# Critical temperatures and the compensation temperatures of disordered and amorphous ferrimagnetic Ising systems

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The concentration dependences of the transition temperature and compensation temperature in a disordered binary ferrimagnetic alloy and its amorphization are investigated by the use of effective-field theory with correlations. For the amorphization process, a number of interesting phenomena, such as the possibility of the reentrant ferrimagnetic phase, are obtained which arise from the random distribution of exchange bonds in sign. While new amorphous ferrimagnetic alloys may reveal other new phenomena, at the present time the main interest of experimentalists is directed towards obtaining an amorphous ferrimagnetic alloy which has a compensation point in the vicinity of room temperature, because of its potential device applications.

### I. INTRODUCTION

Over the past few years the theory of phase transitions in disordered binary magnetic systems has received considerable attention from both a bond and a site perspective. However, the existing theories are essentially confined to ferromagnetics and antiferromagnets. In both cases all the magnetic atoms have equal spins (e.g., their absolute values); 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 positive (or negative) values. In the site model, the lattice sites are randomly occupied by two different species of magnetic ions with equal absolute spin values, and the interaction between two ions is determined entirely by the species of those ions.<sup>1</sup>

On the other hand, amorphous ferrimagnetic rare-earth (RE)-transition-metal (TM) alloys<sup>2-5</sup> are currently of considerable interest because of their potential device applications. The alloys provide an excellent system for investigating basic magnetic phenomena in amorphous magnetic materials. The magnetic properties of the alloys are sensitive to slight compositional changes. The available experimental data confirm their untypical magnetic behavior. Of special interest are two physical parameters, the Curie temperature  $T_c$ , and the compensation temperature  $T_{\rm comp}$ , at which the macroscopic magnetizations of both components are identical and cancel each other. In particular, amorphous ferrimagnetic Gd-Co films have compensation points which vary with composition from approximately 40 to 500 K. The compositions of amorphous RE-TM alloys which lead to compensation points in the vicinity of room temperature have been investigated by many authors, following the suggestion of Chaudhari et al. that thermomagnetic writing and erasing can be carried out at  $T = T_{\rm comp}$  in these films.<sup>2</sup>

Theoretically, in order to analyze the magnetic properties of disordered ferrimagnetic alloys and their temperature and concentration dependences, mean-field theory has been extensively applied in the recent past. The theory can be in fact relied on for an appropriate description of the major aspects of the phenomena being studied. However, it is well known that the mean-field approximation (MFA) has some deficiencies, due to the neglect of correlations, when compared with experiments. Moreover, it is not so clear whether the simple MFA theory can be correctly applied to amorphous ferrimagnetic systems.

For discussing amorphous magnetic systems, there exist a great number of sophisticated techniques. Because of the difficulties inherent in the theoretical description of such complicated magnetic systems, it is sometimes necessary to make some simplifications. For studying such systems, therefore, the lattice model has often been applied, in which the structural disorder is replaced by a random distribution of the exchange integral. In a series of works we have investigated the amorphization of pure and diluted crystalline Ising ferromagnets by using both the effective-field theory (EFT) with correlations introduced by Kaneyoshi and co-workers<sup>6</sup> and the lattice model of amorphous magnets.<sup>7</sup> Due to amorphization, some interesting effects on the relevant thermodynamic quantities appeared in the thermal behavior. The EFT which substantially improves on the standard MFA has been successfully applied to a variety of other physical problems, such as pure systems,8 dilute ferromagnets,9 and systems with competing interactions,<sup>10</sup> as well as surface systems.11

In this paper, we investigate the transition temperatures and the compensation temperatures for two (disordered and amorphous) ferrimagnetic, binary, Ising alloys within the frameworks of EFT and the lattice model of amorphous magnets. As far as we know, those quantities have not been analyzed theoretically over the standard MFA as functions of both concentration and fluctuation of exchange interaction which normally appear in amorphous magnetic materials, except in one incomplete work.<sup>12</sup> The purpose of this work is thus to gain some qualitative insights on the above noted problems, whether the standard MFA can be applied successfully to disordered and amorphous ferrimagnetic binary alloys, and the effects of concentration and exchange fluctuation on these quantities.

The outline of this paper is as follows. In Sec. II we present the formulation of disordered and amorphous ferrimagnetic binary Ising alloys in the EFT. In Sec. III the theory is applied to a disordered ferrimagnetic, binary, square lattice, in order to discuss the effect of concentration and bond mixture on the transition temperatures and the compensation temperatures, and to compare the results of EFT with those of standard MFA. In Sec. IV the effects of amorphization on those quantities are investigated. We find that for the amorphization these quantities in particular exhibit some interesting behavior, such as the reentrant phenomena for ferrimagnetic phase.

# **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. Let the A and B atoms have different spins  $(s_A = \frac{1}{2} \text{ and } s_B = 1)$ , respectively. The interaction between spins is of the Ising form with the exchange interaction different for different pairs of spins. The Hamiltonian of the systems is then

$$H = -\sum_{i < j} [J_{AA}\delta_{iA}\delta_{jA} + J_{BB}\delta_{iB}\delta_{jB} + (J_{AB}\delta_{iA}\delta_{jB} + J_{BA}\delta_{iB}\delta_{jA})]s_i^z s_j^z \xi_i \xi_j , \qquad (1)$$

where the  $J_{ij}$ 's are the exchange interaction between type-*i* and type-*j* ions 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. Performing the random configurational average denoted by  $\langle \cdots \rangle_r$ , the averaged value of  $\xi_i$  has a restriction

$$\langle \xi_{i=A} \rangle_r + \langle \xi_{i=B} \rangle_r = 1$$
, (2)

where  $\langle \xi_{i=A} \rangle_r = p$  is the concentration of A atoms. For the amorphization of the system, moreover, it is necessary to take account of the fluctuation of exchange integral; the nearest-neighbor exchange interactions are given by independent random variables as

$$P(J_{AA}) = \frac{1}{2} [\delta(J_{AA} - J - \Delta J) + \delta(J_{AA} - J + \Delta J)],$$
  

$$P(J_{AB}) = P(J_{BA})$$
  

$$= \frac{1}{2} [\delta(J_{AB} - aJ - \Delta J') + \delta(J_{AB} - aJ + \Delta J')], \quad (3)$$
  

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

where parameters a and b are constants. The total magnetization per site of a binary alloy is then defined by

$$\langle s_i^z \rangle = \xi_{i=A} \langle s_{i=A}^z \rangle + \xi_{i=B} \langle s_{i=B}^z \rangle , \qquad (4)$$

where the angular brackets denote the usual thermal averages.

The main problem is now the evaluation of the mean values,  $\langle s_A^z \rangle$  and  $\langle s_B^z \rangle$ . As has been discussed in a series of works,<sup>13</sup> the starting point for the evaluation of  $\langle s_{i=A}^z \rangle$  with  $s_A = \pm \frac{1}{2}$  is the exact Callen identity<sup>14</sup>

$$\langle s_{i=A}^{z} \rangle = \frac{1}{2} \left\langle \tanh \left( \frac{\xi_{i=A} \beta \theta_{i=A}}{2} \right) \right\rangle,$$
 (5)

with

$$\theta_{i=A} = \sum_{j} (J_{AA}\delta_{jA} + J_{AB}\delta_{jB})\xi_{j}s_{j}^{z},$$

where  $\beta = 1/k_B T$ . On the other hand, the mean value of  $\langle s_{i=B}^z \rangle$  with  $s_B = \pm 1$  and 0 is also given by the exact relation<sup>15</sup>

$$\langle s_{i=B} \rangle = \left\langle \frac{2 \sinh(\xi_{i=B} \beta \theta_{i=B})}{2 \cosh(\xi_{i=B} \beta c_{i=B}) + 1} \right\rangle, \tag{6}$$

with

$$\theta_{i=B} = \sum_{j} (J_{BB}\delta_{jB} + J_{BA}\delta_{jA})\xi_{j}s_{j}^{z}$$

