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Effect of three-body interactions on the ordering of bcc binary alloys

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The composition dependence of the order–disorder critical temperature is investigated for a model binary alloy equivalent to an Ising system with both the nearest-neighbor interactions (J) and the three-particle interactions (P) present. Using a real-space renormalization-group method for calculating the phase diagram, we find that for small three-body forces, the maximum critical temperature shifts from the 50-50 atomic percent composition proportionally to P/J. The proportionality constant is determined both from the renormalization-group method and from Griffith's smoothness postulate. The results of the two calculations agree with each other, and they are used to estimate the relative strength of the three-body potentials in iron cobalt. The estimate $P/J = 0.11 \pm 0.06$ indicates that three-body forces are small, so that the nearest-neighbor Ising model is a good first approximation for the description of ordering in FeCo.

I. INTRODUCTION

Studies of the Ising model have contributed much to our understanding of second-order phase transitions. This model is thought to give a realistic description for a variety of systems, the simplest among them being the binary alloys which undergo critical order–disorder transformations. As far as the critical properties such as exponents and amplitude ratios are concerned, experiments on β brass,[1–3] Fe_3Al,[4,5] and FeCo[6,7] confirm the nearest-neighbor (NN) Ising model predictions. The measured quantities are universal, however; so the agreement between theory and experiment means only that the order parameter of binary alloys in one dimensional and the interactions are of short range.

Of course, the NN Ising model is only a first approximation, and to find out the details of the interactions which are responsible for the phase transformation, one must look at the nonuniversal aspects of the ordering. An example of nonuniversal quantities is the ordering temperature. Its composition and pressure dependence has been investigated extensively for β brass,[8–10] and the findings do not agree with the corresponding NN Ising-model predictions.[11] This discrepancy led to the conclusion that the next-nearest-neighbor interactions play an important role in the ordering process.[9]

Although this conclusion is probably correct, the quantitative analysis of β brass is quite difficult. In order to compare the measurements with the rigid Ising-model calculations, one must correct for the thermal expansion of the lattice. This correction turns out to be significant but quite uncertain since its calculation involves the use of unverified assumptions about the composition and temperature dependence of the thermal expansion coefficient. Another source of inaccuracy is that the ordering curve exists only in a narrow composition range (44–49 at.% Zn) where no maximum critical temperature (T_0) is observed. So, the analysis of the data is necessarily biased by the usual assumption that T_0 is at the stoichiometric composition. Since the results are sensitive to the possible asymmetry of the ordering curve, the present-day experiments allow only qualitative conclusions about interactions other than the NN ones.

The situation in iron-cobalt alloys is much clearer since ordering occurs over a wide composition range[7,12] (30–70 at.% Co), and the correction for thermal expansion seems to be negligible.[13] The phase diagram (Fig. 1) is obviously not symmetric about the stoichiometric composition. Within the framework of Ising-model calculations, this fact can only be explained by taking into account the interatomic potentials among odd numbers of atoms. Another solution would be to make the NN interactions concentration dependent, but if the dependence is analytic this approach is equivalent to the introduction of a series of special, concentration-independent multisite interactions.

The asymmetry of the phase diagram is relatively small; it can be characterized by the composition n_0 ($n \equiv \frac{1}{100}$ at.% Co) at which the maximum critical temperature T_0 is attained. The shift of n_0 from the stoichiometric composition,

$$\delta n = n_0 - 0.5 \quad , \tag{1}$$

will be called the asymmetry parameter and in the following we show how this shift can be explained in terms of the simplest multisite interactions, namely the three-body ones. We obtain thereby an estimate

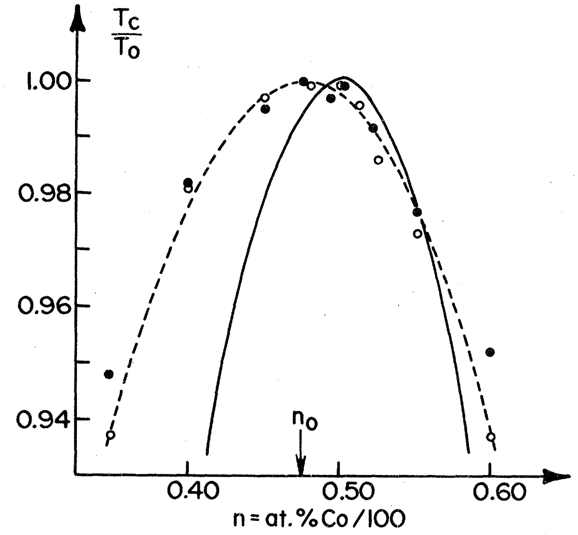

FIG. 1. Order–disorder transition temperature of FeCo as a function of composition. The ordering curve is normalized by the maximum critical temperature T_0. The solid and open circles are, respectively, the data of Oyedele and Collins (Ref. 7) and Asano *et al.* (Ref. 12). The nearest-neighbor Ising-model result of Bienenstock and Lewis is shown by the solid line, while the dashed curve is a parabolic fit to the experimental points.

of the strength of the three-body potentials in iron cobalt.

