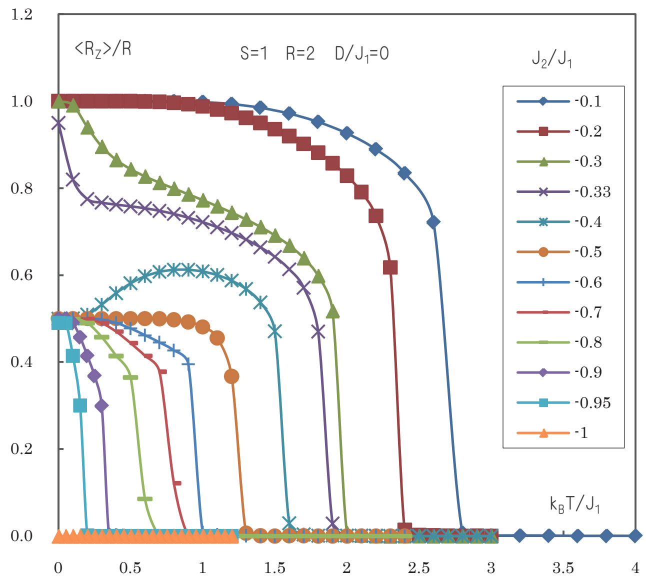



**Fig. 3 Temperature dependence of $\langle R_z \rangle / R$ of the mixed spin system with $S$=1 and $R$=2 calculated by the MC simulation for various values $J_2$ of in the range of $-1.0 \leqq J_2/J_1 \leqq -0.1$.**

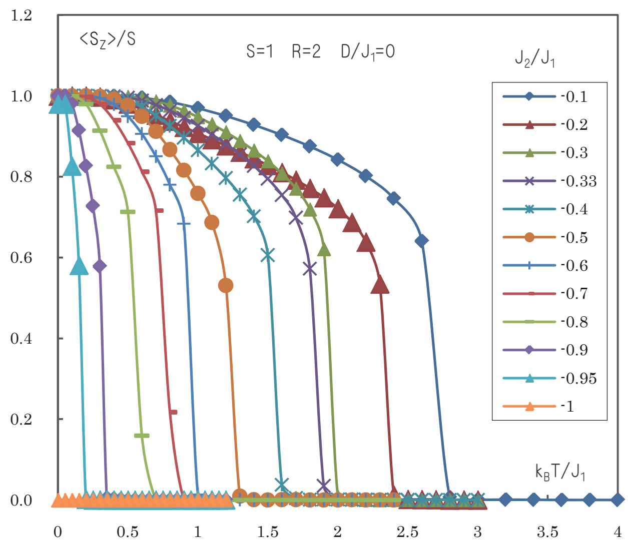



**Fig.4 Temperature dependence of $\langle S_z \rangle / S$ of the mixed spin system with $S$=1 and $R$=2 calculated by the MC simulation for various values $J_2$ of in the range of $-1.0 \leqq J_2/J_1 \leqq -0.1$.**

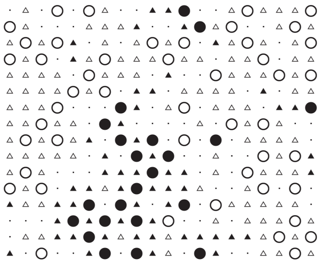
Next, we have investigated the changes of spin structure with the GS spin structures S(a) and S(b) for various values of temperature by making use of the MC simulation and visualized the change of this structure. The changes of spin structure S(a) with $J_2/J_1$=-0.3 are shown in Fig.5 for various temperatures in the range of $0.1 \leqq k_BT/J_1 \leqq 2.0$. As the Curie temperature for $J_2/J_1$= -0.3 is $k_BT_c/J_1$ =1.95, the spin structure (a) is at a paramagnetic state and the spin structures (b), (c), (d) are at an ordered state.

The rates of occupation of a spin $S_z$=1 on S-spin site in ordered state below $T_c$ are 72%, 95%, 100% at temperatures $k_BT/J_1$=1.8, 1.0, 0.1, respectively. On the other hand, the rates of occupation of a spin $R_z$=2 on R-spin site are 40%, 55%, 98% at temperatures $k_BT/J_1$=1.8, 1.0, 0.1, respectively. It is interesting that the rates of the occupation of $S_z$=1 on S-spin site are larger than those of $R_z$=2 on R-spin site at any temperature below $T_c$.

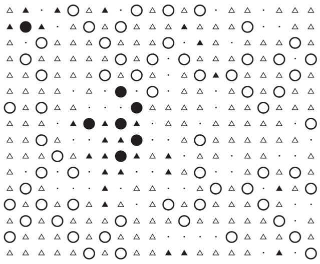
The changes of spin structure S(b) with $J_2/J_1$= -0.36 are shown in Fig.6 for various temperatures in the range of $0.1 \leqq k_BT/J_1 \leqq 2.0$. As the Curie temperature for $J_2/J_1$=-0.36 is $k_BT_c/J_1$ =1.8, the spin structure (a) is at a paramagnetic state and the spin structures (b), (c), (d) are at an ordered state.

The rates of occupation of a spin $S_z$=1 on S-spin site in ordered state below $T_c$ are 66%, 93%, 100% at temperatures $k_BT/J_1$=1.7, 0.9, 0.1, respectively. On the other hand, the rates of occupation of a spin $R_z$=1 on R-spin site are 40%, 64%, 96% at temperatures $k_BT/J_1$=1.7, 0.9, 0.1, respectively. It is remarkable that the rates of the occupation of $S_z$=1 on S-spin site are larger than those of $R_z$=1 on R-spin site at any temperature below $T_c$. The rates of occupation of a spin $R_z$=2 on R-spin site are 38%, 35%, 4% at temperatures $k_BT/J_1$=1.7, 0.9, 0.1, respectively.

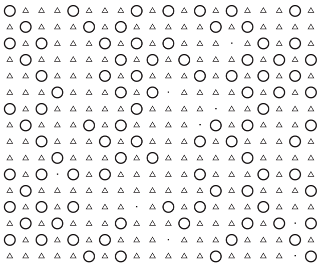
Therefore, the rise just below $T_c$ of the $\langle R_z \rangle$ curve may be understood by considering the facts that a large number of spins with $R_z$=2 exist just below $T_c$ and decrease gradually with decreasing temperature.