At this stage, in order to write identities (5) and (6) in a form which is particularly amenable to approximation, we have introduced the differential operator technique<sup>6</sup> as follows:

$$\langle s_{i=A}^{z} \rangle = \langle e^{D(\xi_{i=A}\theta_{i=A})} \rangle f(x) |_{x=0}$$
<sup>(7)</sup>

and

$$\langle s_{i=B}^{z} \rangle = \langle e^{D(\xi_{i=B}\theta_{i=B})} \rangle g(x) |_{x=0} , \qquad (8)$$

where  $D = \partial/\partial x$  is a differential operator. The functions f(x) and g(x) are defined by

$$f(x) = \frac{1}{2} \tanh\left[\frac{\beta}{2}x\right]$$
(9)

and

$$g(x) = \frac{2\sinh(\beta x)}{2\cosh(\beta x) + 1} .$$
 (10)

Using the relation  $\xi_i^n = \xi_i$  (*n*=integer) and the identities<sup>6,15</sup>

$$\exp(\gamma s_A^z) = \cosh\left[\frac{\gamma}{2}\right] + 2s_A^z \sinh\left[\frac{\gamma}{2}\right]$$
(11)

and

$$\exp(\gamma s_B^z) = (s_B^z)^2 \cosh(\gamma) + s_B^z \sinh(\gamma) + 1 - (s_B^z)^2,$$

the expectation values appearing in Eqs. (7) and (8) are generally rewritten as

$$\langle e^{D(\xi_{i=a}\theta_{i=a})}\rangle = \left\langle \prod_{j} \left\{ \xi_{j} \delta_{jA} \left[ \cosh\left(\frac{D_{aA}^{i}}{2}\right) + 2s_{j}^{z} \sinh\left(\frac{D_{aA}^{i}}{2}\right) \right] + \xi_{j} \delta_{jB} \left[ (s_{j}^{z})^{2} \cosh(D_{aB}^{i}) + s_{j}^{z} \sinh(D_{aB}^{i}) + 1 - (s_{j}^{z})^{2} \right] \right\} \right\rangle,$$
(12)

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where  $\alpha = A$  or B and the parameters  $D_{\alpha A}^{i}$  and  $D_{\alpha B}^{i}$  are defined by

$$D_{\alpha B}^{i} = D\xi_{i=\alpha}J_{\alpha B}$$
 and  $D_{\alpha A}^{i} = D\xi_{i=\alpha}J_{\alpha A}$ 

For a disordered system with random bonds and random occupation of magnetic atoms, we must perform the random-configurational average for Eqs. (4), (7), and (8); the averaged total magnetization per site is given by

$$m = \langle \langle s_i^z \rangle \rangle_r = pm_A + (1-p)m_B , \qquad (13)$$

where  $m_A$  and  $m_B$  are defined by

$$m_{A} = \frac{\langle \xi_{i=A} \langle s_{i=A}^{z} \rangle \rangle_{r}}{\langle \xi_{i=A} \rangle_{r}} , \qquad (14)$$

$$m_B = \frac{\langle \xi_{i=B} \langle s_{i=B}^z \rangle \rangle_r}{\langle \xi_{i=B} \rangle_r} . \tag{15}$$

Here, it is clear that if we try to treat exactly all the spinspin correlations appearing in the sublattice magnetizations through the expansion of Eq. (12) and to perform properly the random configurational average, the problem becomes mathematically untractable. Therefore, in the previous papers, within the EFT, the decoupling approximation, or

$$\langle \langle x_j x_k(x_l)^2 \cdots x_n \rangle \rangle_r$$
  

$$\simeq \langle \langle x_j \rangle \rangle_r \langle \langle x_k \rangle \rangle_r \langle \langle (x_l)^2 \rangle \rangle_r \cdots \langle \langle x_n \rangle \rangle_r , \quad (16)$$

with  $j \neq k \neq l \neq \cdots \neq n$  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>16</sup> as discussed in Refs. 6 and 13. The approximation has been successfully applied to a great number of disordered magnetic systems.<sup>6-11,15</sup>

Taking account of the facts that the exchange interactions and the random occupation of magnetic atom sites are given by independent random variables, the sublattice magnetizations  $m_A$  and  $m_B$  for the EFT reduce to, upon performing the random average,

$$m_{A} = \left\{ p\left[\left\langle \cosh\left(\frac{1}{2}DJ_{AA}\right)\right\rangle_{r} + 2m_{A}\left\langle \sinh\left(\frac{1}{2}DJ_{AA}\right)\right\rangle_{r}\right] + (1-p)\left[q_{B}\left\langle \cosh\left(DJ_{AB}\right)\right\rangle_{r} + m_{B}\left\langle \sinh\left(DJ_{BB}\right)\right\rangle_{r} + 1-q_{B}\right]\right\}^{z} f(x) \big|_{x=0}$$
(17)

and

$$m_{B} = \left\{ p\left[\left\langle \cosh\left(\frac{1}{2}DJ_{BA}\right)\right\rangle_{r} + 2m_{A}\left\langle \sinh\left(\frac{1}{2}DJ_{BA}\right)\right\rangle_{r}\right] + (1-p)\left[q_{B}\left\langle \cosh(DJ_{BB}\right)\right\rangle_{r} + m_{B}\left\langle \sinh(DJ_{BB}\right)\right\rangle_{r} + 1-q_{B}\right]\right\}^{z}g(x) \mid_{x=0},$$
(18)

where z is the number of nearest neighbors. The parameter  $q_B$  is defined by

$$q_B = \frac{\langle \xi_{i=B} \langle (S_{i=B}^z)^2 \rangle \rangle_r}{\langle \xi_{i=B} \rangle_r} .$$
<sup>(19)</sup>

In order to obtain the sublattice magnetizations, therefore, it is necessary to calculate the parameter  $q_B$ . As is understood from the discussions of Ref. 15, we can easily get, in the same way as the evaluation of  $m_B$ ,

$$q_{B} = \{p[\langle \cosh(\frac{1}{2}DJ_{BA})\rangle_{r} + 2m_{A}\langle \sinh(\frac{1}{2}DJ_{BA})\rangle_{r}] + (1-p)[q_{B}\langle \cosh(DJ_{BB})\rangle_{r} + m_{B}\langle \sinh(DJ_{BB})\rangle_{r} + 1-q_{B}]\}^{z}h(x)|_{x=0}, \qquad (20)$$

with

$$h(x) = \frac{2\cosh(\beta x)}{2\cosh(\beta x) + 1} .$$
(21)

In this section we have discussed the effective-field theory with correlations in a binary Ising alloy with random bonds. We are in a position to examine the transition temperatures and the compensation temperatures of a disordered crystalline ferrimagnetic, binary, Ising alloy and its amorphization. In the following sections, within this framework, we shall study the physical quantities. At this point, it is worth commenting that for particular cases the above equations reduce to those already discussed in previous work; for the case with  $J_{BB}=J_{AB}=J_{BA}=0$ ,  $m_B=0$ , and  $M_A$  is nothing but the equation discussed in Ref. 13 for the amorphization of diluted crystalline Ising ferromagnet. In the case of p=0, Eqs. (18) and (20) also reduce to those with zero uniaxial constant in Ref. 15 for the pure (nonrandom bond) problem.

# III. DISORDERED FERRIMAGNET IN SQUARE LATTICE

In this section we investigate a disordered binary ferrimagnet on a square lattice; the term "disordered" means that the distribution of all atoms is assumed to be completely random, but the interaction between two ions is determined entirely by the species of those ions. Therefore, it is not necessary to take account of the effect of the random distributions of bonds [namely, Eq. (3)] in Eqs. (17), (18), and (20).

