

## Phase diagrams of a disordered, ferromagnetic, binary Ising system

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Phase diagrams of a binary ferromagnetic Ising system with randomly distributed exchange parameters are investigated by use of the effective-field theory with correlations. Some interesting results for the effect of structural fluctuations on phase diagrams are found.

### I. INTRODUCTION

Over the last several years the magnetic properties of binary random substitutional alloys have been studied intensively from both bond and site perspectives. The bond model considers all lattice sites to be equivalent, but the interaction energy between each pair of adjacent sites is randomly assigned one of a set of possible values. In the site model, however, the lattice sites are randomly occupied by two different species of magnetic ions,  $A$  and  $B$ , and the interaction between two ions is determined entirely by the species of those ions. The Hamiltonian for the system is then given by

$$\mathcal{H} = \sum_{i,j} [J_{AA} \delta_{iA} \delta_{jA} + J_{BB} \delta_{iB} \delta_{jB} + J_{AB} (\delta_{iA} \delta_{jB} + \delta_{iB} \delta_{jA})] S_i^z S_j^z \xi_i \xi_j, \quad (1)$$

where the  $J_{ij}$ 's are the interaction energies between type- $i$  and type- $j$  ions, the  $S^z$  are the spin variables with  $S^z = \pm 1$ , and the sum is over all nearest-neighbor pairs.  $\xi_i$  is a random variable which takes the value of unity or zero, depending on whether the site  $i$  is occupied by a magnetic atom or not.

On the other hand, there has been some interest in the phase diagrams of binary ferromagnetic alloys with  $A_p B_{1-p}$ , with a transition temperature  $T_c(p)$  as a function of concentration  $p$ .<sup>1-3</sup> When all the exchange interactions in (1) have the same ferromagnetic signs and we suppose without loss of generality that  $T_c(1) > T_c(0)$ , the simplest possible phase boundary may be a straight-line extrapolation for  $T_c(p)$  between  $T_c(0)$  and  $T_c(1)$ . As discussed in Ref. 1, however, nine phases may be possible from the three behaviors of the initial slope ( $\partial \ln T_c / \partial p$ ) of the transition temperature with  $p$  at the two points  $p=0$  and  $p=1$ . For special low-dimensional crystalline lattices, such as the Bethe and honeycomb lattices, six phases have been found by using the exact calculations at  $p=1$  and  $p=0$  from the possible nine phases.<sup>1</sup> The standard molecular field theory (MFT) predicts only four possible phases. The MFT generally overestimates  $T_c$  and underestimates the number of phases in the mixed alloy with low coordination numbers. However, as discussed in Ref. 2, it may be expected that the phase diagrams for lattices

with these low coordination numbers  $z$  approach that given by the standard MFT once  $z$  is increased.

In amorphous ferromagnetic alloys with the general formula  $(A_p B_{1-p})_c N_{1-c}$ , where  $N$  represents some metalloid, the concentration dependences of  $T_c$  have been intensively investigated, in order to determine one of the most important parameters (or  $J_{AA}$ ,  $J_{AB}$ , and  $J_{BB}$ ) for understanding magnetic properties.<sup>4</sup> In amorphous magnets, however, it has been discussed that the fluctuation of exchange interactions (or the structural fluctuation) is one of the underlying causes for the changes of physical quantities, in comparison with those of crystalline alloys. In fact, experiments of the Mössbauer effect and the magnetization of amorphous ferromagnets indicate that, at least in some materials, there are large fluctuations in the exchange interactions.<sup>5</sup> Theoretically, for studying such systems, the lattice model of amorphous magnets has often been applied, in which the structural disorder is replaced by the random distribution of the exchange integral; the exchange interactions in (1) are assumed to be distributed randomly about their mean values.

In this paper, the phase diagrams of a binary ferromagnetic Ising system with randomly distributed exchange parameters are investigated by using the effective-field theory with correlations,<sup>6</sup> in order to clarify the effects of the structural fluctuation on the phase diagrams for its corresponding crystalline binary alloy. We find some interesting results for the phase diagrams. In particular, for an anisotropic structural fluctuation a new phase may appear, which is not found in a crystalline binary alloy.

The outline of our paper is as follows. In Sec. II, we briefly review the basic points of the effective-field theory with correlations<sup>6</sup> when it is applied to a disordered ferromagnetic binary Ising system. In Sec. III, the general formulas of the initial slope ( $\partial \ln T_c / \partial p$ ) at the two points  $p=0$  and  $p=1$  are derived. For the lattice model of an amorphous ferromagnet in a square lattice, the numerical results of the phase diagram are studied and discussed in Sec. IV.

### II. FORMULATION

We consider a binary alloy of the type,  $A_p B_{1-p}$ , randomly occupied by two different species of magnetic ions,

$A$  and  $B$ , where the  $A$  and  $B$  atoms have the same spins ( $S_A^z = \pm 1$  and  $S_B^z = \pm 1$ , respectively). The Hamiltonian of the system is given by (1). Moreover, to describe the structural disorder of amorphous magnets in a simple way, the nearest-neighbor exchange interactions are assumed to be given by three independent random variables, i.e.,  $p(J_{AA})$ ,  $p(J_{AB})$ , and  $p(J_{BB})$ , where  $p(x)$  is a probability distribution function.

