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Magnetic-environment model of Ni-Pt and Ni-Pd alloys

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A model previously developed for alloys of nickel with nonmagnetic impurities, in which the magnetic moment of nickel depends on its magnetic and chemical environment, has been extended to the case of nickel alloys with impurities that acquire a magnetic moment such as Ni-Pd and Ni-Pt. The fitting of the data of average moments versus concentration in the Ni-Cu, Ni-Pd, and Ni-Pt alloys gives the parameters of the model which, without additional assumptions, describes the diffuse scattering neutron measurements that have been obtained for these alloys. With these parameters, we have also calculated the range parameters of the moment disturbances of the dilute alloys Fe-Pd and Fe-Pt. The model describes with a few parameters the magnetic behavior of Ni-based alloys and shows that only first-neighbor interactions are necessary to explain the long-range-moment disturbances measured with neutrons.

I. INTRODUCTION

The magnetic properties of Ni-based alloys have been widely discussed for a long time. Many years ago it was discovered that by alloying Ni and nonmagnetic metals the average moment is decreased proportionally to the impurity concentration[1] and this was considered as an important success of the rigid-band model.[2] However, theoretical considerations[3,4] showed that the charge transfers required by this theory could not be of the right magnitude. Moreover, neutron measurements[5–8] have shown large magnetic inhomogeneities in Ni-based alloys, implying a dependence of the magnetic moments on their local environment. Comly *et al.*[5] tried to explain these inhomogeneities in magnetization with a theory based on a nonlocal susceptibility. This theory, however, could not explain neutron and magnetization results for Ni-Cu and Ni-Zn. Later, Hicks[9] proposed a magnetic environment model for Ni-Cu near the critical concentration and Medina and Cable[10] developed a similar model for the whole ferromagnetic region. These models assumed that the moment on a Ni atom is a function not only of its chemical environment but also of the magnetic moments of the surrounding atoms. Their results explained the Ni-Cu neutron observations by means of two parameters, namely, a magnetic effect parameter Γ related to the range of the perturbations of the nickel magnetic moment and a chemical environment effect parameter ρ. Medina and Cable also applied this model successfully to other dilute alloys of Ni with nonmagnetic impurities (including Ni-Zn). In this paper we extend the magnetic environment model to the cases of Ni-Pd and Ni-Pt in which the impurities acquire induced moments and behave magnetically like Ni.[11,12]

II. MODEL

Assuming a random alloy, we take the moment at each magnetic site to be a function of the number of its impurity neighbors and of an exchange field produced by its nearest neighbors.[10] The moment at site $\vec{n}$ is then

$$\mu_{\vec{n}} = (1-p_{\vec{n}})F_i(H^h_{\vec{n}}, \nu_{\vec{n}}) + p_{\vec{n}}F_h(H^i_{\vec{n}}, \nu_{\vec{n}}) \ , \qquad (2.1)$$

where h and i denote host and impurity atoms, respectively, $\nu_{\vec{n}}$ is the number of impurity neighbors, $H_{\vec{n}}$ is the exchange field, and $p_{\vec{n}}$ is a site occupation operator which is unity if there is an impurity atom at $\vec{n}$ and is zero otherwise.

$$H^h_{\vec{n}} = \sum_{\vec{\delta}} [J_{hh}(1-p_{\vec{n}+\vec{\delta}}) + J_{ih}p_{\vec{n}+\vec{\delta}}]\mu_{\vec{n}+\vec{\delta}} \ , \qquad (2.2)$$

$$H^i_{\vec{n}} = \sum_{\vec{\delta}} [J_{ii}p_{\vec{n}+\vec{\delta}} + J_{ih}(1-p_{\vec{n}+\vec{\delta}})]\mu_{\vec{n}+\vec{\delta}} \ , \qquad (2.3)$$

$$\nu_{\vec{n}} = \sum_{\vec{\delta}} p_{\vec{n}+\vec{\delta}} \ . \qquad (2.4)$$

Here the sums are over the $\vec{\delta}$ nearest neighbors. In the subsequent treatment we will not determine the absolute values of the exchange constants J_{mn} but only their relative values, therefore it is convenient to define the following normalized fields:

$$h^h_{\vec{n}} = (Z_1 J_{hh})^{-1} H^h_{\vec{n}} = \frac{1}{Z_1} \sum_{\vec{\delta}} \mu_{\vec{n}+\vec{\delta}} (1 - p_{\vec{n}+\vec{\delta}} + \alpha p_{\vec{n}+\vec{\delta}}) \ , \tag{2.5}$$

$$h^i_{\vec{n}} = (Z_1 J_{ii})^{-1} H^i_{\vec{n}} = \frac{1}{Z_1} \sum_{\vec{\delta}} \mu_{\vec{n}+\vec{\delta}} [p_{\vec{n}+\vec{\delta}} + \beta (1 - p_{\vec{n}+\vec{\delta}})] \ , \tag{2.6}$$

where Z_1 is the coordination number and α and β are given by

$$\alpha = J_{ih}/J_{hh} \tag{2.7}$$

and

$$\beta = J_{ih}/J_{ii} \ . \tag{2.8}$$

The moment of an atom, therefore, depends on its chemical environment and, through an exchange field, on its magnetic environment.

