Role of single-ion anisotropy in amorphous ferrimagnetic alloys

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The effects of single-ion anisotropy on transition and compensation temperatures in the amorphization of binary ferrimagnetic square lattice are investigated by the use of effective-field theory with correlations. A number of interesting phenomena are obtained for the behavior of the two quantities, which arise from both the single-ion anisotropy and the random distribution of exchange bonds in sign. Further research on new amorphous ferrimagnetic alloys may reveal such interesting behavior.

I. INTRODUCTION

Amorphous rare-earth-transition-metal (RE-TM) alloys have proved extremely suitable for thermomagnetic recording and magneto-optical readout applications. The magnetic properties of the alloys are sensitive to slight compositional changes. Experimental data confirm their untypical behavior. Because of the potential device applications, the compositional dependencies of the Curie temperature T_c and the compensation temperature $T_{\rm comp}$ have been extensively investigated; being ferrimagnetic, some of them possess a compensation-point temperature which varies in the vicinity of room temperature by proper choice of composition. The thermomagnetic writing and erasing can be carried out at the compensation point near the room temperature, because of the high coercivity around the compensation point.¹

Theoretically, in order to analyze the magnetic properties of amorphous ferrimagnetic alloys, namely, the temperature and the concentration dependencies of T_c and $T_{\rm comp}$, mean-field theory has been applied in the recent past; the theory provides a simple solution to the problems.² However, as discussed in previous works,^{3,4} the usefulness of this has been marred in the past by the existence of too many adjustable parameters and by the neglect of the fluctuation of exchange interactions. In amorphous magnets, the fluctuation of exchange interactions (or the structural fluctuation) is in fact considered an important ingredient for the appearance of characteristic behavior, such as the depression of the reduced magnetization curve, reentrant phenomena, and the occurrence of a spin-glass phase.⁵ On the other hand, when we use the mean-field theory, it is known that the effect of the structural fluctuation on T_c cancels out and does not appear, and its effect on $T_{\rm comp}$ is very small.⁶ Thus, it is necessary to investigate the effects of the structural fluctuation on the magnetic properties within the framework over the standard mean-field approximation (MFA).

Very recently, the present author^{3,4} has discussed the concentration variation of the transition temperature T_c and of $T_{\rm comp}$ for two (disordered and amorphous) ferrimagnetic, binary, Ising alloys within the effective-field

theory with correlations (EFT).^{7,8} The EFT, which substantially improves on the standard MFA, has been successfully applied to a variety of other physical problems.⁸ In the previous works,^{3,4} the lattice model of amorphous magnets was used, in which the structural disorder is replaced by random distributions of exchange interactions.⁹ In contrast with disordered ferrimagnetic alloys, in which RE and TM ions are randomly distributed in a lattice, we have found that a number of interesting phenomena coming from the random distribution of exchange bonds, such as the reentrant ferrimagnetic phase, may be expected in the new amorphous ferrimagnetic alloys, although 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.

In this paper, we assume that the rare-earth element is subject to single-ion anisotropy of the simplest kind, and we study the effects of single-ion anisotropy on T_c and $T_{\rm comp}$ in the lattice model of amorphous magnets within the same framework, as in the previous works.^{3,4} We have found that the effects of the negative anisotropy constant on T_c are dramatic, although the changes of T_c and $T_{\rm comp}$ with a positive anisotropy constant show rather unique behavior, depending on the value of the structural fluctuation.

The outline of this paper is as follows. In Sec. II, we briefly review the formulation of amorphous ferrimagnetic binary Ising alloys in the EFT. The formulation is applied to the square lattice with random bonds. In Sec. III, the effects of single-ion anisotropy on T_c and T_{comp} in the square lattice model of amorphous magnets are numerically studied. For the special case with only RE ions, the system reduces to the so-called Blume-Capel model, and the system is especially investigated in the Appendix, in order to obtain the tricritical point within the EFT.