We are now interested in studying the transition temperatures and the compensation temperatures of the disordered ferrimagnet. In order to determine the transition temperature, the usual argument that the sublattice magnetizations tend to zero as the temperature approaches a critical temperature allows us to consider only terms linear in the sublattice magnetizations for Eqs. (17) and (18). For the ferrimagnetic square lattice (z=4) with negative values of  $J_{AB}=J_{BA}$ , the sublattice magnetizations then reduce to

$$(A_1 - 1)m_A + B_1 m_B = 0,$$
  

$$A_2 m_A + (B_2 - 1)m_B = 0,$$
(22)

with

$$A_{1} = 8p^{4}K_{1} + 24p^{3}(1-p)[K_{3}q_{B} + K_{7}(1-q_{B})] + 24p^{2}(1-p)^{2}[K_{10}q_{B}^{2} + 2K_{13}q_{B}(1-q_{B}) + K_{12}(1-q_{B})^{2}] + 8p(1-p)^{3}[K_{20}q_{B}^{3} + 3K_{22}(1-q_{B})q_{B}^{2} + 3K_{24}(1-q_{B})^{2}q_{B} + K_{25}(1-q_{B})^{3}],$$
(23)

$$B_{1} = 4p^{3}(1-p)K_{5} + 12p^{2}(1-p)^{2}[K_{8}q_{B} + K_{9}(1-q_{B})] + 12p(1-p)^{3}[K_{16}q_{B}^{2} + 2K_{18}(1-q_{B})q_{B} + K_{19}(1-q_{B})^{2}] + 4(1-p)^{4}[K_{26}q_{B}^{3} + 3K_{28}(1-q_{B})q_{B}^{2} + 3K_{30}(1-q_{B})^{2}q_{B} + K_{31}(1-q_{B})^{3}],$$
(24)

$$A_{2} = 8p^{4}L_{1} + 24p^{3}(1-p)[L_{3}q_{B} + L_{7}(1-q_{B})] + 24p^{2}(1-p)^{2}[L_{10}q_{B}^{2} + 2L_{13}q_{B}(1-q_{B}) + L_{12}(1-q_{B})^{2}] + 8p(1-p)^{3}[L_{20}q_{B}^{2} + 3L_{22}(1-q_{B})q_{B}^{2} + 3L_{24}(1-q_{B})^{2}q_{B} + L_{25}(1-q_{B})^{3}],$$
(25)

$$B_{2} = 4p^{3}(1-p)L_{5} + 12p^{2}(1-p)^{2}[L_{8}q_{B} + L_{9}(1-q_{B})] + 12p(1-p)^{3}[L_{16}q_{B}^{2} + 2L_{18}(1-q_{B})q_{B} + L_{19}(1-q_{B})^{2}] + 4(1-p)^{4}[L_{26}q_{B}^{3} + 3L_{28}(1-q_{B})q_{B}^{2} + 3L_{30}(1-q_{B})^{2}q_{B} + L_{31}(1-q_{B})^{3}].$$
(26)

The critical surface characterizing the ferrimagnetic phase stability limit is determined by

$$(A_1 - 1)(B_2 - 1) = A_2 B_1 . (27)$$

Looking at Eqs. (23)–(26), the parameter  $q_B$  is included in them. In order to obtain the critical temperature  $T_c$  from Eq. (27), it is at first necessary to evaluate the parameter  $q_B$  at  $T = T_c$ , which is given by, from Eq. (20),

$$q_{B} = p^{4}M_{1} + 4p^{3}(1-p)[M_{4}q_{B} + M_{8}(1-q_{B})] + 6p^{2}(1-p)^{2}[M_{10}q_{B}^{2} + M_{12}(1-q_{B})^{2} + 2M_{13}q_{B}(1-q_{B})] + 4p(1-p)^{3}[M_{20}q_{B}^{3} + 3M_{22}(1-q_{B})q_{B}^{2} + 3M_{24}(1-q_{B})^{2}q_{B} + M_{25}(1-q_{B})^{3}] + (1-p)^{4}[M_{30}q_{B}^{4} + 4(1-q_{B})q_{B}^{3}M_{33} + 6M_{35}(1-q_{B})^{2}q_{B}^{2} + 4(1-q_{B})^{3}q_{B}M_{37} + M_{38}(1-q_{B})^{4}].$$
(28)

The coefficients  $K_i$ ,  $L_j$ , and  $M_k$  in Eqs. (23)–(26) and (28) are given in Appendix A.

In ferrimagnetic materials, on the other hand, the sublattice magnetizations do not have the same sign, and there may be a compensation temperature  $T_{\rm comp}$  at which the total magnetization m=0, even though  $m_A \neq 0$  and  $m_B \neq 0$ . The situation arises when, from Eq. (13),

$$pm_A = -(1-p)m_B$$
 (29)

Even for the case of a square lattice (z=4), the sublattice magnetizations have complicated forms, when Eqs. (17) and (18) are expanded. Using the relation (29), however, the sublattice magnetizations at  $T=T_{comp}$  can be written in compact forms as follows:

$$m_A^2 = \frac{1 - C_1}{C_2} \tag{30}$$

and

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$$n_B^2 = \frac{1 - D_1}{D_2} \ . \tag{31}$$

The factors  $C_1$ ,  $C_2$ ,  $D_1$ , and  $D_2$  in Eqs. (30) and (31) are given in Appendix B. In the factors, the parameter  $q_B$  is also included, which is given, on using Eq. (20) for z=4 and relation (29), by

$$q_B = E_1 + E_2 m_A^2 + E_3 m_A^4 , \qquad (32)$$

where coefficients  $E_1$ ,  $E_2$ , and  $E_3$  are given in Appendix B. By using Eqs. (29)–(32), consequently, we can obtain a closed-form, although complicated, expression for determining the compensation temperature. If a compensation temperature exists, it can be obtained by numerically scanning the temperature range between 0 K and the Curie temperature.

We are now able to evaluate the Curie temperatures and the compensation temperatures by the use of the above derived equations. Before discussing the numerical results, we first review the MFA theory of a binary ferrimagnet. In subsection A the results of the MFA are given. The numerical results of the present formulation are given in Sec. III B and compared with those of the MFA.

# A. MFA results

In the MFA the sublattice magnetizations in a disordered binary ferrimagnet are

$$m_{A} = \frac{1}{2} \tanh \left[ \frac{1}{2k_{B}T} [zpJ_{AA}m_{A} + z(1-p)J_{AB}m_{B}] \right],$$

$$m_{B} = B_{1} \left[ \frac{1}{k_{B}T} [zpJ_{BA}m_{A} + z(1-p)J_{BB}m_{B}] \right],$$
(33)

where  $B_1[x]$  is the Brillouin function for s = 1. Near the Curie temperature, Eqs. (33) may be expanded and simplified as, for a square lattice with z = 4,

$$m_{A} = \frac{1}{k_{B}T} [pJ_{AA}m_{A} + (1-p)J_{AB}m_{B}] ,$$
  
$$m_{B} = \frac{8}{3k_{B}T} [pJ_{BA}m_{A} + (1-p)J_{BB}m_{B}] ,$$

from which the Curie temperature is given by

$$\frac{k_B T_c}{J} = \frac{1}{2} \left( p + \frac{8}{3} (1-p)b + \left\{ \left[ p - \frac{8}{3} (1-p)b \right]^2 + \frac{32}{3} p (1-p)a^2 \right\}^{1/2} \right), \quad (34)$$

where we have defined the exchange interactions as

$$J_{AA} = J ,$$

$$J_{AB} = J_{BA} = aJ ,$$

$$J_{BB} = bJ .$$
(35)

In the ferrimagnetic material with a < 0, on the other hand, the compensation temperature is determined by solving Eqs. (29) and (33) numerically in the range of temperature between 0 K and the Curie temperature. In Figs. 1 and 3, some numerical results of Curie temperature and compensation temperature in a ferrimagnetic square lattice are depicted, in order to compare the MFA results with those of the EFT.

