New disordered phase of a ferromagnetic binary Ising system

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The phase diagrams and the temperature (or field) dependences of magnetizations in a binary Ising ferromagnetic alloy consisting of spin-$\frac{1}{2}$ and spin-1 components with a crystal-field interaction are investigated by the use of an effective-field theory with correlations. We find a new disordered phase in which the transition temperature (or magnetization) reduces to zero when the crystal-field interaction takes a large negative value. The magnetization process in the new disordered phase is examined. We find that it exhibits phenomena similar to, but with important differences from, those of metamagnets, even though all the exchange interactions have a positive sign.

I. INTRODUCTION

In the last few decades 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 set of possible values. In the site model the lattice sites are randomly occupied by two different species of magnetic ions, and the interaction between two ions is determined entirely by the species of these ions. In particular, the phase diagrams that may occur in these systems have been examined.[1-3] However, most works have not discussed the effects of crystal-field interactions on the transition temperature (or phase diagram).

On the other hand, a regular spin-1 Ising model with a crystal-field interaction has been extensively investigated. It is well known in the system that there exists a tricritical point at which the phase transition changes from second order to first order, when the crystal-field interaction takes a large negative value.[4,5] The change of phase-transition order is due to the competition between the positive exchange-interaction term and the negative crystal-field interaction; when the value of the crystal field becomes larger than the internal field, each spin in the system is apt to orient perpendicular to the internal field.

In previous work[6] the role of crystal-field interaction in an amorphous ferrimagnetic alloy consisting of spin-$\frac{1}{2}$ and spin-1 components has been examined by the use of the effective-field theory (EFT) with correlations.[7] Then, the effect of crystal-field interactions on transition and compensation temperatures has been studied for the system with a fixed composition. A number of interesting phenomena are observed there for the behavior of the two quantities, which arise from both the crystal-field interaction and the random distribution of exchange bonds.

In this work, within the EFT,[7] we investigate the magnetic properties of a binary substitutional ferromagnetic Ising system consisting of spin-$\frac{1}{2}$ and spin-1 components with a crystal-field interaction. In particular, the change of transition temperature with composition is studied. We find that a new disordered phase may appear in the phase diagrams, when the crystal-field interaction takes a large negative value. The new phase consists of two types of spins: spin-1 atoms go into the $s^z=0$ state and spin-$\frac{1}{2}$ atoms take on the values $s^z=\pm\frac{1}{2}$ randomly. The magnetization process in the disordered phase is examined. It may exhibit phenomena similar to, but with important differences from, those of metamagnets,[8] even though all the exchange interactions have a positive sign.

The outline of this work is as follows. In Sec. II we briefly present the formalism of the EFT,[7] as applied to the binary ferromagnetic alloy in a honeycomb lattice. In Sec. III, we study the phase diagrams of the system. In Sec. IV the temperature (or field) dependences of total and sublattice magnetizations are investigated.

II. FORMULATION

We consider a binary ferromagnetic alloy of the type A_pB_{1-p} in a honeycomb lattice, which is randomly occupied by two different species of magnetic ions A and B ($s_A=\frac{1}{2}$ and $s_B=1$). B ions have the crystal-field interaction. The Hamiltonian of the system is given by

$$\mathcal{H}=-\sum_{i,j}[J_{AA}\delta_{iA}\delta_{jA}+J_{BB}\delta_{iB}\delta_{jB}+J_{AB}(\delta_{iA}\delta_{jB}+\delta_{iB}\delta_{jA})]s_i^z s_j^z\xi_i\xi_j$$
$$+\Delta\sum_i(s_i^z)^2\delta_{iB}\xi_i-H\sum_i(\delta_{iA}+\delta_{jB})s_i^z\xi_i\ , \tag{1}$$

where the J_{ij} are the interaction energies between type-i and type-j sites, the s^z are spin variables, which can take values $s^z=\pm\frac{1}{2}$ for the A ions and $s^z=\pm1,0$ for the B ions, and the first summation 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. Δ is the crystal-field interaction parameter. $\delta_{i\alpha}$ ($\alpha = A$ or B) expresses that a site i is occupied by a type-α ion. H is the applied magnetic field. In the random average denoted by $\langle \cdots \rangle_r$, the averaged value of ξ_i has a restriction

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

where $\langle \xi_{i=A} \rangle_r = p$ is the concentration of A ions.

As discussed in our previous work,[6,7] within the EFT, the averaged total and lattice magnetizations for the honeycomb lattice with a coordination number $z=3$ are given by

$$M = \left\langle\!\!\left\langle \sum_i s_i^z \right\rangle\!\!\right\rangle_r = N[p\sigma + (1-p)m] , \tag{3}$$

$$\begin{aligned}\sigma &= \langle\langle s_{i=A}^z \rangle\rangle_r \\ &= \left\{ p\left[\cosh\left[\frac{J_{AA}}{2} D \right] + 2\sigma \sinh\left[\frac{J_{AA}}{2} D \right] \right] + (1-p)[q\cosh(J_{AB}D) + m\sinh(J_{AB}D) + 1 - q] \right\}^3 F(x+h)\Big|_{x=0} ,\end{aligned} \tag{4}$$