The magnetization per site is

$$\langle S_i^z \rangle = \xi_{i=A} \langle S_{i=A}^z \rangle + \xi_{i=B} \langle S_{i=B}^z \rangle \quad (2)$$

with a restriction

$$\langle \xi_{i=A} \rangle_r + \langle \xi_{i=B} \rangle_r = 1, \quad (3)$$

where  $\langle \cdots \rangle$  expresses the usual thermal average and  $\langle \cdots \rangle_r$  is the random configurational average. The random average  $\langle \xi_{i=A} \rangle_r = p$  is the concentration of  $A$  atoms. The main problem is the evaluation of the mean values,  $\langle S_{i=A}^z \rangle$  and  $\langle S_{i=B}^z \rangle$ . As has been discussed in a

series of works,<sup>6,7</sup> the starting point for the evaluation  $\langle S_{i=A}^z \rangle$  or  $\langle S_{i=B}^z \rangle$  is the exact Callen identity:<sup>8</sup>

$$\xi_{i=\alpha} \langle S_{i=\alpha}^z \rangle = \xi_{i=\alpha} \langle \tanh(\beta \xi_{i=\alpha} \theta_{i=\alpha}) \rangle, \quad \alpha = A \text{ or } B \quad (4)$$

with

$$\theta_{i=\alpha} = \sum_j (J_{\alpha A} \delta_{jA} + J_{\alpha B} \delta_{jB}) S_j^z \xi_j, \quad (5)$$

where  $\beta = 1/k_B T$ .

At this stage, in order to write identities (4) in a form which is particularly amenable to approximation, let us introduce the differential operator technique<sup>6</sup> as follows:

$$\xi_{i=\alpha} \langle S_{i=\alpha}^z \rangle = \xi_{i=\alpha} \langle e^{D \beta \xi_{i=\alpha} \theta_{i=\alpha}} \tanh(x) \big|_{x=0} \rangle, \quad (6)$$

where  $D = \partial/\partial x$  is a differential operator. Using the relation  $\xi_i^n = \xi_i$  ( $n = \text{integer}$ ) and the identity

$$\exp(as_i^z) = \cosh(a) + S_i^z \sinh(a), \quad (7)$$

Eq. (6) reduces to

$$\xi_{i=\alpha} \langle S_{i=\alpha}^z \rangle = \xi_{i=\alpha} \left\langle \prod_j \{ \xi_j \delta_{jA} [\cosh(D_{\alpha A}^i) + S_j^z \sinh(D_{\alpha A}^i)] + \xi_j \delta_{jB} [\cosh(D_{\alpha B}^i) + S_j^z \sinh(D_{\alpha B}^i)] \} \tanh(x) \bigg|_{x=0} \right\rangle, \quad (8)$$

where  $D_{\alpha r}^i = \xi_{i=\alpha} J_{\alpha r} D \beta$  ( $\alpha, r = A$  or  $B$ ).

For a disordered system with random bonds and random occupation of magnetic atoms, we must perform the random configurational average for Eqs. (2) and (8); the averaged total magnetization per site is given by

$$m = \langle \langle S_i^z \rangle \rangle_r = p m_A + (1-p) m_B, \quad (9)$$

where  $m_A$  and  $m_B$  are defined by

$$m_A = \frac{\langle \xi_{i=A} \langle S_{i=A}^z \rangle \rangle_r}{\langle \xi_{i=A} \rangle_r}, \quad (10)$$

$$m_B = \frac{\langle \xi_{i=B} \langle S_{i=B}^z \rangle \rangle_r}{\langle \xi_{i=B} \rangle_r}. \quad (11)$$

Here, it is clear that, if we try to exactly treat all the spin-spin correlations appearing in the partial magnetiza-

tions through the expansion of Eq. (8) and to properly perform the random configurational average, the problem becomes mathematically untractable. In the previous works,<sup>6-7</sup> therefore, the decoupling approximation, or

$$\langle \langle x_j x_k \cdots x_l \rangle \rangle_r \cong \langle \langle x_j \rangle \rangle_r \langle \langle x_k \rangle \rangle_r \cdots \langle \langle x_l \rangle \rangle_r \quad (12)$$

with  $j \neq k \neq \dots \neq l$  and  $x_j = \xi_j S_j^z$ , has been used. In fact, the approximation corresponds essentially to the Zernike approximation in the nonrandom problem,<sup>9</sup> as discussed in Ref. 6. The approximation has been successfully applied to a great number of disordered magnetic systems.

By taking account of the fact that the exchange interactions and the random occupation of magnetic atom sites are given by independent random variables, the average partial magnetization  $m_\alpha$  ( $\alpha = A$  or  $B$ ) is given by, upon performing the random average and introducing the decoupling approximation (12),

$$m_\alpha = \{ p [ \langle \cosh(D \bar{J}_{\alpha A}) \rangle_r + m_A \langle \sinh(D \bar{J}_{\alpha A}) \rangle_r ] + (1-p) [ \langle \cosh(D \bar{J}_{\alpha B}) \rangle_r + m_B \langle \sinh(D \bar{J}_{\alpha B}) \rangle_r ] \}^z \tanh(x) \big|_{x=0} \quad (13)$$

with

$$\bar{J}_{\alpha r} = \beta J_{\alpha r},$$

where  $z$  is the number of nearest neighbors.