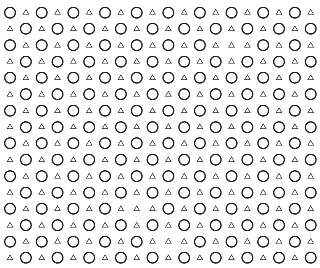
(a)



(b)

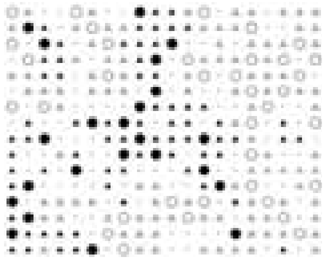


(c)

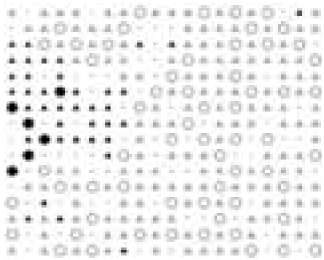


(d)

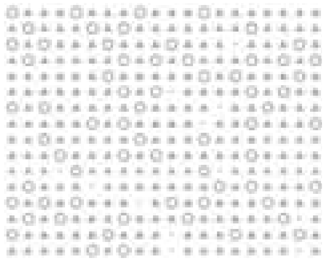
**Fig. 5 Temperature dependence of spin structure S(a) of the mixed spin system of $S$=1 and $R$=2 with $J_2/J_1$=-0.3 calculated by the MC simulation for various temperatures in the range of $0.1 \leqq k_BT/J_1 \leqq 2.0$. (a), (b), (c), (d) represent the spin structures at $k_BT/J_1$= 2.0, 1.8, 1.0, 0.1, respectively. Open and closed circles, and open and closed triangles denote $R_Z = \pm 2$ and $R_Z= S_Z = \pm 1$, and dot denote $R_Z= S_Z$ =0, respectively.**



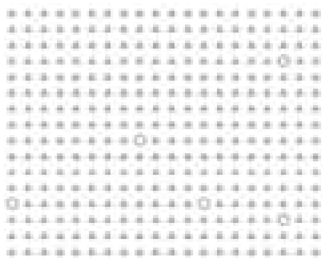
(a)



(b)



(c)



(d)

**Fig. 6 Temperature dependence of spin structure S(b) of the mixed spin system of $S$=1 and $R$=2 with $J_2/J_1$=-0.36 calculated by the MC simulation for various temperatures in the range of 0.1≦$k_BT/J_1$≦2.0. (a), (b), (c), (d) represent the spin structures at $k_BT/J_1$= 2.0, 1.7, 0.9, 0.1, respectively.**

Next, we have investigated the thermal averages $<S_z^2>$ and $<R_z^2>$ of the mixed Ising spin system of $S$=1 and $R$=2 with the GS spin structure S(c). The temperature dependences of $< R_z^2>/R^2$ and $< S_z^2>/S^2(=<S_z^2>)$ of the mixed spin system for various values of interaction $J_2$ in the range of -2.0 ≦ $J_2/J_1$ ≦ -1.0 are shown in Fig.7 and Fig.8, respectively.

As can be seen from Fig.7, the value of $<R_z^2>/R^2$ at $T$=0 are 0.49 for the interaction $J_2$ in the range of -2.0<$J_2/J_1$<-1.0. This fact may suggest that the number of spins of $R_z$=0 are slightly larger than those of $R_z$=±2 or $R_z$=±1. The value of $< R_z^2>/R^2$ at $T$=0 are 0.35 for the interaction $J_2$ with $J_2/J_1$=-1.0. This fact is understood by considering that the number of spins of $R_z$=1 are larger than those of $R_z$=±2 or $R_z$=-1 or $R_z$=0, as the phase transition between S(b) and S(c) occurs at this condition of $J_2/J_1$=-1.0. The interaction $J_2$ turns out not to give a significant effect on the temperature dependence of $< R_z^2>/R^2$.

As can be seen from Fig.8, the value of $<S_z^2>/S^2$ at $T$=0 are 0.005 for the interaction $J_2$ in the range of -2.0<$J_2/J_1$<-1.0. This fact may suggest that the GS spin structure with $S_z$=0 on S-site contains a slight number of spin of $S_z$=1. The value of $<S_z^2>/S^2$ at $T$=0 are 0.14 for the interaction $J_2$ with $J_2/J_1$=-1.0. This fact is understood by considering that a large number of spins of $S_z$=1 exist, as the phase transition between S(b) and S(c) occurs at this condition of $J_2/J_1$=-1.0. The existence of $S_z$=1 on S-site are consistent with that of larger number of spins of $R_z$=0 on R-site. The interaction $J_2$ turns out to give larger effects on the temperature dependence of $<S_z^2>/S^2$ than that of $< R_z^2>/R^2$.

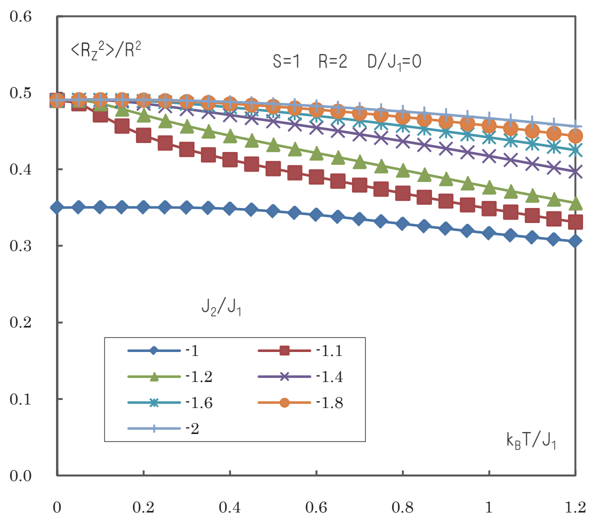


**Fig. 7 Temperature dependence of $<R_z^2>/R^2$ of the mixed spin system with $S$=1 and $R$=2 calculated by the MC simulation for various values $J_2$ of in the range of -2.0≦$J_2/J_1$≦-1.0.**

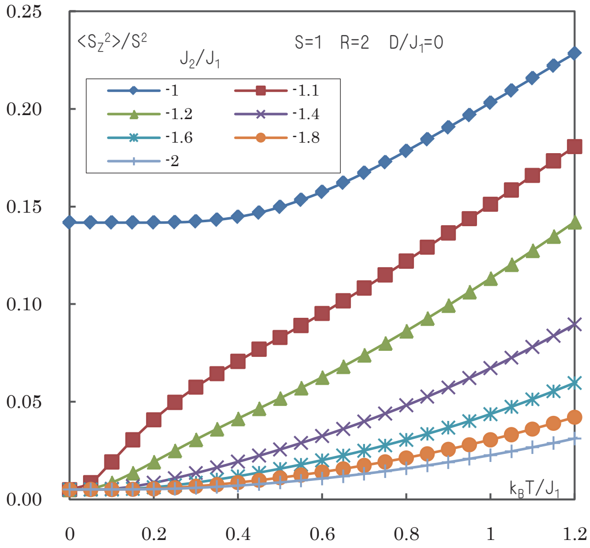
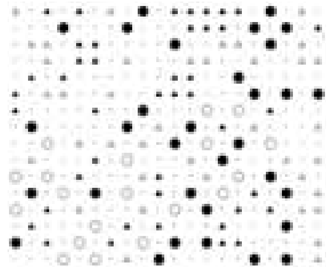


