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Ground state of an Ising ferromagnet with three-site four-spin interaction

Takashi Iwashita† and Norikiyo Uryû‡
† College of General Education, Kyushu Tokai University, Kumamoto 862, Japan
‡ Department of Applied Science, Faculty of Engineering, Kyushu University, Fukuoka 812, Japan

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Abstract. For an Ising ferromagnet with the bilinear exchange interaction J and the three-site four-spin interaction J', the ground-state spin structures are determined from the J'-dependence of the energy of a single spin in various spin structures. It is pointed out that the phase transitions occur at $J' = -J/4$ for the linear-chain Ising ferromagnet of spin $S = 1$ and at $J' = -J/9$ and $J' = -3J/22$ for the Ising ferromagnet of spin $S = 1$ on a two-dimensional square lattice. For the Ising ferromagnet of spin $S = \frac{3}{2}$ on a two-dimensional square lattice, it turns out that the phase transitions also occur at $J' = -2J/45$, $J' = -4J/75$ and $J' = -5J/78$. Furthermore, it is made clear that the phase transitions at $J' = -J/9$ and $J' = -2J/45$ agree well with those obtained from the anomalous behaviour of the magnetization for Ising ferromagnets of $S = 1$ and $S = \frac{3}{2}$, respectively.

1. Introduction

In ferromagnets and antiferromagnets the existence and importance of higher-order exchange interactions such as the biquadratic exchange interaction $(S_i \cdot S_j)^2$, the three-site four-spin interaction $(S_i \cdot S_j)(S_j \cdot S_k)$ and the four-site four-spin interaction $(S_i \cdot S_j)(S_k \cdot S_l)$ have been discussed extensively by many investigators [1].

In previous studies [2], the present authors have discussed the appearance of a higher-order exchange interaction of the types $(S_i \cdot S_j)(S_j \cdot S_k)$ and estimated the order of magnitude of these terms as one tenth or one thousandth of the Heisenberg exchange interaction.

Subsequently, the effects of the interaction $(S_i \cdot S_j)(S_j \cdot S_k)$ on the temperature dependence of the magnetization of the Ising ferromagnet of $S \geqslant 1$ have been investigated, and it has been pointed out that the interaction with a negative value changes the ground state from the ferromagnetic spin structure to a complicated structure [1, 3, 4].

In previous studies [5], we have investigated and determined the ground-state spin structure of the Ising ferromagnet with the biquadratic exchange interaction of negative sign. Therefore, it is interesting to investigate the ground-state spin structure of the Ising ferromagnet with the three-site four-spin interaction of negative sign.

In the present paper, for Ising ferromagnets of $S = 1$ and $S = \frac{3}{2}$ with the three-site four-spin interaction $J'(S_i \cdot S_j)(S_j \cdot S_k)$, the ground-state spin structure is determined from the J'-dependence of the energy for a single spin in various spin structures. Furthermore, the condition for the phase change is obtained for the Ising ferromagnet with the three-site four-spin interaction of negative sign. The ground-state spin structure and the condition for the phase change are discussed for a linear-chain Ising ferromagnet of $S = 1$ and for Ising ferromagnets of $S = 1$ and $S = \frac{3}{2}$ on a two-dimensional square lattice.

In section 2, the Hamiltonian of the present spin systems of $S = 1$ and $S = \frac{3}{2}$ is approximated by making use of the three-spin model approximation [6]. Then, the self-consistent simultaneous equations to calculate the thermal averages of S_z and S_z^3 are derived with the use of the eigenvalues obtained from the above Hamiltonian. Furthermore, the dependence of the magnetization on the three-site four-spin interaction parameter J' is calculated numerically for a negative value of J'. The condition for an occurrence of the phase change is obtained from the numerical results for the Ising ferromagnet on the two-dimensional square lattice. In section 3, the J'-dependence of the energy for a single spin in various spin structures is obtained, and the ground-state spin structure and the condition for the phase change are determined for various values of J'. In the final section, concluding remarks are given.