The exact form of the response functions F_h and F_i is not important for most of the following treatment. At this stage we only assume that they are linear for small h and saturating for large h.

In order to solve the infinite set of coupled nonlinear equations given by Eq. (2.1) one must make some approximations. The first one we make is the linearization of the equations. If the moment fluctuations are small we can expand both response functions about an effective field h_{eff} and the average number of nearest impurity neighbors $Z_1 c$ where c is the impurity concentration

$$F(h_{\vec{n}}, \nu_{\vec{n}}) \approx F(h_{\text{eff}}, Z_1 c) + \frac{\partial F}{\partial h}(h_{\vec{n}} - h_{\text{eff}}) + \frac{\partial F}{\partial \nu}(\nu_{\vec{n}} - Z_1 c) \ . \tag{2.9}$$

The effective fields h^h_{eff} and h^i_{eff} are defined by the following equations:

$$\bar{\mu}_h = F_h(h^h_{\text{eff}}, Z_1 c) \tag{2.10}$$

and

$$\bar{\mu}_i = F_i(h^i_{\text{eff}}, Z_1 c) \ , \tag{2.11}$$

where $\bar{\mu}_h$ and $\bar{\mu}_i$ are the host and impurity average moments. We have assumed that the F's are smooth functions of ν so that we may expand about $Z_1 c$. The moment at site $\vec{n}$ is then given by the equation

$$\mu_{\vec{n}} = (1 - p_{\vec{n}}) \left[\bar{\mu}_h + \Gamma_h (h^h_{\vec{n}} - h^h_{\text{eff}}) + \Gamma_h \rho_h \sum_{\vec{\delta}} (p_{\vec{n}+\vec{\delta}} - c) \right] + p_{\vec{n}} \left[\bar{\mu}_i + \Gamma_i (h^i_{\vec{n}} - h^i_{\text{eff}}) + \Gamma_i \rho_i \sum_{\vec{\delta}} (p_{\vec{n}+\vec{\delta}} - c) \right] \ , \tag{2.12}$$

where

$$\Gamma_h = \frac{\partial F_h}{\partial h} \ , \tag{2.13}$$

$$\Gamma_i = \frac{\partial F_i}{\partial h} \ , \tag{2.14}$$

$$\rho_h = \left(\frac{\partial F_h}{\partial \nu} \right) \Big/ \left(\frac{\partial F_h}{\partial h} \right) \ , \tag{2.15}$$

and

$$\rho_i = \left(\frac{\partial F_i}{\partial \nu} \right) \Big/ \left(\frac{\partial F_i}{\partial h} \right) \ . \tag{2.16}$$

From Eqs. (2.10), (2.11), and (2.12), we obtain the following equations for the effective fields:

$$h^h_{\text{eff}} = \langle (1 - p_{\vec{0}}) h^h_{\vec{0}} \rangle / (1 - c) \tag{2.17}$$

and

$$h^i_{\text{eff}} = \langle p_{\vec{0}} h^i_{\vec{0}} \rangle / c \ . \tag{2.18}$$

To make the second approximation, we use Marshall's[13] expansion

$$\mu_{\vec{n}} = (1 - p_{\vec{n}}) \left[\bar{\mu}_h + \sum_{\vec{l}} g_{\vec{l}} (p_{\vec{n}+\vec{l}} - c) + \cdots \right] + p_{\vec{n}} \left[\bar{\mu}_i + \sum_{\vec{l}} h_{\vec{l}} (p_{\vec{n}+\vec{l}} - c) + \cdots \right] \ . \tag{2.19}$$

Here $g_{\vec{l}}$ and $h_{\vec{l}}$ are the impurity-induced moment disturbances at host sites and impurity sites, respectively, and the dots indicate nonlinear terms. The approximation consists of neglecting the nonlinear terms. The use of Marshall's expansion is also convenient because it is related to measurable quantities, namely the host and impurity average moments $\bar{\mu}_h$ and $\bar{\mu}_i$, and the weighted moment perturbations $\phi_{\vec{n}}$ given for $n \neq 0$ by

$$\phi_{\vec{n}} = (1 - c) g_{\vec{n}} + c h_{\vec{n}} \ . \tag{2.20}$$

The moment perturbations for $l \neq 0$ are given by the equations

$$c(1-c)^2 g_{\vec{l}} = \langle (p_{\vec{l}} - c)(1 - p_{\vec{0}}) \mu_{\vec{0}} \rangle \tag{2.21}$$

and

$$c^2 (1-c) h_{\vec{l}} = \langle (p_{\vec{l}} - c) p_{\vec{0}} \mu_{\vec{0}} \rangle \ ; \tag{2.22}$$

by definition $g_{\vec{0}} \equiv 0$ and $h_{\vec{0}} \equiv 0$.

