

Bragg–Williams Model of CsCl-Type Ordering of the FeCo System in Strong Magnetic Fields

P.R. OHODNICKI Jr., Y. HANLUMYUANG, D.E. LAUGHLIN, and M.E. MCHENRY

A modified Bragg–Williams (B–W) model of α and α' FeCo is extended to estimate the effect of strong magnetic fields on the critical ordering temperature (T_{ORDER}) taking into account long-range chemical and magnetic ordering. The model discussed here is generalized from our previous work in which only the larger average exchange per atom in the chemically ordered state was taken into account. A positive shift of critical temperatures for the higher order $\alpha \rightarrow \alpha'$ order-disorder phase transformation has been predicted in the presence of a strong field. In this work, the experimentally observed dependence of the average magnetic moment of Fe atoms on the degree of chemical order has been accounted for explicitly. The estimated shift in the critical ordering temperature (ΔT_{ORDER}) is larger when the dependence of the Fe moment on the degree of chemical order is taken into account, particularly in the case of Fe-rich compositions (e.g., $\Delta T_{\text{ORDER}} \sim 13$ K vs $\Delta T_{\text{ORDER}} \sim 10$ K for $H \sim 50$ T at equiatomic composition). For most compositions, however, the contribution to ΔT_{ORDER} associated with the larger average exchange per atom in the chemically ordered state accounts for the majority of the shift. The estimated effect remains quite small and is only expected to be experimentally observable in static fields larger than currently available in most laboratories (ΔT_{ORDER} is only predicted to be larger than ~ 2 to 3 K for $H > \sim 10$ T).

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I. INTRODUCTION

THE FeCo-based alloys are technologically important materials, particularly for use in soft magnetic applications that require high saturation inductions as well as high temperature operation. The largest room temperature saturation induction observed to date ($B_s \sim 2.5$ T) is documented for bulk FeCo-based binary alloys. The Curie temperature of the low temperature disordered bcc α -phase is sufficiently high so that ferromagnetic behavior persists to temperatures approaching 1000 °C.^[1] At equiatomic composition, the extrapolated Curie temperature of the α -phase is significantly higher than the temperature above which ferromagnetic behavior is no longer observed.^[2] The origin of the abrupt loss of ferromagnetism is actually the structural phase transformation from chemically disordered ferromagnetic bcc α -FeCo to the high temperature paramagnetic fcc γ -phase. If the low temperature bcc phase could be stabilized relative to the γ -phase, ferromagnetism would persist to even higher temperatures.

In the FeCo system, the chemical order-disorder phase transformation is a higher order transition in which the equilibrium value of the chemical order parameter falls continuously to zero at T_{ORDER} with

increasing temperature. The Bragg–Williams (B–W) model employed here was originally used to describe the FeCo order-disorder transformation in Reference 3, taking into account nearest neighbor chemical ($V \sim 820$ K) and magnetic exchange interactions ($J_{\text{FeFe}} \sim 265$ K, $J_{\text{CoCo}} \sim 360$ K, and $J_{\text{FeCo}} \sim 435$ K) independent of both composition and temperature. A cooperative interaction between chemical and magnetic ordering was observed. A further generalization of this model described in our previous work used the same parameters and assumptions (but with $V \sim 812$ K rather than $V \sim 820$ K)^[4] by considering the influence of an external field on the order-disorder behavior through an additional Zeeman energy term. It was observed that applied magnetic fields tend to stabilize the chemically ordered phase (α' , B2) relative to the disordered phase (bcc, α , A2). However, it was found that this effect was negligible for fields smaller than $H \sim 10$ T at which a shift of approximately 2 to 3 K would be expected. For $H \sim 100$ T, a shift of T_{ORDER} of approximately 18 K was predicted, although complications such as short-range order, magnetostriction, an increase in the magnitude of the atomic moments in the presence of strong magnetic fields, and longer range chemical and magnetic interactions were neglected.^[5,6] In the current work, a further extension of the model is discussed. Here, we explicitly account for the dependence of the magnitude of the magnetic moment of Fe atoms on the degree of chemical order (i.e., the number of Co nearest neighbors) that has been observed experimentally.^[2,7]

II. MODEL

As defined by Eguchi *et al.*,^[3] the probability of occupation of α and β sites by Fe and Co atoms and the