2. Theory

We consider the ferromagnetic Ising spin system in which the bilinear exchange interaction and the three-site four-spin interaction are assumed. Then, the spin Hamiltonian for the present spin system is written as

$$H = -J \sum_{\langle ij \rangle} S_{iz} S_{jz} - 2J' \sum_{\langle ijk \rangle} S_{iz} S_{jz}^2 S_{kz}. \tag{1}$$

The Hamiltonian (1) may be approximated by making use of the three-spin model approximation [6] as given by equations (3) and (4) in [1]. From the approximate Hamiltonian (3) in [1], 27 and 64 eigenvalues are obtained for the ferromagnetic spin systems of $S = 1$ and $S = \frac{3}{2}$, respectively.

With the use of these eigenvalues λ_m, the thermal averages $\langle S_z \rangle$ and $\langle S_z^3 \rangle$ can be calculated as a function of temperature from the following equations:

$$\langle S_z \rangle = \sum_m \tfrac{1}{3}(S_{iz} + S_{jz} + S_{kz}) \exp(-\beta\lambda_m) \Big/ \sum_m \exp(-\beta\lambda_m) \tag{2}$$

$$\langle S_z^3 \rangle = \sum_m \tfrac{1}{4}(S_{iz}^2 S_{jz} + S_{iz} S_{jz}^2 + S_{jz}^2 S_{kz} + S_{jz} S_{kz}^2) \exp(-\beta\lambda_m) \Big/ \sum_m \exp(-\beta\lambda_m) \tag{3}$$

where $\beta = 1/k_B T$ and k_B is the Boltzmann constant.

The J'-dependence of $\langle S_z \rangle$ has been obtained by numerical calculation for negative values of J' in the ranges $-0.12J \leqslant J' \leqslant 0$ and $-0.05J \leqslant J' \leqslant 0$ for the Ising ferromagnets of $S = 1$ and $S = \frac{3}{2}$, respectively. These negative values can appear through various mechanisms [2] in the real magnetic systems. The temperature dependences of $\langle S_z \rangle$ for the ferromagnetic Ising spin systems of $S = 1$ and $S = \frac{3}{2}$ on the square lattice ($z = 4$) have been calculated and the results for $S = 1$ and $S = \frac{3}{2}$ are shown in figure 1 and figure 2, respectively.

As can be seen from figure 1 and figure 2, the effect of the three-site four-spin interaction on the temperature dependence of $\langle S_z \rangle$ is large at low temperatures. It is noticed that the temperature dependence of $\langle S_z \rangle$ for the Ising spin systems of $S = 1$ and $S = \frac{3}{2}$ shows an abnormal behaviour at low temperatures for the three-site four-spin interaction of $J' \leqslant -0.11J$ and $J' \leqslant -0.045J$, respectively. These anomalous behaviours suggest that the ferromagnetic ground state becomes unstable at $J' \simeq -0.11J$ and $J' \simeq$

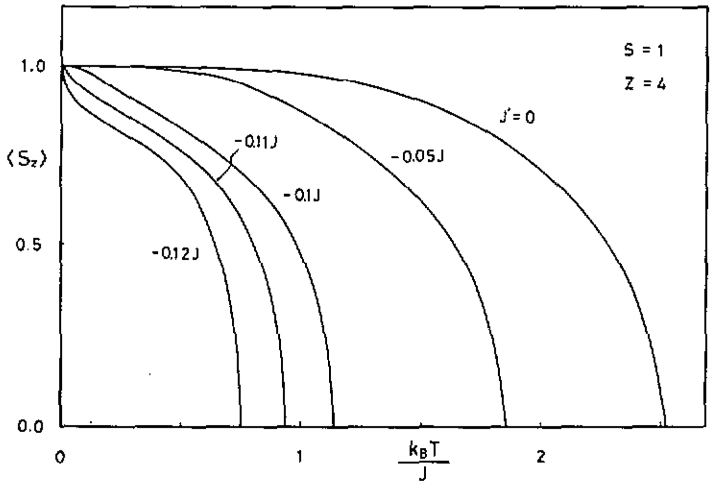

Figure 1. The temperature dependence of $\langle S_z \rangle$ for the ferromagnetic Ising spin system of $S = 1$ on a two-dimensional square lattice ($z = 4$) with various values of the three-site four-spin interaction J' in the range $-0.12J \leqslant J' \leqslant 0$.