We now substitute the moments that appear in the definition of the exchange fields, Eqs. (2.5) and (2.6), by Marshall's expansion [Eq. (2.19)] and use expressions (2.17) and (2.18) to obtain the effective fields

$$h_{\text{eff}}^{h}=(1-c)(\bar{\mu}_h-cg_1)+\alpha c(\bar{\mu}_i-ch_1) \;, \tag{2.23}$$

$$h_{\text{eff}}^{i}=c[\bar{\mu}_i+(1-c)h_1]+\beta(1-c)[\bar{\mu}_h+(1-c)g_1] \;, \tag{2.24}$$

where g_1 and h_1 are the nearest-neighbors moment disturbances.

The moment disturbances are obtained in a similar way. We again substitute the moments that appear in the definition of exchange field by Marshall's expansion and then we use Eqs. (2.12), (2.21), and (2.22). The equations so obtained are Fourier transformed giving two coupled linear equations

$$[1-\Gamma_h(1-c)F_1(\vec{k})]G(\vec{k})-c\alpha\Gamma_hF_1(\vec{k})H(\vec{k})$$
$$=\Gamma_h\{F_1(\vec{k})[-\bar{\mu}_h+cg_1+Z_1\rho_h+\alpha(\bar{\mu}_i-ch_i)]-(1-c)g_1-\alpha ch_1\} \tag{2.25}$$

and

$$-(1-c)\beta\Gamma_iF_1(\vec{k})G(\vec{k})+[1-c\Gamma_iF_1(\vec{k})]H(\vec{k})$$
$$=\Gamma_i(F_1(\vec{k})\{\bar{\mu}_i+(1-c)h_1+Z_1\rho_i-\beta[\bar{\mu}_h+(1-c)g_1]\}-ch_1-\beta(1-c)g_1) \;, \tag{2.26}$$

where the following definitions have been used:

$$F_1(\vec{K})=\frac{1}{Z_1}\sum_{\delta}e^{i\vec{k}\cdot\vec{\delta}} \;, \tag{2.27}$$

$$G(\vec{K})=\sum_{\vec{m}}e^{i\vec{k}\cdot\vec{m}}g_{\vec{m}} \;, \tag{2.28}$$

and

$$H(\vec{K})=\sum_{\vec{m}}e^{i\vec{k}\cdot\vec{m}}h_{\vec{m}} \;. \tag{2.29}$$

Solving the system of equations formed by Eqs. (2.25) and (2.26), a solution for $G(\vec{K})$ and $H(\vec{K})$ is obtained in terms of g_1 and h_1, which are unknown, but which can be determined by the following two additional conditions:

$$g_{\vec{0}}=\frac{1}{V_c^*}\int G(\vec{K})d^3\vec{K}=0 \tag{2.30}$$

and

$$h_{\vec{0}}=\frac{1}{V_c^*}\int H(\vec{K})d^3\vec{K}=0 \;, \tag{2.31}$$

where the integrals are over a reciprocal-lattice unit cell. Solving them, one obtains,

$$g_1=-a_1\bar{\mu}_h-a_2\alpha\bar{\mu}_i-a_3Z_1\rho_h-a_4\alpha Z_1\rho_i \tag{2.32}$$

and

$$h_1=-b_1\bar{\mu}_i-b_2\beta\bar{\mu}_h-b_3Z_1\rho_i-b_4\beta Z_1\rho_h \;. \tag{2.33}$$

The expressions of the c-dependent coefficients a_j and b_j are given in the Appendix.

$G(\vec{K})$ and $H(\vec{K})$ are related to the moment disturbance function measured by neutron scattering[13]

$$M(\vec{K})=\bar{\mu}_i-\bar{\mu}_h+(1-c)G(\vec{K})+cH(\vec{K}) \;. \tag{2.34}$$

The expression for $M(\vec{K})$ is of the following form:

$$M(\vec{K})=\Delta\bar{\mu}+\frac{U+VF_1(\vec{K})+WF_1(\vec{K})^2}{1-\Gamma F_1(\vec{K})+DF_1(\vec{K})^2} \;, \tag{2.35}$$

where

$$\Gamma=(1-c)\Gamma_h+c\Gamma_i \;, \tag{2.36}$$

$$D=c(1-c)\Gamma_i\Gamma_h(1-\alpha\beta) \;, \tag{2.37}$$

and U, V, and W are complicated functions of α, β, Γ_i, Γ_h, ρ_i, ρ_h, μ_i, μ_h, and c, and are given in the Appendix.

