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Curie Temperatures and Tricritical Points in Mixed Ising Ferromagnetic Systems

Takahito KANEYOSHI

Department of Physics, Nagoya University, Nagoya 464

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The critical behavior of a mixed ferromagnetic Ising spin system consisting of spin $-\frac{1}{2}$ and spin-1 with a crystal-field interaction is investigated by the use of the effective-field theory with correlations. The general expressions for evaluating the Curie temperature and the tricritical point are obtained. We find that the tricritical point exists in the system with $Z>3$, where Z is the coordination number.

§1. Introduction

In Ising models there exist many systems exhibiting the tricritical point at which the phase transition changes from second order to first order. In particular, spin-one Ising systems containing a crystal-field interaction or biquadratic exchange interactions are such a typical system and have been extensively investigated by many authors.[1)]

Recently, attention[2,3)] has been directed to two-sublattice mixed spin $-\frac{1}{2}$ and spin-1 Ising ferromagnetic systems with biquadratic exchange interactions, and the effects of the biquadratic exchange interactions on the Curie temperature have been discussed by the use of several techniques. However, it has not been clarified whether or not the systems exhibit a tricritical behavior.

On the other hand, using the differential operator technique introduced by Honmura and Kaneyoshi,[4)] we have studied the general spin-1 Ising model with random bond and crystal-field interactions, and obtained some general expressions for determining the second-order phase transition lines and the tricritical point within the framework of the effective-field theory with correlations (EFT).[5)] The theory is based on the use of rigorous Ising spin identities. Although it is mathematically simple, this approach which is conceptually as simple as the standard mean-field theory, shares with other methods a great versatility.

The purpose of this work is to present some extensions of the referred effective-field approach to nonrandom two-sublattice mixed spin $-\frac{1}{2}$ and spin-1 Ising models with crystal-field interactions. The main aim is to investigate whether or not the mixed Ising systems may exhibit the tricritical behavior. Furthermore, the present model is closely related to the mixed ferromagnetic Ising systems with a biquadratic exchange interaction.[2,3)] From this approach we can also know whether or not the tricritical behavior may exist in the systems.

The outline of this paper is as follows. In §2, we briefly present the general formulation of a mixed Ising spin system with a crystal-field interaction within the EFT. In §3, some general expressions for evaluating the Curie temperature and the tricritical point are obtained. In §4, the change of Curie temperature with a crystal-field interaction and the tricritical point are investigated for honeycomb ($Z=3$), square ($Z=4$) and simple cubic ($Z=6$) lattices, where Z is the coordination number. We find that the tricritical point exists in a mixing Ising spin system with $Z>3$.

§2. Formulation

We consider a two-sublattice spin $-\frac{1}{2}$ and spin-1 mixed ferromagnetic Ising model with a crystal-field interaction Δ. The Hamiltonian of the system is given by

$$\mathcal{H}=-J\sum_{\langle ij\rangle}S_i^z\mu_j^z+\Delta\sum_i(S_i^z)^2, \tag{1}$$

where S_i^z takes the values ± 1 and 0, μ_j^z can be $+1$ or -1, and the first summation is carried out only over nearest-neighbor pairs of spins.

J is the nearest-neighbor exchange interaction parameter.

On the other hand, the model Hamiltonian (1) is closely related to the mixed Ising model with a biquadratic exchange interaction J_1 studied by some authors;[2,3] the Hamiltonian is then given by

$$\mathcal{H} = -2J\sum_{\langle ij\rangle} S_i^z \sigma_j^z - 2J_1 \sum_{\langle ij\rangle} (S_i^z)^2(\sigma_j^z)^2, \quad (2)$$

where σ_j^z takes the values $\pm\frac{1}{2}$. The second term of (2) is reduced to $-(J_1/2)Z\sum_i (S_i^z)^2$, which is a single-ion potential. Z is the number of nearest neighbors. Thus, the formulation for the Hamiltonian (1) which will be studied in the following can easily be extended to the case of (2).