We are now interested in investigating the phase diagram of the system. The usual argument that  $m_\alpha$  tends to zero as the temperature approaches a critical temperature allows us to consider only terms linear in  $m_\alpha$ . Near the critical point, therefore, we have

$$\begin{aligned} A m_A + B m_B &= 0, \\ C m_A + D m_B &= 0 \end{aligned} \quad (14)$$

with

$$\begin{aligned}
 A &= pz[p \langle \cosh(D\bar{J}_{AA}) \rangle_r + (1-p) \langle \cosh(D\bar{J}_{AB}) \rangle_r]^{z-1} \langle \sinh(D\bar{J}_{AA}) \rangle_r \tanh(x) |_{x=0} - 1, \\
 B &= (1-p)z[p \langle \cosh(D\bar{J}_{AA}) \rangle_r + (1-p) \langle \cosh(D\bar{J}_{AB}) \rangle_r]^{z-1} \langle \sinh(D\bar{J}_{AB}) \rangle_r \tanh(x) |_{x=0}, \\
 C &= pz[(1-p) \langle \cosh(D\bar{J}_{BB}) \rangle_r + p \langle \cosh(D\bar{J}_{AB}) \rangle_r]^{z-1} \langle \sinh(D\bar{J}_{AB}) \rangle_r \tanh(x) |_{x=0}, \\
 D &= (1-p)z[(1-p) \langle \cosh(D\bar{J}_{BB}) \rangle_r + p \langle \cosh(D\bar{J}_{AB}) \rangle_r]^{z-1} \langle \sinh(D\bar{J}_{BB}) \rangle_r \tanh(x) |_{x=0} - 1.
 \end{aligned}
 \tag{15}$$

Equation (14) yields the following secular equation

$$\tilde{M} \begin{pmatrix} m_A \\ m_B \end{pmatrix} = \begin{pmatrix} A & B \\ C & D \end{pmatrix} \begin{pmatrix} m_A \\ m_B \end{pmatrix} = 0. \tag{16}$$

The critical ferromagnetic boundaries (or phase diagram) can be derived from the condition  $\det \tilde{M} = 0$ , namely,

$$AD = BC. \tag{17}$$

From this relation we can obtain the transition temperature  $T_c(p)$  as a function of  $p$ .

### III. INITIAL SLOPE

Let us now study the initial slope  $\partial \ln T_c / \partial p$  at the two points  $p=0$  and  $p=1$ . Although the initial slopes by no means provide a complete description of the phase diagram obtained from (16), they do severely restrict what can occur and so can be used at the basis of a classification scheme; these initial slopes determine the overall character of the phase diagram although not the detailed shape, of course. In fact, when all the exchange interactions in (1) have the same ferromagnetic signs and we suppose without loss of generality that  $T_c(1) > T_c(0)$ , the simplest possible phase boundary may be a straight-line extrapolation for  $T_c(p)$  between  $T_c(0)$  and  $T_c(1)$ , as shown in Fig. 1 by a dashed line. As shown in Fig. 1, the initial slopes at  $p=0$  can identify three possible types of behavior: (i) a slope greater than the linear extrapolation, (ii) a slope less than the linear extrapolation but greater than zero, and (iii) a slope less than zero. There are also three similar types of behavior at  $p=1$ . From the three possible behaviors of the initial slope at the two points  $p=0$  and  $p=1$  in Fig. 1, nine phases may be possible. Based on these arguments, Thorpe and McGurn<sup>1</sup> have examined the various phase diagrams which may occur in a mixed  $A_p B_{1-p}$  Ising ferromagnetic crystalline alloy for special low-dimensional lattices, such as the Bethe and honeycomb ones, and found that for these lattices three possible phases ( $T, S$ , and  $S'$  in their notations) do not appear. Honmura *et al.*<sup>2</sup> have also investigated the phase diagrams for mixed  $A_p B_{1-p}$  crystalline Ising alloys with  $z=4$  and  $z=6$  and found that the three phases do not appear, using the relation (17) without random-bond averages in (15).

Within the present formulation discussed in Sec. II, let us now investigate the initial slopes at the two points  $p=0$  and  $p=1$  and derive general relations for determin-

ing the overall character of the phase diagram.

Differentiating both sides of (17) with  $p$ , the initial slopes are given by

$$- \left[ \frac{\partial D}{\partial p} \right]_{T=T_c(B)}^{p=0} = \left[ B \frac{\partial C}{\partial p} \right]_{T=T_c(B)}^{p=0} \tag{18}$$

and

$$- \left[ \frac{\partial A}{\partial q} \right]_{T=T_c(A)}^{q=0} = \left[ B \frac{\partial B}{\partial q} \right]_{T=T_c(A)}^{q=0}, \tag{19}$$

where  $q=1-p$ . The parameters  $T_c(A)$  and  $T_c(B)$  are the transition temperatures for  $p=1$  and  $p=0$ , which are obtained from

$$z[\langle \cosh(D\bar{J}_{AA}) \rangle_r]^{z-1} \langle \sinh(D\bar{J}_{AA}) \rangle_r \tanh(x) |_{x=0} = 1 \tag{20}$$

and

$$z[\langle \cosh(D\bar{J}_{BB}) \rangle_r]^{z-1} \langle \sinh(D\bar{J}_{BB}) \rangle_r \tanh(x) |_{x=0} = 1. \tag{21}$$

Using the relation

$$\frac{\partial \bar{J}_{ar}}{\partial p} = - \frac{\bar{J}_{ar}}{T_c} \frac{\partial T_c}{\partial p} \quad (\alpha, r = A \text{ or } B), \tag{22}$$

we obtain, from (18) and (19),

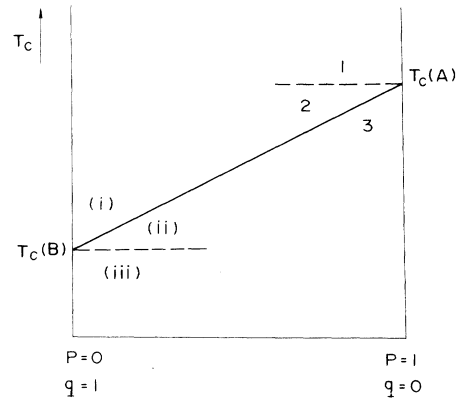


FIG. 1. Nine possible initial derivatives at  $p=1$  and  $p=0$ , and the linear extrapolation of the phase boundary between  $T_c(0)$  and  $T_c(1)$ .