The average moments are functions of the effective fields [Eqs. (2.10) and (2.11)] which are themselves functions of the average moments [Eqs. (2.23), (2.24), (2.32), and (2.33)]. It is therefore possible to obtain the average moments for each concentration if we know the response functions F_h and F_i.

In the case where only the host is magnetic, as in Ni-Cu, Eq. (2.35) reduces to

$$M(\vec{K})=-\bar{\mu}_h-\Gamma\left\{\frac{(1-c)g_1+F_1(\vec{K})(\bar{\mu}_h-cg_1-Z_1\rho_h)}{1-\Gamma F_1(\vec{K})}\right\} \;. \tag{2.38}$$

A result obtained previously for the case of Ni-Cu (Ref. 10) had the same form except for the term cg_1. This discrepancy is due to a different approximation used in the previous treatment. This difference shows up as a small correction to the chemical parameters previously obtained. The formula for g_1, in the case where only the host is magnetic, becomes

$$g_1=\frac{-(\bar{\mu}_h-Z_1\rho_h)[B(\Gamma)-1]}{\Gamma(1-c)B(\Gamma)+c[B(\Gamma)-1]} \;, \tag{2.39}$$

where $B(\Gamma)$ has been defined in the Appendix.

It has been shown that $M(0)$ is related to the concentration derivative of the average moment,[14] namely,

$$M(0)=\frac{d\bar{\mu}}{dc} \; . \tag{2.40}$$

Due to our approximations the proposed method of calculation does not necessarily meet this condition. Thus Eq. (2.40) can be used as a test of the goodness of the approximations. In the case of Ni-Cu, Medina and Cable[10] considered that the form of $M(K)$ is valid even in the case where this condition is not met. The Γ_h parameter was then considered as a free parameter no longer satisfying Eq. (2.13) and to be determined by imposing the condition given by Eq. (2.40). Note that this procedure implies solving a differential equation. In the present application of the model to Ni-Cu we also apply this method, and for Ni-Pd and Ni-Pt a similar treatment will be used.

III. APPLICATION OF THE MODEL TO Ni-Cu, Ni-Pd, AND Ni-Pt ALLOYS

For Ni-Pd and Ni-Pt alloys the response functions F_h and F_i are not known, but since the experimentally obtained values of the moments of Pd and Pt are small[11,12] we make the approximation

$$F_i(h,\nu)=\Gamma_i h \; , \tag{3.1}$$

where we have neglected the chemical environment dependence and the saturation. The same approximations cannot be made for Ni since the saturation effect is important. Although a Ni response function obtained from the density of states has been proposed previously,[10] we will use the following simple and analytical expression, similar to the one used by Hicks[15]:

$$F_h(h,\nu)=\frac{A(\nu)h}{1+B(\nu)h} \; . \tag{3.2}$$

The parameter $A(\nu)$ is some kind of low-field susceptibility while $A(\nu)/B(\nu)$ is the high-field moment. In general, we have to include the atomic environment dependence of these two quantities. We do not know the exact form of the functions $A(\nu)$ and $B(\nu)$ but we can always write

$$A(\nu)=A\phi_1(\nu) \tag{3.3}$$

and

$$A(\nu)/B(\nu)=(A/B)\phi_2(\nu) \; . \tag{3.4}$$

The constants A and B are the pure-Ni values for $\phi_1(0)=\phi_2(0)=1$. From these expressions and Eq. (2.15), one obtains the chemical parameter,

$$\rho_h=h_{\text{eff}}^{h}[\phi_1'/\phi_1-\phi_2'/\phi_2]+(\bar{\mu}_h/\Gamma_h)\phi_2'/\phi_2 \; . \tag{3.5}$$

For simplicity it is convenient to use simple exponentials for ϕ_1 and ϕ_2

$$\phi_1(\nu)=e^{Q_1\nu} \tag{3.6}$$

and

$$\phi_2(\nu)=e^{Q_2\nu} \; . \tag{3.7}$$

With the expression for ρ_h and Eqs. (2.10), (2.11), (2.23), (2.24), (2.32), (2.33), (3.1), and (3.2), one is able to determine the average moments $\bar{\mu}_h$ and $\bar{\mu}_i$.