The problem is now the evaluation of the mean values $\langle S_i^z\rangle$ and $\langle \mu_j^z\rangle$. As discussed in a series of works,[6] the formal identities for the spin correlation functions for the Ising models can be used. The starting point for the evaluation of $\langle S_i^z\rangle$ and $\langle \mu_j^z\rangle$ are the exact identities[6]

$$m = \langle S_i^z\rangle = \left\langle \frac{2\sinh(\beta\theta_i)}{2\cosh(\beta\theta_i)+\exp(\Delta\beta)}\right\rangle, \quad (3)$$

and

$$\sigma = \langle \mu_j^z\rangle = \langle \tanh(\beta\bar{\theta}_j)\rangle, \quad (4)$$

with

$$\theta_i = J\sum_j \mu_j^z$$
$$\bar{\theta}_j = J\sum_i S_i^z, \quad (5)$$

where

$$\beta = \frac{1}{k_B T}.$$

Introducing the differential operator D ($D=(\partial/\partial x)$) into eqs. (3) and (4), one may rewrite them in the following forms

$$m = \langle \pi_\delta \{\cosh(DJ) + \mu_{i+\delta}^z \sinh(DJ)\}\rangle g(x)|_{x=0}, \quad (6)$$

and

$$\sigma = \langle \pi_\delta \{(S_{j+\delta}^z)^2 \cosh(DJ) + S_{j+\delta}^z \sinh(DJ) + 1 - (S_{j+\delta}^z)^2\}\rangle f(x)|_{x=0}, \quad (7)$$

where π indicates the product over all the nearest neighbors of the site i (or the site j). The functions $f(x)$ and $g(x)$ are defined by

$$f(x) = \tanh(\beta x), \quad (8)$$

and

$$g(x) = \frac{2\sinh(\beta x)}{2\cosh(\beta x)+\exp(\Delta\beta)}. \quad (9)$$

Equations (6) and (7) constitute a set of exact relations, according to which we can study the present model. However, if we try to exactly treat all the spin-spin correlations for the set of equations, the problem quickly becomes untractable. A first obvious attempt to deal with it is to ignore correlations; the decoupling approximations

$$\langle \mu_i^z \mu_j^z \cdots \mu_k^z\rangle \cong \langle \mu_i^z\rangle\langle \mu_j^z\rangle\cdots\langle \mu_k^z\rangle,$$
$$\langle S_m^z (S_n^z)^2 \cdots S_l^z\rangle \cong \langle S_m^z\rangle\langle (S_n^z)^2\rangle\cdots\langle S_l^z\rangle, \quad (10)$$

with $i\neq j\neq\cdots\neq k$ and $m\neq n\neq\cdots\neq l$ have been introduced within the effective-field theory with correlations (EFT). In fact, the approximation corresponds essentially to the Zernike approximation for a system with $S=\frac{1}{2}$, and has been successfully applied to a great number of magnetic systems.[6,7] Then, the set of equations reduces to

$$m = [\cosh(DJ) + \sigma\sinh(DJ)]^z g(x)|_{x=0}, \quad (11)$$

$$\sigma = [q\cosh(DJ) + m\sinh(DJ) + 1 - q]^z f(x)|_{x=0}, \quad (12)$$

where parameter q is defined by $q = \langle (S_i^z)^2\rangle$.

At this place, in order to evaluate magnetizations m and σ, it is necessary to calculate the parameter q. Starting from the identity[6]

$$q = \langle (S_i^z)^2\rangle = \left\langle \frac{2\cosh(\beta\theta_i)}{2\cosh(\beta\theta_i)+\exp(\Delta\beta)}\right\rangle, \quad (13)$$

we can easily obtain the equation for q, in the same way as the evaluation of (11);

$$q = [\cosh(DJ) + \sigma\sinh(DJ)]^z h(x)|_{x=0}, \quad (14)$$

with

$$h(x) = \frac{2\cosh(\beta x)}{2\cosh(\beta x)+\exp(\Delta\beta)}. \quad (15)$$

In this section, we have discussed the effective-field theory with correlations (EFT) in a mixed Ising ferromagnet with a crystal-field interaction. We are in a position to examine the second-order phase transition and the tricritical behavior, if it exists in the system. In the following sections, we shall study the physical quantities.