II. FORMULATION

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

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magnetic ions A and B with different spins $(S_A = \frac{1}{2}$ and $S_B = 1$). We assume that the B ions are subject to singleion anisotropy of the simplest kind. The Hamiltonian of the system is then

$$\mathscr{H} = -\sum_{i < j} [J_{AA} \delta_{iA} \delta_{jA} + J_{BB} \delta_{iB} \delta_{jB} + J_{AB} (\delta_{iA} \delta_{jB} + \delta_{iB} \delta_{jA})] S_i^z S_j^z \xi_i \xi_j - D \sum_i (S_i^z)^2 \delta_{iB} \xi_i , \qquad (1)$$

where the J_{ij} 's are the exchange interaction between type-*i* and type-*j* ions, and the sum of the first term is over all nearest-neighbor pairs. ξ_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. *D* is the anisotropy constant. $\delta_{i\alpha}$ ($\alpha = A$ or *B*) expresses that a site *i* is occupied by a type- α ion. Performing the random average denoted by $\langle \cdots \rangle_r$, the average value of ξ_i has a restriction

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

where $\langle \xi_{i=A} \rangle_r = p$ is the concentration of A atoms. For the lattice model of amorphous magnets, the nearestneighbor exchange interactions are given by independent random variables as

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

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

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

 $m = \langle \langle S_i^z \rangle \rangle_r = Pm_A + (1-P)m_B$,

where parameters a and b are constants.^{3,4}

The main problem is now the evaluation of the mean values, $\langle S_A^z \rangle$ and $\langle S_B^z \rangle$, where the angular brackets denote the usual thermal averages. As previously discussed in a series of works,⁸ the starting point for the evaluation of $\langle S_{i=A}^z \rangle$ with $S_A = \pm \frac{1}{2}$ is the exact Callen identity¹⁰

$$\langle S_{i=A}^{z} \rangle = \frac{1}{2} \left\langle \tanh \left[\frac{\xi_{i=A} \beta \theta_{i=A}}{2} \right] \right\rangle,$$
 (4)

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 relation¹¹

$$\langle S_{i=B}^{z} \rangle = \left\langle \frac{2\sinh(\xi_{i=B}\beta\theta_{i=B})}{2\cosh(\xi_{i=B}\beta\theta_{i=B}) + \exp(-\beta D\xi_{i=B})} \right\rangle,$$
(5)

with

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

As discussed in Refs. 3 and 4, within the framework of the EFT, the averaged total and sublattice magnetizations are given by [from Eqs. (4) and (5)]

(6)

$$m_{A} = \left\{ P\left[\left\langle \cosh\left[\frac{J_{AA}}{2}\overline{D}\right] \right\rangle_{r} + 2m_{A} \left\langle \sinh\left[\frac{J_{AA}}{2}\overline{D}\right] \right\rangle_{r} \right] + (1-P)[q_{B} \left\langle \cosh(J_{AB}\overline{D}) \right\rangle_{r} + m_{B} \left\langle \sinh(J_{AB}\overline{D}) \right\rangle_{r} + 1 - q_{B}] \right\}_{r}^{z} f(x) \Big|_{x=0}, \qquad (7)$$

$$m_{B} = \left\{ P\left[\left\langle \cosh\left[\frac{J_{AB}}{2}\overline{D}\right] \right\rangle_{r} + 2m_{A} \left\langle \sinh\left[\frac{J_{AB}}{2}\overline{D}\right] \right\rangle_{r} \right] + (1-P)[q_{B} \left\langle \cosh(J_{BB}\overline{D}) \right\rangle_{r} + m_{B} \left\langle \sinh(J_{BB}\overline{D}) \right\rangle_{r} + 1 - q_{B}] \right\}_{r}^{z} f(x) \Big|_{x=0}, \qquad (8)$$

with

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

$$g(x) = \frac{2\sinh(\beta x)}{2\cosh(\beta x) + \exp(-\beta D)},$$
 (10)

where $\overline{D} = \partial/\partial x$ is a differential operator. Z is the number of nearest neighbors. The parameter q_B is here defined by¹¹

$$q = \frac{\langle \xi_{i=B} \langle (S_{i=B}^{z})^{2} \rangle \rangle_{r}}{\langle \xi_{i=B} \rangle_{r}} , \qquad (11)$$

with

$$\langle (S_{i=B}^{z})^{2} \rangle = \left\langle \frac{2\cosh(\beta\xi_{i=B}\theta_{i=B})}{2\cosh(\beta\xi_{i=B}\theta_{i=B}) + \exp(-D\beta\xi_{i=B})} \right\rangle.$$
(12)