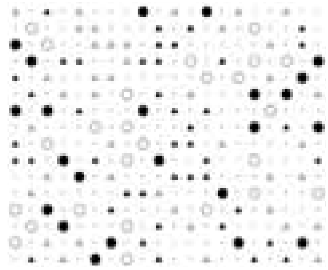

**Fig. 8 Temperature dependence of $<S_z^2>/S^2$ of the mixed spin system with $S$=1 and $R$=2 calculated by the MC simulation for various values $J_2$ of in the range of -2.0≦$J_2/J_1$≦-1.0.**

Next, we have investigated the changes of spin structure with the GS spin structure S(c) for various values of temperature by making use of the MC simulation and visualized the change of this structure. The changes of spin structure S(c) with interaction $J_2/J_1$=-1.2 are shown in Fig.9 for various temperatures in the range of 0.1≦$k_BT/J_1$≦1.2.

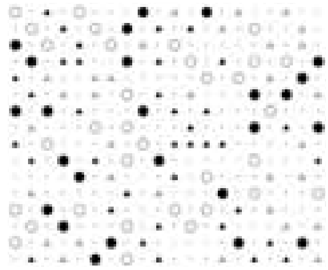
The rates of occupation of a spin $S_z$=0 and $S_z$=1 on S-spin site are 86%, 91%, 96%, 99.5% and 14%, 9%, 4%, 0.5% at temperatures $k_BT/J_1$=1.2, 0.8, 0.4 and 0.1, respectively. On the other hand, the rates of occupation of a spin $R_z$=0 on R-spin site are 40% and 21.6% at temperatures $k_BT/J_1$=1.2 and 0.1, respectively. It is interesting that the increase of the rates of occupation of a spin $S_z$=1 on S-spin site is related with that of a spin $R_z$=0 on R-spin site with increasing temperature.



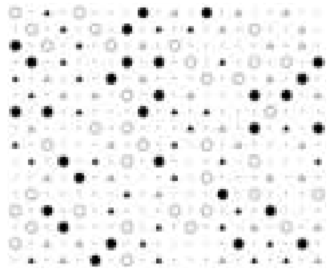
(a)



(b)



(c)



(d)

**Fig. 9 Temperature dependence of spin structure S(c) of the mixed spin system of $S$=1 and $R$=2 with $J_2/J_1$=-1.2 calculated by the MC simulation for various temperatures in the range of 0.1≦$k_BT/J_1$≦1.2. (a), (b), (c), (d) represent the spin structures at $k_BT/J_1$= 1.2, 0.8, 0.4, 0.1, respectively.**

### (B) Magnetic Properties of Spin System with Positive Anisotropy ($D/J_1$>0)

Next, let us investigate the magnetic properties such as the magnetizations $<S_z>$ and $<R_z>$, and the thermal averages $<S_z^2>$ and $<R_z^2>$ of the mixed Ising spin system of $S$=1 and $R$=2 with positive anisotropy $D$>0 by making use of the MC simulation. The temperature dependences of $<R_z>/R$ and $<S_z>/S$ (=$<S_z>$) of the mixed spin system with anisotropy $D$=0.5 for various values of interaction $J_2$ in the range of -0.7≦$J_2/J_1$≦-0.1 are shown in Fig.10 and Fig.11, respectively.

As can be seen from Fig.10, the values of $<R_z>/R$ at $T$=0 are 1.0, 0.5 and 0 for the interaction $J_2$ in the range of -0.45≦$J_2/J_1$, -0.65<$J_2/J_1$<-0.45 and $J_2/J_1$≦-0.65, respectively. These facts suggest that the phase change occurs at $J_2/J_1$=-0.45 and -0.65. It should be noticeable that for the interaction in the range of -0.65<$J_2/J_1$<-0.45 which correspond to the spin structure S(b), the curve of $<R_z>/R$ show rapid increase just below $T_c$ and negative temperature dependence in the middle temperature range.

It is remarkable that the temperature dependence curves of $<S_z>/S$ for $J_4/J_1$=-0.45 at the phase transition show a depression at low temperatures. As can be seen from Fig.11, for the interaction in the range of -0.65<$J_2/J_1$<-0.45, the curve of $<S_z>/S$ show rapid increase just below $T_c$ and linear dependence in the middle temperature range. Furthermore, for the interaction in this range of -0.65<$J_2/J_1$<-0.45, the values of $<S_z>/S$ are constant and take one for the temperature of $k_BT/J_1$<0.3.

It is interesting that the curves of $<S_z>/S$ for the interaction in the range of -0.65<$J_2/J_1$<-0.45 have the same temperature dependence for the temperature of $k_BT/J_1$<0.6. The Curie temperature turns out to disappear suddenly at the same time of disappearing of $<R_z>/R$ and $<S_z>/S$.

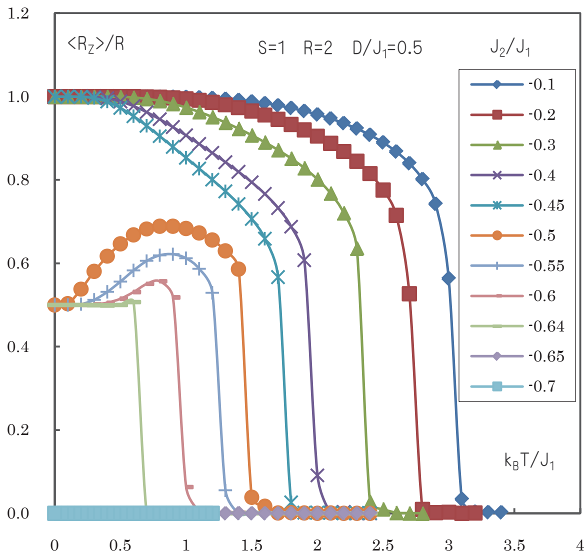



**Fig. 10 Temperature dependence of $<R_z>/R$ of the mixed spin system of $S$=1 and $R$=2 with anisotropy $D$=0.5 calculated by the MC simulation for various values $J_2$ of in the range of -0.7≦$J_2/J_1$≦-0.1.**