#### B. Numerical results of the EFT

For some selected pairs of values (a,b), the behavior of  $T_c$  and  $T_{comp}$  versus concentration in a binary ferrimagnetic square lattice are depicted in Figs. 1–3. In Fig. 1, in order to compare the MFA results with those of the EFT, some results of the quantities are shown for some pairs of values (a,b) with a fixed value of b=0.1. The figure clearly expresses that the MFA results for both quantities deviate somewhat from those of the EFT, although they show the qualitative description of the major aspects of the phenomena being studied; because of its simplicity, the MFA theory has been used by a great number of experimentalists for analyzing their experimental data of ferrimagnetic alloys. The results of Fig. 1, however, imply that the MFA theory must be applied with caution for analyzing the experimental data.

In Figs. 2 and 3 the concentration dependences of  $T_c$  and  $T_{\rm comp}$  obtained from the EFT are depicted for some selected pairs of values (a,b). In Fig. 3 the MFA results of  $T_c$  are also shown for comparison. In the figure, the



FIG. 1. Reduced plots of  $T_c$  and  $T_{comp}$  as a function of p of atoms with  $s = \frac{1}{2}$  for some selected pairs of values (a, b = 0.1): (a) (-0.01, 0.1), (b) (-0.1, 0.1), (c) (-0.5, 0.1), and (d) (-1.0, 0.1). The solid lines are the MFA results, and the dashed lines are the results of the EFT.



FIG. 2. Plots of  $T_c$  and  $T_{comp}$  vs p for selected pairs of values (a,b=0.1): (a) (-0.01,0.1), (b) (-0.1,0.1), (c) (-0.25,0.1), (d) (-0.5,0.1), (e) (-0.75,0.1), and (f) (-1.0,0.1). The solid lines are the concentration variation of  $T_c$  and the dashed lines are the concentration variation of  $T_{comp}$ .

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FIG. 3. Plots of  $T_c$  and  $T_{comp}$  vs p for selected pairs of values (a = -0.135, b): (a) (-0.135, 0.014), (b) (-0.135, 0.1), (c) (-0.135, 0.3), and (d) (-0.135, 0.5). The solid lines are the concentration variation of  $T_c$  and the dashed lines are those of  $T_{comp}$ . The dot-dashed lines are the MFA results.

transition temperature of the EFT for p=1 is given by  $k_BT_c/J=0.7724$ , which is nothing but the result obtained from the Zernike approximation,<sup>13,11</sup> although the MFA leads to  $k_BT_c/J=1.0$  for p=1. As is seen from Figs. 1–3, on the other hand, the values of  $T_{\rm comp}$  are found in a narrow range of p, when the value of a (or b) under a fixed value of b (or a) increases. The concentration at which  $T_{\rm comp}=0$  in the figures is given by  $p^*=\frac{2}{3}$ , since at T=0,  $m_A=0.5$  and  $m_B=-1$ , and the concentration for m=0 is determined from  $\frac{1}{2}p-(1-p)=0$ . In curve (a) of Fig. 3 numerical values of a and b are especially chosen as a=-0.135 and b=0.014, since these values may be consistent with experimental data for amorphous Gd-Co alloys.<sup>12</sup>

In Figs. 4 and 5 the concentration dependences of  $T_c$  for a given value of a (or b) obtained from the present formulation are depicted, on changing the value of b (or a). A particularly interesting result appears in the figures; for a=0 (or b=0) the ferromagnetic (or ferrimagnetic) phase cannot be obtained in a range of p, which just corresponds to the dilution problem of exchange bonds. Such a behavior cannot be obtained from the MFA theory, for instance, as shown in Fig. 5.

In this section, we have studied the behavior of  $T_c$  and  $T_{\rm comp}$  versus the change of p in a disordered binary ferrimagnetic square lattice. In amorphous magnetic materials, on the other hand, it has been discussed that the fluctuations in the exchange interactions are the underlying causes for the changes of physical quantities, in compar-



FIG. 4. Concentration dependences of  $T_c$  for selected pairs of values (a, b = 1.0): (a) (0.0, 1.0), (b) (-0.2, 1.0), (c) (-0.4, 1.0), (d) (-0.6, 1.0), (e) (-0.8, 1.0), (f) (-1.0, 1.0), (g) (-1.2, 1.0), (h) (-1.4, 1.0), (i) (-1.6, 1.0), (j) (-1.8, 1.0), and (k) (-2.0, 1.0).



FIG. 5. Concentration dependences of  $T_c$  for selected pairs of values (a = -1.5, b): (a) (-1.5, 0.0), (b) (-1.5, 0.1), and (c) (-1.5, 0.3). The dot-dashed line is the MFA result for curve (a).

ison with those of crystalline magnetic alloy.<sup>7,17,18</sup> In the next section the effects of exchange fluctuations on  $T_c$  and  $T_{\text{comp}}$  will be investigated.

### IV. AMORPHIZATION OF FERRIMAGNETIC SQUARE LATTICE

To describe the structural disorder in a simple way, the lattice model has been often used in amorphous magnets; the nearest-neighbor exchange interactions are given by independent random variables (3). The random-bond averages in Eqs. (17) and (18) are then given by

$$\left\langle \cosh\left[D\frac{J_{AA}}{2}\right]\right\rangle_{r} = \cosh\left[\frac{J}{2}D\right] \cosh\left[\frac{J}{2}\frac{\Delta J}{J}D\right],$$

$$\left\langle \sinh\left[D\frac{J_{AA}}{2}\right]\right\rangle_{r} = \sinh\left[\frac{J}{2}D\right] \cosh\left[\frac{J}{2}\frac{\Delta J}{2}D\right],$$

$$\left\langle \cosh(DJ_{AB})\right\rangle_{r} = \cosh(aJD) \cosh\left[\frac{J}{2}\frac{2\Delta J'}{J}D\right],$$

$$\left\langle \sinh(DJ_{AB})\right\rangle_{r} = \sinh(aJD) \cosh\left[\frac{J}{2}\frac{2\Delta J'}{J}D\right],$$

$$\left\langle \cosh(DJ_{BB})\right\rangle_{r} = \cosh(bJD) \cosh\left[\frac{J}{2}\frac{2\Delta J''}{J}D\right],$$

$$\left\langle \sinh(DJ_{BB})\right\rangle_{r} = \sinh(bJD) \cosh\left[\frac{J}{2}\frac{2\Delta J''}{J}D\right],$$

$$\left\langle \sinh(DJ_{BB})\right\rangle_{r} = \sinh(bJD) \cosh\left[\frac{J}{2}\frac{2\Delta J''}{J}D\right].$$

Here, it is extremely tedious to treat in general the amorphization of the disordered ferrimagnetic square lattice discussed in Sec. III, so that let us take

$$\frac{\Delta J}{J} = \frac{2\,\Delta J'}{J} = \frac{2\,\Delta J''}{J} = \delta , \qquad (37)$$

where  $\delta$  is a dimensionless parameter which measures the amount of fluctuation of exchange interactions. The parameter  $\delta$  is often called the structural fluctuation. The transition temperatures and compensation temperatures of the amorphization in the disordered ferrimagnetic square lattice can be evaluated from Eqs. (27) and (28) for  $T_c$  and from Eqs. (29)–(32) for  $T_{\rm comp}$ , except for the fact that in the coefficients,  $K_i$ ,  $L_j$ , and  $M_k$ , a power of the extra factor  $\cosh[(J/2)\delta D]$  must be included, and the power depends on how many of the functions of sinh and cosh are included in the coefficients given in Appendix A.