$$\begin{aligned}m &= \langle\langle s_{i=B}^z \rangle\rangle_r \\ &= \left\{ p\left[\cosh\left[\frac{J_{AB}}{2} D \right] + 2\sigma \sinh\left[\frac{J_{AB}}{2} D \right] \right] + (1-p)[q\cosh(J_{BB}D) + m\sinh(J_{BB}D) + 1 - q] \right\}^3 G(x+h)\Big|_{x=0} ,\end{aligned} \tag{5}$$

with

$$F(x) = \tfrac{1}{2}\tanh\left[\frac{\beta}{2} x \right] , \tag{6}$$

$$G(x) = \frac{2\sinh(\beta x)}{2\cosh(\beta x) + \exp(\beta\Delta)} , \tag{7}$$

where $\beta = 1/k_B T$, $h = \beta H$, and N is the total number of magnetic atoms. $D = \partial/\partial x$ is a differential operator. The operator q is given by

$$\begin{aligned}q &= \langle\langle (s_{i=B}^z)^2 \rangle\rangle_r \\ &= \left\{ p\left[\cosh\left[\frac{J_{AB}}{2} D \right] + 2\sigma \sinh\left[\frac{J_{AB}}{2} D \right] \right] + (1-p)[q\cosh(J_{BB}D) + m\sinh(J_{BB}D) + 1 - q] \right\}^3 E(x+h)\Big|_{x=0} ,\end{aligned} \tag{8}$$

with

$$E(x) = \frac{2\cosh(\beta x)}{2\cosh(\beta x) + \exp(\beta\Delta)} . \tag{9}$$

As discussed in a great number of works,[3,6-8] these equations can be easily calculated by applying a mathematical relation $e^{\gamma D}\phi(x) = \phi(x+\gamma)$.

III. PHASE DIAGRAMS

Here we are interested in studying the transition temperature of the system. For $H=0$ 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 sublattice magnetizations for Eqs. (4) and (5). The sublattice magnetizations then reduce to

$$(A_1 - 1)\sigma + B_1 m = 0, \quad A_2\sigma + (B_2 - 1)m = 0 , \tag{10}$$

with

$$\begin{aligned}
A_1 &= 6p \sinh\left[\frac{J_{AA}}{2} D \right] \left[p\cosh\left[\frac{J_{AA}}{2} D \right] + (1-p)[q_0\cosh(J_{AB}D) + 1 - q_0] \right]^2 F(x)\Big|_{x=0} , \\
B_1 &= 3(1-p)\sinh(J_{AB}D) \left[p\cosh\left[\frac{J_{AA}}{2} D \right] + (1-p)[q_0\cosh(J_{AB}D) + 1 - q_0] \right]^2 F(x)\Big|_{x=0} , \\
A_2 &= 6p \sinh\left[\frac{J_{AB}}{2} D \right] \left[p\cosh\left[\frac{J_{AB}}{2} D \right] + (1-p)[q_0\cosh(J_{BB}D) + 1 - q_0] \right]^2 G(x)\Big|_{x=0} , \\
B_2 &= 3(1-p)\sinh(J_{BB}D) \left[p\cosh\left[\frac{J_{AB}}{2} D \right] + (1-p)[q_0\cosh(J_{BB}D) + 1 - q_0] \right]^2 G(x)\Big|_{x=0} ,
\end{aligned} \tag{11}$$

where q_0 is determined from

$$q_0=\left[p\cosh\left[\frac{J_{AB}}{2}D\right]+(1-p)[q_0\cosh(J_{BB}D)+1-q_0]\right]^3 E(x)\Big|_{x=0} . \tag{12}$$

The critical boundary characterizing the ferromagnetic-phase stability limit is determined by

$$(A_1-1)(B_2-1)=A_2B_1 . \tag{13}$$

By solving Eq. (13) numerically, we find the phase diagrams shown in the following.

Figure 1 shows the change of transition temperature with p for the system with a selected value of Δ, when the exchange interactions are taken as $J_{AA}=J_{AB}=J_{BB}=J$. The figure clearly expresses some interesting results, when the value of Δ becomes larger than $\Delta=J$; for instance, the curve labeled $\Delta=1.5J$ has no solution for the concentration p below $p=0.48$. Moreover, the curve labeled $\Delta=1.4J$ shows an anomalous behavior, for which no solution of (13) can be obtained in the region of $0.14<p<0.375$. In other words, the transition temperature for the system with $\Delta=1.5J$ (or $\Delta=1.4J$) reduces to zero in these regions of p, indicating that the system is in a new disordered phase with $T_c=0$.

Before discussing the physical background for the characteristic behaviors of Fig. 1, we present another result in Fig. 2; the change of transition temperature with Δ is plotted for the system with $J_{AA}=J_{AB}=J_{BB}=J$, when p is fixed at $p=0.3$.

Now, it is well known that the regular spin-1 Ising model with a crystal field Δ given by the Hamiltonian

$$\mathcal{H}=-J\sum_{i,j}s_i^zs_j^z+\Delta\sum_i(s_i^z)^2 , \tag{14}$$

where $s_i^z=\pm1,0$, exhibits a tricritical point at which the phase transition changes from second order to first order, when the value of Δ takes a critical value $\Delta=\Delta_t$.[4,5] For $H=0$, the Hamiltonian (1) reduces to (14), when p is given by $p=0$. As discussed in the previous works[9,10] (or see the Appendix), for $p=0$ the critical surface characterizing the ferromagnetic-phase-stability limit is determined by

$$m^2=(1-a)/b . \tag{15}$$

The second-order phase-transition line is determined from $a=1$. The right-hand side of (15) must be positive.

