Dilute Ising ferromagnet: Its physical properties

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Within the new type of correlated effective-field theory, which substantially improves the molecular-field approximation, we calculate the transition temperature, the critical concentration for site percolation, magnetization, correlated effective parameter, and initial susceptibility associated with the dilute ferromagnetic Ising system on a square lattice. The results are qualitatively satisfactory; the critical concentration P_c is determined as $P_c = 0.564$. The effect, on initial susceptibility, of the (eventually) coexisting finite and infinite clusters is exhibited. It is shown that the correlated effective parameter exhibits some interesting behavior as a function of temperature for selected values of concentration of magnetic atoms.

I. INTRODUCTION

There is a long history of research on the dilution problem by nonmagnetic atoms in a ferromagnet. It is now well known that the critical temperature for a phase transition decreases monotonically as the concentration of nonmagnetic impurities increases, and that the transition temperature vanishes at some nonzero value of the concentration, which depends on the topology of the lattice structure.¹ Since the pioneering work of Sato et al., much work has been done concerning the behavior of the transition temperature as a function of nonmagnetic impurities. The problem is studied by using a variety of approximations and mathematical techniques. Nevertheless, the physical properties, such as magnetization, susceptibility, specific heat, and short-range order, as functions of concentration and temperature have not been studied extensively in the dilute Ising ferromagnet.

Recently, Kaneyoshi *et al.*³ have developed for the spin- $\frac{1}{2}$ pure Ising model a new type of correlated effective-field theory. The theory yields values for the critical temperature and other thermodynamic properties that are identical to those of the Bethe-Peierls approximation, although the approach is completely different in its formulation from the Bethe-Peierls method. The approach has already been applied to a variety of physical problems, such as pure anisotropic systems,⁴ dilute ferromagnets,⁵ and surface magnetism.⁶ In particular, most of these works have proved that the correlated effective parameter introduced in the theory (originally due to Lines⁷), which is basically a measure of the short-range order, shows some interesting behavior in each problem.

In the present work we study the dilute spin- $\frac{1}{2}$ Ising ferromagnet on a square lattice and calculate the most relevant thermodynamical quantities (transition temperature, magnetization, correlated effective parameter, and zero-field magnetic susceptibility) within the correlated effective-field theory. This approach is in much the same spirit as the early works based on the effective-field concept. While we do not expect the effective-field approximation to yield accurate values in the critical region, due to the absence of long-range fluctuations, we do expect to obtain reasonable values for the critical concentration and the expression for the phase boundary over the entire temperature and concentration range. In fact, we can obtain results that are a significant improvement on other effective-field approximations; for instance, the critical concentration P_c is given by $P_c = 0.564$, which is near to the best value $P_g = 0.590$ obtained from the seriesexpansion method.⁸ For other thermodynamical properties we find some interesting behavior characteristic of the dilute Ising system.

In the next section we introduce the new type of correlated effective-field theory for the dilute Ising model. In Sec. III the framework is applied to the dilute ferromagnet in a square lattice. The analytical forms of the relevant thermodynamical quantities are obtained. In Sec. IV the numerical results of such quantities are studied and discussed.

II. CORRELATED EFFECTIVE-FIELD THEORY

In a dilute ferromagnet magnetic and nonmagnetic atoms are randomly assigned to the sites of a lattice without site correlation, as if an alloy had been quenched from a perfectly disordered configuration at high temperature to a condition in which all atoms are immobile. The Hamiltonian of the dilute Ising model is given by

$$
\mathscr{H} = -\frac{1}{2} \sum_{i,j} J_{ij} \xi_i \xi_j \mu_i \mu_j - H \sum_i \xi_i \mu_i , \qquad (1)
$$

where $\mu_i = \pm 1$ is the usual Ising variable, and J_{ij} is the exchange interaction between spins occupied at sites i and j . H is the external field, and ξ_i is a random variable equal to ¹ if the site is occupied or to zero if it is empty.

Formal identities for the correlation functions of the Ising model have appeared in the literature for some $time.⁹$ The starting point for the statistics of our spin system is the exact relation due to Callen'

$$
\langle \mu_i \{i\} \rangle = \left\langle \{i\} \tanh \left(\beta \sum_j J_{ij} \mu_j \xi_j + \beta H \right) \right\rangle, \tag{2}
$$