In order to obtain the sublattice magnetizations, therefore, it is necessary to calculate the parameter q_B . The parameter q_B is given by, in the same way as the evaluation of m_B ,

$$q_{B} = \left\{ P\left[\left\langle \cosh\left(\frac{J_{AB}}{2}\overline{D}\right) \right\rangle_{r} + 2m_{A} \left\langle \sinh\left(\frac{J_{AB}}{2}\overline{D}\right) \right\rangle_{r} \right] + (1-P)\left[q_{B} \left\langle \cosh(J_{BB}\overline{D}) \right\rangle_{r} + m_{B} \left\langle \sinh(J_{BB}\overline{D}) \right\rangle_{r} + 1 - q_{B} \right] \right\}^{z} h(x) \Big|_{x=0},$$
(13)

with

$$h(x) = \frac{2\cosh(\beta x)}{2\cosh(\beta x) + \exp(-\beta D)} .$$
(14)

As discussed in a great number of works,^{3,4,7,8,11} these equations can be easily evaluated by applying a mathematical relation $e^{\alpha \overline{D}} f(x) = f(x + \alpha)$.

At this point, it is worth commenting that in comparison with the previous works^{3,4} with zero anistropy constant (D=0), the only differences are the definitions of functions g(x) and h(x). Therefore, the equations in Ref. 3 for evaluating T_c and T_{comp} can be easily extended to the present problem.

For the ferrimagnetic square lattice (Z = 4) with random bonds, namely, the amorphization of ferrimagnetic square lattice, the critical surface characterizing the ferrimagnetic phase stability limit is determined by, as discussed in Ref. 3,

$$(A_1 - 1)(B_2 - 1) = A_2 B_1 . (15)$$

The coefficients A_1 , A_2 , B_1 , and B_2 are given only by replacing the functions h(x) and g(x) in Ref. 3 with the new Eqs. (10) and (14). On the other hand, in the ferrimagnetic materials, 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 $m_B \neq 0$. For Z = 4, the effect of D on $T_{\rm comp}$ can be evaluated from Eqs. (30)–(32) in Ref. 3 with new functions (10) and (14), if a compensation temperature exists.

In the next section, we shall investigate the role of D in the behavior of T_c and T_{comp} for the amorphization of ferrimagnetic square lattice by solving these equations numerically.

III. ROLE OF ANISOTROPY CONSTANT IN THE AMORPHIZATION OF FERRIMAGNETIC SQUARE LATTICE

Let us now investigate the behavior of T_c and T_{comp} in the presence of a finite D for the amorphization of ferrimagnetic square lattice. As discussed in previous works,^{3,4} it is extremely tedious to treat the fluctuations of exchange interactions in general, so that we also take

$$\frac{\Delta J}{J} = \frac{2\Delta J'}{J} = \frac{2\Delta J''}{J} = \delta .$$
 (16)

The parameter δ is often called the structural fluctuation in amorphous magnets.

Before showing the numerical results, it is first worth commenting on some relations between the results obtained within the present formulation (Z = 4) and those

obtained 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 gadolinium-based alloys the single-ion anisotropy is believed to be negligible. This is due to the fact that Gd is an s-state ion with little or no interaction with the crystal electric field. Terbium, for instance, is known to couple strongly to the electric field and create large amounts of single-ion anisotropy. It may be expected that the model for amorphous Tb-TM alloys is one that includes both the single-ion anisotropy of Tb and the structural fluctuation.

Figure 1 shows T_c and T_{comp} versus D for an alloy with p=0.5. Exchange interactions are then chosen as



FIG. 1. Curie and compensation temperatures versus singleion anisotropy constant D for a binary $A_p B_{1-p}$ system with p=0.5, in which exchange interactions defined by (3) are fixed as a = -0.12 and b=0.02, and the structural fluctuation δ defined by (16) is varied; curve a is for $\delta=0$, curve b is for $\delta=0.1$, and curve c is for $\delta=0.4$. Solid lines denote T_c and dashed lines are for $T_{\rm comp}$.

 $J_{AA} = J(1\pm\delta)$, $J_{AB} = -J(0.2\pm0.5\delta)$, and $J_{BB} = J(0.02\pm0.5\delta)$, values which may be consistent with the experimental data for amorphous Tb-TM alloys.¹² The \pm sign of exchange interactions are taken with equal probability, according to the definitions (3). As discussed in Ref. 3, the compensation temperature $T_{\rm comp}$ is allowed for a system with p in the region of $p < \frac{2}{3}$, so that for a small value of δ , the change of $T_{\rm comp}$ versus D (D > 0) is found in the figure.