The effects of many-body interactions in alloys have been reviewed by Clapp,[14] and a partial updating of that paper can be found in Ref. 15. Their effects on phase diagrams were studied first by van Baal[16] who used a sophisticated molecular-field approximation for calculating the ordering properties of face-centered cubic (fcc) alloys of the CuAu and Cu_3Au type. He concluded that most of the variations in the actual phase diagrams could be explained by the inclusion of quite weak three- and four-particle interactions.

In contrast with fcc alloys in which the ordering takes place by a first-order phase transition, body-centered cubic (bcc) alloys like CuZn and FeCo undergo critical order–disorder phase transformations.[17,18] Since critical fluctuations are known to invalidate the quantitative predictions of the molecular-field theory, one must use better calculational schemes for bcc alloys. In this paper we proceed in two ways: first (Sec. II), observing that the asymmetry of the phase diagram in Fig. 1 is small, we assume the three-body interactions to be small, and treat them as perturbations to the NN interactions. Since three-body forces do not change the nature of the symmetry breaking in the ordering process, one can use Griffith's smoothness postulate,[19] and it follows that the calculation of the asymmetry parameter is reduced to that of the critical correlations of the NN Ising model, which can be handled by decoupling schemes and by using high-temperature series expansions.

Second, in Sec. III we carry out a real-space renormalization-group calculation for finding the asymmetry parameter when three-body interactions are present. Although the problem of complicated interactions arises naturally in the renormalization-group framework, the phase diagram of binary alloys has been investigated by this method only for the simple case of NN interactions.[20,21] An exception is the work of Schick *et al.*[22] in which the plane-triangular lattice with NN and three-particle interactions was studied, and the results were compared with the experiments on the ordering of helium on graphite. Even in this case, however, the three-particle interactions were used only for calculational purposes; they were not needed for explaining the experimental data.

In our renormalization-group calculation we found that for small three-particle interactions the upper part of the phase diagram shifts proportionally to the strength of those interactions, thus justifying the introduction of the asymmetry parameter. The proportionality constant is close to that determined from the smoothness postulate; so we feel that the estimate of the three-body potentials in FeCo following from these calculations (Sec. IV) is quite reliable.

II. SMOOTHNESS POSTULATE

Using spin terminology, a nonstoichiometric binary alloy is described as an Ising antiferromagnet in a magnetic field.[23] The addition of three-body potentials leads to the following Hamiltonian:

$$H=-H\sum_{i}\sigma_i+J\sum_{\langle ij\rangle}\sigma_i\sigma_j-P\sum_{\langle ijk\rangle}\sigma_i\sigma_j\sigma_k\ , \quad (2)$$

where $\sigma_i=\pm 1$; H, J, and P are, respectively, the magnetic field, the NN, and the three-spin coupling constants. Throughout this paper we shall be concerned mainly with the bcc lattice, but our calculation yields results for the simple cubic (sc) and square (sq) lattices, too. In all cases the summation $\langle ijk\rangle$ runs over the smallest triangles consisting of two NN and one next-nearest-neighbor bonds. Although the terminology of NN and three-spin interactions is used, it must be remembered that the parameters J and P are actually linear combinations of atomic potentials[23]; so they characterize the contribution of NN and three-body potentials to the *ordering* energy of the alloy. The claim of determining the magnitude of three-body potentials in fact means the estimation of their contribution to the ordering energy without actually being able to calculate energies for individual three-body configurations.

Given the Hamiltonian (2), in principle one should calculate the free energy and from its singularity determine the critical temperature $T_c(H,J,P)$. Then the maximum critical temperature T_0 is found at fixed J and P and the magnetization M_0 at the maximum gives the shift of T_0 from the stoichiometric composition by

$$\delta n = \tfrac{1}{2} M_0 \ . \tag{3}$$

For small P, one excepts δn to be proportional to P. Since the only other disposable dimensional parameter is J, for small P we have

$$\delta n \approx -r\frac{P}{J} \ , \tag{4}$$

where r is a dimensionless parameter. Our aim in this section is to determine r. Then, knowing δn from the experiments, the ratio of the strength of the three-body and the pair potentials can be found.