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where the angular bracket indicates the usual ensemble average

$$
\langle \cdots \rangle = \mathrm{Tr}[\exp(-\beta \mathcal{H}) \cdots]/\mathrm{Tr} \exp(-\beta \mathcal{H})
$$

and $\beta = (k_B T)^{-1}$. $\{i\}$ refers to any function of the variables μ_l as long as $l\neq i$. Here, in order to write the identity (2) in a form that is particularly amenable to approximation, let us introduce the differential operator technique proposed by Honmura and Kaneyoshi 11 ; upon set- $\begin{aligned} \text{diag} \{i\} = 1, \end{aligned}$

$$
]/\text{Tr}\exp(-\beta \mathscr{H}) \qquad \langle \mu_i \rangle = \left\{ \left\langle \exp\left[D\left(\sum_j J_{ij} \mu_j \xi_j\right)\right] \right\rangle \tanh(\beta x + h) \right\}_{x=0}, \quad (3)
$$

where $D = \partial/\partial x$ is a differential operator, and $h = \beta H$. For a system with nearest-neighbor interaction J , Eq. (3) can be written as follows, by using the facts that $\xi_i^2=\xi_i$. and $\exp(\alpha \mu_i) = \cosh(\alpha) + \mu_i \sinh(\alpha)$:

$$
\langle \mu_i \rangle = \left\langle \prod_{\delta} \left\{ \xi_{i+\delta}[\cosh(DJ) + \mu_{i+\delta}\sinh(DJ)] + (1 - \xi_{i+\delta}) \right\} \right| \tanh(\beta x + h) \Big|_{x=0}
$$

=
$$
\sum_{n=1}^{z} \begin{bmatrix} z \\ n \end{bmatrix} \left[\prod_{\delta=1}^{n} \xi_{i+\delta} \right] \left[\prod_{\delta'=n+1}^{z} (1 - \xi_{i+\delta'}) \right] \left\langle \prod_{\delta=1}^{n} [\cosh(DJ) + \mu_{i+\delta}\sinh(DJ)] \right\rangle [\tanh(\beta x + h)]_{x=0},
$$
 (4)

where δ runs over the z nearest-neighbor displacements from a central site i.

For the random configurational average, let us assume that the nonmagnetic and magnetic atoms are randomly distributed in the z nearest-neighbor sites about the central site i and ignore important effects due to the environment in which we place the cluster. Then Eq. (4) can be written as

$$
\sigma_i = \langle \langle \mu_i \rangle \rangle_r = \sum_{n=1}^z \binom{z}{n} P^n (1 - P)^{z - n} \langle \langle \prod_{\delta=1}^n \left[\cosh(DJ) + \mu_{i+\delta} \sinh(DJ) \right] \rangle \rangle_r [\tanh(\beta x + h)]_{x=0}
$$
\n(5)

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with $\langle \xi_i \rangle_r = P$, where $\langle \rangle_r$ expresses the random average. P is the concentration of magnetic atoms. This equation is essentially equivalent to that used by Matsudaira¹² initially and later by other authors.^{5,13} In other words, Eq. (5) is an approximation for the random configurational average. Precisely speaking, the random average on the right-hand side of Eq. (5) must be a conditional average, depending on the configuration of the magnetic atoms in the cluster taken. At the present time, however, it is difficult to evaluate the conditional average, so that a number of authors have replaced the conditional average with the usual random average. In the following, we will use the same notation as the usual thermal average, namely $\sigma_i = \langle \mu_i \rangle = \langle \mu_i \rangle$, Then the random configurational average is always taken after the thermal average, since we are dealing with a quenched system.

In order to evaluate Eq. (5) many authors¹²⁻¹⁵ have introduced an approximation; by assuming the statistical independence of lattice sites,

$$
\langle \mu_i \mu_j \cdots \mu_l \rangle \simeq \langle \mu_i \rangle \langle \mu_j \rangle \cdots \langle \mu_l \rangle . \tag{6}
$$

This approximation led, in spite of its simplicity, to quite satisfactory results. In fact, the approximation essentially corresponds to the Zernike approximation in the pure $(P=1)$ system. For the dilute Ising system with a square lattice, for instance, the critical concentration P_c is given by $P_c = 0.428$, a value which is relatively close to the best value of 0.590 obtained by the series-expansion (SE) method.¹⁶ The approximation is also applied to other diluted magnetic systems, such as diluted thin $films¹³$ and diluted surface.¹³

On the other hand, Kaneyoshi et $al.$ ³ have refined the approximation based on (6) by introducing a nearestneighbor correlation parameter λ for the pure Ising model. This is done by correlating the nearest-neighbor site $(i + \delta)$ with the central site (i) of the cluster via

$$
\mu_{i+\delta} = \langle \mu_{i+\delta} \rangle + \lambda(\mu_i - \langle \mu_i \rangle) \tag{7}
$$

Substituting this expression into Eq. (5) yields an equation for the averaged magnetization σ as a function of the single-site average (σ) , the correlated effective parameter (λ) , the concentration (P) , and the reduced temperature (t) , where t is defined by

$$
t = \frac{J}{k_B T} \tag{8}
$$

In order to determine λ uniquely, another equation is needed. In Ref. 16, Honmura very recently has found that the parameter λ can be determined analytically by solving the higher-order correlation function, namely the threesite correlation function $\langle \mu_i \mu_{i+ \delta} \mu_{i+ \delta'} \rangle$ in a square lattice, instead of the pair-correlation function $\langle \mu_i \mu_{i+\delta} \rangle$ introduced in Refs. 3 and 4, and that the thermodynamical properties of the pure Ising model obtained are completely equivalent to those of the Bethe-Peierls method.