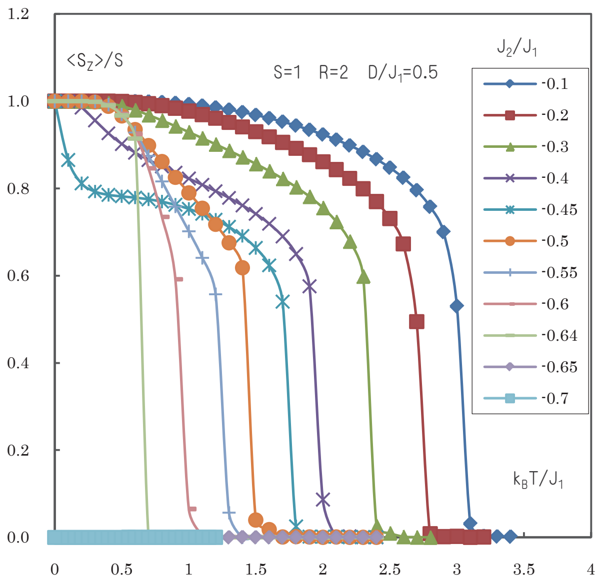
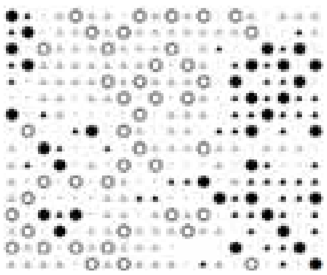



**Fig.11 Temperature dependence of $<S_z>/S$ of the mixed spin system of $S$=1 and $R$=2 with anisotropy $D$=0.5 calculated by the MC simulation for various values $J_2$ of in the range of -0.7≦$J_2/J_1$≦-0.1.**

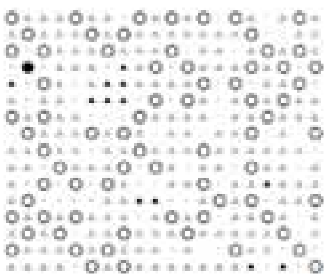
Next, we have investigated the changes of spin structure with the GS spin structures S(b) for various values of temperature by making use of the MC simulation and visualized the change of this structure. The changes of spin structure S(b) with $J_2/J_1$=-0.5 and $D/J_1$=0.5 is shown in Fig.12 for various temperatures in the range of 0.1≦$k_BT/J_1$≦1.6. As the Curie temperature for $J_2/J_1$= -0.3 is $k_BT_c/J_1$ =1.5, the spin structure (a) is at a paramagnetic state and the spin structures (b), (c), (d), (e) are at an ordered state.

The rates of occupation of a spin $S_z$=1 on S-spin site in an ordered state below $T_c$ are 62%,86%, 99%, 100% at temperatures $k_BT/J_1$=1.4, 0.8, 0.4, 0.1, respectively. On the other hand, the rates of occupation of a spin $R_z$=2 on R-spin site are 18%, 38%, 23%, 1% at temperatures $k_BT/J_1$=1.4, 0.8, 0.4, 0.1, respectively. It is interesting that the rates of the occupation of $S_z$=1 on S-spin site are larger than 99% for low temperature range of $k_BT/J_1$≦0.4.

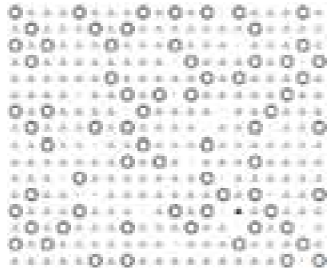
It should be noticeable that the rate of occupation of a spin $R_z$=2 on R-spin site for the GS spin structure S(b) with $S_z$=1 and $R_z$=1 is very large and 38% at the temperature $k_BT/J_1$=0.8. Moreover, this large value of the rate of a spin $R_z$=2 keeps in the wide temperature range. The GS spin structure on R-site appears only at very low temperatures of $k_BT/J_1$<0.05.



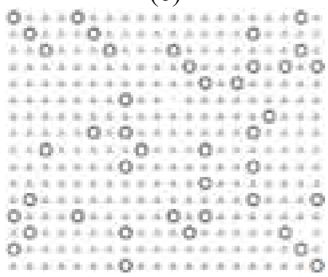
(a)



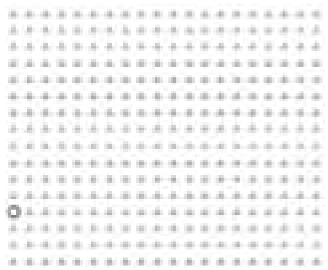
(b)



(c)



(d)



(e)

**Fig. 12 Temperature dependence of spin structure S(b) of the mixed spin system of $S$=1 and $R$=2 with $J_2/J_1$=-0.5 and $D/J_1$=0.5 calculated by the MC simulation for various temperatures in the range of 0.1≦$k_BT/J_1$≦1.6. (a), (b), (c), (d), (e) represent the spin structures at $k_BT/J_1$= 1.6, 1.4, 0.8, 0.4, 0.1, respectively.**

Next, we have investigated the thermal averages $<S_z^2>$ and $<R_z^2>$ of the mixed Ising spin system of $S$=1 and $R$=2 with the GS spin structure S(d). The temperature dependences of $< R_z^2>/R^2$ and $< S_z^2>/S^2(=<S_z^2>)$ of the mixed spin system with $D/J_1$=0.5 for various values of interaction $J_2$ in the range of -1.2≦$J_2/J_1$≦-0.65 are shown in Fig.13 and Fig.14, respectively.

As can be seen from Fig.13, the dependence of $< R_z^2>/R^2$ on the interaction parameter $J_2$ is small even at high temperatures. The value of $< R_z^2>/R^2$ is one in the temperature range of 0≦$k_BT/J_1$≦0.2 regardless of the interaction $J_2$. On the other hand, the interaction $J_2$ gives large effects on the value of $< R_z^2>/R^2$ at high temperatures. Furthermore, the temperature ranges with $<S_z^2>$=0 depend largely on the interaction $J_2$, and are $k_BT/J_1 \leqq 0.35$, 0.45, 0.6, 0.7, 0.8, 0.9, 0.98 for the interaction $J_2/J_1$=-0.65, -0.7, -0.8, -0.9, -1.0, -1.1, -1.2, respectively.