In order to obtain the constants A and B, we have applied the model to magnetization and neutron data of Ni-Cu alloys. In this case, where only the Ni is magnetic, the polarized neutron data[10] for $M(K)$ had already been fitted with a function of the same form as Eq. (2.36), but with a small difference as explained in Sec. II, and values for Γ and ρ were obtained for each concentration. Using the method previously explained, with $\Gamma_i=0$, and the response function of Eq. (3.2), we performed a simultaneous fitting of the magnetization data[7,16–20] and the $\Gamma-\rho$ data.[10] In this way, we detemined the parameters A, Q_1, and Q_2, and calculated the parameter B using $B=(A-1)/0.616$, which is obtained by applying Eq. (3.2) to pure Ni. Figures 1 and 2 show the average Ni moment data, the $\Gamma-\rho$ data, and their calculated values. The dashed curve in Fig. 2 shows the values of Γ calculated using Eq. (2.13) while the continuous curve shows the values of Γ obtained using condition (2.40) as explained in Sec. II. We see that the difference becomes important as the critical concentration is approached.

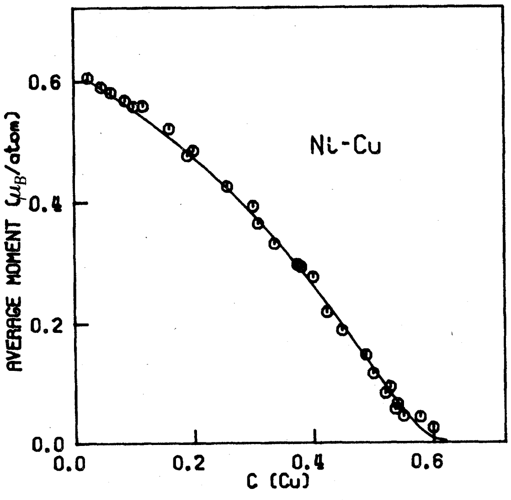

FIG. 1. Data and theoretical calculations of the Ni average moment. Experimental values are from Refs. 7 and 16–20.

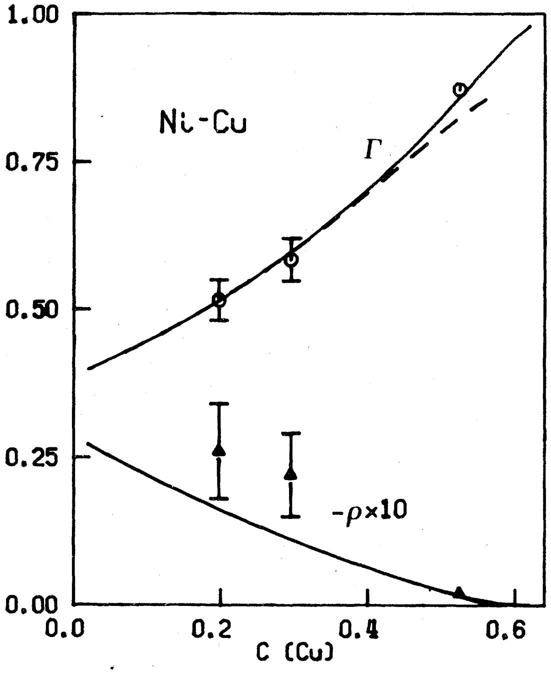

FIG. 2. Experimental and theoretical values of the parameters Γ and ρ of the magnetic-environment model for Ni-Cu. The dashed curve shows the values calculated using Eq. (2.13) while the continuous curve shows the values obtained using condition (2.40) as explained in Sec. II. Experimental values are from Ref. 10.

TABLE I. Parameters from the simultaneous fit of $\bar{\mu}$, Γ, and ρ for Ni-Cu.

A	B	Q_1	Q_2
2.58(12)	2.56(19)	−0.036(7)	−0.0065(30)

Given the difficulty of solving a nonlinear least-squares-fitting problem where the calculation of the function implies solving numerically a differential equation, i.e., condition (2.40), we made the following approximation. Instead of considering Γ_h as an arbitrary function, we expressed it as

$$\Gamma_h = \exp[f(c,b_1,b_2,\ldots,)]\frac{\partial F_h}{\partial h}, \qquad (3.8)$$

where f is a correction function of a fixed form and with free parameters b_i. These parameters enter into the least-squares-fitting problem as additional parameters to be determined, while condition (2.40) enters as additional data. This procedure is justified because the difference between Γ_h and $\partial F_h/\partial h$ is small. For Ni-Cu we used $f = b_i \exp(b_2 c)$. The parameters obtained by the fitting are given in Table I.