§3. Second-order Phase Transition and Tricritical Point

We are interested in studying the transition temperature of the system. Expanding the right-hand side of eqs. (11), (12) and (14), we obtain

$$m=ZA_1\sigma+\frac{Z!}{3!(Z-3)!}A_2\sigma^3+\cdots, \tag{16}$$

$$\sigma=Z\bar{B}_1m+\frac{Z!}{3!(Z-3)!}\bar{B}_2m^3+\cdots, \tag{17}$$

and

$$q=q_0+q_1\sigma^2+\cdots, \tag{18}$$

with

$$A_1=\sinh(DJ)\cosh^{z-1}(DJ)g(x)|_{x=0}, \tag{19}$$

$$A_2=\sinh^3(DJ)\cosh^{z-3}(DJ)g(x)|_{x=0}, \tag{20}$$

$$\bar{B}_1=\sinh(DJ)[q\cosh(DJ)+1-q]^{z-1}f(x)|_{x=0}, \tag{21}$$

$$\bar{B}_2=\sinh^3(DJ)[q\cosh(DJ)+1-q]^{z-3}f(x)|_{x=0}, \tag{22}$$

and

$$q_0=\cosh^z(DJ)h(x)|_{x=0}, \tag{23}$$

$$q_1=\frac{z!}{2!(z-2)!}\sinh^2(DJ)\cosh^{z-2}(DJ)h(x)|_{x=0}. \tag{24}$$

Replacing q in (21) and (22) with the expression of (18), eq. (17) reduces to

$$\sigma=ZB_1m+\frac{Z!}{3!(Z-3)!}B_2m^3+Z(Z-1)q_1B_3\sigma^2m+\cdots, \tag{25}$$

with

$$B_1=\sinh(DJ)[q_0\cosh(DJ)+1-q_0]^{z-1}f(x)|_{x=0}, \tag{26}$$

$$B_2=\sinh^3(DJ)[q_0\cosh(DJ)+1-q_0]^{z-3}f(x)|_{x=0}, \tag{27}$$

$$B_3=\sinh(DJ)[\cosh(DJ)-1][q_0\cosh(DJ)+1-q_0]^{z-2}f(x)|_{x=0}. \tag{28}$$

Substituting eq. (16) into eq. (25), we obtain in general an equation for σ of the form

$$\sigma=a\sigma+b\sigma^3+c\sigma^5+\cdots \tag{29}$$

The second-order phase transition line is then determined by $a=1$, i.e.,

$$1=Z^2A_1B_1 \quad \text{or} \quad 1=Z\sqrt{A_1B_1}. \tag{30}$$

In the vicinity of the second-order phase transition line, the magnetization σ is given by

$$\sigma^2=\frac{1-a}{b}. \tag{31}$$

The right-hand side of (31) must be positive. If this is not the case, the transition is of the first order, and hence the point at which $a=1$ and $b=0$ is the tricritical point.[5] The parameter b is then given by

$$b=\frac{Z^2(Z-1)}{6}[(Z-2)B_1A_2+6q_1B_3A_1+Z^2(Z-2)B_2A_1^3]. \tag{32}$$

Thus, eqs. (30) and (32) are the general expressions of the mixed Ising model with a crystal-field interaction $\varDelta$ for evaluating the second-order phase transition line and the tricritical point within the framework of the EFT. The equations can be easily calculated by the use of a mathematical relation $e^{\alpha D}F(x)=F(x+\alpha)$.