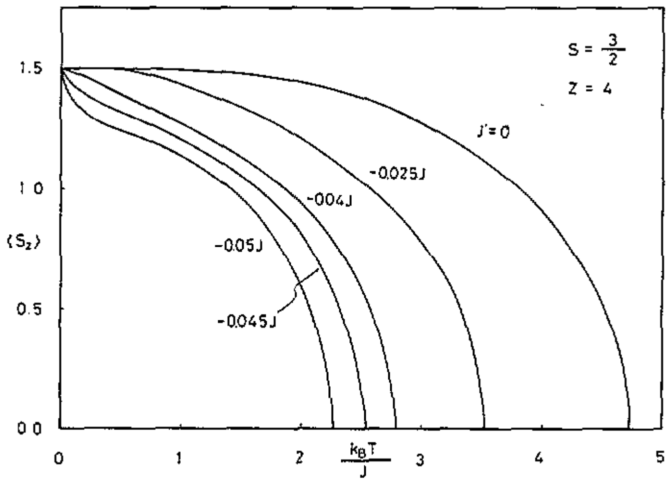

Figure 2. The temperature dependence of $\langle S_z \rangle$ for the ferromagnetic Ising spin system of $S = \frac{3}{2}$ on a two-dimensional square lattice ($z = 4$) with various values of the three-site four-spin interaction J' in the range $-0.05J \leqslant J' \leqslant 0$.

$-0.045J$ for the Ising spin systems of $S = 1$ and $S = \frac{3}{2}$, respectively. Below these values of J', a complicated ferrimagnetic spin structure may appear as the ground state.

3. Spin structure and energy

Let us consider the spin structures of the Ising spin system with the bilinear exchange interaction J and the three-site four-spin interaction J'. It can easily be concluded that

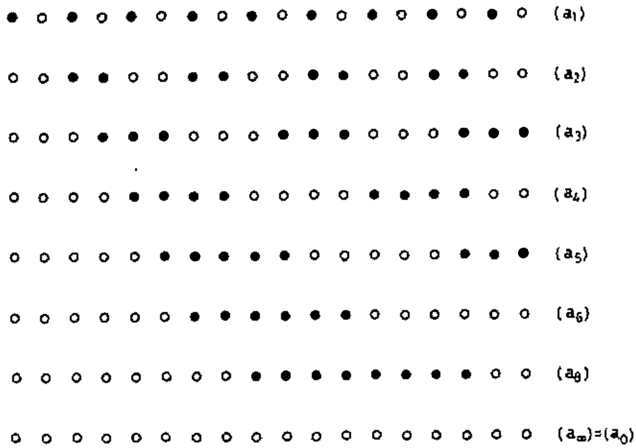

Figure 3. The spin structures of the Ising spin system of $S = 1$ on a linear chain: ○, $S_z = 1$; ●, $S_z = -1$.

the spin system with interaction parameters both of positive sign, i.e. $J > 0$ and $J' > 0$, has a ferromagnetic ground state. Starting from the ferromagnetic spin structure, the excited states may be those in which some spins have turned from the state of $S_z = S$ to those of $S_z = S - 1$ and $S_z = S - 2$ and so on. Here we assume that the lattice sites with excited spins constitute a periodic structure.

3.1. *Ising spin system of S = 1 on a linear chain (z = 2)*

We consider the various spin structures and their energies of the spin system of $S = 1$ on a linear chain. Firstly, the Ising spin system with periodic spin structure consisting of the spins $S_z = 1$ and $S_z = -1$ will be examined. Here, the periodic spin structure consisting of the successive n spins of $S_z = 1$ followed by another successive n spins of $S_z = -1$ will be expressed as (a_n), and the energy for a single spin in this spin structure is denoted as $\varepsilon(a_n)$.

The spin structures for the cases $n = 1$–6, $n = 8$ and $n = \infty$ are shown in figure 3. The spin structure for the case $n = \infty$ is the same as the ferromagnetic spin structure (a_0). The energies $\varepsilon(a_n)$ for various structures (a_n) are as follows:

$$\begin{aligned} &\varepsilon(a_1) = J - 2J' && \varepsilon(a_2) = 2J' \\ &\varepsilon(a_3) = -\tfrac{1}{3}J + \tfrac{2}{3}J' && \varepsilon(a_4) = -\tfrac{1}{2}J \\ &\varepsilon(a_5) = -\tfrac{3}{5}J - \tfrac{2}{5}J' && \varepsilon(a_6) = -\tfrac{2}{3}J - \tfrac{2}{3}J' \\ &\varepsilon(a_8) = -\tfrac{3}{4}J - J' && \varepsilon(a_\infty) = -J - 2J'. \end{aligned} \tag{4}$$

The J'-dependences of $\varepsilon(a_n)$ are shown in figure 4. As can be seen, all lines other than that for $\varepsilon(a_1)$ cross each other at the point $J' = -J/4$. The ferromagnetic spin structure (a_0) turned out to be of the lowest energy in the range $J' \geqslant -J/4$, and the spin structure (a_2) is the lowest in the range $J' < -J/4$.