The calculated saturation moment given by the ratio A/B is 1.01 ± 0.03. This is in contrast with the common belief that the moment of pure Ni is saturated but is in fair agreement with the value of 0.96 that can be obtained from the work of Hicks.[15] The value of Γ_h for $c=0$, a property of pure Ni which in this model equals $A^{-1} = 0.39 \pm 0.02$ is not too different to the estimated values of Medina and Cable[10] (0.31) and Hicks[15] (0.36). Both chemical effect parameters are negative and small. In particular, the very small value of Q_2 shows that the copper atoms do not change the saturation moment of the nearby nickel atoms, which indicates that the charge transfer between cooper and nickel is small. The response function of pure Ni, shown in Fig. 3, is very similar to one previously determined by Medina and Cable,[10] and is drawn in the same figure as a dashed curve. The response function of Ni with 58 at. % Cu is also shown.

We used the values of A and B determined with Ni-Cu to treat the cases of Ni-Pd and Ni-Pt. The average alloy moment of Ni-Pd (Refs. 1, 11, 21–25) and of Ni-Pt (Refs. 12, 26–29) and their average host and impurity moments[11,12,21] were fitted simultaneously, as explained in Sec. II, in order to obtain the parameters Γ_i, α, β, Q_1, and Q_2. Again, Γ_h was considered to be an unknown function of concentra-

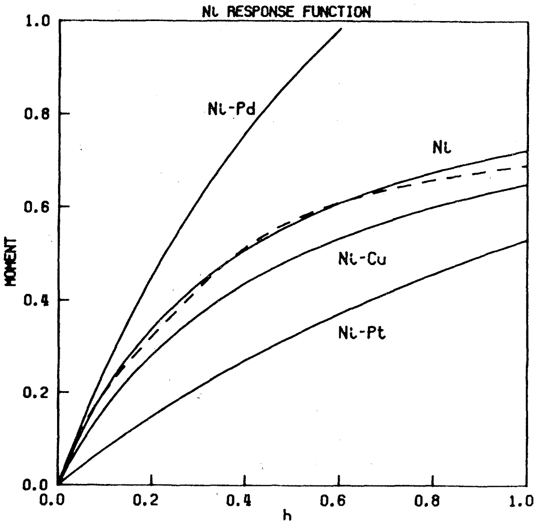

FIG. 3. Response functions of pure Ni, Ni-Cu, Ni-Pd, and Ni-Pt at the critical concentration. The dashed curve corresponds to the response function of pure Ni determined previously by Medina and Cable (Ref. 10).

tion to be determined by imposing condition (2.40). This was done in the same way as in the Ni-Cu case but with a different correction function, namely, a fourth-degree polynomial function. The results of the fitting are shown in Figs. 4 and 5 and the parameters are given in Table II. In general, the values of the parameters are similar for these isoelectronic alloys except for the parameter Q_1, which changes the low-field susceptibility, and which is big and negative for Ni-Pt and positive and much smaller for Ni-Pd. The values of Γ_i are close to, but smaller than one, in accord with the high polarizability of these impurities. The values of α and β indicate that a Pd atom produces an exchange field about five times bigger than that produced by a Ni atom with the same moment, independently of the kind of atom that the field acts upon. A Pt atom produces an exchange field, on a Ni atom, about three times bigger than the one produced by a Ni atom with the same moment and about one and a half times bigger if it acts upon another Pt atom. In both cases the presence of an impurity atom increases the saturation moment of nearby Ni atoms as expressed by the values of Q_2. This probably indicates that Ni transfers electrons to impurities nearby. The Ni response functions for both alloys at the critical concentration are shown in Fig. 3. With the parameters obtained, we calculated the values of the spherical average of $M(\vec{K})$ with Eq. (2.35), and they are compared with the neutron data[11,12,21] in Figs. 6, 7, and 8. Considering the approximations, the agreement is remarkable in the case of Ni-Pt for all concentrations and in the case of Ni-Pd for concentrations near the critical one. The calculated $M(\vec{K})$ for Ni-Pt coincides with the polarized neutron data of Parra and Cable.[12]

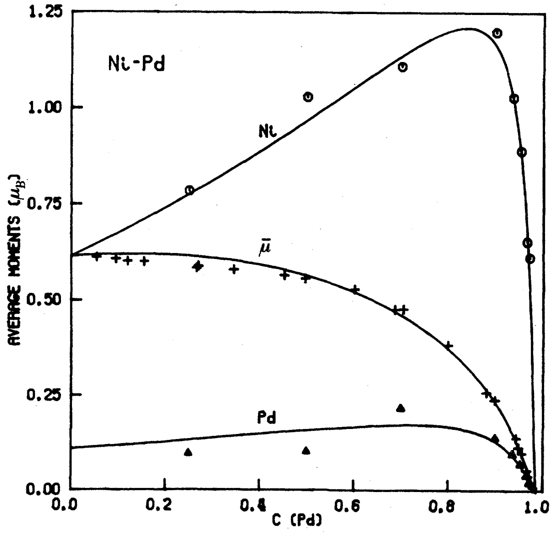

FIG. 4. Average moments of Ni-Pd from Refs. 1, 11, and 21–25. The curve represents the fitted values obtained with the magnetic environment model.