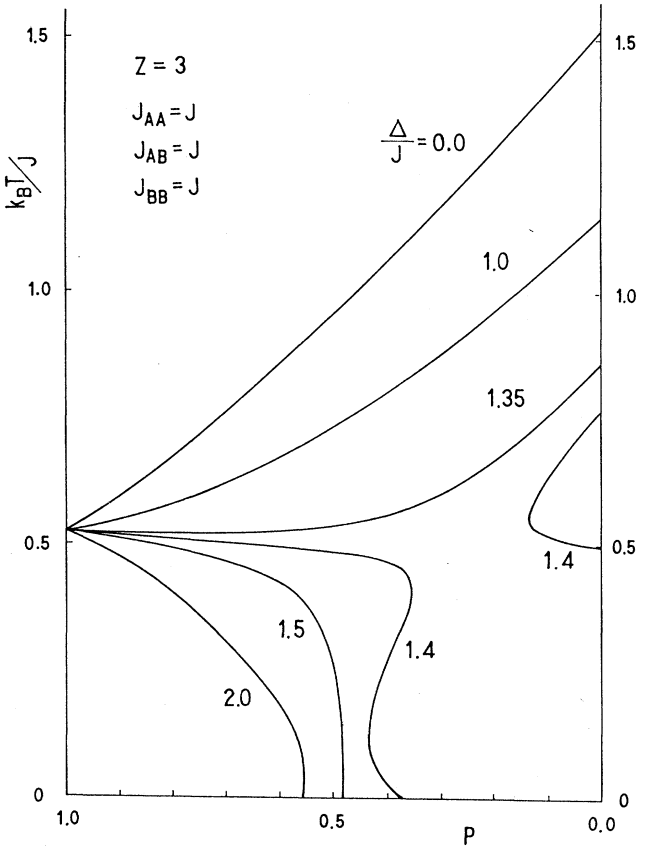

FIG. 1. Phase diagrams in T-p space for the binary ferromagnetic Ising alloy in a honeycomb lattice ($z=3$), when Δ is changed. The exchange interactions are fixed as $J_{AA}=J_{AB}=J_{BB}=J$.

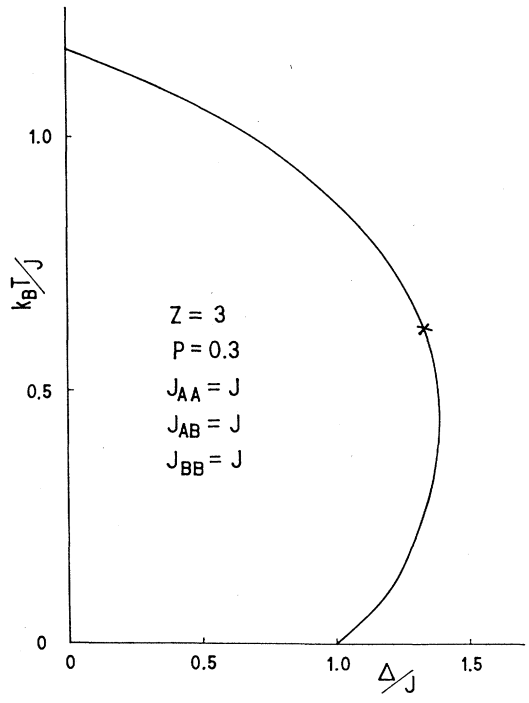

FIG. 2. Behavior of transition temperature vs Δ for the system with $J_{AA}=J_{AB}=J_{BB}=J$, when p is fixed at $p=0.3$.

If this is not the case, the transition is of first order, and hence the point at which $a=1$ and $b=0$ is the tricritical point.

For $p=0$, as shown in Fig. 3, the tricritical point is given by $\Delta=\Delta_t=1.4245J$ and the dashed line denotes the first-order phase transition. The change of order of phase transition at the tricritical point results from the competition between the crystal-field interaction and the internal field at each site; when the crystal field is larger than the internal field, each spin orients perpendicularly to the z direction suddenly at the transition temperature, so that a first-order phase transition (or discontinuity) may appear in the magnetization curve.

As previously discussed, the relation (13) from which Figs. 1 and 2 are obtained just corresponds to the relation $a=1$ of (15). From the relation (13) alone, therefore, we could not determine the tricritical point, even if it exists in the system with a concentration p near $p=0$. On the other hand, for the bond-diluted system with $J_{AA}=J_{AB}=0$ and $J_{BB}=J$ Benayard *et al.*[10] proved that the coupled equations (5) and (8) can be written in the form of (15). Then, they showed that the tricritical point decreases from that of $p=0$ with $1-p$. In this way, for the curve of Fig. 2 there is a possibility of the existence of a tricritical point, especially in the region of Δ larger than $\Delta=J$, although for finite values of J_{AA}, J_{AB}, and J_{BB} we cannot rewrite the coupled equations (4), (5), and (8) in the form of (15). [A preliminary calculation indicates that the cross in Fig. 2, which is obtained by writing (4), (5), and (8) in the form of (15), may approximately correspond to the tricritical point, and that below the cross the curve may have no physical meaning, as in Fig. 3.]