Let us consider the case $J = 0$ in the (a_n)-structures. In this case, the J'-dependences of $\varepsilon(a_n)$ are also obtained from equations (4) by putting $J = 0$. It is noticed that $\varepsilon(a_1)$ and

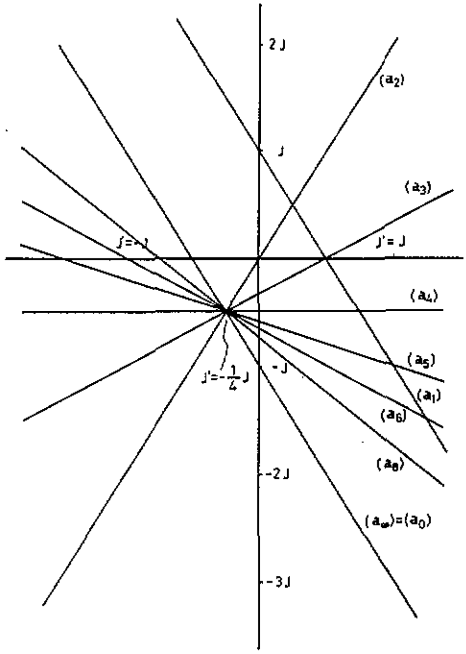

Figure 4. The J'-dependences of the energy for a single spin in the various spin structures of $S = 1$ on a linear chain.

$\varepsilon(a_0)$ coincide with each other, the spin structures (a_1) and (a_0) are the lowest in the range $J' \geqslant 0$ and the spin structure (a_2) is the lowest in the range $J' < 0$.

Next, we consider the other periodic spin structure in which the successive n spins $S_z = 1$ are separated by an isolated single spin $S_z = -1$. This spin structure will be expressed as (b_n), and the energy for a single spin in this structure is denoted as $\varepsilon(b_n)$.

From the calculation of the J'-dependences of $\varepsilon(b_n)$, it is shown that all lines other than that for $\varepsilon(b_1)$ cross each other at the point $J' = -J/2$. In the range $J' < -J/4$, all spin structures (b_n) have a higher energy than the spin structure (a_2).

In the same way, we consider the periodic spin structure in which the successive n spins $S_z = 1$ are separated by an isolated pair of spins $S_z = -1$. This spin structure will be expressed as (c_n) and the energy for a single spin in this structure is denoted as $\varepsilon(c_n)$. It can be shown that all lines other than that for $\varepsilon(c_1)$ cross each other at the point $J' = -J/4$. In the range $J' < -J/4$, the spin structure $(c_2)(=(a_2))$ can be seen to have the lowest energy.

Furthermore, we consider the periodic spin structure in which the successive n spins $S_z = 1$ are separated by an isolated group of three spins $S_z = -1$. This spin structure will be expressed as (d_n), and the energy for a single spin in this structure is denoted as $\varepsilon(d_n)$. Except for the line for $\varepsilon(d_1)$, all lines are shown to cross each other at the point $J' = -J/4$. In the range $J' < -J/4$, all spin structures (d_n) have a higher energy than the spin structure (a_2).

Finally, we consider the periodic spin structure in which the successive n spins $S_z = 1$ are separated by an isolated single spin $S_z = 0$. This spin structure will be expressed as (e_n), and the energy for a single spin in this structure is denoted as $\varepsilon(e_n)$. All lines except that for $\varepsilon(e_1)$ are shown to cross each other at the point $J' = -J/3$. In the range $J' < -J/4$, all structures (e_n) can be said to have a higher energy than the spin structure (a_2).