$$\left[ \frac{1}{T_c} \frac{\partial T_c}{\partial p} \right]_{p=0} = \left[ \frac{K_2 - 1}{K_1} \right]_{T=T_c(B)} \quad (23)$$

$$\left[ \frac{1}{T_c} \frac{\partial T_c}{\partial p} \right]_{q=0} = \left[ \frac{K_4 - 1}{K_3} \right]_{T=T_c(A)}, \quad (24)$$

and

where the coefficients  $K_i$  ( $i=1-4$ ) are defined by

$$\begin{aligned} K_1 &= [\langle \cosh(D\bar{J}_{BB}) \rangle_r]^{z-1} \langle \bar{J}_{BB} \cosh(D\bar{J}_{BB}) \rangle_r \operatorname{sech}^2(x) \Big|_{x=0} \\ &\quad + (z-1) [\langle \cosh(D\bar{J}_{BB}) \rangle_r]^{z-2} \langle \sinh(D\bar{J}_{BB}) \rangle_r \langle \bar{J}_{BB} \sinh(D\bar{J}_{BB}) \rangle_r \operatorname{sech}^2(x) \Big|_{x=0}, \\ K_2 &= z \{ [\langle \cosh(D\bar{J}_{BB}) \rangle_r]^{z-1} \langle \sinh(D\bar{J}_{AB}) \rangle_r \tanh(x) \Big|_{x=0} \} \{ [\langle \cosh(D\bar{J}_{AB}) \rangle_r]^{z-1} \langle \sinh(D\bar{J}_{AB}) \rangle_r \tanh(x) \Big|_{x=0} \} \\ &\quad + (z-1) \langle \cosh(D\bar{J}_{AB}) \rangle_r [\langle \cosh(D\bar{J}_{BB}) \rangle_r]^{z-2} \langle \sinh(D\bar{J}_{BB}) \rangle_r \tanh(x) \Big|_{x=0}, \\ K_3 &= [\langle \cosh(D\bar{J}_{AA}) \rangle_r]^{z-1} \langle \bar{J}_{AA} \cosh(D\bar{J}_{AA}) \rangle_r \operatorname{sech}^2(x) \Big|_{x=0} \\ &\quad + (z-1) [\langle \cosh(D\bar{J}_{AA}) \rangle_r]^{z-2} \langle \sinh(D\bar{J}_{AA}) \rangle_r \langle \bar{J}_{AA} \sinh(D\bar{J}_{AA}) \rangle_r \operatorname{sech}^2(x) \Big|_{x=0}, \\ K_4 &= z \{ [\langle \cosh(D\bar{J}_{AB}) \rangle_r]^{z-1} \langle \sinh(D\bar{J}_{AB}) \rangle_r \tanh(x) \Big|_{x=0} \} \{ [\langle \cosh(D\bar{J}_{AA}) \rangle_r]^{z-1} \langle \sinh(D\bar{J}_{AB}) \rangle_r \tanh(x) \Big|_{x=0} \} \\ &\quad + (z-1) [\langle \cosh(D\bar{J}_{AA}) \rangle_r]^{z-2} \langle \sinh(D\bar{J}_{AA}) \rangle_r \langle \cosh(D\bar{J}_{AB}) \rangle_r \tanh(x) \Big|_{x=0}. \end{aligned} \quad (25)$$

From Fig. 1, on the other hand, the phase boundaries determining the overall behavior of the phase diagram are given by the following relations:

$$\left[ \frac{1}{T_c} \frac{\partial T_c}{\partial p} \right]_{T_c=T_c(B)}^{p=0} = 0, \quad (26)$$

$$\left[ \frac{1}{T_c} \frac{\partial T_c}{\partial p} \right]_{T_c=T_c(B)}^{p=0} = \frac{T_c(A) - T_c(B)}{T(B)}, \quad (27)$$

$$\left[ \frac{1}{T_c} \frac{\partial T_c}{\partial q} \right]_{T_c=T_c(A)}^{q=0} = 0, \quad (28)$$

$$\left[ \frac{1}{T_c} \frac{\partial T_c}{\partial q} \right]_{T_c=T_c(B)}^{q=0} = \frac{T_c(B) - T_c(A)}{T(A)}. \quad (29)$$

Thus, within the present formulation, the general relations which determine the behavior of initial slopes at  $p=1$  and  $p=0$  are given by Eqs. (23)–(29).

Let us now investigate a disordered system in which the probability distribution functions  $p(J_{AA})$ ,  $p(J_{AB})$ , and  $p(J_{BB})$  for exchange interactions  $J_{AA}$ ,  $J_{AB}$ , and  $J_{BB}$  are given by

$$\begin{aligned} p(J_{AA}) &= \frac{1}{2} [\delta(J_{AA} - J_{AA}^0 - \Delta J) + \delta(J_{AA} - J_{AA}^0 + \Delta J)], \\ p(J_{AB}) &= \frac{1}{2} [\delta(J_{AB} - J_{AB}^0 - \Delta J') + \delta(J_{AB} - J_{AB}^0 + \Delta J')], \\ &\quad (30) \end{aligned}$$

$$p(J_{BB}) = \frac{1}{2} [\delta(J_{BB} - J_{BB}^0 - \Delta J'') + \delta(J_{BB} - J_{BB}^0 + \Delta J'')],$$