In practice, the above program cannot be carried out exactly. In three dimensions not even the solution of the NN Ising model is known, although in the last two decades much information has been gathered through approximate methods like high-temperature series.[23] We shall make connection with this information through the application of Griffith's smoothness postulate[19] to the parameters H and P, thereby reducing the calculation of r to that of the susceptibilities of the NN model.

The smoothness postulate has been designed to deal with parameters which do not alter the dimensionality, the range of interactions, and the nature of the symmetry breaking in a system. In renormalization-group language these parameters are called irrelevant. A well-known example is the magnetic field H at the antiferromagnetic critical point. The effect of three-spin interactions is quite similar to that of the magnetic field: they try to increase the magnetization, and they do not favor any of the sublattices of the antiferromagnet. Furthermore, in the renormalization-group calculations of Sec. III, both H and P turn out to be irrelevant parameters. So we shall assume that P also qualifies for the smoothness postulate, which implies that the free energy of the system can be expressed as

$$F(T,H,P) = F_0(T,H,P) + \phi(H,P) f(\theta(H,P)[T - T_c(H,P)]) \ , \tag{5}$$

where F_0, ϕ, θ, and T_c are smooth functions of their respective arguments (the dependence on J is implicit; the temperature is scaled by it), and the singular part of F is proportional to a function of single variable $f(x)$. The phase boundary is given by $T_c = T_c(H,P)$ with $T_N = T(0,0)$ being the Néel temperature. The functions ϕ and θ are normalized at T_N, i.e., $\phi(0,0) = \theta(0,0) = 1$.

It follows from the form of the Hamiltonian (2) that the free energy does not change if the sign of both H and P is reversed. Consequently, for small H and P,

$$T_c(H,P) = T_N + T_c^{(1)} PH + \tfrac{1}{2} T_c^{(2)} H^2 + \cdots \ , \tag{6}$$

with $T_c^{(1)} = (\partial^2 T_c / \partial H \partial P)_N$ and $T_c^{(2)} = (\partial^2 T_c / \partial H^2)_N$, the subscript N indicating that the derivatives are taken at the Néel point with all the other parameters fixed. At fixed P the maximum critical temperature T_0 is attained at

$$H_0 = \frac{T_c^{(1)}}{T_c^{(2)}} P \ , \tag{7}$$

and the ratio $T_c^{(1)}/T_c^{(2)}$ can be found by calculating the singular part of the temperature derivative of the susceptibility $\chi = (\partial M/\partial H)_{H=0,P=0}$ and the "three-point" susceptibility $\chi_3 = (\partial M/\partial P)_{H=0,P=0}$. From Eq. (5) it follows immediately that:

$$\chi' = \frac{d}{dT}\chi\Big|_{\text{sing}} = T_c^{(2)} \frac{d^2}{dT^2} f(T - T_N) \tag{8}$$

and

$$\chi_3' = \frac{d\chi_3}{dT}\Bigg|_{\text{sing}} = T_c^{(1)} \frac{d^2}{dT^2} f(T - T_N) \ ; \tag{9}$$

so the ratio of the amplitudes of the singularities of χ_3' and χ' is $T_c^{(1)}/T_c^{(2)}$.

Since P, and consequently H_0, are small, the magnetization M_0 at (H_0,P) can be obtained as

$$M_0 = \chi_{3N} P + \chi_N H_0 \ , \tag{10}$$

where χ_{3N} and χ_N are, respectively, the values of χ_3 and χ at T_N. Substituting Eqs. (7)–(9) into Eq. (10) we arrive at

$$M_0 = \left(\chi_{3N} - \frac{\chi_3'}{\chi'} \chi_N \right) P \ . \tag{11}$$

This formula contains no other approximation than the smoothness postulate, which is unproven but almost certainly correct. Unfortunately, only χ_N and χ' are known reliably from high-temperature series.[23] For χ_3 we must design an approximation which yields the required proportionality of the singular parts of χ and χ_3, Eqs. (8) and (9). This can be done by writing χ_3 as a sum of four-point correlation functions

$$T\chi_3 = \sum_i \sum_{a,b} \langle \sigma_i \sigma_0 \sigma_a \sigma_b \rangle \ , \tag{12}$$

where i goes over all the lattice sites while in the second sum σ_a and σ_b are next-nearest neighbors to each other and nearest neighbors to the spin at the origin σ_0. Decoupling the four-point correlations into the sum of all possible pair-correlation products, we find

$$T\chi_3 = c(2\langle\sigma_0\sigma_a\rangle + \langle\sigma_a\sigma_b\rangle)\sum_i \langle\sigma_0\sigma_i\rangle \ , \qquad (13)$$

where c is a geometrical factor giving the number of smallest triangles belonging to one point; its values for different lattices are listed in Table I. The NN correlation $\Gamma_1 = \langle\sigma_0\sigma_a\rangle$, the next-nearest-neighbor correlation $\Gamma_2 = \langle\sigma_a\sigma_b\rangle$, and the susceptibility $T\chi = \sum_i \langle\sigma_0\sigma_i\rangle$ have common singular structure, since they are all finite at T_N, and their temperature derivative has the same singularity. If follows then from Eq. (13) that, as required, the singular structures of χ_3 and χ are the same. An additional benefit of the above decoupling scheme is that it correctly reproduces the leading term in the high-temperature series of χ_3.