As can be seen from the behaviours of the J'-dependences of $\varepsilon(a_1)$ in figure 4, the periodic spin structures with an isolated single spin cannot be the ground state. Other than those spin structures with an isolated single spin, all lines of the J'-dependences of the energies turn out to cross at the point $J' = -J/4$ in the spin structures of $S_z = 1$ and $S_z = -1$. Furthermore, it is pointed out that in the spin structures (a_n), (c_n) and (d_n) the corresponding energies $\varepsilon(a_n)$, $\varepsilon(c_n)$ and $\varepsilon(d_n)$ become higher with increase in n in the range $J' < -J/4$. On the other hand, in the case of spin structures consisting of $S_z = 1$ and $S_z = 0$, all lines of the J'-dependences of the energies cross at the point $J' = -J/3(< -J/4)$ other than that with an isolated single spin $S_z = 1$. It can be confirmed by further calculations for other structures consisting of $S_z = 1$ and $S_z = 0$ that the lines of the J'-dependences of the energies cross at a smaller constant value of J' than $J' = -J/4$. Thus, the spin structures consisting of $S_z = 1$ and $S_z = 0$ turn out as unable to be the ground state of the Ising spin system of $S = 1$ on a linear chain. Therefore, we may conclude that the ferromagnetic spin structure (a_0) and the spin structure (a_2) are the ground state of the Ising spin system on a linear chain in the range $J' \geqslant -J/4$ and $J' < -J/4$, respectively.

The energy of the Ising spin system with an aperiodic spin structure is given by a linear combination of those with periodic spin structures. This may be understood by dividing the aperiodic spin structure on the linear chain into various periodic segments. Therefore, the energy of the aperiodic spin structure is expected to have an intermediate value of the energies for the periodic spin structures. Thus, it is concluded that the aperiodic spin structure cannot be the ground state.

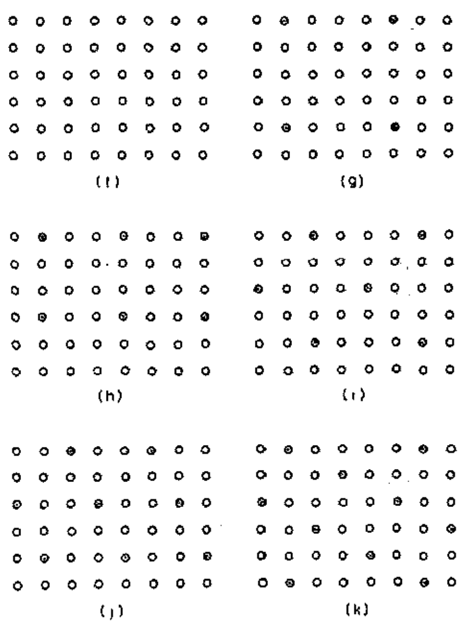

Figure 5. The spin structures of the Ising spin system of $S = 1$ on a two-dimensional square lattice: ○, $S_z = 1$; ⊙, $S_z = 0$.

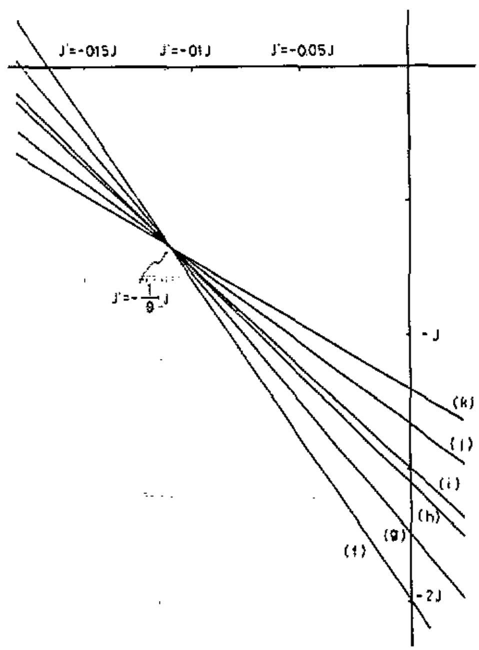

Figure 6. The J'-dependences of the energy for a single spin in the various spin structures of $S = 1$ on a two-dimensional square lattice.