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distribution of the orientation of atomic magnetic moments can be represented in the combined nearest neighbor B–W and Ising model approximation using a total of five long-range order parameters: a chemical order parameter (x) and four magnetic order parameters describing the degree of order in the spin system (p , q , r , and s). The expressions for the occupation probabilities originally presented in Reference 3 for an $\text{Fe}_{1-\varepsilon}\text{Co}_{1+\varepsilon}$ alloy are reproduced in Table I. The maximum degree of chemical order (x_{MAX}) depends on the composition, decreasing monotonically from a value of 1 at the equiatomic composition. It can be shown that $x_{\text{MAX}} = 1 - \varepsilon$ for $\varepsilon > 0$ (Co-rich alloys) or $x_{\text{MAX}} = 1 + \varepsilon$ for $\varepsilon < 0$ (Fe-rich alloys).

The internal energy (E), magnetization (M), Zeeman free energy contribution (G_Z), and configurational entropy (S) of the system calculated using the Stirling approximation^[4] are expressed in terms of the order parameters through the following equations:

$$M = \frac{N}{4}(\mu_{\text{Fe}}(p+r) + \mu_{\text{Co}}(q+s)) \quad [1]$$

$$\begin{aligned} E &= \sum_{ij} \left[-V_{ij}N_{ij} + \frac{J_{ij}}{2}(N_{ij}^{\uparrow\downarrow} + N_{ij}^{\downarrow\uparrow}) - \frac{J_{ij}}{2}(N_{ij}^{\uparrow\uparrow} + N_{ij}^{\downarrow\downarrow}) \right] \Big|_{x,p,q,r,s} \\ &- \sum_{ij} \left[-V_{ij}N_{ij} + \frac{J_{ij}}{2}(N_{ij}^{\uparrow\downarrow} + N_{ij}^{\downarrow\uparrow}) - \frac{J_{ij}}{2}(N_{ij}^{\uparrow\uparrow} + N_{ij}^{\downarrow\downarrow}) \right] \Big|_{x,p,q,r,s=0} \\ &= -\frac{NZ_{\text{AVG}}}{16}(Vx^2 + J_{\text{FeFe}}pr + J_{\text{FeCo}}(pq+rs) + J_{\text{CoCo}}qs) \end{aligned} \quad [2]$$

$$G_Z = -HM = -\frac{NH}{4}(\mu_{\text{Fe}}(p+r) + \mu_{\text{Co}}(q+s)) \quad [3]$$

$$S = -\frac{Nk}{8} \left[\begin{aligned} &(1-\varepsilon+x+p) \ln(1-\varepsilon+x+p) + (1-\varepsilon+x-p) \ln(1-\varepsilon+x-p) + \\ &(1+\varepsilon+x+q) \ln(1+\varepsilon+x+q) + (1+\varepsilon+x-q) \ln(1+\varepsilon+x-q) + \\ &(1-\varepsilon-x+r) \ln(1-\varepsilon-x+r) + (1-\varepsilon-x-r) \ln(1-\varepsilon-x-r) + \\ &(1+\varepsilon-x+s) \ln(1+\varepsilon-x+s) + (1+\varepsilon-x-s) \ln(1+\varepsilon-x-s) \end{aligned} \right] \quad [4]$$

where E , S , and G_Z are calculated with respect to the chemically and magnetically fully disordered state ($x = p = q = r = s = 0$). In the preceding expressions, N is the number of atoms per unit volume, Z_{AVG} is the coordination number (8 for bcc and CsCl type), μ_{Fe} and μ_{Co} are the atomic moments of the respective

elements, and k is the Boltzmann's constant. In contrast with the expressions used to find the characteristic equations in Reference 4, μ_{Fe} is now allowed to vary with composition and degree of order. By minimizing the overall free energy of the system ($G = E + PV - TS - HM$) with respect to the order parameters at a given composition and temperature (fixed ε and T), one can determine the equilibrium state as a function of temperature, composition, and applied magnetic field. We have assumed that both pressure and volume remain fixed for simplicity.