Before discussing the numerical results obtained from eqs. (30) and (32), it is worth commenting on the following facts. If $\varDelta$ goes to negative infinity, from the definitions of (23) and (24), the parameters q_0 and q_1 are given by $q_0=1$ and $q_1=0$, so that the coefficients B_1 and B_2 are equal to A_1 and A_2, respectively, and m and σ reduce to $m=\sigma$; physically, for $\varDelta=-\infty$, the spins with $S=1$ behave like Ising spins with $S=\frac{1}{2}$. The transition temperature for $\varDelta=-\infty$ is determined from $ZA_1=1$, namely

$$Z\sinh(DJ)\cosh^{Z-1}(DJ)f(x)|_{x=0}=1, \quad (33)$$

for which $Z=6$ is equivalent to that obtained by Zernike by the use of another method.[7,8)]

§4. Numerical Results

By solving eqs. (30) and (32) numerically, let us discuss the effect of crystal-field interaction $\varDelta$ on Curie temperature T_c, and also whether a tricritical point may exist or not in a mixed Ising system. For this purpose, we must choose the number of Z. In the following, three lattices, namely with $Z=3$, $Z=4$ and $Z=6$, are investigated.

A Honeycomb lattice (Z=3)

Putting $Z=3$ into the coefficients (19), (20) and (26)–(28), and parameters q_0 and q_1 ((23) and (24)), we obtain

$$A_1=\frac{1}{4}[g(3J)+g(J)],$$

$$A_2=\frac{1}{4}[g(3J)-3g(J)],$$

$$B_1=q_0^2\frac{1}{4}[f(3J)+f(J)]$$
$$+q_0(1-q_0)f(2J)+(1-q_0)^2f(J),$$

$$B_2=\frac{1}{4}[f(3J)-3f(J)],$$

$$B_3=q_0\frac{1}{4}[f(3J)+f(J)-2f(2J)]$$
$$+(1-q_0)\frac{1}{2}[f(2J)-2f(J)], \quad (34)$$

and

$$q_0=\frac{1}{4}[h(3J)+3h(J)],$$

$$q_1=\frac{3}{4}[h(3J)-h(J)].$$

Using eq. (30), the transition temperature is determined from

$$1=3\sqrt{A_1B_1}. \quad (35)$$

The parameter b is given by

$$b=\frac{3}{2}(A_1B_2+6q_1B_3A_1+9B_2A_1^3). \quad (36)$$

In Fig. 1, the change of Curie temperature T_c with $\varDelta$ ($\varDelta\geq 0$) is plotted, by solving eq. (35) numerically. For $\varDelta=0$, T_c is given by $(k_BT_c/J)=1.783$. The dotted line region indicated in the curve (EFT) refers to an extrapolating pro-

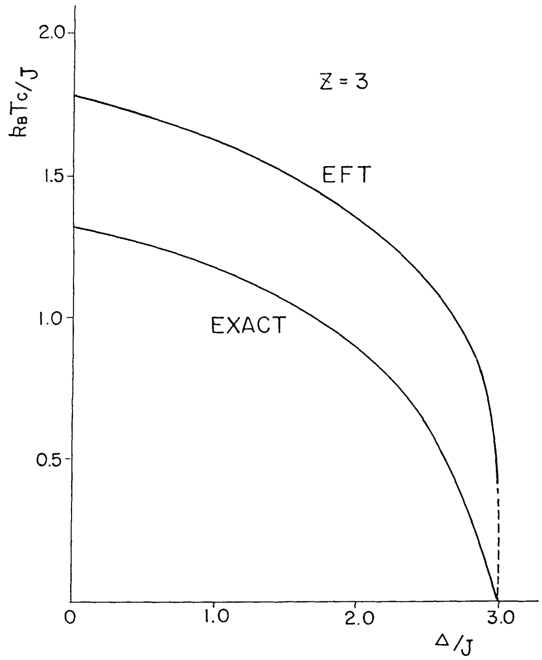

Fig. 1. The change of Curie temperature with a crystal-field interaction for the honeycomb lattice. The curves (EFT) and (EXACT) represent the $\varDelta$ dependences of T_c obtained from eq. (35) and exact solution (37), respectively.

cedure, since in the region the numerical error of (35) becomes very large. Within the studied region of Δ, we could not find any point satisfying the tricritical condition ($a=1$ and $b=0$); for the mixed Ising system on the honeycomb lattice, we may conclude that the tricritical behavior does not exist.