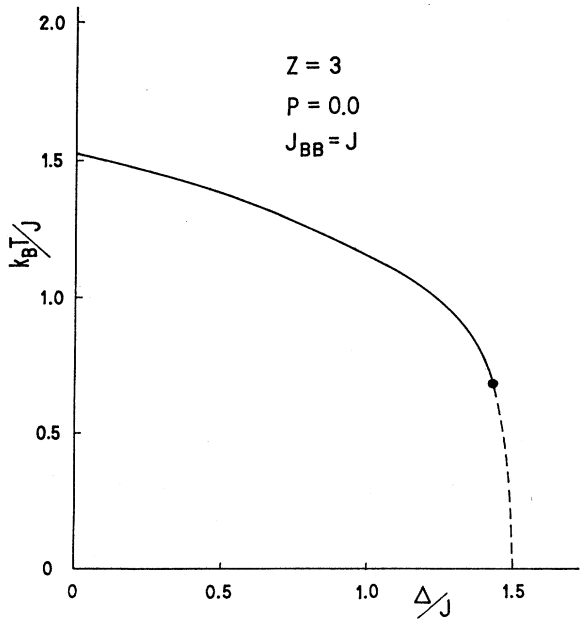

FIG. 3. Curie temperature vs Δ for the regular spin-1 Ising system in a honeycomb lattice ($z=3$). The solid circle on the curve denotes the tricritical point obtained from the framework discussed in the Appendix.

We are now in a position to discuss the results of Fig. 1. The curve labeled 1.5 in the figure has a value larger than $\Delta=\Delta_t$ for the system with $p=0$ (or the regular spin-1 system). When B ions with $s_B=1$ are replaced with A ions with $s_A=\frac{1}{2}$, spins of the remaining B ions then continue to take the $s^z=0$ state, so that spins of A ions are forced to take randomly the states $s^z=\pm\frac{1}{2}$ because of the exchange interaction J_{AB}. Until the concentration $p=0.48$, therefore, the transition temperature reduces to zero. Above that concentration, the exchange interaction J_{AA} becomes dominant, spins of A ions are apt to take the z direction, and hence the transition temperature has a finite value.

On the other hand, looking at Fig. 2, the cross is given by $\Delta=1.325J$. The curve labeled $\Delta=1.35J$ in Fig. 1 has a value larger than $\Delta=1.325J$, which implies that the point at $p=0.3$ in the curve labeled 1.35 may not have any physical meaning, since the point may correspond to that of a first-order phase transition. That is to say, the curves labeled 1.35 and 1.4 in Fig. 1 are simply obtained from the relation (13) [or the condition $a=1$ of (15)] and hence at some concentrations near $p=0$ the possibility of satisfying the tricritical condition remains in the curves, although we could not determine the points.

In Fig. 4 the phase boundaries of the alloy with

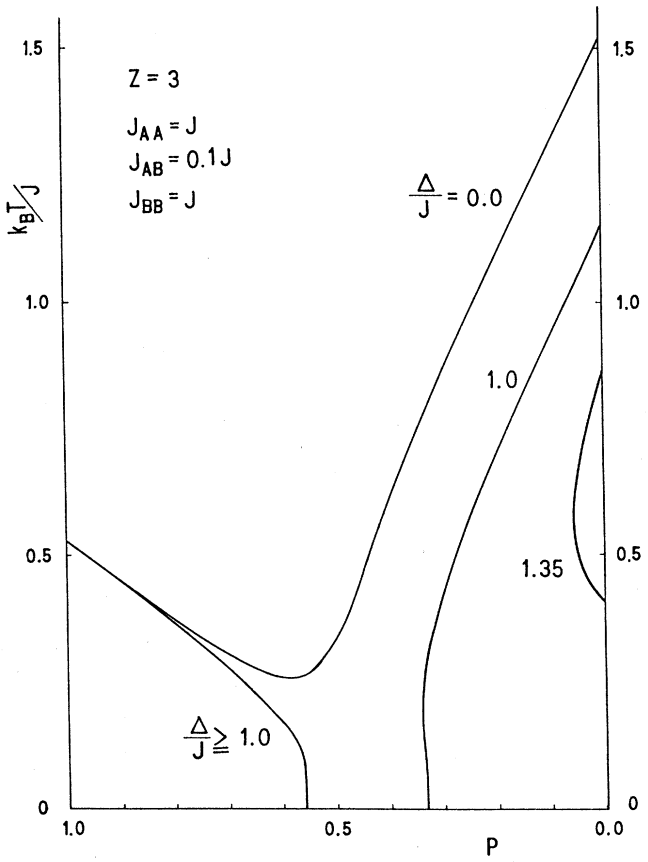

FIG. 4. Phase diagrams of a binary ferromagnetic alloy of the type A_pB_{1-p} with $z=3$, when Δ is changed. The exchange interactions are then taken as $J_{AA}=J_{BB}=J$ and $J_{AB}=0.1J$.