The dependence of μ_{Fe} on the number of Co nearest neighbors in $\alpha\text{-FeCo}$, reported on both theoretically^[2] and experimentally,^[7] is taken into account in the preceding Zeeman energy term. The value of μ_{Co} has been shown to be relatively independent of the local environment of the Co atoms, and hence it is taken to be fixed at $1.8 \mu_{\text{B}}$. In the B–W approximation, the atomic moment of Fe (μ_{Fe}) is expressed as a function of the composition and the long-range chemical order parameter ($\mu_{\text{Fe}}(\varepsilon, x)$). At the equiatomic composition, for $x = 1$, each Fe atom would be surrounded by eight Co atoms, while for $x = 0$, the number of Co nearest neighbors would be only four on average. For the fully chemically disordered state ($x = 0$), the average value of μ_{Fe} as a function of composition was estimated from the data in Reference 7 under the assumption that each Fe atom has a nearest neighbor coordination identical to the average. The difference between the average μ_{Fe} for maximum chemical order and the fully chemically disordered states ($\Delta\mu_{\text{Fe}}(\varepsilon) = \mu_{\text{Fe}}(\varepsilon, x_{\text{MAX}}) - \mu_{\text{Fe}}(\varepsilon, 0)$) was estimated using the same data set under the same assumptions. The estimated value of $\Delta\mu_{\text{Fe}}$ used here is plotted as a function of composition in Figure 1.

Because the minimization of the preceding free energy expressions requires a continuous expression for $\Delta\mu_{\text{Fe}}$ at intermediate values of chemical order, the average Fe moment was assumed to vary with the square of the chemical order parameter as in Reference 10:

$$\mu_{\text{Fe}}(\varepsilon, x) = \mu_{\text{Fe}}(\varepsilon, 0) + \frac{x^2}{(x_{\text{MAX}})^2} \Delta\mu_{\text{Fe}}(\varepsilon, x) \quad [5]$$

Here, x_{MAX} is the maximum degree of long-range chemical order at a given composition, as defined

Table I. Distribution of A and B Atoms on α and β Sites and Orientation of Moments as Calculated in the Combined B–W and Ising Model Approximation¹⁹

| Sites | Atoms | | | | Sum |
|----------|----------------------------------|----------------------------------|----------------------------------|----------------------------------|---------------|
| | A ⁺ | A ⁻ | B ⁺ | B ⁻ | |
| α | $\frac{N}{8}(1+x+p-\varepsilon)$ | $\frac{N}{8}(1+x-p-\varepsilon)$ | $\frac{N}{8}(1-x+s+\varepsilon)$ | $\frac{N}{8}(1-x-s+\varepsilon)$ | $\frac{N}{2}$ |
| β | $\frac{N}{8}(1-x+r-\varepsilon)$ | $\frac{N}{8}(1-x-r-\varepsilon)$ | $\frac{N}{8}(1+x+q+\varepsilon)$ | $\frac{N}{8}(1+x-q+\varepsilon)$ | $\frac{N}{2}$ |
| Sum | $\frac{N}{2}$ | | $\frac{N}{2}$ | | N |

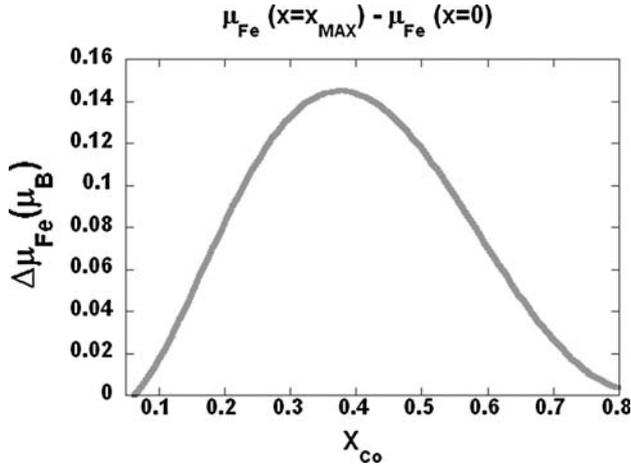


Fig. 1—Estimated value of $\Delta\mu_{\text{Fe}}(X_{\text{Co}}) = \mu_{\text{Fe}}(X_{\text{Co}}, X_{\text{MAX}}) - \mu_{\text{Fe}}(X_{\text{Co}}, 0)$ based on the moment vs composition data for α -FeCo found in Ref. 7. X_{Co} is the atomic fraction of cobalt in the FeCo alloy.