Now, in order to evaluate the three-site correlation function in the dilute square Ising model discussed in the following section, let us assume that the central site i is again occupied and that at least two nearest-neighbor sites are occupied with the probability P . Consequently, the remaining $z-2$ sites are then occupied randomly. The averaged expression can be written as, upon setting $\{i\} = \mu_{i+\delta}\mu_{i+\delta'}$ in Eq. (2) and following the same process as that derived in Eq. (5),

$$
\langle \mu_i \mu_{i+\delta} \mu_{i+\delta'} \rangle = \sum_{n=0}^{z-2} \binom{z-2}{n} P^n (1-P)^{z-2-n} \Big\langle \mu_{i+\delta} \mu_{i+\delta'} \prod_{\delta''(\neq \delta,\delta')}^{n} [\cosh(DJ) + \mu_{i+\delta''} \sinh(DJ)] \Big\rangle [\tanh(\beta x + h)]_{x=0} . \tag{9}
$$

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Substituting (7) into Eq. (9), we can obtain another expression including σ , λ , P , and t . Thus we can determine the parameter λ analytically by means of Eqs. (5), (7), and (9), as will be shown in the next section.

In this section, we have briefly reviewed the correlated effective-field theory in the dilute Ising ferromagnet. In the following sections, we shall study the physical properties of the dilute ferromagnetic square lattice with the use of these formulas. On the other hand, Taggart⁵ also has applied the correlated effective-field theory to the dilute Ising systems and has discussed the critical concentration and transition temperature. However, in order to determine the parameter λ , an inverse function, instead of (9), was then introduced:

$$
1 = \sum_{n=1}^{z-1} \binom{z-1}{n} P^{n+1} (1-P)^{z-1-n} \Big\langle \mu_i \coth\left[t \sum_{f=1}^{n+1} \mu_{i+f}\right] \Big\rangle,
$$

which can be obtained by substituting $\{i\}$ $= \coth(\beta \sum_i J_{ij} \mu_i \xi_j)$ into Eq. (2). From the beginning, this equation neglects the possible spin configurations that result in $\sum_j J_{ij} \mu_j \xi_j = 0$, for which the equation cannot be defined. Accordingly, it does not seem reasonable to use the inverse function.

III. DILUTE FERROMAGNET IN A SQUARE LATTICE

In this section let us study the dilute Ising ferromagnet in a square lattice by using the formulas given in Sec. II. For the case of four nearest neighbors, Eq. (5) can be expanded, i.e.,

$$
\sigma = \sigma_i
$$

= $K_1 \sigma + K_2 \langle \mu_{i+1} \mu_{i+2} \mu_{i+3} \rangle$
+ $h(K_3 + K_4 \langle \mu_{i+1} \mu_{i+2} \rangle + K_5 \langle \mu_{i+1} \mu_{i+2} \mu_{i+3} \mu_{i+4} \rangle)$, (10)

where the coefficients K_i ($i = 1-5$) are given in Appendix. Equation (10) was then derived by expanding $tanh(\beta x + h)$ in Eq. (5) with h and retaining the terms linear to h.

Applying the correlated approximation (7) to the spincorrelation functions in Eq. (10), we obtain
 $0 = (B\sigma^2 - A)\sigma + hC$

$$
0 = (B\sigma^2 - A)\sigma + hC \tag{11}
$$

with

$$
A = 1 - K_1 - K_2 + B \t\t(12)
$$

$$
B = K_2 (1 - \lambda)^2 (1 + 2\lambda) , \qquad (13)
$$

$$
C = K_3 + K_4[\sigma^2 + \lambda^2(1 - \sigma^2)]
$$

+ K_5[\sigma^4 + 6\sigma^2(1 - \sigma^2)\lambda^2 - 8\sigma^2(1 - \sigma^2)\lambda^3
+ \lambda^4(1 + 2\sigma^2 - 3\sigma^4)]. (14)