$J_{AA}=J_{BB}=J$ and $J_{AB}=0.1J$ are plotted as a function of p, by selecting the value of Δ. As is seen from the figure, for the curve labeled $\Delta=1.0J$, the transition temperature reduces to zero in the region of $0.33<p<0.56$. With the increase of Δ, the region at which the transition temperature reduces to zero becomes wider, but the curve on the left-hand side labeled $\Delta/J\geq 1.0$ means that the curve takes almost the same values for this whole range of Δ. For the curve on the right-hand side with Δ/J larger than unity, the possibility of a tricritical point remains, as previously discussed. On the other hand, when Δ/J is smaller than unity, the region in which the transition temperature reduces to zero does not appear.

In Fig. 5 the change of critical boundary with p is depicted for the system with $J_{AA}=J_{BB}=J$ and $J_{AB}=5J$, changing the value of Δ. In this case, because of the strong exchange interaction J_{AB}, the region at which the transition temperature reduces to zero may appear when the value of Δ takes a value larger than $\Delta=\Delta_t$, as shown for the curve labeled $\Delta=2.0J$.

Thus, an important conclusion of this section is as follows, although we have not studied the possibility of a tricritical point: In a binary ferromagnetic alloy of the type A_pB_{1-p} a new disordered phase, in which the transition temperature reduces to zero, may appear when spin-1 ions with a crystal-field interaction larger than $\Delta=J$ is randomly mixed with spin-$\frac{1}{2}$ ions. The region of p in which the new disordered phase appears is strongly dependent on the value of exchange interaction J_{AB}. In the new disordered phase, the spins of the B ions go into the $s^z=0$ state and the spins of the A ions are forced into random occupation of the states $s^z=\pm\frac{1}{2}$.

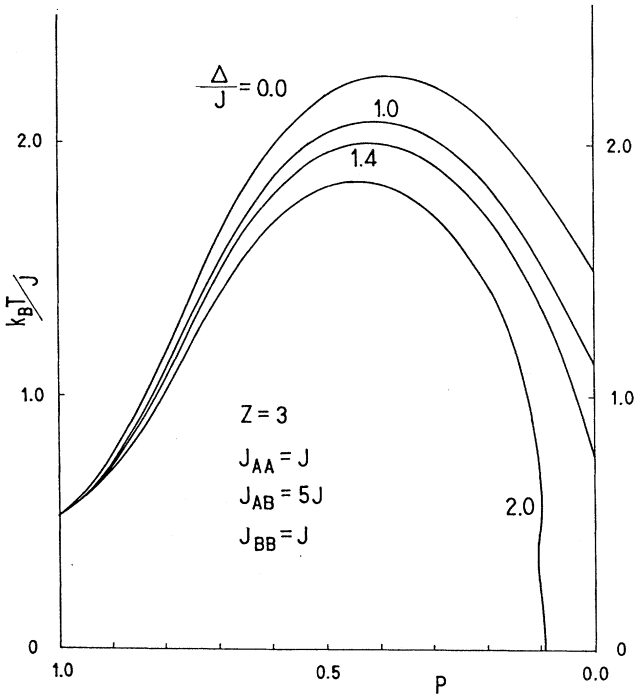

FIG. 5. Phase diagrams in the T-p space for the binary ferromagnetic alloy with $z=3$, when Δ is changed. The exchange interactions are then taken as $J_{AA}=J_{BB}=J$ and $J_{AB}=5J$.

IV. MAGNETIZATION

In this section let us study the magnetization curves and the magnetization process by solving Eqs. (3)–(5) and (8) numerically, especially for the binary ferromagnetic system with $J_{AA}=J_{AB}=J_{BB}=J$ (Fig. 1).

In Fig. 6 the temperature dependences of total and sublattice magnetizations under $H=0$ are shown for the system with $p=0.7$. As is seen from the curves labeled a and b, the saturation magnetizations of σ and m for $\Delta=0$ and $\Delta=J$ are given by $\sigma=\frac{1}{2}$ and $m=1$, so that the total magnetization at $T=0$ K is given by $M=0.65N$. On the other hand, when the value of Δ is larger than $\Delta=J$, the total magnetization at $T=0$ K decreases from $M=0.65N$, as in curves c and d. In particular, as is seen from the dashed curve labeled c, the sublattice magnetization m at $T=0$ K is depressed greatly from $m=1$. It implies that the spins of the B ions with $s_B=1$ deviate greatly from the z direction because of the large value of Δ.

In Fig. 7 the temperature dependences of total and sublattice magnetizations for the system with $p=0.3$ are depicted for the two values of Δ ($\Delta=0$ and J), when H is taken as 0 or $0.1J$. As seen from the curves, the behavior of M in finite applied field is normal.