previously. The chosen form of Eq. [5] is based on the assumption that the average atomic moment of Fe, μ_{Fe} , scales linearly with the average number of Co nearest neighbors per Fe atom in the B–W approximation. Because the number of Co nearest neighbors is proportional to the total number of Fe-Co pairs, this quantity is expected to vary with the square of the order parameter, as can be determined from the occupation probabilities of Table I. Recent experimental results confirm the proportionality between the average atomic moment per atom and the square of the order parameter in Fe-Co alloys.^[12]

Now that the model and its assumptions have been described in detail, the characteristic equations calculated by minimizing the free energy of the system with respect to each of the order parameters are presented:

$$\left(1 + \frac{4(p+r)H\Delta\mu_{\text{Fe}}}{VZ_{\text{AVG}}(x_{\text{MAX}})^2}\right)x - \frac{kT}{VZ_{\text{AVG}}} \ln \left(\frac{(1-\varepsilon+x+p)(1-\varepsilon+x-p)(1+\varepsilon+x+q)(1+\varepsilon+x-q)}{(1-\varepsilon-x+r)(1-\varepsilon-x-r)(1+\varepsilon-x+s)(1+\varepsilon-x-s)} \right) = 0 \quad [6]$$

$$\frac{H\mu_{\text{Co}}}{2} + \frac{Z_{\text{AVG}}}{8}(J_{\text{CoCo}}q + J_{\text{FeCo}}r) - \frac{kT}{4} \ln \left(\frac{1+\varepsilon-x+s}{1+\varepsilon-x-s} \right) = 0 \quad [7]$$

$$\frac{H\mu_{\text{Fe}}}{2} + \frac{Z_{\text{AVG}}}{8}(J_{\text{FeFe}}p + J_{\text{FeCo}}s) - \frac{kT}{4} \ln \left(\frac{1-\varepsilon-x+r}{1-\varepsilon-x-r} \right) = 0 \quad [8]$$

$$\frac{H\mu_{\text{Fe}}}{2} + \frac{Z_{\text{AVG}}}{8}(J_{\text{FeFe}}r + J_{\text{FeCo}}q) - \frac{kT}{4} \ln \left(\frac{1-\varepsilon+x+p}{1-\varepsilon+x-p} \right) = 0 \quad [9]$$

$$\frac{H\mu_{\text{Co}}}{2} + \frac{Z_{\text{AVG}}}{8}(J_{\text{CoCo}}s + J_{\text{FeCo}}p) - \frac{kT}{4} \ln \left(\frac{1+\varepsilon+x+q}{1+\varepsilon+x-q} \right) = 0 \quad [10]$$

III. MODEL RESULTS

Despite the additional complexity of the characteristic equations presented here, the solutions as a function of temperature are qualitatively the same as presented in Reference 4. The major difference observed after taking into account the variation of the Fe moment with respect to the degree of chemical order is an additional

contribution to the shift in the chemical ordering temperatures in the presence of applied fields.

In Figure 2, the calculated order parameters as a function of temperature are presented for equiatomic composition ($\epsilon = 0$), and the shift in the chemical ordering temperature (ΔT_{ORDER}) as a function of applied field first discussed in Reference 4 is illustrated (inset). Notice that the variation of ΔT_{ORDER} illustrated by the dashed line in the inset of Figure 2 corresponds to the assumption of an Fe moment independent of the degree of chemical order ($\Delta\mu_{\text{Fe}}(\epsilon, x) = 0$). This relationship is identical to that presented in Reference 4. The solid line corresponds to the value of ΔT_{ORDER} , taking into account the variation in μ_{Fe} with the degree of chemical order, and it can be seen to result in an additional contribution to the shift in the chemical ordering temperature in the presence of applied fields ($\Delta T_{\text{ORDER}} \sim 24$ K vs $\Delta T_{\text{ORDER}} \sim 18$ K for $H \sim 100$ T at equiatomic). Although the authors realize that a field of $H = 100$ T is unreasonably large compared to static fields currently available, this value is used in figures presented here so that predicted effects of the applied fields can be easily observed. It is interesting to note that although the contribution to ΔT_{ORDER} associated with the larger average exchange interaction in the ordered state relative to the disordered state (discussed previously in Reference 4) is relatively symmetric about the equiatomic composition, the contribution due to the larger average μ_{Fe} is more pronounced for Fe-rich alloys, as illustrated in Figure 3. For most compositions, it is observed that the larger average exchange interaction in the ordered state is estimated to be the primary mechanism responsible for the predicted shift.