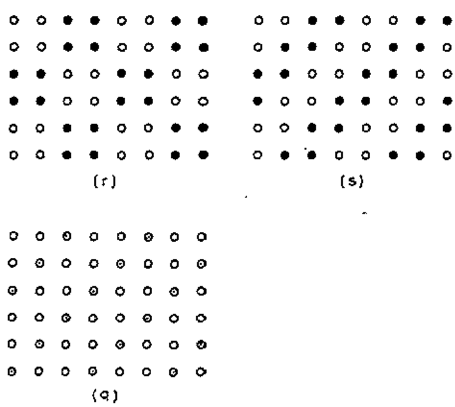

Figure 7. The spin structures of the Ising spin system of $S = 1$ on a two-dimensional square lattice: ○, $S_z = 1$; ⊙, $S_z = 0$; ●, $S_z = -1$.

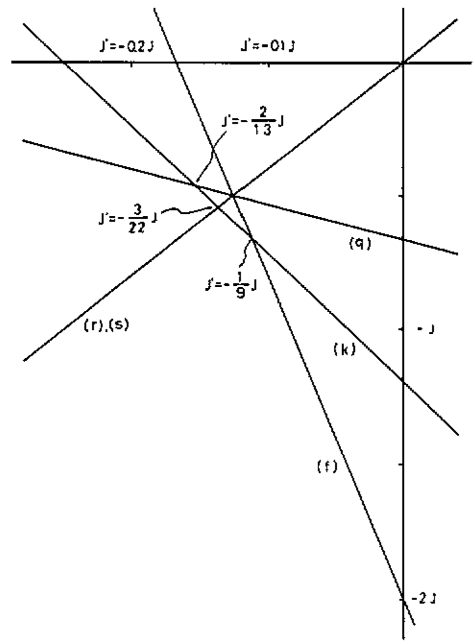

Figure 8. The J'-dependences of the energy for a single spin in the spin structures with the lowest energy on a two-dimensional square lattice.

3.2. *Ising spin system of S = 1 on a two-dimensional square lattice (z = 4)*

We consider the various spin structures and their energies of the spin system of $S = 1$ on a two-dimensional square lattice. The spin structure arranged ferromagnetically with spins $S_z = 1$ is shown by (f) in figure 5. The energy for a single spin in this spin structure is denoted as ε_f and is given by $\varepsilon_f = -2J - 12J'$. The spin structure in which a single spin in a cluster of α spins has $S_z = 0$ and the rest $\alpha - 1$ spins have $S_z = 1$ will be designated as the α-structure. The spin structures for the cases $\alpha = 16, 9, 8, 6$, and 5 are shown by (g), (h), (i), (j) and (k), respectively, in figure 5. The energy for a single spin in the α-structure is denoted by $\varepsilon(\alpha)$ and is given as follows:

$$\begin{aligned} &\varepsilon_g(16) = -\tfrac{7}{4}J - \tfrac{39}{4}J' \qquad && \varepsilon_h(9) = -\tfrac{14}{9}J - 8J' \\ &\varepsilon_i(8) = -\tfrac{3}{2}J - \tfrac{15}{2}J' && \varepsilon_j(6) = -\tfrac{4}{3}J - 6J' \\ &\varepsilon_k(5) = -\tfrac{4}{3}J - 6J'. \end{aligned} \tag{5}$$

In the above spin structures, the sites with spin $S_z = 0$ are beyond the limit of the interaction J' of each other. The J'-dependences of the energy for a single spin in the various spin structures (f)–(k) are shown in figure 6. As can be seen, all lines (f)–(k) cross each other at the point $J' = -J/9$. It turns out that the spin structure (k) has the lowest energy in the range $J' < -J/9$.

Next, we consider the spin structure in which the sites with spin $S_z = 0$ are within the limit of the interaction J' of each other. In the spin structures which satisfy this condition, the spin structure (q) shown in figure 7 turns out to have the lowest energy in the range

$J' < -2J/13$. The J'-dependence of the energy for a single spin in this spin structure is shown by the line (q) in figure 8, and this line (q) crosses with the one for the spin structure (k) at $J' = -2J/13$.

In the previous section, we have investigated the ground state of the linear chain Ising ferromagnet. The spin structure (a_2) has been concluded to be the ground state in the negative region of $J'(J' < -J/4)$. Now, let us apply this result of linear chain Ising ferromagnet to the case of square lattice. In the two spin structures (r) and (s) shown in figure 7, the spin arrangements of both longitudinal and transverse components agree with that of spin structure (a_2). In any other spin structures, the spin arrangement of both components does not agree at the same time with the spin structure (a_2). The energy for a single spin in the spin structures (r) and (s) is given by

$$\varepsilon_r(2) = \varepsilon_s(2) = 4J'. \tag{6}$$

The J'-dependence of the energy for a single spin in these spin structures is shown by the same line (r) and (s) in figure 8. As can be seen from this figure, the line for the spin structures (r) and (s) crosses with the one for the spin structure (k) at the point $J' = -3J/22$ which is larger than the value of $J' = -2J/13$.