Denoting the singular part of $d\Gamma_1/dT$ and $d\Gamma_2/dT$ by Γ_1' and Γ_2' and substituting Eq. (13) into Eq. (11), we obtain

$$M_0 = \frac{c(2\Gamma_1' + \Gamma_2')\chi_N^2}{\chi'} P \ . \qquad (14)$$

In the case of the square lattice, Γ_1' and Γ_2' are related by $\Gamma_1' = -\rho\Gamma_2'$ with $\rho = \sqrt{2}$. A similar situation exists in sc lattices where high-temperature series[25] indicate $\rho = 1.42$. We shall arbitrarily adopt $\rho = 1.4$ for all the lattices, although ρ might have a geometrical meaning (the ratio of the distances between the next-nearest neighbors and the nearest neighbors), and then for the bcc lattice we should use $\rho = 2/\sqrt{3} \approx 1.2$. This difference is, however, not significant, especially in view of the decoupling approximation which probably introduces more uncertainty in our final result.

Since $\Gamma_1' = 2C_s/zJ$, with C_s being the singular part of the heat capacity and z the number of nearest neighbors, Eq. (14) simplifies to

$$M_0 = -\frac{2c(2-\rho)C_s\chi_N^2}{z\chi'} \frac{P}{J} \ . \qquad (15)$$

TABLE I. Values of the parameters for calculating r in Eq. (17). The meaning of these parameters is described in the text in connection with formulas (13)–(16). r_{RG} is the renormalization-group result of Sec. III.

	bcc	sc	sq
z	8	6	4
c	12	12	4
ρ	1.4(1.2?)	1.4	1.4
q	4.3	4.7	18
r	0.21	0.26	0.033
r_{RG}	0.25	0.33	0.17

Further simplification occurs by noting that, for the case of the NN Ising antiferromagnet in a magnetic field, the smoothness postulate predicts a parabolic phase boundary near T_N (Refs. 19 and 24)

$$\frac{T_c}{T_N} = 1 - \tfrac{1}{2}qM^2 + \cdots \ , \qquad (16)$$

with $q = \chi'/C_s\chi_N^2$. Using q in Eq. (15) and remembering how M_0 is related to the shift in composition, we arrive at

$$\delta n = -\frac{c(2-\rho)}{zq}\frac{P}{J} = -r\frac{P}{J} \ . \qquad (17)$$

Here c and z are geometrical factors while ρ and q are found from high-temperature series. The results for each lattice are presented numerically in Table I.

The above calculation suffers from the approximation introduced for calculating the four-point correlations. In order to assess the accuracy of our results, in Sec. III we determine r by using a completely different calculational scheme, namely, the real-space renormalization group.

III. RENORMALIZATION-GROUP METHOD

The calculation of phase boundaries by the real-space renormalization-group[26] method is now well established. The lattice is divided into cells and, by deriving an effective interaction between the cells, a transformation is generated in the space of coupling constants of the Hamiltonian. From the fixed points and flow properties of this renormalization-group transformation, one determines the critical surfaces which are directly related to the phase boundaries. The various renormalization-group schemes are different in their choice of cells and in the approximation which is used to obtain the effective interaction between the cells.

Apart from computational conveniences, the choice of cells is governed by the requirement that the renormalization-group transformation should leave invariant the ground states associated with the competing phases.[27] In our case there is competition between the antiferromagnetic phase, favored by the NN interaction, and the ferromagnetic phase, which is favored by the magnetic field and the three-spin interaction. In order to leave both the ferro- and antiferromagnetic ground-state invariant, we divide the lattices into two sublattices and for each sublattice a square (for sq lattice) or a tetrahedral (for sc and bcc lattices) arrangement is chosen as a basic cell (Fig. 2). This type of cell arrangement has been successfully used for investigating the ordering of NN antiferromagnets in a magnetic field.[20,21]

Using the above choice of cells, one is faced with the difficulty that, under the renormalization-group