As is seen from the figure, curves a and b with $\delta = 0.0$ and $\delta = 0.1$ express typical behavior for the change of D; for D > 0, T_c and T_{comp} increase at first, and then approach constant values upon increasing the value of D. For D < 0, T_c at first decreases rapidly and approaches to a finite value, on decreasing the value of D. The $T_{\rm comp}$ in curve a cannot be defined at the point near D/J = -0.1. Therefore, in curve b, T_{comp} versus D is only plotted for $D \ge 0$. The results labeled b express that the exchange interaction J_{BB} takes positive and negative values randomly, so that T_c and T_{comp} for $\delta = 0.1$ have small values in comparison with those of curve *a*. Physically, the increases of T_c and T_{comp} for D > 0 come from the fact that for D > 0, spins at B sites more preferably take the direction parallel to the z axis. On the other hand, for D < 0, spins at B sites are apt to direct perpendicularly to the z axis and hence the T_c decreases rapidly at first and then approaches to the value of T_c mainly determined from the contribution of A atoms.

A particularly interesting result is obtained for curve cin Fig. 1. The structural fluctuation δ is chosen as $\delta = 0.4$. The exchange interactions J_{AB} and J_{BB} can then take positive and negative values randomly. The behavior of T_c versus D is in contrast with a and b in the figure. In this case, T_{comp} cannot be obtained for any value of D, because of the large value of δ . The result may be understood as follows. Upon decreasing the value of D from D=0, as mentioned above, spins at B sites behave like "nonmagnetic atoms" for the estimation of T_c . The most important aspect of the "dilution" is to break out the frustration in A spins, which comes mainly from J_{AB} . Therefore, the value of T_c for D=-2J becomes larger than that for D=2J, where the frustration due to J_{AB} and J_{BB} is fully taken into account.

In Figure 2, the exchange interactions are chosen as $J_{AA} = J(1\pm\delta)$, $J_{AB} = -J(0.25\pm0.5\delta)$, and $J_{BB} = J(0.1\pm0.5\delta)$. The values of T_c and T_{comp} for D=0 in the curves labeled *a* are equivalent to those of Fig. 6 in Ref. 3. As shown in Ref. 3, when we take the values of the exchange interactions and the value of δ is not so large, the compensation temperature is found only for a system with *p* in the region $0.5 . When <math>\delta = 0.4$ is chosen as for curves *a'*, *b'*, and *c'* in Fig. 2, the exchange interaction *J*_{BB} can take positive and negative values with equal probability.

As is seen from the curves labeled a in Fig. 2, the behavior of T_c and T_{comp} versus D are very similar to those labeled a in Fig. 1. For the system with p=0.6 and $\delta=0.4$, however, the increase of T_{comp} with positive D is rather insignificant, although the change of T_c with D is also similar to that of curve b in Fig. 1. Thus, it seems that the effect of D on T_{comp} is reduced for a large value



FIG. 2. The behavior of T_c (solid line) and T_{comp} (dashed line) versus D for two systems with $\delta = 0$ and 0.4, in which curves a, b, and c are for $\delta = 0$, and curves a', b', and c' are for $\delta = 0.4$. Exchange interactions defined by (3) are fixed as a = -0.25 and b = 0.1. Concentration p is varied; curve a is for p = 0.6, curve b is for p = 0.43, and curve c is for p = 0.4. Black point in curves b', c and c' denotes a measure for the tricritical point.

of δ .

In Fig. 2, the behavior of T_c versus D is furthermore depicted for two selected values of p, namely p=0.43 and p=0.4. Curve b with p=0.43 and $\delta=0$ changes continuously with D, but for curve c with p=0.4 and $\delta=0$, any solution of T_c cannot be obtained below a critical value of D (or the black point in the curve) by solving Eq. (15) numerically. The abrupt change of T_c behavior versus negative D is in fact observed in the concentration range 0.42 . The result reminds us the following fact:When we take $J_{AB} = 0$ and $J_{BB} = 0$, namely, the bonddilution problem of A atoms, the critical percolation concentration p_c is given by $p_c = 0.4284$ within the present formulation (EFT).⁸ Therefore, as the concentration p approaches p_c , the contribution of A atoms to T_c reduces, and for the value of p below p_c , the effect of B atoms on T_c becomes dominant. On the other hand, the special case with p = 0 and $\delta = 0$, namely the "pure" system with only B atoms (pure means $\delta = 0$), just corresponds to the so-called two-dimensional Blume-Capel model. It is well-known that for a negative value of D, the Blume-Capel model exhibits a tricritical point at which the system changes from the second-order phase transition to the first-order one. Thus, the black point in curve c may indicate the tricritical point, although the point is obtained by solving Eq. (15) numerically. Therefore, it is