Therefore, we may conclude that the ferromagnetic spin structure (f) and the spin structure (k) are the ground state in the range of $J' \geqslant -J/9$ and $-3J/22 \leqslant J' \leqslant -J/9$, respectively. Furthermore, the spin structures (r) and (s) are the ground states in the range $J' < -3J/22$.

3.3. *Ising spin system of $S = \frac{3}{2}$ on a two-dimensional square lattice ($z = 4$)*

We consider the various spin structures and their energies of the spin system of $S = \frac{3}{2}$ on a two-dimensional square lattice. In this case, S_z takes four values $\pm\frac{3}{2}$ and $\pm\frac{1}{2}$. We have calculated the energies for a single spin in the various spin structures with $S = \frac{3}{2}$. The J'-dependences of these energies will be compared with each other in the same way as has been done in the case of $S = 1$ in the previous section.

The various spin structures with the lowest energy are shown by (a)–(e) in figure 9. The energies for a single spin in these spin structures are given as follows:

$$\begin{aligned} \varepsilon_a(\infty) &= -\tfrac{9}{2}J - \tfrac{243}{4}J' \\ \varepsilon_b(5) &= -\tfrac{33}{10}J - \tfrac{135}{4}J' \\ \varepsilon_c(3) &= -\tfrac{5}{2}J - \tfrac{75}{4}J' \\ \varepsilon_d(2) &= \varepsilon_e(2) = \tfrac{81}{4}J'. \end{aligned} \tag{7}$$

The J'-dependences of the energy for a single spin in the various spin structures ((a)–(e)) are shown in figure 10. The lines (a) and (b), and the lines (b) and (c) cross each other at the point $J' = -2J/45$ and $J' = -4J/75$, respectively. Furthermore, lines (c)–(e) cross each other at $J' = -5J/78$. All lines of the J'-dependences of the energy for the other spin structures are located higher than these lines ((a)–(e)).

Therefore, we may conclude that the ferromagnetic spin structure (a) and the spin structures (b) and (c) are the ground state in the ranges $J' \geqslant -2J/45$, $-4J/75 \leqslant J' < -2J/45$ and $-5J/78 \leqslant J' < -4J/75$, respectively. Furthermore, the spin structures (d) and (e) are the ground states in the range $J' < -5J/78$.

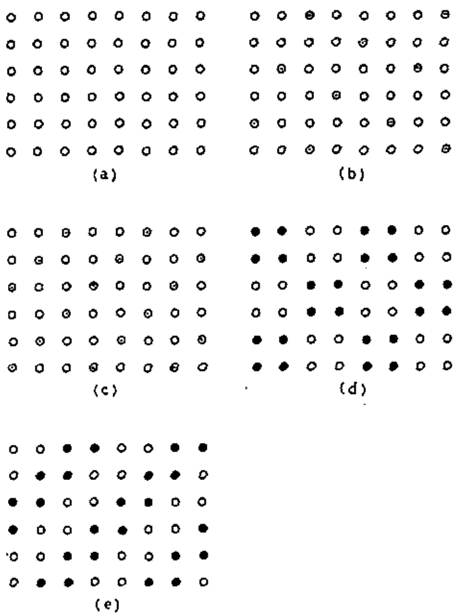

Figure 9. The spin structures of the Ising spin system of $S = \frac{3}{2}$ on a two-dimensional square lattice: ○, $S_z = \frac{3}{2}$; ⊙, $S_z = \frac{1}{2}$; ●, $S_z = -\frac{3}{2}$.