The variation of the chemical and magnetic exchange interaction parameters (V , J_{FeFe} , J_{CoCo} , and J_{FeCo})

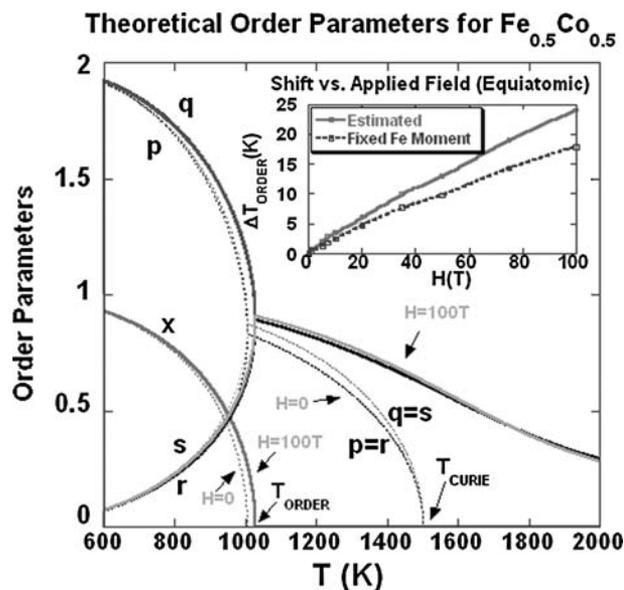


Fig. 2—Theoretical order parameters in zero applied field (dotted lines) and a field of $H = 100$ T (solid lines) for bulk ($Z_{\text{AVG}} = 8$) equiatomic FeCo. The inset compares ΔT_{ORDER} due to an applied field assuming a fixed Fe moment (dashed) as in Ref. 4 and a varying $\mu_{\text{Fe}}(\epsilon, x)$ (solid), as described in the text.

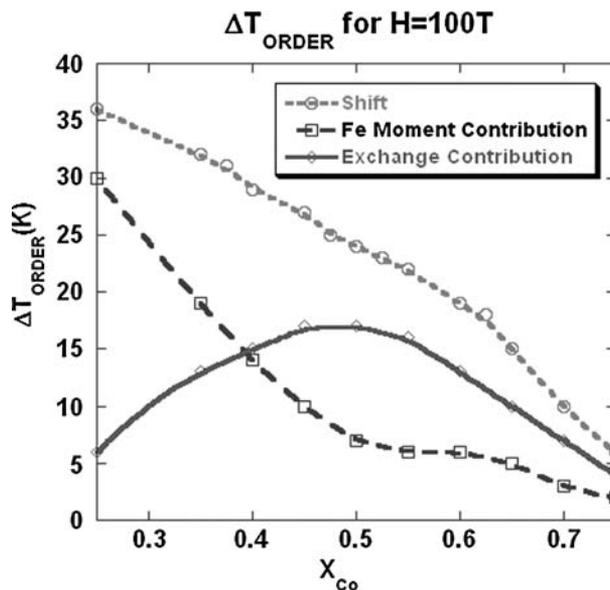


Fig. 3— ΔT_{ORDER} as well as the contributions due to the dependence of average exchange (T_{Curie}) and μ_{Fe} on the degree of chemical order.

between adjacent atoms with composition, temperature, and degree of order has been neglected. Taking into account such complications would affect the predicted values of T_{ORDER} even in the absence of the field.

The origin of the stabilization of chemical order in the presence of a field illustrated in Figures 2 and 3 is the larger reduction in the free energy of the chemically ordered phase in the presence of a magnetic field relative to the chemically disordered phase. Because of the larger average exchange per atom (hence, a higher calculated Curie temperature) and a larger average Fe moment of the chemically ordered phase, the magnetization of the chemically ordered phase is larger at a given temperature and applied magnetic field than that of the chemically disordered phase. As a result, the magnetization is expected to increase with the degree of chemical order. Hence, the application of a magnetic field is predicted to stabilize a greater degree of chemical order at temperatures below the zero-field T_{ORDER} and a nonzero degree of chemical order at slightly higher temperatures, resulting in the predicted ΔT_{ORDER} .