On the other hand, from Eq. (9) we can also obtain the

following, by following the same procedure as that which led from Eq. (5) to Eq. (10):

$$
0 = (B'\sigma^2 - A')\sigma + hC'
$$
 (15)

with

$$
A'=1-K'_1-K'_2+B'
$$
\n(16)

$$
B'=K_2'(1-\lambda)^2(1+2\lambda)-(1-\lambda)^2,
$$
 (17)

$$
C' = K'_3 + K'_4 [\sigma^2 + \lambda^2 (1 - \sigma^2)]
$$

+ K'_5 [\sigma^4 + 6\sigma^2 (1 - \sigma^2)\lambda^2 - 8\sigma^2 (1 - \sigma^2)\lambda^3
+ \lambda^4 (1 + 2\sigma^2 - 3\sigma^4)], \qquad (18)

where the coefficients K_i' ($i = 1-5$) are also given in the Appendix. From Eqs. (11) and (15) we can evaluate some thermodynamical quantities of the dilute Ising ferromagnet in a square lattice.

A. Magnetization

For $h = 0$, the averaged magnetization σ is given by, from Eq. (11),

$$
\sigma^2 = \frac{A}{B} = 1 + \frac{1 - K_1 - K_2}{B} \tag{19}
$$

or from Eq. (15)

$$
\sigma^2 = \frac{A'}{B'} = 1 + \frac{1 - K_1' - K_2'}{B'} \tag{20}
$$

Inspecting the coefficients K_1 and K_2 , or K'_1 and K'_2 given in the Appendix, one can easily prove that the magnetization for the pure system is well defined at $T=0$, namely, $\sigma = 1$ at $T = 0$ for each case.

B. Correlated effective parameter

Upon setting Eq. (19) equal to Eq. (20), the correlated effective parameter can be determined as a function of P and t:

$$
\frac{1-K_1-K_2}{B}=\frac{1-K'_1-K'_2}{B'}.
$$

Solving the equation, we can easily prove that the parameter λ is given by

$$
\lambda = \frac{1}{2} \frac{1 - K_2' + aK_2}{K_2' - aK_2} \tag{21}
$$

with

$$
a = \frac{1 - K_1' - K_2'}{1 - K_1 - K_2} \tag{22}
$$

However, the parameter (21) is only valid below a transition temperature $T_c(P)$, since in order to determine λ we used the averaged magnetizations. For temperatures

above $T_c(P)$, another equation will be obtained later.

For the pure system, Eqs. (21) and (22) reduce to the following by the use of the coefficients (K_1, K_2, K'_1) , and K_2') given in the Appendix:

$$
\lambda = \frac{1}{2} \frac{1 - \tanh(2t)}{\tanh(2t)} \quad \text{for } P = 1 \tag{23}
$$

and

$$
a = 1 \quad \text{for } P = 1 \tag{24}
$$

The result (23) is nothing but that of the special case $(J_1 = J_2)$ discussed in Ref. 16.

C. Transition temperature

At the transition temperature $T=T_c(P)$, the magnetization reduces to zero; the critical line as a function of concentration is given by $A = 0$. Consequently, we have

$$
1 - K_1 - K_2 + K_2 (1 - \lambda_c)^2 (1 + 2\lambda_c) = 0 , \qquad (25)
$$

where λ_c is the value at $T=T_c(P)$. By solving the coupled equations (25) and (21), the transition temperature $T_c(P)$ can be determined as a function of P.

For the special case of $P=1$, Eq. (25) can be rewritten as the following, with the use of the coefficients $(K_1, K_2,$ K'_1 , and K'_2) and Eq. (23):

$$
(3-5x)(1+x)=0
$$

with

$$
x=\tanh(2t_c),
$$

where $t_c = J/k_B T_c$. Therefore, the value of λ_c at $P = 1$ is given by, from Eq. (23),

$$
\lambda_c = \frac{1}{3} \quad \text{for } P = 1 \tag{26}
$$

which is nothing but that derived in Ref. 3. The transition temperature T_c is then given by

$$
\frac{k_B T_c}{J} = \frac{2}{\ln 2} \approx 2.885 \text{ for } P = 1 , \qquad (27)
$$

which is equivalent to that of the Bethe-Peierls method.