On the other hand, as shown in Fig. 1, when the value of Δ becomes larger than $\Delta=J$, the phase diagram may exhibit a number of characteristic behaviors, although the forms of the phase-boundary lines near $p=0$ cannot

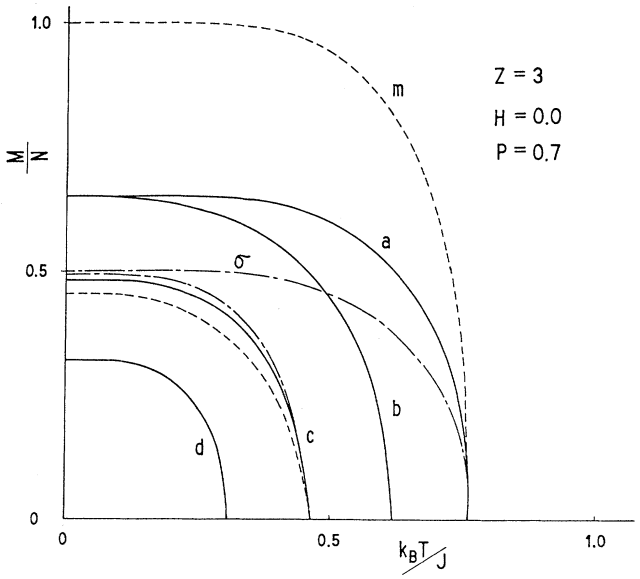

FIG. 6. Magnetization curves for $H=0$ in a binary ferromagnetic alloy of the type A_pB_{1-p} with $z=3$ and $p=0.7$, when Δ is changed as follows: a, $\Delta=0$; b, $\Delta=J$; c, $\Delta=1.5J$; d, $\Delta=2J$. The exchange interactions are taken as $J_{AA}=J_{AB}=J_{BB}=J$ (see Fig. 1). Solid lines denote the total magnetization. Dashed and short-dashed–long-dashed lines are sublattice magnetizations m and σ for $\Delta=0$ and $1.5J$.

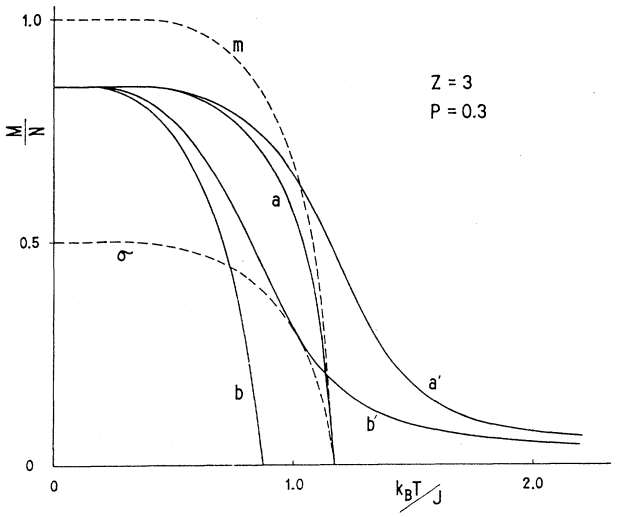

FIG. 7. Temperature dependences of total (solid lines) and sublattice (dashed lines) magnetizations for the binary ferromagnetic A_pB_{1-p} alloy with $p=0.3$ and $z=3$, when Δ and H are selected as follows: a, $\Delta=0$ and $H=0$; a', $\Delta=0$ and $H=0.1J$; b, $\Delta=J$ and $H=0$; b', $\Delta=J$ and $H=0.1J$. The exchange interactions are then taken as $J_{AA}=J_{AB}=J_{BB}=J$.

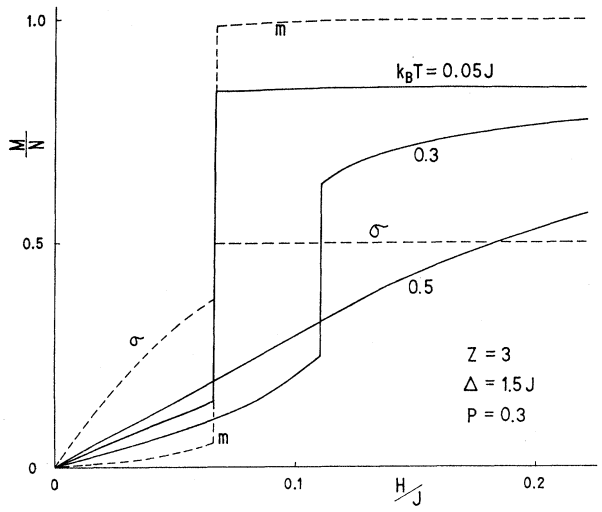

FIG. 8. Magnetization processes in the new disordered phase of the binary ferromagnetic A_pB_{1-p} alloy with $p=0.3$, $\Delta=1.5J$, and $z=3$, when T is changed as $k_BT=0.05J$, $0.3J$, or $0.5J$. The exchange interactions are then taken as $J_{AA}=J_{AB}=J_{BB}=J$. See Fig. 1.

be believed because of the possibility of a tricritical point. However, the appearance of the new disordered phase in which the transition temperature reduces to zero is correct. In order to examine the physical content of the disordered phase, the magnetization process is examined in Fig. 8, especially for the system with $p=0.3$ and $\Delta=1.5J$, which is in the new disordered phase of Fig. 1.