The argument outlined previously is illustrated by the magnetization and ΔG (with respect to full chemical and magnetic disorder $x = p = q = r = s = 0$) vs temperature curves plotted in Figures 4(a) and (b) for the chemically ordered phase ($x = 1$ and $p, q, r, \text{ and } s$ varying), the chemically disordered phase ($x = 0$ and $p, q, r, \text{ and } s$ varying), and the theoretically predicted equilibrium FeCo alloy at equiatomic composition ($x, p, q, r, \text{ and } s$ varying). As stated previously, in the vicinity of T_{ORDER} , the magnetization of the fully ordered state is larger than that of the disordered state. The free energy of the fully ordered state is therefore reduced by a larger amount than that of the disordered state due to the application of a strong magnetic field. It is also interesting to note that the contributions to the relative reduction in free energy of the ordered phase associated

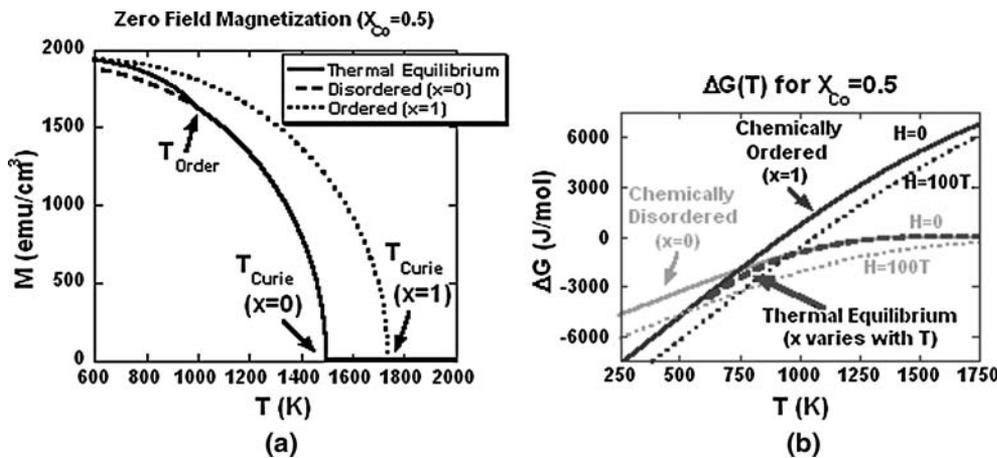


Fig. 4—(a) Zero field calculated M vs T curves for the fully chemically ordered state ($x = 1$), the fully chemically disordered state ($x = 0$), and the predicted thermal equilibrium state (x varying with T below T_{ORDER}). (b) Calculated ΔG vs T curves with the reference state corresponding to the fully chemically and magnetically disordered state ($x = p = q = r = s = 0$). Solid lines correspond to zero field free energy curves for fixed values of x ($x = 0$ and $x = 1$). Dotted lines correspond to the free energy curves in the presence of a uniform field of $H = 100$ T. The thick dashed line corresponds to thermal equilibrium in zero applied field.

with the average exchange interaction and the variation of μ_{Fe} with degree of order are approximately proportional to the calculated contributions to ΔT_{ORDER} presented previously.

Figure 4(a) illustrates the larger theoretical magnetization of the maximally chemically ordered phase relative to the chemically disordered phase arising from a larger average exchange interaction and a larger average Fe moment per atom. Experimentally, a sharp drop in the magnetization associated with the loss of ferromagnetism is observed due to the ferromagnetic bcc α -phase to paramagnetic fcc γ -phase transformation ($T \sim 1250$ K near equiatomic) before the Curie temperature of the disordered bcc phase is reached. As a result, an accurate measurement of the Curie temperature for both chemically disordered and chemically ordered Fe-Co alloys is not possible for most compositions. The values of T_{Curie} calculated by the model cannot be compared directly with experimental data. Although the Curie temperatures calculated here for equiatomic composition are significantly larger than those obtained through a fitting technique in Reference 7, they are comparable to the values obtained from a similar technique in Reference 8. Without accurate experimental data for comparison, errors in the values of the assumed exchange interactions may potentially affect the validity of the predictions of this model. Although quantitative predictions of the model are to be treated with caution, such errors are not expected to be so large that they affect the qualitative predictions.