The temperature dependences of $< R_z^2>/R^2$ and $< S_z^2>/S^2(=<S_z^2>)$ of the mixed spin system with $D/J_1$=1.0 for various values of interaction $J_2$ in the range of $-1.0 \leqq J_2/J_1 \leqq -0.57$ are shown in Fig.15 and Fig.16, respectively.

As can be seen from Fig.15, the values of $< R_z^2>/R^2$ don't show effective dependence on the interaction parameter $J_2$ even at high temperatures. The value of $< R_z^2>/R^2$ is one in the temperature range of $0 \leqq k_BT/J_1 \leqq 0.4$ regardless of the interaction $J_2$. On the other hand, the interaction $J_2$ gives large effects on the value of $< S_z^2>/S^2$ at high temperatures. Furthermore, the temperature ranges with $<S_z^2>=0$ depend largely on the interaction $J_2$, and are $k_BT_c/J_1 \leqq 0.01$, 0.15, 0.45, 0.75, 0.95, 1.1 for the interaction $J_2/J_1$=-0.57, -0.6, -0.7, -0.8, -0.9, -1.0, respectively. Therefore, the complete spin ordering with $S_z$=0 on S-site appears at higher temperature than that with $R_z$=2 on R-site for the interaction $J_2/J_1 \leqq -0.7$.

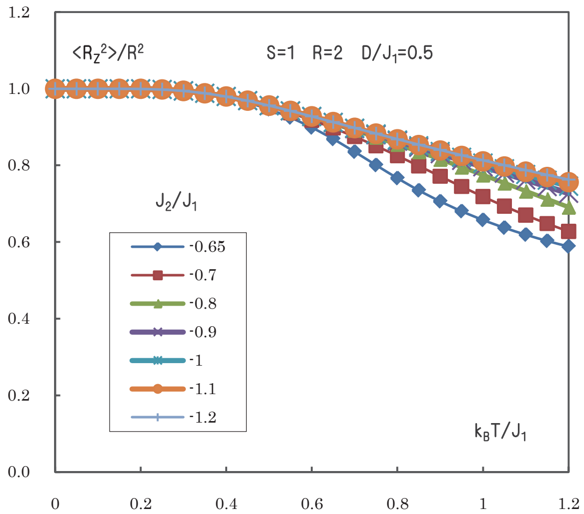



**Fig. 13 Temperature dependence of $<R_z^2>/R^2$ of the mixed spin system of $S$=1 and $R$=2 with $D/J_1$=0.5 calculated by the MC simulation for various values $J_2$ of in the range of $-1.2 \leqq J_2/J_1 \leqq -0.65$.**

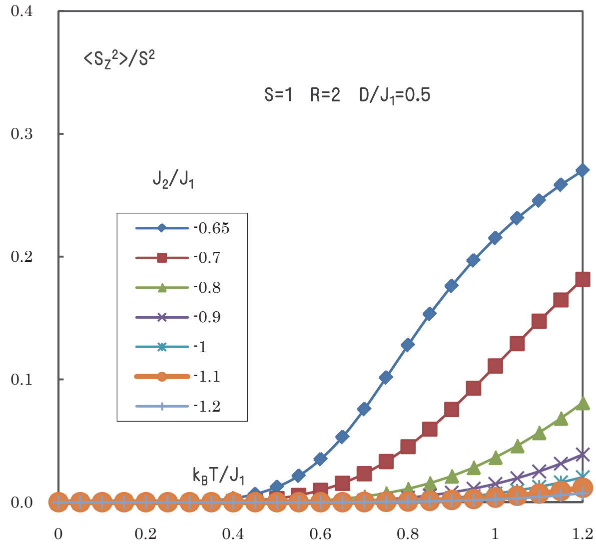



**Fig. 14 Temperature dependence of $<S_z^2>/S^2$ of the mixed spin system of $S$=1 and $R$=2 with $D/J_1$=0.5 calculated by the MC simulation for various values $J_2$ of in the range of $-1.2 \leqq J_2/J_1 \leqq -0.65$.**

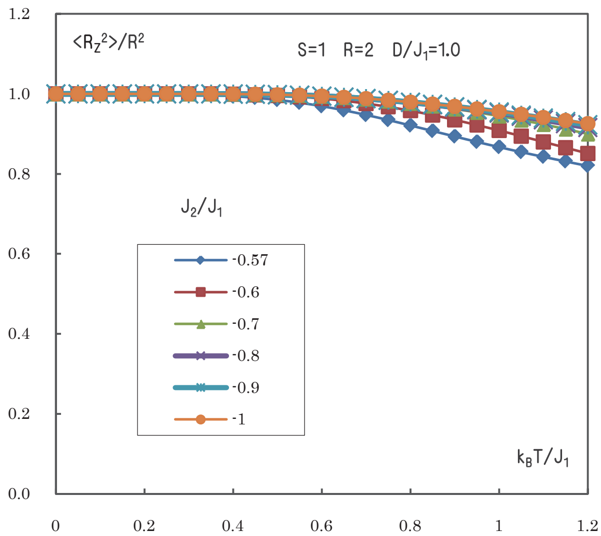



**Fig. 15 Temperature dependence of $<R_z^2>/R^2$ of the mixed spin system of $S$=1 and $R$=2 with $D/J_1$=1.0 calculated by the MC simulation for various values $J_2$ of in the range of $-1.0 \leqq J_2/J_1 \leqq -0.57$.**

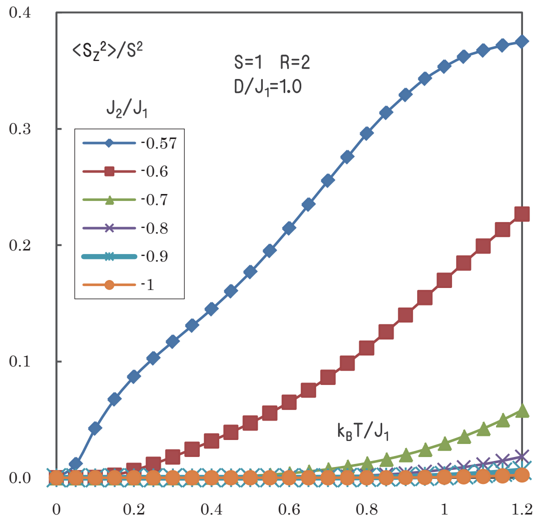


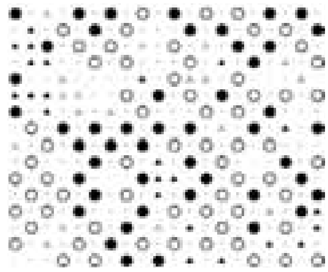

**Fig. 16 Temperature dependence of $\langle S_z^2\rangle/S^2$ of the mixed spin system of *S*=1 and *R*=2 with $D/J_1$=1.0 calculated by the MC simulation for various values $J_2$ of in the range of -1.0≦$J_2/J_1$≦-0.57.**

Next, we have investigated the changes of spin structure with the GS spin structure S(d) for various values of temperature by making use of the MC simulation and visualized the change of this structure. The changes of spin structure S(d) with $J_2/J_1$=-0.8 and $D/J_1$=0.5 are shown in Fig.17 for various temperatures in the range of 0.2≦$k_BT/J_1$≦1.2.