$$z \cosh^z(D\bar{J}_{\alpha\alpha}^0 \delta_{\alpha\alpha}) \cosh^{z-1}(D\bar{J}_{\alpha\alpha}^0) \sinh(D\bar{J}_{\alpha\alpha}^0) \tanh(x) \Big|_{x=0} = 1 \quad (\alpha = A \text{ or } B) \quad (33)$$

by which the transition temperature  $T_c(A)$  or  $T_c(B)$  is determined. Then, by applying a mathematical relation  $e^{aD}f(x) = f(x+a)$ , Eq. (33) can be expressed as a sum of transcendental functions  $\tanh x$  with an appropriate argument  $x$ . For  $z=4$ , Eq. (33) has already been solved nu-

merically as a function of  $\delta$  in Ref. 11 (the definition  $\delta$  in Ref. 11 corresponds to  $2\delta = \delta_{\alpha\alpha}$  in the present formulation). In Fig. 2, the result of (33) for  $z=4$  is once more depicted as a function of  $\delta_{\alpha\alpha}$ . For  $\delta_{\alpha\alpha}=0$ , the transition temperature  $t_c = k_B T_c(\alpha) / J_{\alpha\alpha}^0$  ( $\alpha = A$  or  $B$ ) is then given

$$\begin{aligned} \langle \cosh(D\bar{J}_{\alpha\eta}) \rangle_r &= \cosh(D\bar{J}_{\alpha\eta}^0) \cosh(D\bar{J}_{\alpha\eta}^0 \delta_{\alpha\eta}) \\ \langle \sinh(D\bar{J}_{\alpha\eta}) \rangle_r &= \sinh(D\bar{J}_{\alpha\eta}^0) \cosh(D\bar{J}_{\alpha\eta}^0 \delta_{\alpha\eta}), \end{aligned} \quad (\alpha, \eta = A \text{ or } B) \quad (31)$$

where  $\bar{J}_{\alpha\eta}^0 = \beta J_{\alpha\eta}^0$ . The parameters  $\delta_{\alpha\eta}$  are dimensionless parameters which are often called structural fluctuations in the lattice model of amorphous magnets and are defined by

$$\begin{aligned} \delta_{AA} &= \frac{\Delta J}{J_{AA}^0}, \\ \delta_{AB} &= \frac{\Delta J'}{J_{AB}^0}, \\ \delta_{BB} &= \frac{\Delta J''}{J_{BB}^0}. \end{aligned} \quad (32)$$

Substituting the relations (31) into (20) or (21), we obtain

merically as a function of  $\delta$  in Ref. 11 (the definition  $\delta$  in Ref. 11 corresponds to  $2\delta = \delta_{\alpha\alpha}$  in the present formulation). In Fig. 2, the result of (33) for  $z=4$  is once more depicted as a function of  $\delta_{\alpha\alpha}$ . For  $\delta_{\alpha\alpha}=0$ , the transition temperature  $t_c = k_B T_c(\alpha) / J_{\alpha\alpha}^0$  ( $\alpha = A$  or  $B$ ) is then given

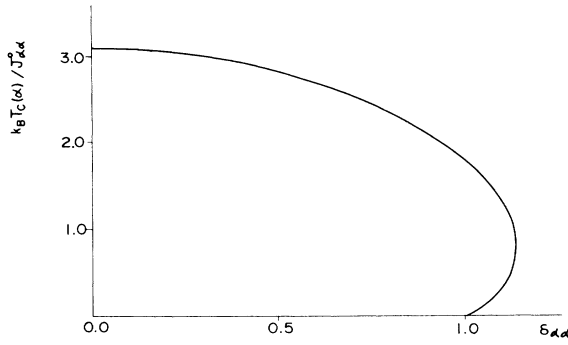


FIG. 2. Transition temperature for square lattice ( $z=4$ ) plotted as a function of structural fluctuation. The possibility of reentrant phenomenon may be found in the range  $1.0 < \delta_{\alpha\alpha} < 1.13$ .

by  $k_B T_c(\alpha)/J_{\alpha\alpha}^0 = 3.0898$ , as obtained in Refs. 2, 6, 7, and 11. As is easily understood from the definition of  $\delta_{\alpha\alpha}$ , the probability distribution function  $p(J_{\alpha\alpha})$  can take positive and negative values randomly, when  $\delta_{\alpha\alpha}$  becomes larger than  $\delta_{\alpha\alpha} = 1.0$ . For  $\delta_{\alpha\alpha} > 1.0$ , therefore, the so-called ef-

fect of frustration appears in Fig. 2; the possibility of reentrant phenomenon is obtained in the range  $1.0 < \delta < 1.13$ . In the following, however, we are interested in the overall character of the phase diagram shown in Fig. 1, so that we restrict the value of  $\delta$  in the range  $0 < \delta_{\alpha\alpha} < 1.0$ .

Substituting (31) into (25), and using the relations

$$\xi = \frac{J_{AB}^0}{(J_{AA}^0 J_{BB}^0)^{1/2}}, \quad \eta = \frac{J_{BB}^0}{J_{AA}^0},$$

and

$$t_c = \frac{J_{\alpha\alpha}^0}{k_B T_c(\alpha)},$$

Eqs. (23) and (24) reduce to

$$\left[ \frac{1}{T_c} \frac{\partial T_c}{\partial p} \right]_{p=0} \frac{\bar{K}_2 - 1}{\bar{K}_1} \quad (35)$$

and

$$\left[ \frac{1}{T_c} \frac{\partial T_c}{\partial q} \right]_{q=0} \frac{\bar{K}_4 - 1}{\bar{K}_3} \quad (36)$$