FIG. 3. Curie temperatures versus D for a system with p=0.9, in which exchange interactions defined in (3) are fixed as a = -1.0 and b=1.0. Structural fluctuation δ is changed; curve a is for $\delta=0$, curve b is for $\delta=1.0$, and two curves c_1 and c_2 are for $\delta=1.2$. Between the two curves c_1 and c_2 , the reentrant ferrimagnetic phase is possible.

worthwhile to investigate whether the black point obtained from (15) corresponds to the tricritical point. In the Appendix, the special case with p=0 and $\delta=0$, namely, the two-dimensional Blume-Chapel model, is investigated in detail within the present formulation. The result expresses that the critical (or black) point below which any value of T_c cannot be obtained from Eq. (15) is a measure for the tricritical point. The critical point is also seen in the curves b' and c'. In contrast with curve b with $\delta=0$, the curve b' with $\delta=0.4$ exhibits such a critical point. A peculiar result is obtained in curve c': Upon increasing the value of D from D=0, the value of T_c decreases to a finite constant value.

In Fig. 3, exchange interactions are chosen as $J_{AA} = J(1\pm\delta)$, $J_{AB} = -J(1\pm0.5\delta)$, and $J_{BB} = J(1\pm0.5\delta)$, and p is fixed at p=0.9. In order to study the effect of D on T_c , three values of δ are selected there. For curves a and b with $\delta=0$ and 1.0, the behavior of T_c versus D changes smoothly in comparison with those labeled a and b in Fig. 1, and any critical value of D (or tricritical point) is not obtained, since the concentration of B atoms is very small, and hence the effect of D on T_c is weakened.

In the previous works,^{3,4} on the other hand, we have discussed that the reentrant ferrimagnetic phase is possible for $\delta = 1.2$; the phase was obtained between two transition temperatures at D=0 of two curves c_1 and c_2 in Fig. 3, since for $\delta = 1.2$, exchange interaction J_{AA} can take positive and negative value randomly. Contrary to the expectation mentioned above, the effect of D on curves c_1 and c_2 , where the reentrant ferrimagnetic phase is ob-

FIG. 4. Curie temperatures versus D for a system with p=0.5, in which exchange interactions defined in (3) are fixed as a = -1.0 and b = 1.0. δ is changed as follows: curve a is for $\delta = 0$ and curve b is for $\delta = 1.2$. Black point in curve b is a measure for the tricritical point.

tained between the two curves, is remarkable, especially for some negative values of D; the reentrant ferrimagnetic region for the values near D = -0.5J becomes wider than that of D = 0.

In Fig. 4, the exchange interactions are chosen as the same values as those of Fig. 3, and p is then fixed at p=0.5. In this case, the compensation temperature cannot be defined even for $\delta = 0$, since the exchange interaction J_{AB} is too strong. For $\delta = 1.2$, as mentioned above, only exchange interaction J_{AA} becomes randomly distributed in sign, and the tricritical behavior (or black point) is observed in the studied region of D. Even for D=0, the transition temperature for $\delta = 1.2$ is pronouncedly reduced from that for $\delta = 0$, and the ratio $T_c(\delta=1.2)/T_c(\delta=0)$ amounts to 0.75.

IV. CONCLUSION

We have studied the effect of single-ion anisotropy on transition and compensation temperatures in the amorphization of binary ferrimagnetic square lattice, using the same framework (EFT) as that of the previous works.^{3,4} As shown in Figs. 1–3, the obtained results exhibit very interesting phenomena for both T_c and T_{comp} . Especially for negative values of D, the characteristic of T_c is more enhanced.