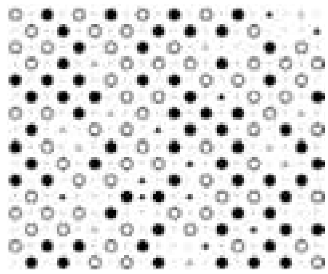
The rates of occupation of a spin $S_z$=0 and $S_z$=1 on S-spin site are 92%, 99%, 100%, 100% and 8%, 1%, 0%, 0% at temperatures $k_BT/J_1$=1.2, 0.8, 0.4 and 0.2, respectively. On the other hand, the rates of occupation of spins $R_z$=±2 on R-spin site are 68%, 87%, 97% and 100% at temperatures $k_BT/J_1$=1.2, 0.8, 0.4 and 0.1, respectively. It is interesting that the rates of occupation of a spin $S_z$=0 on S-spin site are much larger than that of a spin $R_z$=±2 on R-spin site at any temperatures.

The changes of spin structure S(d) with $J_2/J_1$=-0.8 and $D/J_1$=1.0 are shown in Fig.18 for various temperatures in the range of 0.4≦$k_BT/J_1$≦1.2.

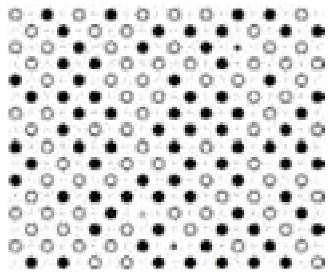
The rates of occupation of a spin $S_z$=0 and $S_z$=1 on S-spin site are 98%, 100%, 100%, and 2%, 0%, 0% at temperatures $k_BT/J_1$=1.2, 0.8, and 0.4, respectively. On the other hand, the rates of occupation of spins $R_z$=±2 on R-spin site are 91%, 97% and 100% at temperatures $k_BT/J_1$=1.2, 0.8 and 0.4, respectively. The rates of occupation of a spin $S_z$=0 on S-spin site and of a spin $R_z$=±2 on R-spin site for $D/J_1$=1.0 turn out to be much larger than those for $D/J_1$=0.5 at any temperatures.



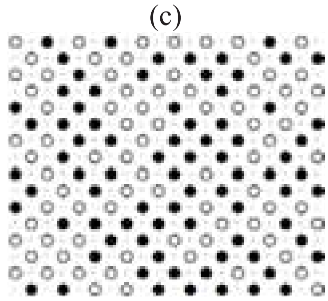
(a)



(b)

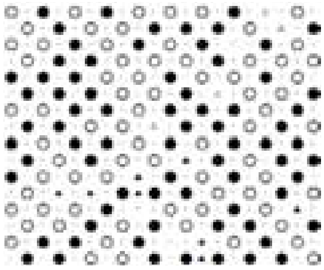


(c)

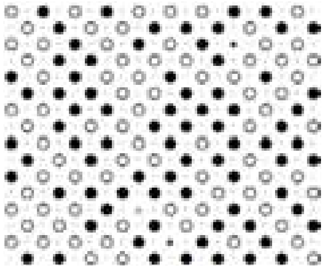


(d)

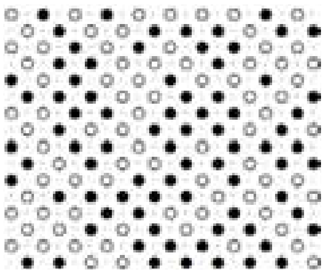
**Fig. 17 Temperature dependence of spin structure S(d) of the mixed spin system of *S*=1 and *R*=2 with $J_2/J_1$=-0.8 and $D/J_1$=0.5 calculated by the MC simulation for various temperatures in the range of 0.2≦$k_BT/J_1$≦1.2. (a), (b), (c), (d) represent the spin structures at $k_BT/J_1$= 1.2, 0.8, 0.4, 0.2, respectively.**



(a)



(b)



(c)

**Fig. 18 Temperature dependence of spin structure S(d) of the mixed spin system of $S$=1 and $R$=2 with $J_2/J_1$= -0.8 and $D/J_1$=1.0 calculated by the MC simulation for various temperatures in the range of 0.4≦$k_BT/J_1$≦1.2. (a), (b), (c) represent the spin structures at $k_BT/J_1$= 1.2, 0.8, 0.4, respectively.**

## (C) Magnetic Properties of Spin System with Negative Anisotropy ($D/J_1$<0)

Next, let us investigate (C) Magnetic properties such as the magnetizations $\langle S_z\rangle$ and $\langle R_z\rangle$, and the thermal averages $\langle S_z^2\rangle$ and $\langle R_z^2\rangle$ of the mixed Ising spin system of $S$=1 and $R$=2 with negative anisotropy $D$<0 by making use of the MC simulation. The temperature dependences of $\langle R_z\rangle/R$ and $\langle S_z\rangle/S$ (=$\langle S_z\rangle$) of the mixed spin system with anisotropy $D$=-0.5 for various values of interaction $J_2$ in the range of -0.8≦$J_2/J_1$≦-0.1 are shown in Fig.19 and Fig.20, respectively.

As can be seen from Fig.19, the values of $\langle R_z\rangle/R$ at $T$=0 are 1.0, 0.5 and 0 for the interaction $J_2$ in the range of -0.21≦$J_2/J_1$, -0.75<$J_2/J_1$<-0.21 and $J_2/J_1$≦-0.75, respectively. These facts suggest that the phase change occurs at the conditions of $J_2/J_1$=-0.21 and -0.75. It should be noticeable that for the interaction in the range of -0.35<$J_2/J_1$< -0.21 which correspond to the spin structure S(b), the curve of $\langle R_z\rangle/R$ show rapid increase just below $T_c$ and negative temperature dependence in the middle range below $T_c$.