with

$$\begin{aligned} \bar{K}_1 &= t_c \cosh^{z-1}(Dt_c) \cosh^{z-1}(Dt_c \delta_{BB}) [\cosh(Dt_c) \cosh(Dt_c \delta_{BB}) + \delta_{BB} \sinh(Dt_c) \sinh(Dt_c \delta_{BB})] \operatorname{sech}^2(x) \Big|_{x=0} \\ &+ (z-1) t_c \cosh^{z-2}(Dt_c) \sinh(Dt_c) \cosh^{z-1}(Dt_c \delta_{BB}) \\ &\times [\sinh(Dt_c) \cosh(Dt_c \delta_{BB}) + \delta_{BB} \cosh(Dt_c) \sinh(Dt_c \delta_{BB})] \operatorname{sech}^2(x) \Big|_{x=0}, \\ \bar{K}_2 &= z \left[ \cosh^{z-1}(Dt_c \delta_{BB}) \cosh \left[ Dt_c \frac{\xi}{\sqrt{\eta}} \delta_{AB} \right] \cosh^{z-1}(Dt_c) \sinh \left[ Dt_c \frac{\xi}{\sqrt{\eta}} \right] \tanh(x) \Big|_{x=0} \right] \\ &\times \left[ \cosh^z \left[ Dt_c \frac{\xi}{\sqrt{\eta}} \delta_{AB} \right] \cosh^{z-1} \left[ Dt_c \frac{\xi}{\sqrt{\eta}} \right] \sinh \left[ Dt_c \frac{\xi}{\sqrt{\eta}} \right] \tanh(x) \Big|_{x=0} \right] \\ &+ (z-1) \cosh^{z-1}(Dt_c \delta_{BB}) \cosh \left[ Dt_c \frac{\xi}{\sqrt{\eta}} \delta_{AB} \right] \cosh \left[ Dt_c \frac{\xi}{\sqrt{\eta}} \right] \cosh^{z-2}(Dt_c) \sinh(Dt_c) \tanh(x) \Big|_{x=0}, \\ \bar{K}_3 &= t_c \cosh^{z-1}(Dt_c) \cosh^{z-1}(Dt_c \delta_{AA}) [\cosh(Dt_c) \cosh(Dt_c \delta_{AA}) + \delta_{AA} \sinh(Dt_c) \sinh(Dt_c \delta_{AA})] \operatorname{sech}^2(x) \Big|_{x=0} \\ &+ (z-1) t_c \cosh^{z-2}(Dt_c) \sinh(Dt_c) \cosh^{z-1}(Dt_c \delta_{AA}) \\ &\times [\sinh(Dt_c) \cosh(Dt_c \delta_{AA}) + \delta_{AA} \cosh(Dt_c) \sinh(Dt_c \delta_{AA})] \operatorname{sech}^2(x) \Big|_{x=0}, \\ \bar{K}_4 &= z [\cosh^{z-1}(Dt_c \delta_{AA}) \cosh(Dt_c \xi \sqrt{\eta} \delta_{AB}) \cosh^{z-1}(Dt_c) \sinh(Dt_c \xi \sqrt{\eta}) \tanh(x) \Big|_{x=0}] \\ &\times [\cosh^z(Dt_c \xi \sqrt{\eta} \delta_{AB}) \cosh^{z-1}(Dt_c \xi \sqrt{\eta}) \sinh(Dt_c \xi \sqrt{\eta}) \tanh(x) \Big|_{x=0}] \\ &+ (z-1) \cosh^{z-1}(Dt_c \delta_{AA}) \cosh(Dt_c \xi \sqrt{\eta} \delta_{AB}) \cosh(Dt_c \xi \sqrt{\eta}) \cosh^{z-2}(Dt_c) \sinh(Dt_c) \tanh(x) \Big|_{x=0}, \end{aligned} \quad (37)$$

where the coefficients  $\bar{K}_i$  ( $i=1-4$ ) can be also evaluated by the use of the mathematical relation  $e^{aD} f(x) = f(x+a)$ .

We are now in a position to examine phase diagrams for the lattice model of amorphous ferromagnets, which

can be obtained by solving Eqs. (35), (36), and (26)–(29), numerically. In the next section, the results are shown for a square lattice ( $z=4$ ). Before discussing the results, it is worthwhile to notice a general fact for the overall character of the phase diagram; for  $\delta_{AA} = \delta_{BB} = \delta_{AB} = \bar{\delta}$  the fol-

lowing relations are always satisfied for the initial slopes (26) and (28):

$$\xi = \sqrt{\eta} \tag{38}$$

and

$$\xi = \frac{1}{\sqrt{\eta}} \tag{39}$$

For the phase diagrams in the  $(\xi, \eta)$  space, therefore, the phase boundaries (38) and (39) are independent of  $\bar{\delta}$ , although the phase boundaries obtained from (27) and (29), may change with  $\bar{\delta}$ . The results (38) and (39) are also obtained in Refs. 1 and 2 and from the standard MFT (Ref. 5) for a mixed Ising ferromagnetic alloy with  $\bar{\delta}=0$ .