Let us now discuss the numerical results for the amorphization of the disordered ferrimagnetic square lattice. In Figs. 6 and 7, the effects of the amorphization on the  $T_c$  and  $T_{\rm comp}$  are shown. As is seen from the figures, the effect of amorphization on  $T_c$  and  $T_{\rm comp}$  is for them to fall below the values of  $T_c$  and  $T_{\rm comp}$  in the disordered ferrimagnetic crystalline alloy. Such a phenomenon is generally found in amorphous RE-TM ferrimagnetic alloys.<sup>4</sup>

At this point some interesting phenomena of amorphization are observed in the figures. For the weak amorphization ( $\delta = 0.2$ ) of the curve (a) with a = -0.01 and b = 0.1 shown in Fig. 6, the value of  $T_c$  disappears in the



FIG. 6. Concentration variations of  $T_c$  and  $T_{comp}$  for selected pairs of values (a, b = 0.1): (a) (-0.01, 0.1), (b) (-0.25, 0.1), (c) (-0.5, 0.1), and (d) (-0.1, 0.1). The solid lines are for the weak amorphization  $(\delta=0.2)$ . The dashed lines are for the disordered crystalline alloys with  $\delta=0.1$  in Fig. 2.



FIG. 7. Concentration variations of  $T_c$  and  $T_{comp}$  for the three values of  $\delta$ : (a)  $\delta = 0.0$ , (b)  $\delta = 0.2$ , and (c)  $\delta = 0.5$ , when the pair of values (a = -0.135, b) are chosen as (-0.135, 0.1) solid line, (-0.135, 0.3) dashed line, and (-0.135, 0.5) dot-dashed line. The insert is the variation of  $p^*$  for the system with (-0.135, 0.3), when  $\delta$  is changed.

region of p given by 0.175 and also a value of $T_{\rm comp}$  cannot be obtained; the exchange interaction  $J_{AB}$ then can take positive and negative values randomly because of the fact that a = -0.01 and  $\delta = 0.2$ . The effect of so called "frustration" appears in the region of p in which the phase may be spin-glass-like. For the curve (a) of Fig. 3 with a = -0.135 and b = 0.014, similar behavior can be also obtained for weak amorphization ( $\delta = 0.2$ ). For instance, the solid curve (c) with a = -0.135 and b=0.1 in Fig. 7 does not have a value of  $T_c$  in the range of  $p \ (0 and the value of <math>T_{\text{comp}}$  cannot be obtained (Fig. 7) for the amorphization of  $\delta = 0.5$ , since  $J_{AB}$ and  $J_{BB}$  then can take positive and negative values randomly. On the other hand, nontypical behavior is also obtained in the concentration dependence of  $T_{\rm comp}$  for the amorphization, the solid curve (b) with a = -0.135 and b=0.1 in Fig. 7. As is seen from Figs. 6 and 7, for weak amorphization ( $\delta = 0.2$ ) the concentration for  $T_{\rm comp} = 0$  is also given by  $p^* = \frac{2}{3}$ . Increasing the structural fluctuation, however, the concentration of  $T_{\rm comp} = 0$  decreases rapidly from  $p^* = \frac{2}{3}$  at a value of  $\delta$ , such as the inset of Fig. 7 obtained for the case with a = -0.135 and b = 0.3. The value of  $\delta$  at which the value of  $p^*$  rapidly decreases from  $p^* = \frac{2}{3}$  depends on the values of a and b. The change of  $p^*$  is due to the decreases from  $m_A = \frac{1}{2}$  and  $m_B = -1$  of sublattice magnetizations at T = 0 K in the sense that it becomes impossible for some spins to take any preferable direction because of the random distribution of  $J_{ii}$  in sign, although in this case the ferrimagnetic order is not destroyed completely, as is seen from Fig. 7. Thus, if the difference of the values of  $p^*$  exists between a disordered ferrimagnetic crystalline alloy and its corresponding amorphous ferrimagnetic alloy with the same composition, it may indicate that the exchange interactions are in sign distributed randomly in the amorphous alloy.

Now, it is here worth commenting on some relations between the results obtained within the present formulation (z=4) and those in real amorphous RE-TM ferrimagnetic alloys which may be assumed to be z = 12. In real systems  $J_{AA}$ ,  $J_{AB}$ , and  $J_{BB}$  correspond to TM-TM, RE-TM, and RE-RE interactions, respectively. The magnitudes of exchange interactions are usually taken as  $J_{BB} < -J_{AB} < J_{AA}$ . In order to analyze the experimental data of amorphous RE-TM ferrimagnetic alloys, all experimentalists have taken a model discussed in Sec. III; RE and TM ions are randomly distributed on a lattice with z = 12 and the exchange interaction between two ions is determined entirely by the species of those ions. Except for the authors of Ref. 12, the sublattice magnetizations  $m_A$  and  $m_B$  are then assumed to follow Brillouin functions (or the MFA theory) for the spin values  $s_{RE}$  and  $s_{TM}$  ( $s_{TM} < s_{RE}$ ), like Eq. (33). In order to explain the concentration dependence of  $T_c$  in amorphous binary ferrimagnets, in some works the spin value of TM is not a constant value, but the variation of the TM moment in terms of a charge transfer model with the addition of a moment induced by the RE is taken into account within the framework of the MFA.<sup>19</sup> On the other hand, Zagorski and Nazarewicz<sup>12</sup> have tried to extend the MFA theory of the model to a better approximate theory, although their work seems to be incomplete in comparison with the discussions of Sec. III. The exchange interactions are then taken as  $J_{AA} = J$ ,  $J_{AB} = -0.135J$ , and  $J_{BB} = 0.014J$ , which values may be consistent with the experimental data for amorphous Gd-Co alloys. They obtained the concentration dependence of  $T_c$  and  $T_{comp}$  for the fcc lattice structure (z = 12) with constant spin values ( $s_A = \frac{1}{2}$  and  $s_B = 1$ ); their results are similar to those of curve (a) in Fig. 3 of Sec. III for z = 4. The results are not in contradiction with the experimental data, although a close comparison is not possible.

To our knowledge, however, the effects of the structural fluctuation  $\delta$  on these quantities in amorphous ferrimagnets have not been discussed in the previous works. Theoretically, when we use the MFA theory, it is known that the effect of the structural fluctuation on  $T_c$  cancels out and does not appear, and its effect on  $T_{\text{comp}}$  is very small.<sup>20</sup> However, the results are characteristic of the MFA. As discussed in this section, when we use an approximation better than the MFA, a number of interesting effects of  $\delta$  come up in the behavior of  $T_c$  and  $T_{comp}$ . The results imply that the MFA theory not including the structural fluctuation may give incorrect analyses of the experimental data for  $T_c$  and  $T_{comp}$ . Experimentally, in amorphous TM-metalloid alloys, the fluctuation of exchange interaction  $J_{AA}$  is considered as an important ingredient for the appearance of characteristic behavior, such as the depression of reduced magnetization curves, reentrant phenomena, and spin-glass phase. In amorphous Gd noble-metal ferromagnetic alloys, on the other hand, reentrant phenomena and spin-glass phase are also found; the exchange interaction  $J_{BB}$  is considered to be fluctuating around a mean value. Thus, in amorphous **RE-TM** ferrimagnetic alloys, it may be reasonable to take account of the structural fluctuation, like Eqs. (3) and (37), although it is not certain whether the exchange interaction  $J_{AB}$  fluctuates around a mean value.