D. Susceptibility

The initial susceptibility is defined by

$$
\chi = \lim_{H \to 0} \frac{\partial \sigma}{\partial H} = \frac{t}{J} \frac{\partial \sigma}{\partial h} \bigg|_{h=0} \,. \tag{28}
$$

Differentiating both sides of Eqs. (11) and (15) with h, we obtain

$$
C = (A - 3B\sigma^2) \left[\frac{\partial \sigma}{\partial h} \bigg|_{h=0} \right] + \sigma (1 - \sigma^2) \left[\frac{\partial B}{\partial \lambda} \bigg| \left[\frac{\partial \lambda}{\partial h} \bigg|_{h=0} \right] \right]
$$
(29)

$$
C' = (A' - 3B'\sigma^2) \left(\frac{\partial \sigma}{\partial h}\bigg|_{h=0}\right)
$$

+ $\sigma(1-\sigma)^2 \left(\frac{\partial B'}{\partial \lambda}\bigg)\left(\frac{\partial \lambda}{\partial h}\bigg|_{h=0}\right)$. (30)

Therefore the inverse initial susceptibility is given by

$$
(J\chi)^{-1} = \frac{(A - 3B\sigma^2) + D(A' - 3B'\sigma^2)}{t(C - DC')}, \quad T \le T_c \tag{31}
$$

with

$$
D = \frac{3K_2\lambda}{3K_2'\lambda - 1} \tag{32}
$$

On the other hand, the inverse paramagnetic susceptibility is given by, from Eqs. (29) or (30},

$$
(\textit{J}\chi_{\text{para}})^{-1} = \frac{A}{tC}
$$
\n⁽³³⁾

or

$$
(J\chi_{\text{para}})^{-1} = \frac{A'}{tC'} \tag{34}
$$

In Eq. (21) the correlated effective parameter λ was defined only for the region of $T \leq T_c$. In order to extend the parameter to the region of $T \geq T_c$, let us here impose a condition, namely,

$$
\frac{C}{A} = \frac{C'}{A'},\tag{35}
$$

from which we can obtain an equation determining the parameter λ for the region of $T \geq T_c$:

$$
\frac{1-K_1-K_2+K_2(1-\lambda)^2(1+2\lambda)}{K_3+K_4\lambda^2+K_5\lambda^4}
$$

=
$$
\frac{1-K_1'-K_2'+K_2'(1-\lambda)^2(1+2\lambda)-(1-\lambda)^2}{K_3'+K_4'\lambda^2+K_5'\lambda^4}
$$
(36)

The parameter λ is then determined as a function of P and t. By the use of λ , the inverse paramagnetic susceptibilit (33) can be evaluated.

We are now in a position to examine the physical properties of the dilute Ising ferromagnet in a square lattice numerically. The numerical results will be given in the next section.

IV. NUMERICAL RESULTS AND DISCUSSIONS

By solving the coupled equations (25) and (21), in Fig. ¹ the transition temperature and the critical correlated effective parameter $\lambda_c(P)$ are plotted as a function of P. The T_c and λ_c for the pure system are given by (26) and (27). In the limit that $T_c(P) \rightarrow 0$ and $\sigma \rightarrow 0$, we can obtain the critical concentration P_c as $P_c = 0.5642$. The result of P_c is extremely reasonable, since the best result of SE in square lattice is $P_c = 0.590$.⁸ On the other hand, the $\lambda_c(P)$ rapidly increases from the value $\lambda_c = \frac{1}{3}$ at $P = 1$ to the

and

FIG. 1. Critical temperature T_c and critical correlated effective parameter λ_c as a function of the concentration of magnetic atoms. The critical concentration P_c for $Z=4$ is given by 0.5642.

critical value $\lambda_c=0.734$ at $P=P_c$, as if the effective coordination number z^* decreases; as discussed in Ref. 3, the λ_c for pure systems is given by

$$
\lambda_c = \frac{1}{z-1} \; .
$$

If we apply the argument to the present system, z^* at $P=P_c$ is given by $z^* \approx 2.362$, which means that our system is quasi-one-dimensional at $P = P_c$.

In Fig. 2, the reduced magnetization curves obtained by solving the coupled equations (19) and (21) numerically are shown for the selected values of P. The effect of decreasing the concentration of magnetic atoms is an increase in the depression of magnetization over the entire temperature range for $T \leq T_c$, a phenomenon which is generally observed in dilute and amorphous ferromagnets. Very near the critical concentration, however, the behavior of the reduced magnetization curve is rather different. The curve of $P=0.6$ is over that of $P=0.7$ and

FIG. 2. Reduced magnetization curves for selected values of P. The concentration dependence of spontaneous magnetization is also depicted for the range of $0.6 \le P \le 1$.

shows a rather slow decrease of magnetization on increasing temperature. The result also reminds us of that of the reduced magnetization curve of a quasi-one-dimensional ferromagnet; as discussed in Ref. 15, near the critical concentration the magnetic behavior of a diluted twodimensional ferromagnet becomes similar to that of a one-dimensional system. In the inset of Fig. 2, the concentration dependence of saturation moment at $T=0$ is also shown for the region of $0.6 \le P \le 1$.