IV. NUMERICAL RESULTS AND DISCUSSIONS

At first, the phase diagrams of square lattice ( $z=4$ ) for  $\delta_{AA}=\delta_{BB}=\delta_{AB}=\bar{\delta}$  are shown in Fig. 3 by solving (27) and (29) numerically. As mentioned in Sec. III, the phase boundaries (38) and (39) (solid lines) are independent of  $\bar{\delta}$ . The phase diagram for  $\bar{\delta}=0$  is equivalent to that found in Fig. 1 of Ref. 2 for the mixed ferromagnetic Ising square lattice. In the following, we use the same notations in describing the phase diagrams as those of Thorpe and McGurn.<sup>1</sup> In Fig. 3, only six kinds of the phase ( $A, A', S, S', B$  and  $B_1$ ) are permitted, although the nine phases ( $T, A, A', B, B_1, S, S', S_1$ , and  $S'_1$ ) compatible with the initial slopes are possible (see Fig. 1 of Ref. 1). As is seen from the figure, the phase boundary obtained from (27) is very sensitive to the change of  $\bar{\delta}$  in the region

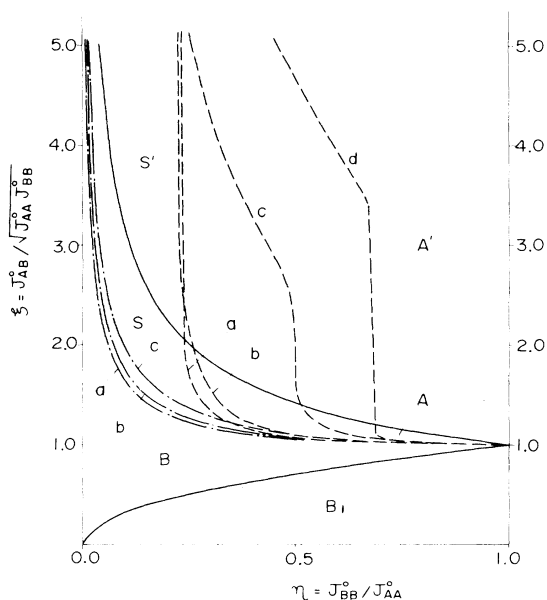


FIG. 3. Phase diagrams of square lattice ( $z=4$ ) for  $\delta_{AA}=\delta_{BB}=\delta_{AB}=\bar{\delta}$  with selected values of  $\bar{\delta}$ : (a)  $\bar{\delta}=0.0$ , (b)  $\bar{\delta}=0.50$ , (c)  $\bar{\delta}=0.90$ , (d)  $\bar{\delta}=0.05$ . Solid lines show the phase boundaries  $\xi=\sqrt{\eta}$  and  $\xi=1/\sqrt{\eta}$ . The dashed lines are obtained from the relation (27). The dotted-dashed lines are determined from the relation (29).

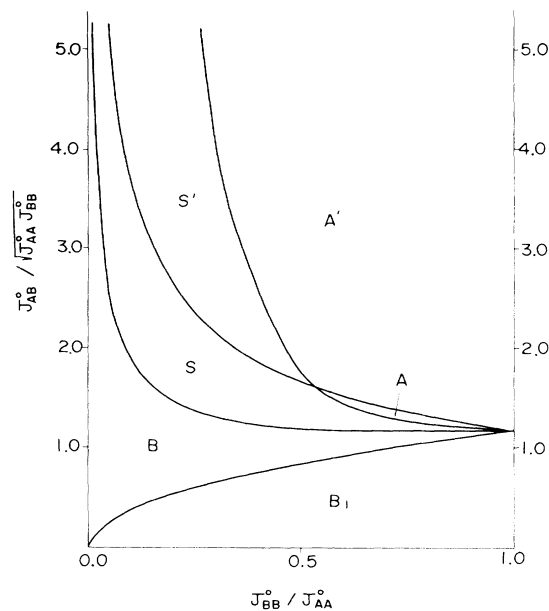


FIG. 4. Phase diagrams of square lattice ( $z=4$ ) for  $\delta_{AA}=\delta_{BB}=\delta_0=1.5, \delta_{AB}=1.5\delta_0$ .

$0.8 < \bar{\delta} < 1.0$ . On the other hand, the phase boundary obtained from (29) changes slightly upon increasing the value of  $\bar{\delta}$ . Especially for the value of  $\bar{\delta}$  in the range  $0.8 < \bar{\delta} < 0.9$ , the region of parameter space where the  $S$  and  $S'$  occur are extremely expanded, while the  $A$  and  $A'$  regions become narrower in comparison with each region

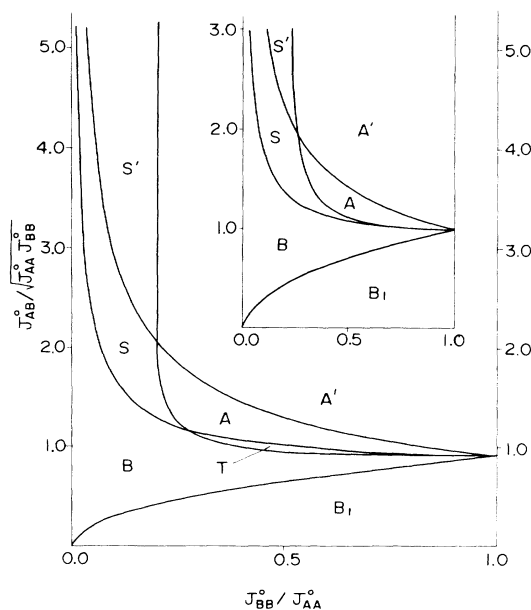


FIG. 5. Phase diagrams of square lattice ( $z=4$ ) for  $\delta_{AA}=\delta_{BB}=\delta_0=0.5, \delta_{AB}=0.1\delta_0$ . There is a new phase  $T$ . The figure inside corresponding the phase diagrams for  $\delta_{AA}=\delta_{BB}=\delta_{AB}=0.5$ .