In Fig. 8 the field dependences of total and sublattice magnetizations are shown for the three fixed temperatures ($k_BT=0.05J$, $0.3J$, and $0.5J$). At $H=0$, as expected, their magnetizations are given by zero. The total and sublattice magnetizations at $k_BT=0.05J$ increase with the increase of H, exhibit a discontinuity at $H=0.066J$, and then approach their saturation values. For the temperature $k_BT=0.3J$, the total magnetization exhibits a discontinuity at $H=0.115J$, although it is smaller than that for $k_BT=0.05J$. For the temperature $k_BT=0.5J$, however, such a discontinuity disappears from the magnetization process. The results are very similar to those found in metamagnetic materials, although in the usual metamagnets the critical field at which the magnetization changes discontinuously does not depend on the temperature.[8] In this way an interesting characteristic of Fig. 8 is the temperature dependence of the critical field. The result implies that in the new disordered state the spins of the A and B ions are more tightly bound in the $s_A^z=\pm\frac{1}{2}$ and $s_B^z=0$ states for the temperature $k_BT=0.3J$ than for the temperature $k_BT=0.05J$, against thermal agitation. On the other hand, the result for the temperature $k_BT=0.5J$ indicates that the states are easily broken under the application of an applied field. The discontinuity of magnetization in Fig. 8 comes from the spin flip from the $s_B^z=0$ state to the $s_B^z=1$ state, similar to that of metamagnets. On the other hand, the solid solution $CO(S_xSe_{1-x})_2$ can be antiferromagnetic, paramagnetic, or ferromagnetic, when $0\le x\le 0.5$, $0.5<x<0.88$, or $0.88\le x\le 1$, respectively.[11] Adachi *et al.*[11] have suggested that the paramagnetic state for $0.5<x<0.88$ results from the competition of ferromagnetic and antiferromagnetic exchange interactions. In the paramagnetic state they have observed an unusual magnetization process; the transition field at which the total magnetization shows a discontinuity depends on the temperature, as in Fig. 8. Adachi *et al.* have also noted that the dependence of transition field on temperature resembles the phase diagram of liquid-gas systems.

At first sight the magnetization process of Fig. 8 seems to be similar to that of $CO(S_xSe_{1-x})_2$ in the paramagnetic state. However, as shown in Fig. 9, the magnetic behavior is completely different from that of $CO(S_xSe_{1-x})_2$. In Fig. 9 the dependences of critical field H_c and gap width $\Delta M/N$ with temperature are plotted, where $\Delta M/N$ and H_c are the values of gap width and field at the point where the total magnetization of Fig. 8 exhibits a discontinuity. The change of H_c with temperature is dramatic; with increasing T it increases abruptly to the maximum value near $T=0.16J$ and then decreases, although the change of H_c in $CO(S_xSe_{1-x})_2$ exhibits a monotonic increase. Moreover, the gap width $\Delta M/N$ exhibits a decrease in two steps. The change of curvature is observed at the point of the maximum value of H_c. The value $\Delta M/N$ reduces to zero at $k_BT=0.32J$.

Thus, the new disordered phase exhibits unusual magnetization behavior. Experimentally, as far as we know, such a disordered phase as well as the unusual magnetization process has not been observed.

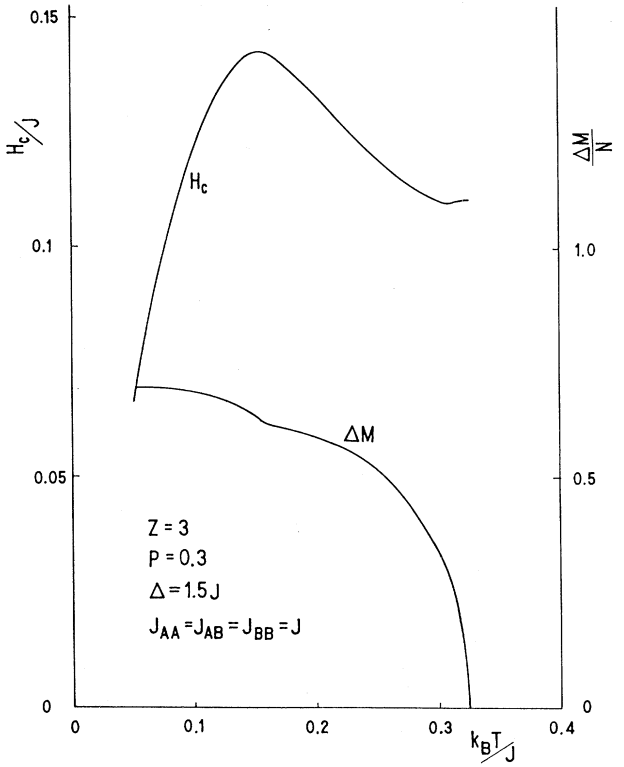

FIG. 9. Temperature dependences of field H_c and gap width $\Delta M/N$ at the point where the total magnetization of Fig. 8 exhibits a discontinuity.

V. CONCLUSIONS

We have studied the phase diagrams and the temperature (or field) dependences of magnetization of a binary Ising ferromagnetic alloy A_pB_{1-p} consisting of spin-$\frac{1}{2}$ ions A and spin-1 ions B with a (negative) crystal-field interaction where all the exchange interactions have the same sign, using the effective-field theory with correlations. We find that a new disordered phase in which the total and sublattice magnetizations are given by zero may appear, when the crystal-field interaction becomes larger than $\Delta=J$. The region of p in which the disordered phase appears is strongly dependent on the value of the exchange interaction J_{AB}. In the disordered phase the spins of A and B ions are occupied by $s_A^z=\pm\frac{1}{2}$ and $s_B^z=0$ states, respectively. The magnetization process in the disordered phase exhibits phenomena similar to but with important differences from those of metamagnets.