In previous work on the amorphization of a crystalline Ising ferromagnet on a square lattice with  $s = \frac{1}{2}$ , <sup>13</sup> we have discussed that reentrant phenomena can be obtained for large structural fluctuation in the range of  $\overline{\delta}(0.5 < \overline{\delta} < 0.565)$  and for  $\overline{\delta} > 0.565$  the system may be in the spin-glass phase (the definition of  $\delta$  in the work corresponds to  $\delta = 2\overline{\delta}$  in the present formulation). The results can also be obtained from Eq. (17), when we put p = 1 and z = 4 into it and use the distribution function (3) and (36). Therefore, in Figs. 8 and 9, the effects of large fluctuations  $\delta$  on  $T_c$  are depicted for three selected pairs of values (a,b) in Figs. 4 and 5. In Fig. 8 the exchange interactions are taken as  $J_{AA} = -J_{AB} = J_{BB} = J$  (or a = -1and b=1). Increasing the structural fluctuation, as mentioned above, the transition temperature at p = 1 decreases monotonically until the value of  $\delta = 1$  and in the range  $1.0 < \delta < 1.13$  the reentrant ferromagnetic phase is obtained and for  $\delta > 1.13$  the system becomes spin-glass-like. Even for the amorphization, the concentration variation of  $T_c$  changes almost linearly (except the region near p=1) until the value of  $\delta = 1$  [or curve (c)]. For the curve (d) with  $\delta = 1.2$ , the possibility of a reentrant ferrimagnetic phase is seen in the region 0.86 , since the system for p = 1 is already spin-glass-like. For a little larger



FIG. 8. Concentration dependences of  $T_c$  in the system with a = -1.0 and b = 1.0 for the amorphization: (a)  $\delta = 0.0$ , (b)  $\delta = 0.5$ , (c)  $\delta = 1.0$ , (d)  $\delta = 1.2$ , and (e)  $\delta = 1.5$ .



FIG. 9. The concentration dependences of  $T_c$  in the two systems with solid line (a = -1.5, b = 0.3) and dashed line (a = -1.5, b = 0.0) for the amorphization: (a)  $\delta = 0.0$ , (b)  $\delta = 0.5$ , (c)  $\delta = 1.0$ , and (d)  $\delta = 1.2$ .

value of  $\delta$  [or curve (e)], however, the reentrant ferrimagnetic phase is found only in a very narrow region of p. On the other hand, in Fig. 9 the exchange interaction  $J_{AB}$ is fixed at a = -1.5 and two values of  $J_{BB}$  (or b = 0.0 and b=0.3) are chosen. For large amorphization, a reentrant ferrimagnetic phase is also observed in the figure: for the solid curve (c) with  $\delta = 1.0$ , in the region 0.382because of randomly distributed positive and negative values of  $J_{BB}$ , and for the solid curve (d) with  $\delta = 1.2$ , in two regions, 0.46 and <math>0.885 , because of the frustration coming from  $J_{AA}$  and  $J_{BB}$ . As shown in Figs. 6-9, in this way a number of interesting phenomena coming from the random distribution of exchange bonds may be expected in amorphous ferrimagnetic alloys, although at the present time we do not have any experimental result.

### **V. CONCLUSION**

We have studied the concentration dependence of transition and compensation temperatures in a disordered binary ferrimagnetic square lattice and its amorphization using effective-field theory with correlations. As discussed in Sec. IV, the MFA theory especially must be applied with caution to amorphous binary ferrimagnetic alloys because of the serious effects of structural fluctuation on  $T_c$  and  $T_{comp}$ , even though all experimentalists have neglected these effects in analyzing their experimental data. At the present time, the main interest of experimentalists is directed towards obtaining an amorphous ferrimagnetic alloy which has a compensation point in the vicinity of room temperature and a wide region with a large and stable coercive field near the point because of its potential device applications, such as thermomagnetic recording.<sup>21</sup> As shown in Sec. IV, on the other hand, a number of interesting phenomena coming from the random distribution of exchange bonds may be also expected in amorphous ferrimagnetic alloys, although they may not be of technical, but only of academic importance.

Finally, it may be worth commenting on the following facts; in order to discuss the concentration change of the compensation point, we have used the Ising model. In actual materials, however, there exist transverse components in the spin Hamiltonian which may be important for the compensation effect, although they are not effective for discussing the transition temperature, since only the counting of the statistical weights of the system is effective near  $T_c$ . The concentration dependence of compensation, therefore, may need more detailed study than that of the present work. The problem is left as future work.

### APPENDIX A

The coefficients  $K_i$ ,  $L_j$ , and  $M_k$  in Eqs. (23)–(26), (28), and (A1)–(A7) are given as follows:

$$K_{1} = \cosh^{3}(\frac{1}{2}J_{AA}D)\sinh(\frac{1}{2}J_{AA}D)f(x)|_{x=0},$$

$$K_{2} = \cosh(\frac{1}{2}J_{AA}D)\sinh^{3}(\frac{1}{2}J_{AA}D)f(x)|_{x=0},$$

$$K_{3} = \cosh^{2}(\frac{1}{2}J_{AA}D)\sinh(\frac{1}{2}J_{AA}D)\cosh(J_{AB}D)f(x)|_{x=0},$$

$$K_{4} = \sinh^{3}(\frac{1}{2}J_{AA}D)\cosh(J_{AB}D)f(x)|_{x=0},$$

 $K_5 = \cosh^3(\frac{1}{2}J_{AA}D) \sinh(J_{AB}D)f(x) |_{x=0},$  $K_6 = \cosh(\frac{1}{2}J_{AA}D)\sinh^2(\frac{1}{2}J_{AA}D)\sinh(J_{AB}D)f(x) |_{x=0},$  $K_7 = \cosh^2(\frac{1}{2}J_{AA}D) \sinh(\frac{1}{2}J_{AA}D) f(x) |_{x=0},$  $K_8 = \cosh^2(\frac{1}{2}J_{AA}D) \cosh(J_{AB}D) \sinh(J_{AB}D) f(x) |_{x=0},$  $K_9 = \cosh^2(\frac{1}{2}J_{AA}D) \sinh^2(J_{AB}D)f(x) |_{x=0}$  $K_{10} = \cosh(\frac{1}{2}J_{AA}D) \sinh(\frac{1}{2}J_{AA}D) \cosh^2(J_{AB}D)f(x) |_{x=0},$  $K_{11} = \cosh(\frac{1}{2}J_{AA}D)\sinh(\frac{1}{2}J_{AA}D)\sinh^2(J_{AB}D)f(x) |_{x=0},$  $K_{12} = \cosh(\frac{1}{2}J_{AA}D)\sinh(\frac{1}{2}J_{AA}D)f(x)|_{x=0},$  $K_{13} = \cosh(\frac{1}{2}J_{AA}D)\sinh(\frac{1}{2}J_{AA}D)\cosh(J_{AB}D)f(x) |_{x=0},$  $K_{14} = \sinh^2(\frac{1}{2}J_{AA}D) \cosh(J_{AB}D) \sinh(J_{AB}D) f(x) |_{x=0},$  $K_{15} = \sinh^2(\frac{1}{2}J_{AA}D) \sinh(J_{AB}D)f(x) |_{x=0}$ ,  $K_{16} = \cosh(\frac{1}{2}J_{AA}D)\cosh^2(J_{AB}D)\sinh(J_{AB}D)f(x)\big|_{x=0},$  $K_{17} = \cosh(\frac{1}{2}J_{AA}D)\sinh^3(J_{AB}D)f(x) \mid_{x=0},$  $K_{18} = \cosh(\frac{1}{2}J_{AA}D)\cosh(J_{AB}D)\sinh(J_{AB}D)f(x)|_{x=0},$  $K_{19} = \cosh(\frac{1}{2}J_{AA}D) \sinh(J_{AB}D)f(x) |_{x=0},$  $K_{20} = \sinh(\frac{1}{2}J_{AA}D)\cosh^2(J_{AB}D)f(x) \mid_{x=0},$  $K_{21} = \sinh(\frac{1}{2}J_{AA}D)\cosh(J_{AB}D)\sinh^2(J_{AB}D)f(x) |_{x=0},$  $K_{22} = \sinh(\frac{1}{2}J_{AA}D)\cosh^2(J_{AB}D)f(x) \mid_{x=0},$  $K_{23} = \sinh(\frac{1}{2}J_{AA}D)\sinh^2(J_{AB}D)f(x)|_{x=0}$  $K_{24} = \sinh(\frac{1}{2}J_{AA}D)\cosh^2(J_{AB}D)f(x) \mid_{x=0},$  $K_{25} = \sinh(\frac{1}{2}J_{AA}D)f(x)|_{x=0}$ ,  $K_{26} = \cosh^3(J_{AB}D) \sinh(J_{AB}D) f(x) \mid_{x=0},$  $K_{27} = \cosh(J_{AB}D) \sinh^3(J_{AB}D) f(x) \mid_{x=0},$  $K_{28} = \cosh^2(J_{AB}D) \sinh(J_{AB}D) f(x) |_{x=0},$  $K_{29} = \sinh^3(J_{AB}D)f(x) |_{x=0}$ ,  $K_{30} = \cosh(J_{AB}D) \sinh(J_{AB}D) f(x) |_{x=0},$  $K_{31} = \sinh(J_{AB}D)f(x) \mid_{x=0},$  $K_{32} = \sinh^3(\frac{1}{2}J_{AA}D)f(x)|_{x=0}$ .