In Fig. 3 the temperature dependences of the correlated effective parameter are shown for selected values of P, which are obtained by solving Eqs. (21) and (36). The curve for the pure $(P=1)$ system is equivalent to that obtained in Refs. 3 and 4. The critical value λ_c for $P=1$ is given by (26). The curve for $P=1$ shows a sharp maximum at the transition temperature, Decreasing the concentration of magnetic atoms, on the other hand, the correlated effective parameter shows anomalous behavior, especially below the transition temperature. For the concentration less than unity, even at $T=0$, the parameter has a finite value which increases on decreasing the concentration of magnetic atoms. This anomalous behavior may be related to the following fact: As discussed by some authors, 17 the correlated effective-field concept is closely related to that of reaction field introduced by Onsager. Namely, the Weiss field acting on a given spin is the sum of two fields, the cavity field, which is the field in the absence of the spin in question, and the reaction field due to the polarization, by the spin, of its neighbors. Usually this reaction field is small compared to the cavity field and one can identify the cavity field as the Weiss field, excluding the critical region. The fact is clearly seen in the curve for $P=1$; at a very low temperature the correlated effective parameter for $P=1$ has a very small value. Increasing the number of nonmagnetic atoms, on the other hand, the cavity field simply decreases, but the fluctuation of the central spin due to the reaction coming from the random sitting of neighbors may have a finite value even at $T=0$. In other words, it seems that the effect of dilution on the Weiss field is to increase the importance of reaction field. Thus the correlated effective parameter may increase upon decreasing the concentration of magnetic atoms. Especially near the critical concentration, the correlated effective parameter has a large value and behaves almost independent of temperature below $T_c(P)$. This phenomenon may have some relationship to the problem of spin-glasses; the spin-glass phase may be characterized by a reaction field, which can be larger than the cavity field, as proposed by Cyrot.¹⁸

Above the transition temperature, on the other hand, the correlated effective parameter decreases monotonically with increasing the temperature, as shown in Fig. 3. Replotting the values of λ at $T=T_c(P)$, it seems that the critical values λ_c for selected values of P just lie on a curve, as depicted by the solid line in Fig. 4. For the concentration near $P=1$, the temperature dependence of λ above T_c follows the same curve, within numerical errors, but near the critical concentration the dependence shows a clear difference, as depicted by the dotted line in the figure.

The temperature dependence of the inverse initial sus-

FIG. 3. Temperature dependences of λ for selected values of $P.$

ceptibility χ^{-1} is shown in Fig. 5 for selected values of P. Only for the pure $(P = 1)$ system does the initial susceptibility vanish in the limit of $T\rightarrow 0$. For the concentration of $P_c < P < 1$, the susceptibility diverges twice, once at the critical point (infinite cluster contribution) and again at $T=0$ (finite cluster contribution). Thus we observe the coexistence of ^a Curie-Weiss —type law and ^a Curie-type law within one formalism. In order to observe the behavior of χ above the transition temperature clearly, the inverse paramagnetic susceptibility is depicted in Fig. 6 for selected values of P, which is obtained by solving Eqs. (33) and (36). Near the critical temperatures, the results of χ_{para}^{-1} have all downward curvatures. A characteristic behavior is that the deviation from the Curie-Weiss law is observed in a wider region than that of the pure $(P=1)$ system, upon decreasing the concentration of magnetic atoms. In view of the result, we evaluate the effective exponent $\gamma(T)$ of the paramagnetic susceptibility defined by

$$
\gamma(T) = (T - T_c) \chi_{\text{para}} \frac{d\chi_{\text{para}}^{-1}}{dT} , \qquad (37)
$$

which was first introduced by Kouvel and Fisher.¹⁹ By the use of Eqs. (33), (35), and (36), we can solve Eq. (37) numerically. The results for selected values of P are depicted in Fig. 7. At $T = T_c$, the results all reduce to the value of unity, since our approach is essentially a molecular field approximation. For higher temperatures, the ef-

FIG. 4. Concentration dependence of λ_c . The solid and dashed lines are the temperature dependences of λ above the transition temperatures for $P = 1$ and $P = 0.6$, respectively.

FIG. 5. Thermal dependence of the inverse initial susceptibility for typical values of P .

fective exponent also approaches gradually to the value of unity. The effective exponent shows a maximum at a temperature T_{max} . A characteristic feature of the result is that both the maximum value and the T_{max} increase with decreasing concentration of magnetic atoms. For crystalline systems, however, it is well known that the effective exponent decreases monotonically with increasing temperature. Accordingly, the result for the $P=1$ system cannot have any physical meaning, especially for the "real" critical range, since our theory is a molecular field approximation. In amorphous and dilute ferromagnets, on the other hand, the effective exponent runs through a maximum and the real critical range is expected to become narrower than that for the pure system.²⁰ Thus our result may have some relation to the experimental results.