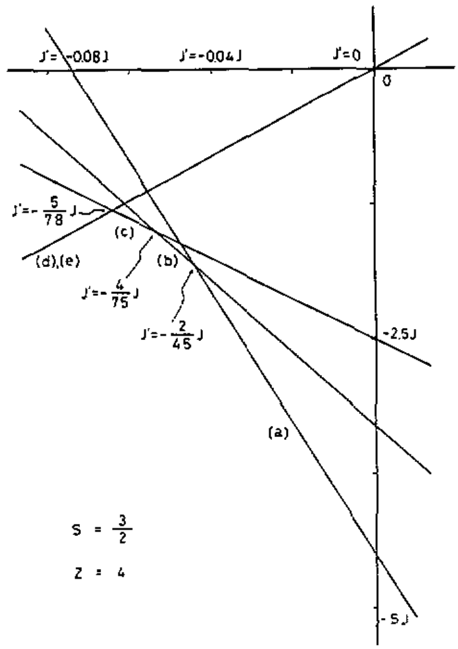

Figure 10. The J'-dependences of the energy for a single spin in the spin structures with the lowest energy on a two-dimensional square lattice.

4. Concluding remarks

In the previous section, for the Ising ferromagnets of spin $S = 1$ and $S = \frac{3}{2}$ with the bilinear exchange interaction J and the three-site four-spin interaction J', the ground-state spin structure has been obtained from the behaviour of the J'-dependences of the energy for a single spin in the various spin structures.

Summarizing the present results, we may conclude as follows.

(1) The phase transition occurs at $J' = -J/4$ for the spin-1 linear-chain Ising ferromagnet with J and J'. The spin structures (a_0) and (a_2) are the ground-state spin structures in the linear-chain Ising ferromagnet in the range $J' \geqslant -J/4$ and $J' < -J/4$, respectively. On the other hand, the phase transition occurs at $J' = 0$ for the linear-chain Ising ferromagnet only with J'. The spin structures (a_0) and (a_2) are the ground states in the linear-chain Ising ferromagnet only with J' in the range of $J' \geqslant 0$ and $J' < 0$, respectively.

(2) The phase transitions occur at $J' = -J/9$ and $J' = -3J/22$ for spin-1 Ising ferromagnet with both interactions J and J' on the two-dimensional square lattice. The condition $J' = -J/9$ of the phase transition obtained from this method agrees well with that obtained from the behaviour of the temperature dependence of $\langle S_z \rangle$ for the spin-1 Ising ferromagnet at low temperatures. The spin structures (f) and (k) are the ground state in the ranges $J' \geqslant -J/9$ and $-3J/22 \leqslant J' < -J/9$, respectively. Furthermore, the spin structures (r) and (s) are the ground states in the range $J' < -3J/22$.

(3) The phase transition occurs at $J' = 0$ for the Ising ferromagnet only with J' on the two-dimensional square lattice. The result can easily be obtained by replacing the

term of J in ε_f, and ε_g–ε_k in equation (5), and ε_r and ε_s in equation (6) by zero. The spin structure (f) and the spin structures (r) and (s) are the ground states in the ranges $J' \geqslant 0$ and $J' < 0$, respectively.

(4) The phase transitions occur at $J' = -2J/45$, $J' = -4J/75$ and $J' = -5J/78$ for the Ising ferromagnet of $S = \frac{3}{2}$ with both interactions J and J' on the two-dimensional square lattice. The condition $J' = -2J/45$ of the phase transition obtained from this method agrees well with that obtained from the behaviour of the temperature dependence of $\langle S_z \rangle$ for the Ising ferromagnet of $S = \frac{3}{2}$ at low temperatures. The spin structures (a), (b) and (c) are the ground states in the ranges $J' \geqslant -2J/45$, $-4J/75 \leqslant J' < -2J/45$ and $-5J/78 \leqslant J' < -4J/75$, respectively. Furthermore, the spin structures (d) and (e) are the ground states in the range of $J' < -5J/78$. In the case of $J = 0$, the ground-state spin structure changes from (a) to (d) and (e) at $J' = 0$.

(5) At the point of the phase change such as $J' = -J/4$, $J' = -J/9$, $J' = -3J/22$, $J' = -2J/45$, $J' = -4J/75$ and $J' = -5J/78$, many spin structures coexist as the ground state.

(6) It has been made clear that the magnitude of the three-site four-spin interaction due to various mechanisms amounts to about $|J'/J| = 10^{-1}$–10^{-3}. Therefore, it may be possible to detect the effect of the three-site four-spin interaction on the phase transition in the Ising ferromagnets of spin $S = 1$ and $S = \frac{3}{2}$.

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