Several features of the ΔG curves of Figure 4(b) are worth mentioning. The slopes of all of the ΔG curves are positive due to the fact that the reference state is chosen to be fully chemically and magnetically disordered ($x = p = q = r = s = 0$). In addition, the slopes decrease with increasing temperature as a result of the decrease in the order parameters. According to the thermodynamic relation $\partial G/\partial T = -S$ at constant pressure, the slope of the curves in Figure 4(b) correspond to the expression $\partial \Delta G/\partial T = -(S - S_{DIS})$, where S_{DIS}

corresponds to the entropy of the disordered reference state. Because the configurational entropy of the fully disordered state is assumed to be independent of temperature and a maximum for this system, $S - S_{DIS}$ is negative in the case where any degree of chemical or magnetic order is present ($x, p, q, r, \text{ or } s$ not equal to zero) and $S - S_{DIS}$ is zero when no order is present in the system ($x = p = q = r = s = 0$). These features are observed in Figure 4(b), because the ΔG curves of the fully chemically disordered and thermal equilibrium states have zero slope at temperatures above T_{Curie} of the disordered phase ($T_{Curie} > T_{ORDER}$) in zero applied field. It is observed that the slope of the ΔG curve of the chemically ordered phase is always larger than that of the chemically disordered phase. In addition, the slope of the ΔG curve for the alloy in thermal equilibrium flattens gradually as the critical ordering temperatures are approached (T_{ORDER} and T_{Curie}) due to the increase in configurational entropy and the increase in entropy in the magnetic spin system. The gradual change in slope is consistent with the higher-order nature of the phase transition that results in disordering over a range of temperatures below the critical temperature. Another consequence of the higher-order phase transition is that T_{ORDER} does not occur at the intersection of the ΔG curves of the fully chemically ordered and fully chemically disordered alloys. Instead, the continuous transition between the two states results in a value of T_{ORDER} at significantly higher temperatures.

The calculated zero field order-disorder phase boundary is plotted in Figure 5 along with the predicted phase boundary in the presence of a uniform field of $H = 50$ T and $H = 100$ T. This plot is similar to the one presented in Reference 4, but the shift in the critical ordering temperature is more pronounced for Fe-rich compositions because of the additional contribution due to the variation of the Fe moment with the degree of order. Even though short-range order effects have been neglected in the present work, this additional complication is not expected to significantly change the

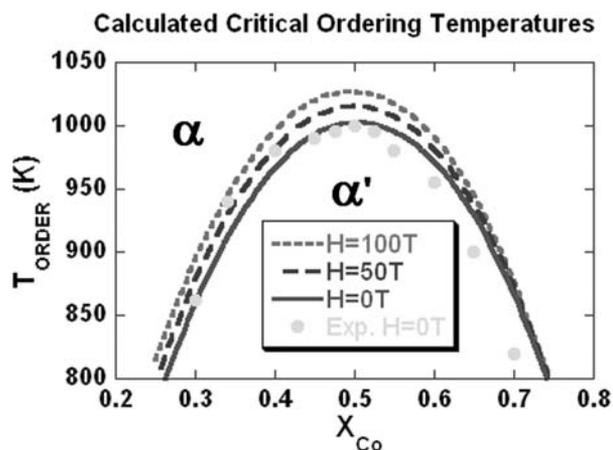


Fig. 5—Calculated order-disorder phase boundary in the zero field and a uniform field of $H = 50$ T and $H = 100$ T. Experimental data taken from Ref. 19.