V. CONCLUSION

We have discussed the dilute spin- $\frac{1}{2}$ Ising ferromagne in a square lattice. Within the correlated effective field theory, we calculated the most relevant thermodynamical quantities, namely critical temperature, critical concentration, magnetization, correlated effective parameter, and initial susceptibility. Some interesting effects of dilution come up in the thermal behaviors, especially for the corre-

FIG. 6. Thermal dependence of the inverse paramagnetic susceptibility for selected values of P .

FIG. 7. Temperature dependence of $\gamma(T)$ for selected values of P.

lated effective parameter and susceptibility. The susceptibility shows the effect of the eventual coexistence, in the system, of an infinite cluster with finite ones. The correlated effective parameter exhibits some characteristic behaviors for the concentration less than unity. The result may be attributed to the increasing importance of the reaction field in the Weiss field, on decreasing the concentration of magnetic atoms. It is important to remark here that the characteristic behavior (shown in Fig. 3) of the correlated effective parameter below the transition temperature is not explicitly reflected in the observable thermodynamical quantities. In Sec. III, the correlated effective parameter was defined separately in the regions of $T \leq T_c(P)$ and $T_c(P) \leq T$. Although not explicitly stated there, it is also important to remark that the parameters so determined take the same value at $T=T_c(P)$.

Finally, we have studied for simplicity the physical properties of the dilute Ising ferromagnet in a square lattice. Of course, the formulation discussed in Sec. II can be applied to other dimensional dilute ferromagnetic Ising systems. However, the framework suffers from faults characteristic of any effective-field theory, particularly in the critical regions and depending only on z, although it does provide a straightforward method of determining the thermodynamical quantities over the entire temperature range.

APPENDIX

The coefficients
$$
K_i
$$
 ($i = 1-5$) and K'_i ($i = 1-5$) in Eqs. (10) and (15) are given as follows:
\n
$$
K_1 = \begin{pmatrix} 4 \\ 1 \end{pmatrix} P(1-P)^3 \sinh(DJ) [\tanh(\beta x)]_{x=0} + 2 \begin{pmatrix} 4 \\ 2 \end{pmatrix} P^2(1-P)^2 \sinh(DJ) \cosh(DJ) [\tanh(\beta x)]_{x=0} \n+3 \begin{pmatrix} 4 \\ 3 \end{pmatrix} P^3(1-P) \cosh^2(DJ) \sinh(DJ) [\tanh(\beta x)]_{x=0} + 4 \begin{pmatrix} 4 \\ 4 \end{pmatrix} P^4 \sinh(DJ) \cosh^3(DJ) [\tanh(\beta x)]_{x=0},
$$
\n
$$
K_2 = 4 \begin{pmatrix} 4 \\ 4 \end{pmatrix} P^4 \cosh(DJ) \sinh^3(DJ) [\tanh(\beta x)]_{x=0} + \begin{pmatrix} 4 \\ 3 \end{pmatrix} P^3(1-P) \sinh^3(DJ) [\tanh(\beta x)]_{x=0},
$$
\n
$$
K_3 = \begin{pmatrix} 4 \\ 1 \end{pmatrix} P(1-P)^3 \cosh(DJ) [\operatorname{sech}^2(\beta x)]_{x=0} + \begin{pmatrix} 4 \\ 2 \end{pmatrix} P^2(1-P)^2 \cosh^2(DJ) [\operatorname{sech}^2(\beta x)]_{x=0} \n+ \begin{pmatrix} 4 \\ 3 \end{pmatrix} P^3(1-P) \cosh^3(DJ) [\operatorname{sech}^2(\beta x)]_{x=0} + \begin{pmatrix} 4 \\ 4 \end{pmatrix} P^4 \cosh^4(DJ) [\operatorname{sech}^2(\beta x)]_{x=0},
$$
\n
$$
K_4 = \begin{pmatrix} 4 \\ 2 \end{pmatrix} P^2(1-P)^2 \sinh^2(DJ) [\operatorname{sech}^2(\beta x)]_{x=0} + 3 \begin{pmatrix} 4 \\ 3 \end{pmatrix} P^3(1-P) \sinh^2(DJ) \cosh(DJ) [\operatorname{sech}^2(\beta x)]_{x=0}
\n+ 6 \begin{pmatrix} 4 \\ 4 \end{pmatrix} P^4 \cosh^2(DJ) \sinh^2(DJ) [\operatorname{sech}
$$