On the other hand, the Curie temperature of this mixed Ising spin system can be related to that of Ising spin system of $S=\frac{1}{2}$ on the triangular lattice with the use of the "star-triangle" transformation,[9] and can be shown to be given exactly as a solution of the equation

$$\cosh(3\beta J)-3\cosh(\beta J)=\exp(\Delta\beta), \quad (37)$$

as is discussed in ref. 2b). The exact result of (37) is also shown in the figure, in order to compare with the result of the EFT. The exact value of T_c for $\Delta=0$ is given by $(k_B T_c/J)=1.320$.

As commented in §2, the present model can be connected to the mixed Ising systems with a biquadratic exchange interaction J_1. For this system on a honeycomb lattice, Iwashita and Uryu (*I-U*)[2] obtained the Curie temperature as $(k_B T_c/J)=1.631$ for $J_1=0$ in their Bethe-Peierls treatment. However, when their result is extrapolated, the value of J_1 at which T_c reduces to zero is given by $J_1=-1.82J$; in comparison with our result, their value of Δ at which T_c becomes zero is given by $\Delta=2.73J$. Thus, our calculation (EFT) reproduces the exact value ($\Delta=3J$) for the lowest value of Δ at which $T_c=0$, although the value of T_c for $\Delta=0$ is larger than that of *I-U*. The same result as ours has also been obtained by Siqueira and Fittipaldi (S-F)[3] using almost the same framework as our EFT except the discussions on the tricritical behavior.

B Square lattice ($Z=4$) and simple cubic lattice ($Z=6$)

For $Z=4$ and $Z=6$, we can obtain the expressions of coefficients (A_1, A_2, B_1, B_2 and B_3) and parameters q_0 and q_1, like the part A for $Z=3$. Using these expressions, the changes of T_c with Δ can be obtained by solving eqs. (30) and (32) numerically. The results for $Z=4$ and $Z=6$ are plotted in Fig. 2. The values of T_c at $\Delta=0$ are given in Table I.

In contrast with the case of $Z=3$, increasing the value of Δ from $\Delta=0$, in each case T_c decreases from the value of T_c for $\Delta=0$ and satisfies the tricritical condition ($a=1$ and $b=0$) at the point $((k_B T_t/J, \Delta_t/J))$: (0.9936,

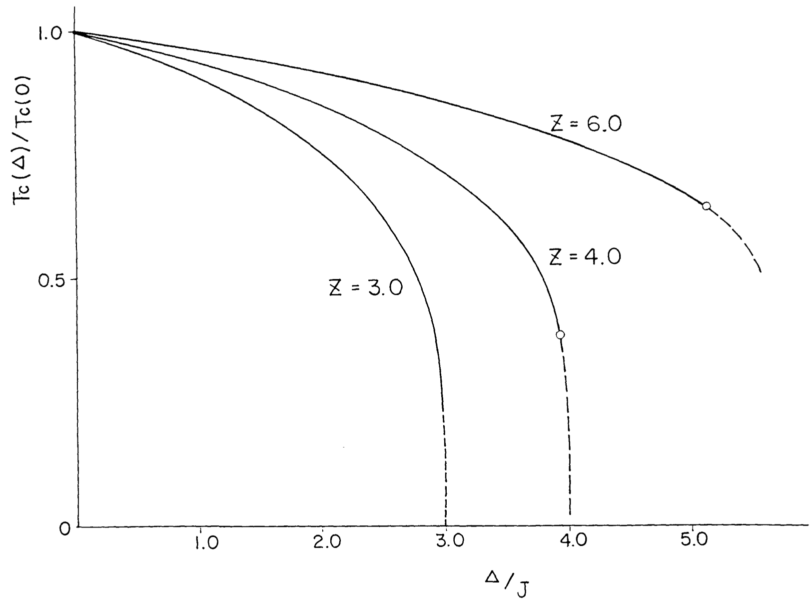

Fig. 2. Reduced critical temperature $T_c(\Delta)/T_c(0)$ as a function of Δ for three lattices with $Z=3$, $Z=4$, and $Z=6$. Open circles denote the tricritical points.