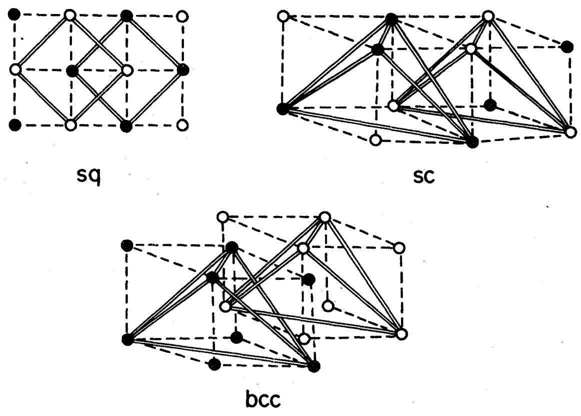

FIG. 2. Sublattice cell arrangements for sq, sc, and bcc lattices.

transformation, spatially inhomogeneous couplings are generated.[28] Kinzel[29] has devised an approximation for calculating the interaction between the cells which avoids the problem of inhomogeneity. In his method the interactions ($\mathcal{H}$) are separated into intracell ($\mathcal{H}_0$) and intercell (V) parts. The intercell part is not considered to be small as in the cumulant expansion[26]; instead it is treated in the mean-field approximation ($V \rightarrow V_{\mathrm{MF}}$), and the effective Hamiltonian of the cell spins (σ_i') is calculated from

$$\mathcal{H}'(\sigma') = \ln \sum_{\{\sigma\}} T(\sigma', \sigma) \exp(\mathcal{H}_0 + V_{\mathrm{MF}}) \ , \qquad (18)$$

where $T(\sigma', \sigma)$ is the cell-spin weight factor (we shall use the Niemeijer–van Leeuwen majority rule[26,29]) and the spin expectation values in the mean-field form of V are calculated self-consistently

$$\langle \sigma_i \rangle_{\sigma'} = \sum_{\{\sigma\}} T(\sigma', \sigma) \sigma_i \times \exp(H_0 + V_{\mathrm{MF}}) / \exp[\mathcal{H}'(\sigma')] \ . \qquad (19)$$

At this point the problem is still very complicated. Equation (19) should be solved for all the cell-spin configurations. Substituting the results into Eq. (18), one expects that all possible interactions are generated in $\mathcal{H}'(\sigma')$. The crucial assumption to be made here is that only a few of those interactions matter, and they can be obtained from a few cell-spin configurations. For instance, if only the NN interactions are kept, Eqs. (18) and (19) are calculated for the ferromagnetic (F) and antiferromagnetic (AF) cell-spin configurations, and the transformation is defined as $K' = [\mathcal{H}'(\mathrm{F}) - \mathcal{H}'(\mathrm{AF})]/zN'$ with $K' = -J'/kT$ and N' being the number of cell spins. Now the calculation is greatly simplified because the high symmetry of the F and AF configurations reduces Eq. (19) to the solution of two independent transcendental equations and the construction of those equations involves the spins in one cell only. With this remarkably simple approximation, Kinzel obtained reasonable results (10–20% accuracy) for the critical properties of the two-, three-, and four-dimensional Ising models and for the phase boundary of the two-dimensional Ising antiferromagnet in a magnetic field.

In the choice of the cell-spin configurations which are to be kept for evaluating Eqs. (18) and (19), one is guided by the symmetries and ground-state properties of the system. In our case the renormalization-group transformation is in the space of the coupling constants $K = -J/kT$, $\tilde{H} = -H/kT$, and $\tilde{P} = -P/kT$, and the basic symmetry in this space is the invariance of the renormalization-group flow under the mapping $(K, \tilde{H}, \tilde{P}) \leftrightarrow (K, -\tilde{H}, -\tilde{P})$ which follows from the reversal of all spins under the trace of the partition function. The competing ground states of the system are the ferro- and antiferromagnetic configurations but, because of the three-spin interactions, the ferromagnetic configurations with all spins up or down should be distinguished. In order to incorporate the above properties and have a renormalization-group transformation which does not enlarge the space of coupling constants, we shall use the following cell-spin configurations: antiferromagnetic (AF), ferromagnetic with all cell spins up ($\mathrm{F}\!\uparrow\!+$), ferromagnetic with the cell spins down ($\mathrm{F}\!\downarrow\!+$), the same ferromagnetic configurations in a magnetic field of the same magnitude but pointing in the opposite direction ($\mathrm{F}\!\uparrow\!-$) and ($\mathrm{F}\!\downarrow\!-$). Then the renormalization-group equations are as follows:

$$\begin{aligned} K' &= [\mathcal{H}'(\mathrm{F}\!\uparrow\!+) + \mathcal{H}'(\mathrm{F}\!\downarrow\!+) + \mathcal{H}'(\mathrm{F}\!\uparrow\!-) \\ &\quad + \mathcal{H}'(\mathrm{F}\!\downarrow\!-) - 4\mathcal{H}'(\mathrm{AF})]/4zN' \ , \\ \tilde{H}' &= [\mathcal{H}'(\mathrm{F}\!\uparrow\!+) - \mathcal{H}'(\mathrm{F}\!\downarrow\!+) \\ &\quad + \mathcal{H}'(\mathrm{F}\!\downarrow\!-) - \mathcal{H}'(\mathrm{F}\!\uparrow\!-)]/4N' \ , \qquad (20) \\ \tilde{P}' &= [\mathcal{H}'(\mathrm{F}\!\uparrow\!+) - \mathcal{H}'(\mathrm{F}\!\downarrow\!-) \\ &\quad + \mathcal{H}'(\mathrm{F}\!\uparrow\!-) - \mathcal{H}'(\mathrm{F}\!\downarrow\!+)]/4cN' \ , \end{aligned}$$

where z and c are given in Table I and the number of cell spins N' enters Eq. (20) only formally, since the calculation of $\mathcal{H}'$ reduces to the calculation of independent cells and so all $\mathcal{H}'$ are proportional to N'.

Having set up the recursion relations (20), one next finds the fixed points of the transformation. As expected, there exists a ferro- and an antiferromagnetic fixed point ($K^* = \pm K_c$, $\tilde{P}^* = 0$, and $\tilde{H}^* = 0$), and the fields $\tilde{H}$ and $\tilde{P}$, which break the up–down symmetry, are relevant at the ferromagnetic fixed point while they are irrelevant at the antiferromagnetic one. This result supports the smoothness postulate explored in Sec. II, although it should not be taken as a proof, since in several real-space renormalization-group calculations $\tilde{H}$ turned out to be relevant at the antiferromagnetic fixed point.[20,21,30]

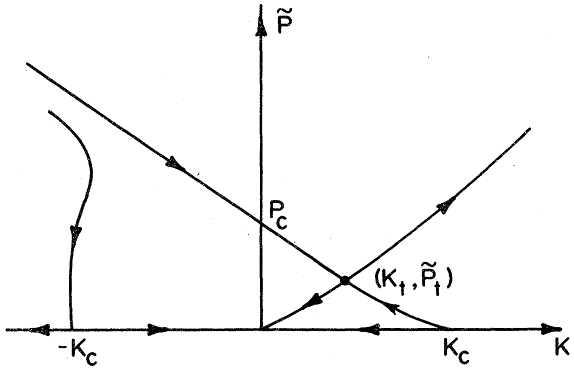

FIG. 3. Renormalization-group flow in the $(K,\tilde{P},\tilde{H}=0)$ plane. Only the $\tilde{P}>0$ part is shown, since the picture is symmetric about the K axis.

Other fixed points of interest are located at $(K^*=K_t,\ P^*=\pm\tilde{P}_t$, and $H^*=0)$. As can be seen from Fig. 3, they govern the phase transition in the model with pure three-spin interactions. This model has been investigated[31] by deriving low-temperature series for the magnetization and analyzing it under the assumption that the phase transition is connected with the disappearance of the magnetization. The nature of the transition is not clear, however, since it has been proved that in the bcc lattice the magnetization persists up to arbitrary high temperatures.[32] (The proof can easily be extended to the sc and sq lattices, too.) Our simple approximation indicates that, although the magnetization exists at any temperature, its fluctuations diverge at the critical point $\tilde{P}=P_c$, i.e., the magnetization versus $\tilde{P}$ curve has an inflection point at P_c with $dM/d\tilde{P}$ being infinite. This picture might be simpler than the real situation, since the above calculational scheme can treat only ferro- and antiferromagnetic orderings; it would be nevertheless interesting to reanalyze the series along the lines of the above findings.

In the following we shall restrict ourselves to the neighborhood of the Néel point, where an idea about the accuracy of our results can be obtained by comparing our results for K_c and the thermal (y_T) and "sublattice" magnetic (y_SH) exponents with the available exact and high-temperature-series[23] results (Table II). The discrepancies are of the order of 20%, indicating that the method compares with the first-order cumulant expansion and with the simple-cluster[26] and sublattice[20] approximations.