It should be remarkable that the temperature dependence curves of $\langle S_z\rangle/S$ for $J_2/J_1$=-0.21 at the phase transition show a depression at low temperatures. As can be seen from Fig.20, for the interaction in the range of -0.75<$J_2/J_1$<-0.21, the curve of $\langle S_z\rangle/S$ show rapid increase just below $T_c$ and swift dependence. Furthermore, with approaching the interaction $J_2/J_1$ to -0.75, the values of $\langle S_z\rangle/S$, $\langle R_z\rangle/R$ and the Curie temperature $T_c$ turns out to disappear at the same time.

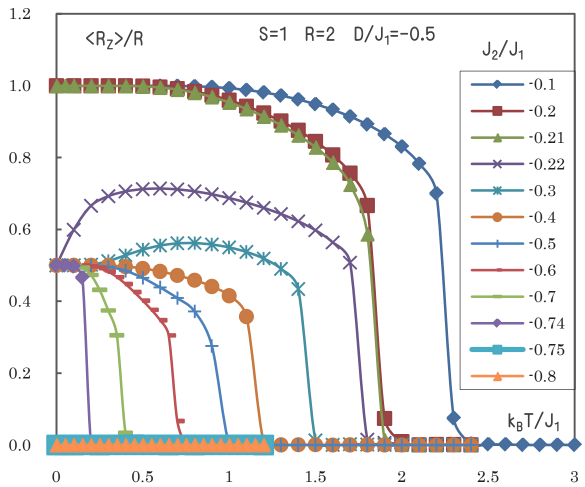



**Fig. 19 Temperature dependence of $\langle R_z\rangle/R$ of the mixed spin system of $S$=1 and $R$=2 with anisotropy $D$=-0.5 calculated by the MC simulation for various values $J_2$ of in the range of -0.7≦$J_2/J_1$≦ -0.1.**

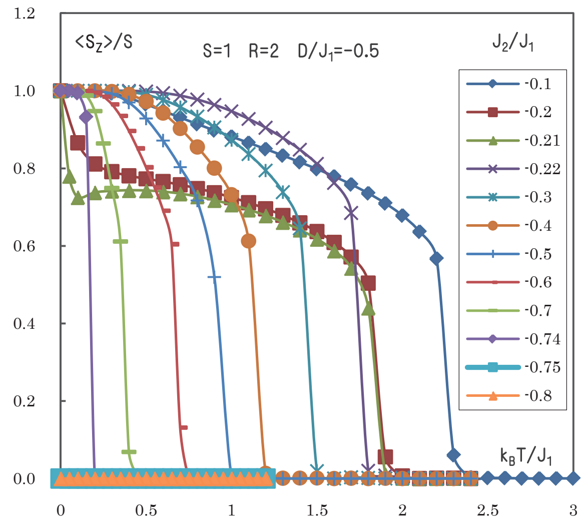



**Fig.20 Temperature dependence of $<S_z>/S$ of the mixed spin system of $S$=1 and $R$=2 with anisotropy $D$=-0.5 calculated by the MC simulation for various values $J_2$ of in the range of -0.7≦$J_2/J_1$≦ -0.1.**

Next, we have investigated the thermal averages $<S_z^2>$ and $<R_z^2>$ of the mixed Ising spin system of $S$=1 and $R$=2 with the GS spin structure S(e). The temperature dependences of $< R_z^2>/R^2$ and $< S_z^2>/S^2(=<S_z^2>)$ of the mixed spin system with $D/J_1$=-0.5 for various values of interaction $J_2$ in the range of -1.5≦$J_2/J_1$≦-0.75 are shown in Fig.21 and Fig.22, respectively.

As can be seen from Fig.21, the dependence of $< R_z^2>/R^2$ on the interaction parameter $J_2$ is small especially at high temperatures. For the interaction $J_2$ in the range of -0.8≦$J_2/J_1$≦-0.75, the value of $< R_z^2>/R^2$ shows rapid decrease with decreasing temperature and becomes zero at $k_BT/J_1$=0.08. It is quite interesting that the value of $< R_z^2>/R^2$ is zero in the temperature range of 0 ≦ $k_BT/J_1$ ≦ 0.08 regardless of the interaction $J_2$.

It can be seen from Fig.22 that the dependence of $< S_z^2>/S^2$ on the interaction parameter $J_2$ is large at all temperatures. For all interaction $J_2$ in the range of $J_2/J_1$≦-0.75, the value of $< S_z^2>/S^2$ shows rapid decrease with decreasing temperature and becomes zero at $k_BT/J_1$=0.08. It is noticeable that both values of $< R_z^2>/R^2$ and $< S_z^2>/S^2$ is zero in the temperature range of 0≦$k_BT/J_1$≦0.08 regardless of the interaction $J_2$.

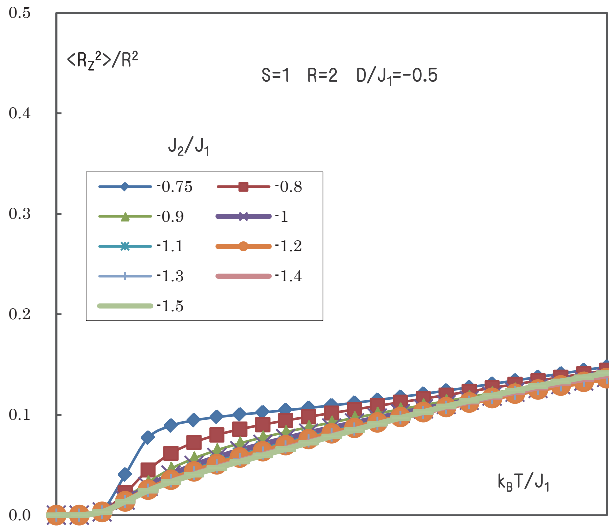



**Fig. 21 Temperature dependence of $<R_z^2>/R^2$ of the mixed spin system of $S$=1 and $R$=2 with $D/J_1$=-0.5 calculated by the MC simulation for various values $J_2$ of in the range of -1.3≦$J_2/J_1$≦-0.75.**

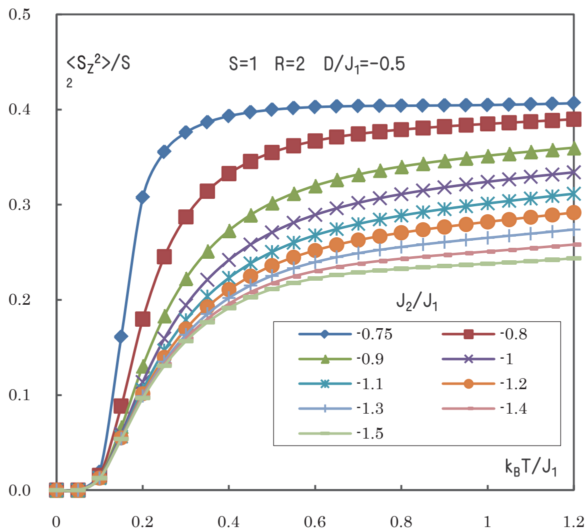
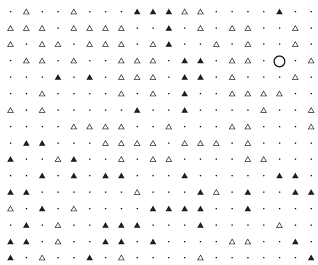