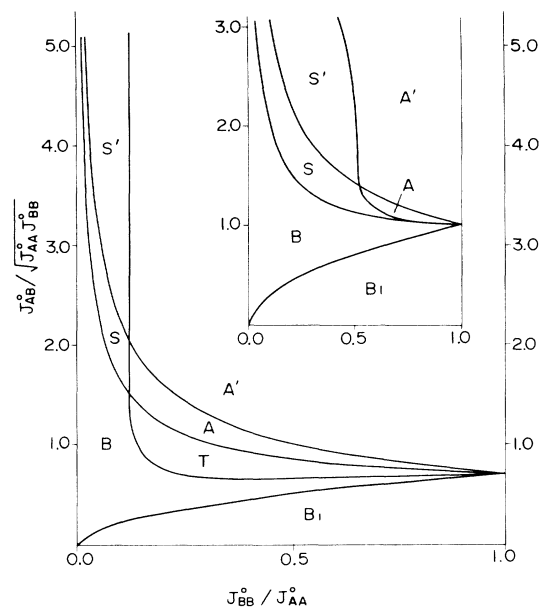


FIG. 6. Phase diagrams of a square lattice ( $z=4$ ) for  $\delta_{AA}=\delta_{BB}=\delta_0=0.9$ ,  $\delta_{AB}=0.1\delta_0$ . A new phase  $T$  appears. The figure inside corresponding the phase diagrams for  $\delta_{AA}=\delta_{BB}=\delta_{AB}=\delta_0=0.9$ .

for  $\bar{\delta}=0$ . Physically, the exchange interactions in the region can take a large value and a very small value with an equal probability, which implies that there appear some weakly-coupled spins in the system. Thus, the result suggests an interesting fact; a mixed Ising ferromagnetic crystalline alloy which expresses the concentration dependence of the  $A$  (or  $A'$ ) type may change the overall character of  $T_c$  versus  $p$  to the  $S$  (or  $S'$ ) type, when it becomes an amorphous state.

As mentioned in Sec. III, only for  $\delta_{AA}=\delta_{BB}=\delta_{AB}=\bar{\delta}$  the relations (38) and (39) are valid. Therefore, it is interesting to investigate the phase diagram of a system which does not satisfy the restriction  $\delta_{AA}=\delta_{BB}=\delta_{AB}=\bar{\delta}$ . For simplicity, let us now take the parameters as  $\delta_{AA}=\delta_{BB}=\delta_0$  and  $\delta_{AB}=a\delta_0$  ( $a\neq 1$ ). For three selected pairs of values ( $a, \delta_0$ ) the phase diagrams are depicted in Figs. 4–6. In Fig. 4, the values of  $a$  and  $\delta_0$  are chosen as  $a=1.5$  and  $\delta_0=0.5$ , in which the same six phases as those of Fig. 3 are also found. On the other hand, when we take  $a=0.1$  and  $\delta_0=0.5$ , a new phase  $T$  appears in Fig. 5 in addition to the six phases. The phase  $T$  is also found in Fig. 6, even when a pair of values ( $a=0.1$  and  $\delta_0=0.9$ ) is selected. Thus, this seventh-phase  $T$  seems to be characteristic of  $a \ll 1$ ; the structural fluctuation  $\delta_{AB}$  is very small in comparison with  $\delta_{AA}$  and  $\delta_{BB}$ . As discussed in Refs. 1 and 2, on the other hand, this phase has not been

found in mixed Ising ferromagnetic crystalline alloys, since the phase boundaries obtained from (26) and (28) are always given by the relations (38) and (39) in the systems.

## V. CONCLUSIONS

We have studied the initial slopes of a disordered binary Ising ferromagnetic alloy where all the exchange interactions have the same sign, using the effective-field theory with correlations introduced by Kaneyoshi *et al.*<sup>7</sup>

The initial slopes at  $p=1$  and  $p=0$  may determine the overall character of the phase diagram although not the detailed shape of course. From the three possible types of behavior at  $p=0$  and  $p=1$ , nine phases are compatible with initial slopes. As discussed in Refs. 1 and 2, in fact, only six kinds of phases have been permitted, although the standard MFT predicts only four kinds of phases. In classifying the possible nine phases compatible with the initial slopes at  $p=1$  and  $p=0$ , we have used the same notations as those of Thorpe and McGurn,<sup>1</sup> in order to avoid the confusion of readers.

In amorphous magnets, the fluctuation of exchange interactions (or the structural fluctuation) is one of underlying causes for the changes of physical quantities, in comparison with those of crystalline magnetic alloys. To our knowledge, the effects of the structural fluctuation on the phase diagrams (or the initial slopes) have not been studied. As shown in Sec. IV, the effects of structural fluctuations on their phase diagrams exhibit some interesting characteristics; a mixed Ising ferromagnetic crystalline alloy which expresses the concentration dependence of the  $A$  (or  $A'$ ) type may change the overall character of  $T_c$  versus  $p$  to the  $S$  (or  $S'$ ) type, when it becomes an amorphous state. A new seventh phase  $T$  appears in Figs. 5 and 6 in addition to the six phases, although such a phase has not been found in mixed Ising ferromagnetic crystalline alloys.<sup>1–2</sup>

Finally, for crystalline mixed alloys there is surprisingly little experimental data to compare with the phase diagrams predicted theoretically. In fact, many transition-metal (TM) compounds are of the Heisenberg type, and most of the insulating magnetic alloys are antiferromagnetic. On the other hand, most amorphous TM-metalloid alloys are ferromagnetic, and they can be fabricated with wider compositions than corresponding crystalline alloys. In Ref. 5, therefore, one of the present authors has compared some of the experimental data with the phase diagram predicted from the MFT, since many real amorphous ferromagnetic alloys usually have a high coordination number equal to  $z=12$ . We have found that most of the data lie in the  $A'$  phase. However, there may be also a possibility of finding through some characteristic discussed in this work, new amorphous ferromagnetic alloys such as an amorphous superlattice alloy with well-controlled composition.

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