The coefficients  $L_j$  (j=1-32) can be obtained by doing the following replacements in  $K_i$  (i=1-32):  $J_{AA}/2 \rightarrow J_{AB}/2$ ,  $J_{AB} \rightarrow J_{BB}$ , and  $f(x) \rightarrow g(x)$ . We have

$$M_{1} = \cosh^{4}(\frac{1}{2}J_{AB}D)h(x) |_{x=0},$$
  

$$M_{2} = \cosh^{2}(\frac{1}{2}J_{AB}D) \sinh^{2}(\frac{1}{2}J_{AB}D)h(x) |_{x=0},$$
  

$$M_{3} = \sinh^{4}(\frac{1}{2}J_{AB}D)h(x) |_{x=0},$$
  

$$M_{4} = \cosh^{3}(\frac{1}{2}J_{AB}D) \cosh(J_{BB}D)h(x) |_{x=0},$$
  

$$M_{5} = \cosh(\frac{1}{2}J_{AB}D) \sinh^{2}(\frac{1}{2}J_{AB}D) \cosh(J_{BB}D)h(x) |_{x=0},$$
  

$$M_{6} = \cosh^{2}(\frac{1}{2}J_{AB}D) \sinh(\frac{1}{2}J_{AB}D) \sinh(J_{BB}D)h(x) |_{x=0},$$

 $M_7 = \sinh^3(\frac{1}{2}J_{AB}D)\sinh(J_{BB}D)h(x)|_{x=0}$  $M_8 = \cosh^3(\frac{1}{2}J_{AB}D)h(x) |_{x=0}$  $M_{9} = \cosh(\frac{1}{2}J_{AB}D)\sinh^{2}(\frac{1}{2}J_{AB}D)h(x)|_{x=0}$  $M_{10} = \cosh^2(\frac{1}{2}J_{AB}D) \cosh^2(J_{BB}D)h(x) |_{x=0},$  $M_{11} = \cosh^2(\frac{1}{2}J_{AB}D) \sinh^2(J_{BB}D)h(x) |_{x=0}$  $M_{12} = \cosh^2(\frac{1}{2}J_{AB}D)h(x) |_{x=0},$  $M_{13} = \cosh^2(\frac{1}{2}J_{AB}D) \cosh(J_{BB}D)h(x) |_{x=0}$  $M_{14} = \cosh(\frac{1}{2}J_{AB}D)\sinh(\frac{1}{2}J_{AB}D)$  $\times \cosh(J_{BB}D) \sinh(J_{BB}D)h(x) |_{x=0}$ ,  $M_{15} = \cosh(\frac{1}{2}J_{AB}D)\sinh(\frac{1}{2}J_{AB}D)\sinh(J_{BB}D)h(x)|_{x=0},$  $M_{16} = \sinh^2(\frac{1}{2}J_{AB}D)\cosh^2(J_{BB}D)h(x)|_{x=0}$ ,  $M_{17} = \sinh^2(\frac{1}{2}J_{AB}D) \sinh^2(J_{BB}D)h(x) |_{x=0}$  $M_{18} = \sinh^2(\frac{1}{2}J_{AB}D)h(x) |_{x=0}$ ,  $M_{19} = \sinh^2(\frac{1}{2}J_{AB}D) \cosh(J_{BB}D)h(x) |_{x=0}$ ,  $M_{20} = \cosh(\frac{1}{2}J_{AB}D)\cosh^{3}(J_{BB}D)h(x)|_{x=0}$  $M_{21} = \cosh(\frac{1}{2}J_{AB}D) \cosh(J_{BB}D) \sinh^2(J_{BB}D)h(x) |_{x=0},$  $M_{22} = \cosh(\frac{1}{2}J_{AB}D)\cosh^2(J_{BB}D)h(x) |_{x=0},$  $M_{23} = \cosh(\frac{1}{2}J_{AB}D)\sinh^2(J_{BB}D)h(x)|_{x=0}$  $M_{24} = \cosh(\frac{1}{2}J_{AB}D)\cosh(J_{BB}D)h(x)|_{x=0}$  $M_{25} = \cosh(\frac{1}{2}J_{AB}D)h(x)|_{x=0}$  $M_{26} = \sinh(\frac{1}{2}J_{AB}D)\cosh^2(J_{BB}D)\sinh^2(J_{BB}D)h(x)|_{x=0}$  $M_{27} = \sinh(\frac{1}{2}J_{AB}D)\sinh^{3}(J_{BB}D)h(x)|_{x=0}$  $M_{28} = \sinh(\frac{1}{2}J_{AB}D)\cosh^2(J_{BB}D)\sinh(J_{BB}D)h(x)|_{x=0}$  $M_{29} = \sinh(\frac{1}{2}J_{AB}D)\sinh(J_{BB}D)h(x)|_{x=0}$  $M_{30} = \cosh^4(J_{BB}D)h(x) |_{x=0}$  $M_{31} = \cosh^2(J_{BB}D) \sinh^2(J_{BB}D)h(x) |_{x=0}$ ,  $M_{32} = \sinh^4(J_{BB}D)h(x) |_{x=0}$ ,  $M_{33} = \cosh^3(J_{BB}D)h(x) |_{x=0}$ ,  $M_{34} = \cosh(J_{BB}D) \sinh^2(J_{BB}D)h(x) |_{x=0}$ ,  $M_{35} = \cosh^2(J_{BB}D)h(x) |_{x=0}$ ,  $M_{36} = \sinh^2(J_{BB}D)h(x) |_{x=0}$ ,  $M_{37} = \cosh(J_{BB}D)h(x) |_{x=0}$ ,  $M_{38} = h(0)$ .

The coefficients  $K_i$ ,  $L_j$ , and  $M_k$  can easily be calculated by applying a mathematical relation,  $e^{\alpha D} f(x) = f(x+\alpha)$ .