The critical surface near the Néel point is traced out by watching the renormalization-group flow. Since both $\tilde{H}$ and $\tilde{P}$ are irrelevant variables, one simply finds the points which are taken by the renormalization-group transformation to the antiferromagnetic fixed point. The only complication is that the critical surface must be found accurately since we want T_c as a function of M, and typically the magnetization varies rapidly at the phase boundary. The magnetization itself is determined as the derivative of the free energy $M=\partial F/\partial\tilde{H}$, F being calculated as a sum along the renormalization path[33] $F=\sum g_n/4^n$. In our case the contribution in the nth step is

$$g_n=\tfrac{1}{32}[\mathcal{H}_n'(\mathrm{F}\uparrow+)+\mathcal{H}_n'(\mathrm{F}\downarrow+)+\mathcal{H}_n'(\mathrm{F}\uparrow-)+\mathcal{H}_n'(\mathrm{F}\downarrow-)+4\mathcal{H}_n'(\mathrm{AF})]\ , \tag{21}$$

where $\mathcal{H}_n'$ are the $\mathcal{H}'$ of Eq. (18) evaluated in the nth step of the transformation. We note that one can also derive recursion relations directly for the magnetization. Usually they converge rapidly and give better accuracy; however, these recursion relations are too lengthy to write out here.

Having determined the magnetization, and consequently the composition on the critical surface, one can draw the phase boundaries at fixed $\tilde{P}/K=P/J$ ratios. We find that for small P/J ($\lesssim 0.5$), the upper part of the phase boundary practically retains its parabolic shape, but it shifts in composition proportionally to P/J. The proportionality constant r_RG gives the shift of the maximum critical temperature; its value for different lattices is listed in Table I. One can see that for the bcc and sc lattices the agreement between the smoothness postulate and renormalization-group

TABLE II. Critical parameters at the antiferromagnetic fixed point. RG refers to our renormalization-group results, while "exact" labels the values obtained from high-temperature series or exact calculations (Ref. 23).

	K_c		y_T		y_SH	
	RG	"exact"	RG	"exact"	RG	"exact"
bcc	0.205	0.157	1.22	1.6	2.80	2.5
sc	0.273	0.222				
sq	0.410	0.441	0.816	1.0	1.87	1.875

calculations is good. For the sq lattice, however, there is a factor of five difference between the two results, indicating that one or both of the calculational schemes break down. This could be due to the larger fluctuations in two dimensions which might invalidate the decoupling scheme employed in Sec. II. Another possibility, however, is that the quite irregular variation of M on the phase boundary observed for the sq lattice in the series analysis[11] is a real feature. The different renormalization-group approximations[21] do not seem to be able to reproduce this detail properly.

IV. COMPARISON WITH THE EXPERIMENT ON FeCo AND CONCLUDING REMARKS

For the three-dimensional lattices the results of the above two calculations are consistent with each other; so we feel that they can be used for estimating P/J in FeCo. To do this, let us look at Fig. 4, which shows the phase boundaries for the bcc case obtained from the high-temperature series, from our renormalization-group calculation, and from a parabola fit to the experimental points. One can see that the shift in composition ($\delta n \approx -0.025$) of the maximum critical temperature T_0 can be explained by adding three-body interactions of relative strength

$$\frac{P}{J} \approx 0.10 \quad , \tag{22}$$

or if we use the value of r obtained from the smoothness postulate,

$$\frac{P}{J} \approx 0.12 \quad . \tag{23}$$

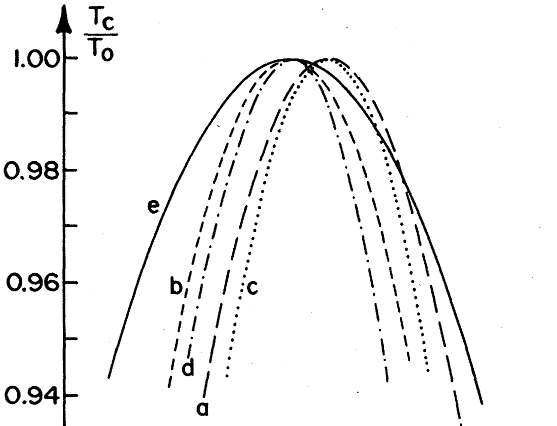

FIG. 4. Composition dependence of the ordering temperature of a bcc binary alloy. The following curves are drawn: (a) High-temperature series for $P/J=0$ (————); (b) the same curve but shifted by $\delta n = -0.025$ (----); renormalization-group results for (c) $P/J=0$ (·····) and for (d) $P/J=0.1$ (— — — — — —); and (e) parabola fit to the experimental results FeCo (———).

These numbers support the view that the NN Ising model is a good first approximation for describing the ordering. But in order to believe in them, it remains to be explained why the lines (d) and (e) have different curvatures at their common maximum.