On the other hand, real existing heavy rareearth-transition-metal alloys may not have the single-ion anisotropy of the simplest kind such as that studied in the present work. For instance, amorphous TbFe alloys are sperimagnetic, in which the direction of single-ion anisotropy at each site may be distributed randomly.¹³ Nevertheless, apart from at very low temperature, their magnetic behavior is qualitatively described by a ferri-





magnetic model with randomly distributed exchange interactions among neighboring iron atoms, as suggested in the previous works and the present work. That is to say, when we compare the transition temperature of crystalline ferrimagnetic TbFe₂ alloy with that of amorphous TbFe₂, the reduction in ordering temperature amounts to $T_c(\text{amorphous})/T_c(\text{crystal})=0.57$,¹⁴ a value too large to be explained directly in terms of a reduction in the effective Tb-Fe exchange coupling. As discussed in Fig. 4, on the other hand, the ratio for $\delta = 1.2$ is 0.75. However, when we take $\delta = 1.5$ in the case of Fig. 4, the ratio is given by 0.59, even for D = 0. Thus, the large reduction in ordering temperature can be easily attained from our model, although the true situation for the reduction of T_c in the amorphous Tb-Fe alloy may be thought to be random directions of the anisotropy axis.

Finally, 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, such as thermomagnetic recording. As shown in Sec. III, a number of interesting phenomena coming from the random distribution of exchange bonds and the single-ion anisotropy of the simplest kind may be also expected in new amorphous ferrimagnetic alloys, although they may not be of technical importance, but only important academically.

APPENDIX

In this part, we study the spin-1 Ising model with crystal-field interactions on a square lattice (Z=4), namely, the two-dimensional Blume-Capel model, by the use of the EFT, in order to clarify whether the critical (or black) point obtained from Eq. (15) is a measure for the tricritical point. The Hamiltonian is given by

$$\mathscr{H} = -J \sum_{i,j} S_i^z S_j^z - D \sum_i (S_i^z)^2 , \qquad (A1)$$

where the first summation runs over all pairs of nearest neighbors.

Using Eq. (8) with p=0, the magnetization m is given by (within the EFT)

$$m = 4\overline{A}_{1}m + 4\overline{B}_{1}m^{3} + 12\overline{C}_{1}mq + 12\overline{D}_{1}mq^{2} + 4\overline{E}_{1}mq^{3} + 4\overline{F}_{1}m^{3}q , \qquad (A2)$$

with

$$\overline{A}_{1} = K_{6} ,$$

$$\overline{B}_{1} = K_{4} ,$$

$$\overline{C}_{1} = K_{5} - K_{6} ,$$

$$\overline{D}_{1} = K_{6} - 2K_{5} + K_{3} ,$$

$$\overline{E}_{1} = K_{1} - 3K_{3} + 2K_{5} - K_{6} ,$$

$$\overline{F}_{1} = K_{2} - K_{6} ,$$
(A3)

where the coefficients K_i (i = 1 to 6) are defined by

$$K_{1} = \cosh^{3}(\overline{D}J)\sinh(\overline{D}J)g(x) \mid_{x=0},$$

$$K_{2} = \cosh(\overline{D}J)\sinh^{3}(\overline{D}J)g(x) \mid_{x=0},$$

$$K_{3} = \cosh^{2}(\overline{D}J)\sinh(\overline{D}J)g(x) \mid_{x=0},$$

$$K_{4} = \sinh^{2}(\overline{D}J)g(x) \mid_{x=0},$$

$$K_{5} = \cosh(\overline{D}J)\sinh(\overline{D}J)g(x) \mid_{x=0},$$

$$K_{6} = \sinh(\overline{D}J)g(x) \mid_{x=0}.$$
(A4)

On the other hand, the parameter q defined by $q = \langle (S_i^z)^2 \rangle$ is given by [from Eq. (13) with p = 0]

$$q = \overline{A}_2 + 4\overline{B}_2 q + 6\overline{C}_2 q^2 + 4\overline{D}_2 q^3 + \overline{E}_2 q^4 + 6\overline{F}_2 m^2 + 12\overline{G}_2 m^2 q + 6\overline{H}_2 m^2 q^2 + \overline{I}_2 m^4 , \qquad (A5)$$

with

$$A_{2} = L_{9} ,$$

$$\overline{B}_{2} = L_{8} - L_{9} ,$$

$$\overline{C}_{2} = L_{6} - 2L_{8} + L_{9} ,$$

$$\overline{D}_{2} = L_{4} - 3L_{6} + 3L_{8} - L_{9} ,$$

$$\overline{E}_{2} = L_{1} - 4L_{4} + 6L_{6} - 4L_{8} + L_{9} ,$$

$$\overline{F}_{2} = L_{7} ,$$

$$\overline{G}_{2} = L_{5} - L_{7} ,$$

$$\overline{H}_{2} = L_{2} - 2L_{5} + L_{7} ,$$

$$\overline{I}_{2} = L_{3} ,$$
(A6)