# APPENDIX B

The factors  $C_1$ ,  $C_2$ ,  $D_1$ ,  $D_2$ ,  $E_1$ ,  $E_2$ , and  $E_3$  in Eqs. (39), (31), and (32) are defined as follows:

$$C_{1} = 8p^{4}K_{1} + 4p^{3}(1-p) \left[ 6K_{3}q_{B} - \left[ \frac{p}{1-p} \right] K_{5} + 6K_{7}(1-q_{B}) \right] + 12p^{2}(1-p)^{2} \left[ 2K_{10}q_{B}^{2} + 2K_{12}(1-q_{B})^{2} + 4K_{13}q_{B}(1-q_{B}) - K_{8} \left[ \frac{p}{1-p} \right] q_{B} - K_{9} \left[ \frac{p}{1-p} \right] (1-q_{B}) \right] + 4p(1-p)^{3} \left[ 2K_{20}q_{B}^{3} + 6K_{22}(1-q_{B})q_{B}^{2} + 6K_{24}(1-q_{B})^{2}q_{B} + 2K_{25}(1-q_{B})^{3} - 3K_{16} \left[ \frac{1-p}{p} \right] q_{B}^{2} - 6K_{18} \left[ \frac{p}{1-p} \right] q_{B} - 3K_{19} \left[ \frac{p}{1-p} \right] (1-q_{B})^{2} \right] + 4(1-p)^{4} \left[ K_{26} \left[ \frac{p}{1-p} \right] q_{B}^{3} + 3K_{28} \left[ \frac{p}{1-p} \right] (1-q_{B})q_{B}^{2} + 3K_{30} \left[ \frac{p}{1-p} \right] (1-q_{B})^{2}q_{B} + K_{31} \left[ \frac{p}{1-p} \right] (1-q_{B})^{3} \right], \quad (B1)$$

$$C_{2} = 32K_{2}p^{4} + 16p^{3}(1-p) \left[ 2K_{4}q_{B} + 2K_{32}(1-q_{B}) - 3K_{6} \left[ \frac{1-p}{p} \right] \right] + 24p^{2}(1-p)^{2} \left[ K_{11} \left[ \frac{p}{1-p} \right]^{2} - 2K_{14} \left[ \frac{p}{1-p} \right] q_{B} - 2K_{15} \left[ \frac{p}{1-p} \right] (1-q_{B}) \right] + 4p(1-p)^{3} \left[ 6K_{21} \left[ \frac{p}{1-p} \right]^{2}q_{B} + 6K_{23} \left[ \frac{p}{1-p} \right]^{2} (1-q_{B}) - K_{17} \left[ \frac{p}{1-p} \right]^{3} \right] - 4(1-p)^{4} \left[ K_{27} \left[ \frac{p}{1-p} \right]^{3}q_{B} + K_{29} \left[ \frac{p}{1-p} \right]^{3} (1-q_{B}) \right], \quad (B2)$$

$$= -6L_{24} \left[ \frac{1-p}{p} \right] + p \left(1-p\right) \left[ \frac{1-p}{p} \right] q_B^2 - 2L_{12} \left[ \frac{1-p}{p} \right] (1-q_B)^2 - 4L_{13} \left[ \frac{1-p}{p} \right] q_B(1-q_B) \right]$$

$$+ 4p(1-p)^3 \left[ 3L_{16}q_B^2 + 6L_{18}(1-q_B)q_B + 3L_{19}(1-q_B)^2 - 2L_{20} \left[ \frac{1-p}{p} \right] q_B^3 - 6L_{22} \left[ \frac{1-p}{p} \right] (1-q_B)q_B^2 \right]$$

$$- 6L_{24} \left[ \frac{1-p}{p} \right] q_B(1-q_B)^2 - 2L_{25} \left[ \frac{1-p}{p} \right] (1-q_B)^3 \right]$$

$$+ 4(1-p)^4 [L_{26}q_B^3 + 3L_{28}(1-q_B)q_B^2 + 3L_{30}(1-q_B)^2 q_B + L_{31}(1-q_B)^2 ],$$
(B3)

$$D_{2} = -32L_{2}p^{4} \left[ \frac{1-p}{p} \right]^{3} + 16p^{3}(1-p) \left[ 3L_{6} \left[ \frac{1-p}{p} \right]^{2} - 2L_{4} \left[ \frac{1-p}{p} \right]^{3} q_{B} - 2L_{32} \left[ \frac{1-p}{p} \right]^{3}(1-q_{B}) \right] + 24p^{2}(1-p)^{2} \left[ 2L_{14} \left[ \frac{1-p}{p} \right]^{2} q_{B} + 2L_{15} \left[ \frac{1-p}{p} \right]^{2}(1-q_{B}) - L_{11} \left[ \frac{1-p}{p} \right] \right] + 4p(1-p)^{3} \left[ L_{17} - 6L_{21} \left[ \frac{1-p}{p} \right] q_{B} - 6L_{23} \left[ \frac{1-p}{p} \right] (1-q_{B}) \right] + 4(1-p)^{4} [L_{27}q_{B} + L_{29}(1-q_{B})] , \qquad (B4)$$

$$E_{1} = p^{4}M_{1} + 4p^{3}(1-p)[M_{4}q_{B} + M_{8}(1-q_{B})] + 6p^{2}(1-p)^{2} [M_{10}q_{B}^{2} + M_{12}(1-q_{B})^{2} + 2M_{13}q_{B}(1-q_{B})]$$

$$+4p(1-p)^{3}[M_{20}q_{B}^{3}+3M_{22}(1-q_{B})q_{B}^{2}+3M_{24}(1-q_{B})^{2}q_{B}+M_{25}(1-q_{B})^{3}]$$
  
+(1-p)^{4}[M\_{30}q\_{B}^{4}+4M\_{33}(1-q\_{B})q\_{B}^{3}+6M\_{35}(1-q\_{B})^{2}q\_{B}^{2}+4M\_{37}q\_{B}(1-q\_{B})^{3}+M\_{38}(1-q\_{B})^{4}], (B5)

$$E_{2} = 24p^{4}M_{2} + 12p^{3}(1-p) \left[ 5M_{5}q_{B} - 2M_{6} \left[ \frac{p}{1-p} \right] + 4M_{9}(1-q_{B}) \right] + 6p^{2}(1-p)^{2} \left[ M_{11} \left[ \frac{p}{1-p} \right]^{2} - 8M_{14} \left[ \frac{p}{1-p} \right] q_{B} - 8M_{15} \left[ \frac{p}{1-p} \right] (1-q_{B}) + 4M_{16}q_{B}^{2} + 4M_{18}(1-q_{B})^{2} + 8M_{19}(1-q_{B})q_{B} \right] + 4p(1-p)^{3} \left[ 3M_{21} \left[ \frac{p}{1-p} \right]^{2}q_{B} + 3M_{23} \left[ \frac{p}{1-p} \right]^{2} (1-q_{B}) - 6M_{26} \left[ \frac{p}{1-p} \right] q_{B}^{2} - 12M_{28} \left[ \frac{p}{1-p} \right] q_{B}(1-q_{B}) - 6M_{29} \left[ \frac{p}{1-p} \right] (1-q_{B})^{2} \right] + 6(1-p)^{4} \left[ M_{31} \left[ \frac{p}{1-p} \right]^{2}q_{B}^{2} + 2M_{34} \left[ \frac{p}{1-p} \right]^{2} (1-q_{B})q_{B} + M_{36} \left[ \frac{p}{1-p} \right]^{2} (1-q_{B})^{2} \right] , \qquad (B6)$$

$$E_{3} = 16p^{4}M_{3} - 32p^{3}(1-p)M_{7} \left[ \frac{p}{1-p} \right] + 24p^{2}(1-p)^{2}M_{17} \left[ \frac{p}{1-p} \right]^{2} - 8p(1-p)^{3}M_{27} \left[ \frac{p}{1-p} \right]^{3} + (1-p)^{4}M_{32} \left[ \frac{p}{1-p} \right]^{4} . \qquad (B7)$$

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