On the other hand, the finding of the new disordered phase is cause for further caution in studying the magnetic phases of a binary alloy. Some authors[2,3] have examined the various kinds of phase diagrams which may occur in binary A_pB_{1-p} Ising ferromagnetic alloys with $\Delta=0$ under $H=0$, in terms of the initial slope $[\partial(\ln T_c)/\partial p]$ of the transition temperature T_c with concentration p at the two points $p=0$ and 1. However, the results obtained in this work clearly show that from the initial slopes alone we cannot determine the overall character of the phase diagram, when a (negative) crystal-field interaction is included.

In Sec. III we investigated the phase diagrams of a binary Ising ferromagnetic alloy in terms of the relation (13) [or $a=1$ in Eq. (15)]. However, in the phase diagrams obtained there, the possibility of a tricritical point at which the phase transition changes from second order to first order remains, especially for the curves near $p=0$, when the crystal-field interaction becomes larger than $\Delta=J$.

In this work we have used the effective-field framework. As discussed in Refs. 3, 6, 7, and 10, the approximation essentially corresponds to the Zernike approximation.[12] This formalism is, from an analytical standpoint, almost as simple as the standard mean-field approximation, and, because of neglect of multispin correlations, shares with it the fact that the critical exponents are all of Landau type. Nevertheless, we verify that its results are quite superior to other effective-field theories, as shown in Figs. 1–9.

Finally, for crystalline mixed alloys there is surprisingly little experimental data to compare with the phase diagram predicted theoretically. In fact, many transition-metal compounds are of the Heisenberg type, and most insulating magnetic alloys are antiferromagnetic. However, there may be a possibility of finding some characteristics discussed in this work, namely the new disordered phase. We hope that this work will stimulate further experimental and theoretical work on the magnetic properties of a binary alloy.

APPENDIX

In this appendix we briefly review the spin-1 Ising model with a crystal-field interaction on a honeycomb lattice ($z=3$). As discussed in Sec. III, when $p=0$, the Hamiltonian (1) for $H=0$ reduces to the Hamiltonian (14). Then, Eqs. (5) and (8) are given by, with $J_{BB}=J$,

$$m=[q\cosh(JD)+m\sinh(JD)+1-q]^3G(x)|_{x=0} \quad \text{(A1)}$$

and

$$q=[q\cosh(JD)+m\sinh(JD)+1-q]^3E(x)|_{x=0}\,. \quad \text{(A2)}$$

We are now interested in studying the transition temperature of the system. Expanding the right-hand sides of (A1) and (A2), we obtain

$$m=Am+Bm^3\,, \quad \text{(A3)}$$

$$q=A'+B'm^2\,, \quad \text{(A4)}$$

with

$$A=3[q\cosh(JD)+1-q]^2\sinh(JD)G(x)|_{x=0}\,, \quad \text{(A5)}$$

$$B=\sinh^3(JD)G(x)|_{x=0}\,, \quad \text{(A6)}$$

$$A'=[q\cosh(JD)+1-q]^3E(x)|_{x=0}\,, \quad \text{(A7)}$$

$$B'=3[q\cosh(JD)+1-q]\sinh^2(JD)E(x)|_{x=0}\,. \quad \text{(A8)}$$

If we replace q in (A5) with the expression of (A4), we obtain an equation for m of the form

$$m = am + bm^3 + \cdots . \tag{A9}$$

The second-order phase-transition line is then determined by $a=1$. In the vicinity of the second-order phase-transition line, the magnetization is given by

$$m^2 = (1-a)/b . \tag{A10}$$

The right-hand side must be positive. If this is not the case, the transition is of the first order, and hence the point at which $a=1$ and $b=0$ is the tricritical point.

At this point, the parameter a is given by

$$a = 3[q_0 \cosh(JD) + 1 - q_0]^3 \sinh(JD) G(x)|_{x=0} , \tag{A11}$$

where q_0 is the solution of

$$q_0 = [q_0 \cosh(JD) + 1 - q_0]^3 E(x)|_{x=0} . \tag{A12}$$

The parameter b is defined by

$$b = 6q_1[q_0 \cosh(JD) + 1 - q_0] \sinh(JD) \times [\cosh(JD) - 1] G(x)|_{x=0} , \tag{A13}$$

with

$$q_1 = f/(1-e) , \tag{A14}$$

where the parameters e and f are given by

$$e = 3[q_0 \cosh(JD) + 1 - q_0]^2 [\cosh(JD) - 1] E(x)|_{x=0} , \tag{A15}$$

$$f = 3[q_0 \cosh(JD) + 1 - q_0] \sinh^2(JD) E(x)|_{x=0} . \tag{A16}$$

These equations can be easily calculated by the use of a mathematical relation $e^{\gamma D}\phi(x) = (x+\gamma)$. The expression of (A10) with (A11) and (A13) for $z=3$ has been derived by Benayard *et al.*[9] and Kaneyoshi.[8]

In Fig. 3 the ferromagnetic critical line is plotted as a function of Δ. The result has been obtained by Benayard *et al.* and Kaneyoshi. In Fig. 3 solid and dashed lines denote second-order and first-order transition lines, respectively. The solid circle denotes the tricritical point ($k_B T_t = 0.6780J$ and $\Delta_t = 1.4245J$).

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