where the coefficients L_i (i=1 to 9) are defined by

$$L_{1} = \cosh^{4}(DJ)h(x) |_{x=0},$$

$$L_{2} = \cosh^{2}(\overline{D}J)\sinh^{2}(\overline{D}J)h(x) |_{x=0},$$

$$L_{3} = \sinh^{4}(\overline{D}J)h(x) |_{x=0},$$

$$L_{4} = \cosh^{3}(\overline{D}J)h(x) |_{x=0},$$

$$L_{5} = \cosh(\overline{D}J)\sinh^{2}(\overline{D}J)h(x) |_{x=0},$$

$$L_{6} = \cosh^{2}(\overline{D}J)h(x) |_{x=0},$$

$$L_{7} = \sinh^{2}(\overline{D}J)h(x) |_{x=0},$$

$$L_{8} = \cosh(\overline{D}J)h(x) |_{x=0},$$

$$L_{9} = h(0).$$
(A7)

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

At this point, when we replace q in (A2) by its expression taken from (A5), we obtain an equation for

$$m = \overline{a}m + \overline{b}m^2 + \overline{c}m^5 + \cdots$$
 (A8)

The second-order phase transition line in the *T*-*D* plane is determined from $\bar{a} = 1$, i.e.,

$$1 = 4\overline{A}_1 + 12\overline{C}_1 q_0 + 12\overline{D}_1 (q_0)^2 + 4\overline{E}_1 (q_0)^3 , \qquad (A9)$$

where q_0 is the solution of

$$q_0 = \overline{A}_2 + 4\overline{B}_2 q_0 + 6\overline{C}_2 (q_0)^2 + 4\overline{D}_2 (q_0)^3 + \overline{E}_2 (q_0)^4 .$$
(A10)



FIG. 5. Curie temperature versus D for the Blume-Capel model in square lattice (z=4). The point \triangle in the curve denotes the tricritical point obtained from the framework discussed in Appendix. Black point in the curve corresponds to a critical point obtained from Eq. (15), below which any solution cannot be obtained from Eq. (15).

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

$$m^2 = \frac{1 - \overline{a}}{\overline{b}} . \tag{A11}$$

The right-hand side must be positive. If this is not the case, the transition is of the first order and in the T-D plane, the point at which $\bar{a} = 1$ and $\bar{b} = 0$ is the tricritical point.¹⁵ In order to obtain the expression for \bar{b} , we must solve (A5) for small *m*. The solution is of the form

$$q = q_0 + q_1 m^2 , (A12)$$

where q_1 is given by

$$q_{1} = \frac{6\overline{F}_{2} + 12\overline{G}_{2}q_{0} + 6\overline{H}_{2}(q_{0})^{2}}{1 - 4\overline{B}_{2} - 12\overline{C}_{2}q_{0} - 12\overline{D}_{2}(q_{0})^{2} - 4\overline{E}_{2}(q_{0})^{3}}$$
(A13)

The parameter \overline{b} is then given by

$$b = 4\overline{B}_1 + 12\overline{C}_1q_1 + 24\overline{D}_1q_0q_1 + 12\overline{E}_1(q_0)^2q_1 + 4\overline{F}_1q_0 .$$
(A14)

By solving (A9), (A10), and (A14) numerically, the behavior of T_c versus D is depicted in Fig. 5. The tricritical point (Δ in the figure) is given by $D_t = -1.8804J$ and $k_B T_t = 1.0336J$. When we use the reduced units defined by $\delta_t = |D_t|/zJ$ and $\theta_t = k_B T_t/zJ$, δ_t and θ_t are given by $\delta_t = 0.470$ and $\theta_t = 0.258$, which values are in quite good agreement with those from high-temperature series expansion method and represent a remarkable improvement on the MFA predictions (see Table I in Ref. 16). On the other hand, the results obtained from Eq. (15) for the system with p=0 and $J_{BB}=J$ also gives the same curves until the black point in Fig. 5 below which any solution cannot be obtained from Eq. (15). Thus, the critical (or black) point in Figs. 2 and 4 obtained from Eq. (15) is a good measure for the tricritical point.

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