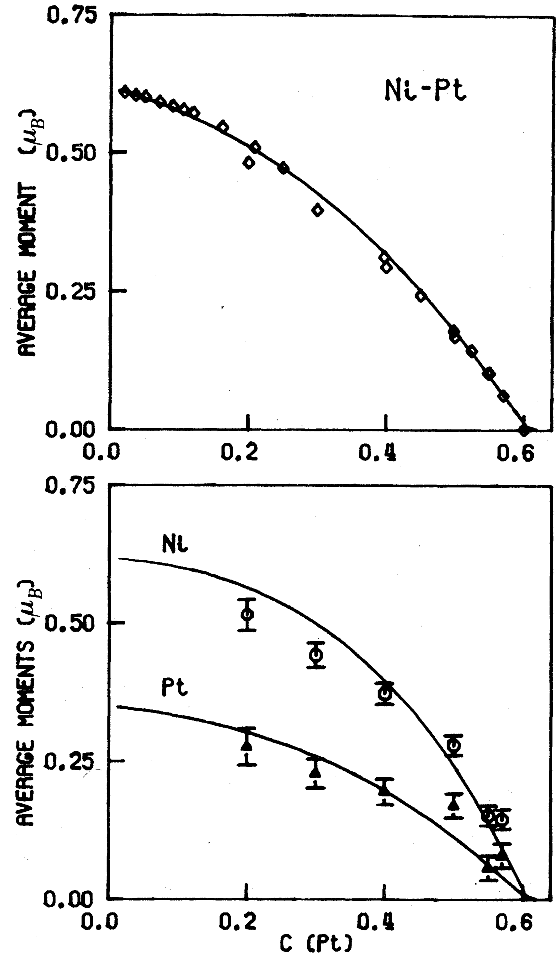

FIG. 5. Average moments of Ni-Pt from Refs. 12 and 26–29 fitted with the magnetic environment model.

There is, however, a larger difference between data and calculated values for Ni-Pd at smaller concentrations, particularly at 50 and 70 at.% Pd. We think that this difference is due to the fact that the $M(\vec{K})$

TABLE II. Parameters from the fit of $\bar{\mu}_i$, $\bar{\mu}_h$, and $\bar{\mu}$ vs c for Ni-Pd and Ni-Pt.

	Ni-Pd	Ni-Pt
Γ_i	0.947(9)	0.875(227)
α	5.6(2.7)	3.10(47)
β	0.182(19)	0.65(23)
Q_1	0.0072(369)	−0.163(25)
Q_2	0.0726(66)	0.056(28)