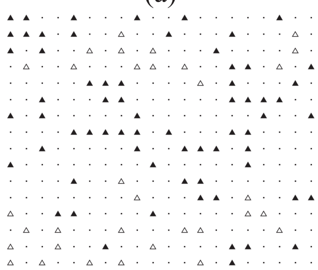

**Fig. 22 Temperature dependence of $<S_z^2>/S^2$ of the mixed spin system of $S$=1 and $R$=2 with $D/J_1$=-0.5 calculated by the MC simulation for various values $J_2$ of in the range of -1.3≦$J_2/J_1$≦-0.75.**

Next, we have investigated the changes of spin structure with the GS spin structures S(e) for various values of temperature by making use of the MC simulation and visualized the change of this structure. The changes of spin structure S(e) with $J_2/J_1$=-0.8 and $D/J_1$=-0.5 is shown in Fig.23 for various temperatures in the range of 0.05≦ $k_BT/J_1$≦0.8.

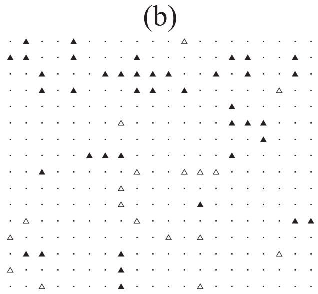
The rates of occupation of a spin $S_z$=0 on S-spin site are 62%, 67%, 82%, 91%, 98%, 100% at temperatures $k_BT/J_1$=0.8, 0.4, 0.2, 0.15, 0.1, 0.05, respectively. On the other hand, the rates of occupation of a spin $R_z$=0 on R-spin site are 54%, 65%, 82%, 91%, 98%, 100% at temperatures $k_BT/J_1$=0.8, 0.4, 0.2, 0.15, 0.1, 0.05, respectively. It is interesting that the rates of the occupation of $S_z$=0 on S-spin site and $R_z$=0 on R-spin site are the same for low temperature range of $k_BT/J_1$≦0.4.



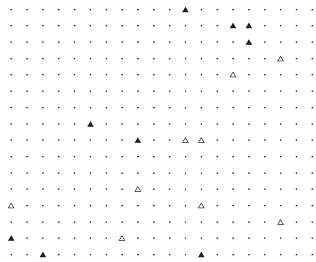
(a)



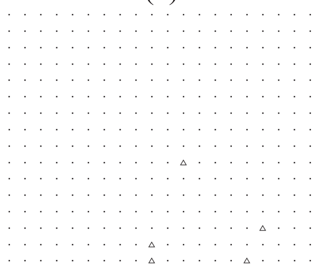
(b)



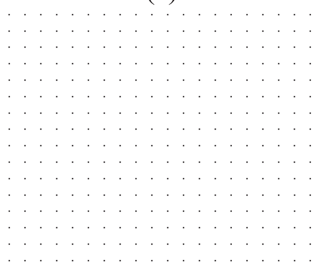
(c)



(d)



(e)



(f)

**Fig. 23 Temperature dependence of spin structure S(d) of the mixed spin system of *S*=1 and *R*=2 with $J_2/J_1$=-0.8 and $D/J_1$=-0.5 calculated by the MC simulation for various temperatures in the range of 0.05≦$k_BT/J_1$≦0.8. (a), (b), (c), (d), (e) represent the spin structures at $k_BT/J_1$= 0.8, 0.4, 0.2, 0.15, 0.1, 0.05, respectively.**

## 4. Concluding Remarks

In the previous section, for the mixed Ising spin system of $S$=1 and $R$=2 with the bilinear exchange interaction $J_1S_{iz}R_{jz}$, the Biquadratic exchange interaction $J_2S_{iz}^2R_{jz}^2$ and a single-ion anisotropy $D(S_{iz}^2+R_{jz}^2)$, the magnetization $<S_z>$ and $<R_z>$, the Curie temperature $T_c$ and the GS spin structures have been calculated for two-dimensional square lattice 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 mixed Ising spin system of *S*=1 and *R*=2 with interaction $J_2/J_1$ and anisotropy $D/J_1$ is obtained by the MC simulation. The conditions of phase transition and the GS spin structures determined by this MC simulation show good agreements with those calculated from the comparison of energies per one spin for various spin structures with low energy except the one between S(b) and S(d).

(2) The new five magnetic phases S(a) ～S(e) are found for parameters $J_2/J_1$ and anisotropy $D/J_1$. Especially, three phases S(c), S(d) and S(e) correspond to zero, positive, negative values of anisotropy term *D*, respectively.

(3) The anisotropy term *D* does not give large effects on the temperature dependence of $<R_z>/R$, but give significant effects on the temperature dependence of $<S_z>/S$. The effects of the anisotropy term *D* on $<S_z>/S$ are large in the middle range for $D>0$ and just below $T_c$ for $D<0$.

(4) The temperature dependences of $<R_z>/R$ and $<S_z>/S$ for S(b) show unique behaviors in the case of positive anisotropy *D*. From the extrapolation of $T_c$ shown in Fig.10 and in Fig.11, $T_c$ turns out to disappear at the condition of $J_2/J_1$ =-0.75. This condition agree well with the one obtained from energy comparison.

(5) For *D*=0, the values of $<R_z^2>/R^2$ and $<S_z^2>/S^2$ at *T*=0 are 0.49 and 0.005, but not 0.5 and 0, respectively. These facts suggest that the number of $R_z$=0 is slightly larger than that $R_z=\pm2$ or $\pm1$, and the number of $S_z=\pm1$ slightly exists in the case of *D*=0.

(6) For positive anisotropy (D>0), the complete arrangement of $S_z$=0 on S-site is accomplished at higher temperatures than that of $R_z=\pm2$ on R-site. On the other hand, for negative anisotropy (D<0), the complete arrangements of $S_z$=0 on S-site and of $R_z$=0 on R-site are accomplished at the same temperature.

**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 **20**, 101 (2010).
15. T. Iwashita and N. Uryu, *J. Phys*. Condens. Matter **3**, 8257 (1991).