qualitative conclusions of the present work. Because the value of T_{ORDER} is suppressed if short-range order effects are taken into account for the standard B–W model of chemical ordering in binary alloys where magnetic interactions are neglected for fixed values of the chemical interaction parameter V ,^[11] such effects are also expected to reduce the predicted values of ΔT_{ORDER} in the present model for fixed values of the chemical and magnetic interaction parameters. In addition, the authors of Reference 15 demonstrated that accounting for short-range correlations in the Bethe approximation can result in an asymmetric zero field order-disorder boundary, indicating that perhaps short-range order effects may result in an additional asymmetry in ΔT_{ORDER} as well. A detailed description of the potential effects of applied fields on the order-disorder boundary would take such short-range order effects into account. However, such an extension of the current model would require five additional short-range order parameters and a more complicated free energy minimization problem. Because of the small magnitude of the predicted ΔT_{ORDER} using the B–W model employed here and the difficulty of experimental verification, the extension of the current model to include the short-range ordering effects is unlikely to result in a dramatic modification of the main conclusions of this work. Further, it is well established that the B–W approximation provides a reasonable first approximation to the description of the chemical ordering behavior if magnetic interactions are neglected in CsCl-type binary ordering alloys.^[17]

A series of works aimed at investigating the interplay between chemical and magnetic ordering have been previously published in References 13 through 15. In References 13 and 14, the same B–W model as employed here was used. In Reference 14, it was applied to the FeCo system as originally done by Eguchi *et al.*^[9] In Reference 15, an extension was made to account for short-range chemical and magnetic ordering in the Bethe approximation, and the asymmetry in the experimental order-disorder phase boundary that is not accounted for using the B–W model was successfully reproduced.

Based on this result, the authors of Reference 15 concluded that short-range ordering effects are important to consider to properly model the order-disorder transformation in the Fe–Co alloys. However, the application of both the B–W model and the Bethe approximation to the FeCo system in References 14 and 15 used values of the magnetic exchange parameters (J_{FeFe} , J_{CoCo} , and J_{FeCo}) extracted from a compositional Curie temperature dependence of disordered bcc FeCo that is concluded to be less consistent with experiment by the authors of Reference 16 than the one employed by Eguchi in Reference 9 and reproduced here through the chosen values of the magnetic exchange parameters. This conclusion was based on careful extrapolation of magnetization vs temperature measurements as well as Mössbauer measurements. By fitting the assumed Curie temperature dependence of composition, the authors of References 14 and 15 obtained a value of $J_{CoCo} < J_{FeFe}$, while the authors of References 9 and 18, and recent band structure calculations,^[2] use a value of $J_{CoCo} > J_{FeFe}$ in agreement with the experimental compositional trend of T_{Curie} in Reference 16. In the case of the Bethe approximation of Reference 15, which includes short-range order effects, the authors claim that the peak in the order-disorder phase boundary on the Fe-rich side of the phase diagram is “driven by the magnetic interactions”. We postulate that the source of the asymmetry must come from the relative values of J_{FeFe} and J_{CoCo} . If this postulate is indeed correct, it is important to point out that if $J_{CoCo} > J_{FeFe}$ were used in the model of Reference 15, the peak in the order-disorder phase boundary would likely have occurred in the Co-rich region of the phase diagram, which would have resulted in poorer agreement with experiment. Because of the difficulty in experimentally measuring the compositional trend of T_{Curie} in the present alloy system, conclusions of phenomenological models that are highly sensitive to the values of the exchange interaction parameters must be regarded with caution.

Although the validity of the quantitative estimates presented here are uncertain without experimental verification due to the assumptions and simplifications described, the proposed model predicts the stabilization of α with respect to α' in an applied field as a result of both the larger exchange interaction between Fe and Co atoms and the dependence of μ_{Fe} on the degree of chemical order. This conclusion is expected to be robust even if short-range order effects are taken into account.

IV. CONCLUSIONS AND IMPLICATIONS

A modified B–W model of the FeCo order-disorder transformation taking into account nearest neighbor chemical and magnetic interactions has been further extended from its original form in References 3 and 9 and our first generalization in Reference 4. In addition to the larger average exchange interaction, it is estimated that the dependence of the average μ_{Fe} on the degree of chemical order also results in a theoretically predicted shift in the critical ordering temperature in strong applied magnetic fields, particularly for Fe-rich compositions.

The magnitude of the predicted ΔT_{ORDER} remains quite small even at very large fields making experimental verification of such an effect quite difficult. In this work, magnetostrictive effects, the dependence of atomic moments on the strength of the applied field, long-range interactions, and short-range order are some of the potential complicating factors that have been neglected. Experimental investigation would be required in order to demonstrate the validity of the simplifying assumptions made in arriving at the predictions outlined here.

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