Table I. The values of $(k_B T_c/J)$ at $\Delta=0$ (or $J_1=0$).

	EFT	S–F[3]	R.G[10]	I–U[2]	EXACT[9]
$Z=3$	1.783	—	—	1.631	1.320
$Z=4$	2.598	2.522	2.372 2.748	2.478	—
$Z=6$	4.223	—	—	—	—

3.9376) for $Z=4$ and (2.7154, 5.1388) for $Z=6$, shown as open circles in the figure. Thus, we may conclude that a mixed Ising spin system with a crystal-field interaction Δ (or a biquadratic exchange interaction J_1) exhibits a tricritical point on the T_c versus Δ (or T_c versus J_1) curve, when the coordination number Z becomes larger than $Z=3$, although previous works[2,3] have never considered such a possibility of tricritical behavior.

For the mixed Ising spin system on square lattice, on the other hand, I–U and S–F have examined the change of T_c with J_1. They have obtained the values of T_c at $\Delta=0$ (or $J_1=0$). The results are also collected in Table I, as well as the results of renormalization group (R.G) analysis,[10] in order to compare our result for $Z=4$ with theirs. However, when their values of T_c are simply extrapolated, the value of T_c in I–U reduces to zero at $\Delta=3.70$ J (or $J_1=-1.85$ J), in contrast with $\Delta=4.0$ J (or $J_1=-2.0$ J) of our EFT and S–F, although the dashed line in Fig. 2 denotes the first-order phase transition.

§5. Conclusions

In this work, we have developed some general formulas for evaluating the second-order phase transition line and the tricritical point in a mixed $S-\frac{1}{2}$ and S-1 Ising model with a crystal-field interaction using the effective-field theory with correlations. The formulas have been applied to some lattices, and the obtained results are all reasonable in comparison with the exact result and other approximate results. In §4, we have found that the mixed Ising spin system with a crystal-field interaction may exhibit a tricritical point when the coordination number Z becomes larger than $Z=3$, although previous works have completely neglected such a fact. Thus, the present framework provides qualitative and, to a certain extent, quantitative confidence for the use of this method in this class of systems. We should also mention that the methodology developed here can be applied to more complex situations, such as disordered mixed Ising spin systems with random bond and crystal-field inteactions, as well as the mixed ferrimagnetic system with $J<0$.

References

1) M. Blume: Phys. Rev. **141** (1966) 517; H. W. Capel: Physica **32** (1966) 966; M. Nauciel-Bloch, G. Sarma and A. Castests: Phys. Rev. **B5** (1972) 4603.
2a) T. Iwashita and N. Uryu: Phys. Lett. **96**A (1979) 311; J. Phys. Soc. Jpn. **53** (1984) 721.
2b) Phys. Status Solidi (b) **125** (1984) 551.
3) A. F. Siqueira and I. P. Fittipaldi: J. Magn. & Magn. Mater. **54–57** (1986) 678.
4) R. Honmura and T. Kaneyoshi: J. Phys. C **12** (1979) 3979.
5) T. Kaneyoshi: J. Phys. C **19** (1986) L557; J. Phys. Soc. Jpn. **56** (1987) 933.
6) T. Kaneyoshi: Phys. Rev. B**33** (1986) 7688; J. Phys. Soc. Jpn. **55** (1986) 1430; Phys. Rev. B **34** (1986) 7866.
7) T. Kaneyoshi, I. Tamura and E. F. Sarmento: Phys. Rev. B **28** (1983) 6491; E. F. Sarmento and C. Tsallis: Phys. Rev. B **27** (1983) 5784.
8) F. Zernike: Physica **7** (1940) 565.
9) C. Domb: Adv. Phys. **9** (1960) 149.
10) S. L. Schafield and R. G. Bowers: J. Phys. A **13** (1980) 3697.