and

$$
K'_{1} = 2\begin{bmatrix} 2 \\ 0 \end{bmatrix} (1-P)^{2} \sinh(DJ) \cosh(DJ) [\tanh(\beta x)]_{x=0} + 2\begin{bmatrix} 2 \\ 1 \end{bmatrix} P(1-P) \cosh^{2}(DJ) \sinh(DJ) [\tanh(\beta x)]_{x=0} + \begin{bmatrix} 2 \\ 1 \end{bmatrix} P(1-P) \sinh^{3}(DJ) [\tanh(\beta x)]_{x=0} + 2\begin{bmatrix} 2 \\ 2 \end{bmatrix} P^{2} \cosh^{3}(DJ) \sinh(DJ) [\tanh(\beta x)]_{x=0} + 2\begin{bmatrix} 2 \\ 2 \end{bmatrix} P^{2} \sinh^{3}(DJ) \cosh(DJ) [\tanh(\beta x)]_{x=0} ,
$$

$$
K'_{2} = \begin{bmatrix} 2 \\ 1 \end{bmatrix} P(1-P) \cosh^{2}(DJ) \sinh(DJ) [\tanh(\beta x)]_{x=0} + 2\begin{bmatrix} 2 \\ 2 \end{bmatrix} P^{2} \cosh^{3}(DJ) \sinh(DJ) [\tanh(\beta x)]_{x=0} + 2\begin{bmatrix} 2 \\ 2 \end{bmatrix} P^{2} \sinh^{3}(DJ) \cosh(DJ) [\tanh(\beta x)]_{x=0} ,
$$

$$
K'_{3} = \begin{bmatrix} 2 \\ 0 \end{bmatrix} (1-P)^{2} \sinh^{2}(DJ) [\operatorname{sech}^{2}(\beta x)]_{x=0} + \begin{bmatrix} 2 \\ 1 \end{bmatrix} P(1-P) \sinh^{2}(DJ) \cosh(DJ) [\operatorname{sech}^{2}(\beta x)]_{x=0} + \begin{bmatrix} 2 \\ 2 \end{bmatrix} P^{2} \sinh^{2}(DJ) \cosh^{2}(DJ) [\operatorname{sech}^{2}(\beta x)]_{x=0} ,
$$

$$
K_4' = \binom{2}{0} (1 - P)^2 \cosh^2(DJ) [\operatorname{sech}^2(\beta x)]_{x=0} + \binom{2}{1} P(1 - P) \cosh^3(DJ) [\operatorname{sech}^2(\beta x)]_{x=0}
$$

$$
+2\begin{bmatrix}2\\1\end{bmatrix}P(1-P)\cosh(DJ)\sinh^{2}(DJ)[\mathrm{sech}^{2}(\beta x)]_{x=0}+\begin{bmatrix}2\\2\end{bmatrix}P^{2}\cosh^{4}(DJ)[\mathrm{sech}^{2}(\beta x)]_{x=0}
$$

$$
+4\begin{bmatrix}2\\2\end{bmatrix}P^{2}\sinh^{2}(DJ)\cosh^{2}(DJ)[\mathrm{sech}^{2}(\beta x)]_{x=0}+\begin{bmatrix}2\\2\end{bmatrix}P^{2}\sinh^{4}(DJ)[\mathrm{sech}^{2}(\beta x)]_{x=0},
$$

$$
K_{5}'=\begin{bmatrix}2\\2\end{bmatrix}P^{2}\cosh^{2}(DJ)\sinh^{2}(DJ)[\mathrm{sech}^{2}(\beta x)]_{x=0}.
$$

The coefficients can easily be calculated by applying a mathematical relation, $e^{aD}f(x)=f(x+\alpha)$. For instance, the coefficients, K_1, K_2, K'_1 , and K'_2 , are given by

$$
K_1=4P(1-P)^3\tanh(t)+6P^2(1-P)^2\tanh(2t)+3P^3(1-P)[\tanh(3t)+\tanh(t)]+\frac{1}{2}P^4[\tanh(4t)+2\tanh(2t)]
$$
,

$$
K_2 = \frac{1}{2}P^4[\tanh(4t) - 2\tanh(2t)] + P^3(1-P)[\tanh(3t) - 3\tanh(t)],
$$

$$
K_1' = (1 - P)^2 \tanh(2t) + \frac{1}{2} P(1 - P) [3 \tanh(3t) - \tanh(t)] + \frac{1}{2} P^2 \tanh(4t) ,
$$

$$
K_2' = \frac{1}{2} P(1-P) [\tanh(3t) + \tanh(t)] + \frac{1}{2} P^2 \tanh(4t).
$$

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