The discrepancy is only partly due to the approximation used. Figure 4 shows that some of the discrepancy would remain even if a method would reproduce the presumably accurate high-temperature-series curve (a) for $P/J=0$, and then this curve would be shifted into (b). For small P/J one cannot expect that during the shift the shape of the curve would change significantly; it follows from the symmetry of the critical surface discussed in Sec. III that the parameter q characterizing the parabolic phase boundary in the neighborhood of T_0 (Sec. II) changes from its NN Ising value proportionally to $(P/J)^2$.

The parameter q can be effectively changed by next-nearest-neighbor ($J_{(2)}$), four-spin (J_4), or in general by some even-spin interactions (J_{2k}). For small J_{2k}/J the change in q is proportional to J_{2k}/J, but the shift of T_0 is effected only in second order ($\sim PJ_{2k}/J^2$). Thus to first order the effects of P and J_{2k} are independent and Eqs. (22) and (23) are estimates of the three-body interactions under the assumption that small additional even-spin interactions take care of the discrepancy between the theoretical and experimental curves in Fig. 4.

An idea about the relative strength of the even-spin interactions can be obtained from examining the effect of next-nearest-neighbor interactions. Harbus and Stanley[34] used high-temperature series to determine the phase boundary for $J_{(2)}/J = -0.5$. Although the calculation is for sc lattice, the result is probably also representative for the bcc case, since for the NN Ising model the phase boundaries for the two lattices are practically identical.[11] For the NN Ising model $q \approx 4.7$ (Table I), while in the case of $J_{(2)}/J = -0.5$ we deduce $q = 2.4$ from the high-temperature series.[34] The latter value is close to the experimentally observed $q \approx 2.0$; so $J_{(2)}/J = -0.5$ is about the relative strength of the next-nearest-neighbor interaction needed to explain the discrepancy between the theoretical and experimental curves in Fig. 4. The above argument puts an error bar on our results [Eqs. (22) and (23)]: since the second-order contribution to the shift of T_0 would be of the order $|PJ_{(2)}/J^2| \approx 0.05$, our final estimate of P/J is

$$\frac{P}{J} = 0.11 \pm 0.06 \quad . \tag{24}$$

An independent estimate of P/J could be obtained by measuring the composition and temperature dependence of the short-range order in Fe_xCo_{1-x} well above the ordering temperature. This measurement would be important because it could decide the correctness of the very recent idea[35] that the asymmetry of the ordering curve of Fe_xCo_{1-x} is caused by magnetic interactions. If this is the right description, then one expects that at high temperatures ($T \approx 1250\,^\circ K$) where the magnetic phase transition takes place, the asymmetry in the composition dependence of the short-range correlations should change quite rapidly. If, on the other hand, the asymmetry is caused by three-body potentials, one should expect smooth variation through the magnetic phase boundary.

If the three-body description turns out to be correct, the short-range-order measurements will probably yield an estimate of P/J which is more accurate than Eq. (24). This is because the experimental results can be compared with the high-temperature-series estimates of the short-range correlations. At high temperatures a few terms of the series should be sufficient for an accurate description, and they can easily be derived even with the many-particle interactions present.[36] A project along these lines has recently been carried out for Cu_xNi_{1-x} systems. Vrijen *et al.*[37] found strong asymmetry in the composition dependence of the short-range order and their preliminary calculation indicates that any quantitative comparison with the Ising model must involve at least the NN, next-nearest-neighbor, and three-particle interactions.

One can see that the explanation of relatively simple details of the ordering phenomena requires distant-neighbor and multisite interactions to be included in the Ising model. Their strength is small as compared to the NN interactions, but nevertheless they make the calculations tedious, and one might wonder if instead of the phenomenological parametrization of the problem, the same effort could yield a microscopic calculation, i.e., a calculation based on the electronic properties of the alloys.

An interesting work in this direction has been undertaken by Kittler and Falicov.[38] They calculated the short- and long-range-order dependence of the alloy internal energy from a tight-binding Hartree Hamiltonian by using an extended cluster-Bethe-lattice method. The configurational entropy was determined by applying the cluster-variation method and then the thermodynamic properties were obtained by minimizing the free energy with respect to the order parameters. In this way Kittler and Falicov found ordering temperatures of CuAu, Cu_3Au, and $CuAu_3$ alloys that were in reasonable agreement with experiments. The fact that they did not use adjustable parameters shows that their method is powerful. It has, however, a shortcoming: it is basically a mean-field theory, so any attempt to apply it to critical order–disorder transformations would encounter the difficultly that the critical exponents would not agree with the experiments.

It would be most interesting, but perhaps extremely difficult, to combine the above electronic theory approach with the renormalization-group ideas. Until this problem can be solved, the Ising model remains the best description of critical ordering transformations. A problem that remains for the future is the development of renormalization-group schemes which are easily applicable to the three-dimensional case and give more reliable results than those given by the present methods.

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