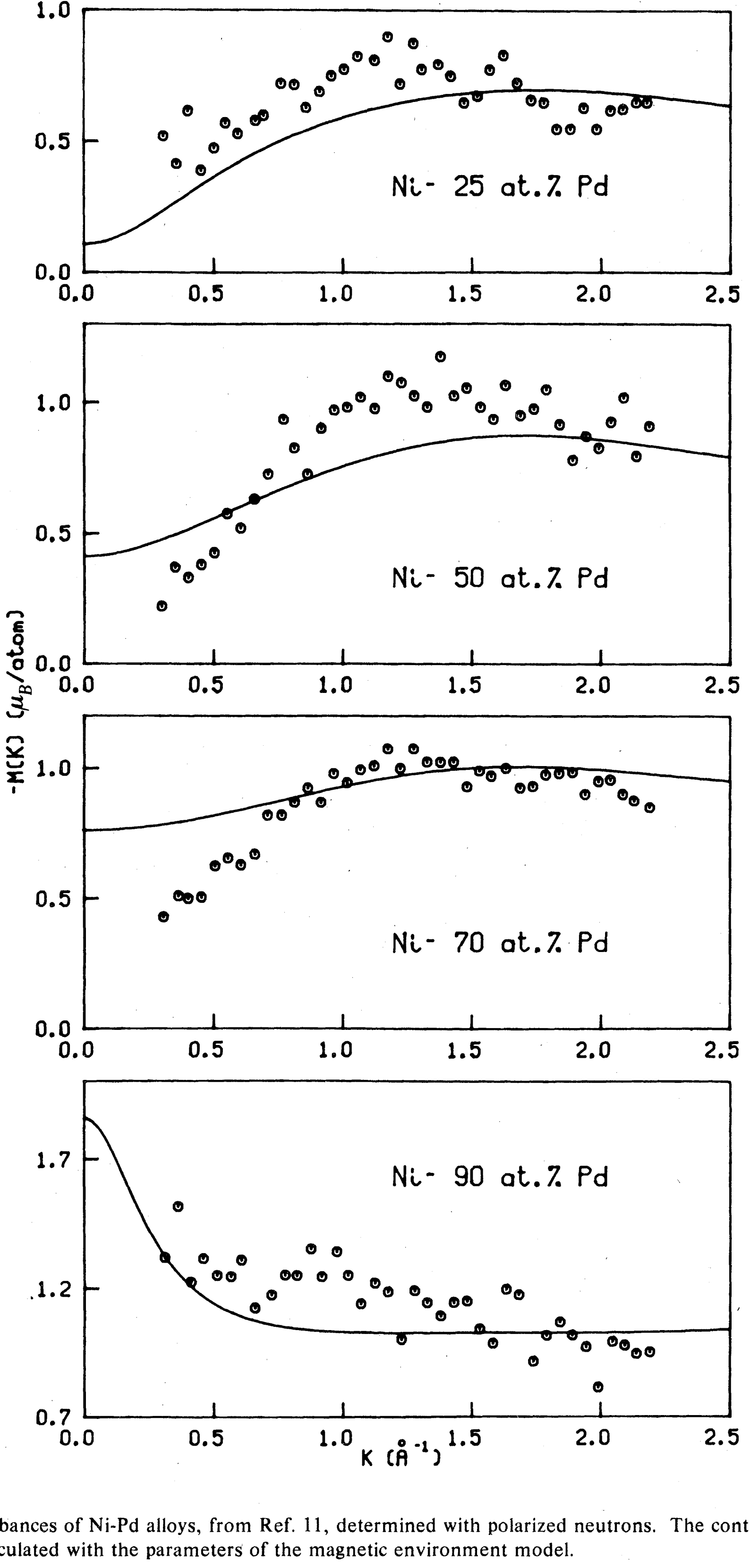

FIG. 6. Moment disturbances of Ni-Pd alloys, from Ref. 11, determined with polarized neutrons. The continuous curve corresponds to the values calculated with the parameters of the magnetic environment model.

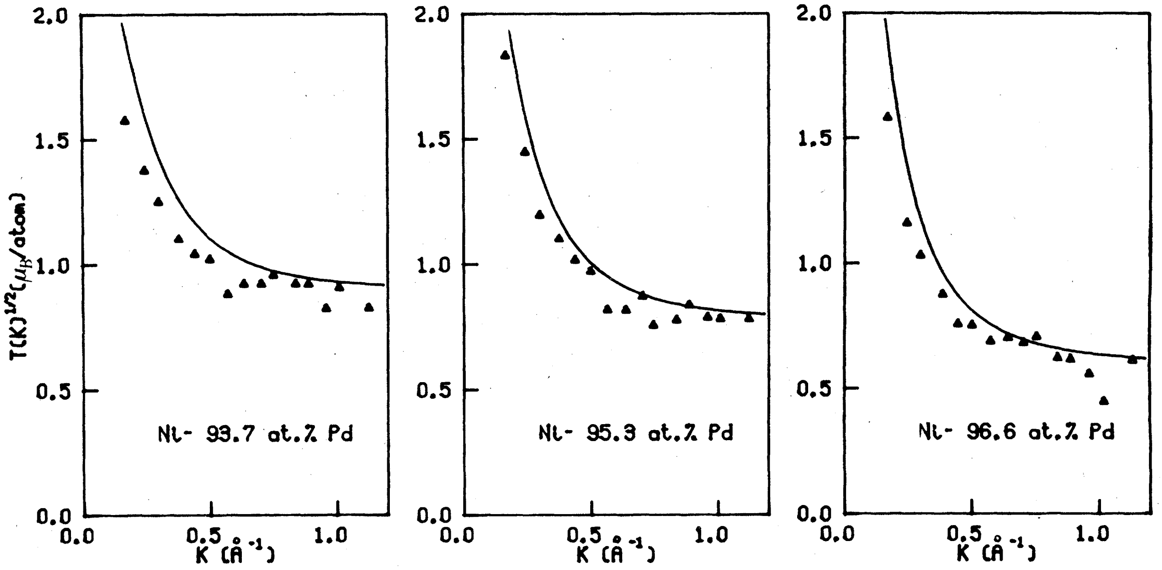

FIG. 7. Ni-Pd moment disturbances, from Ref. 21, determined with unpolarized neutrons (dots), and values calculated with the model (continuous curve).

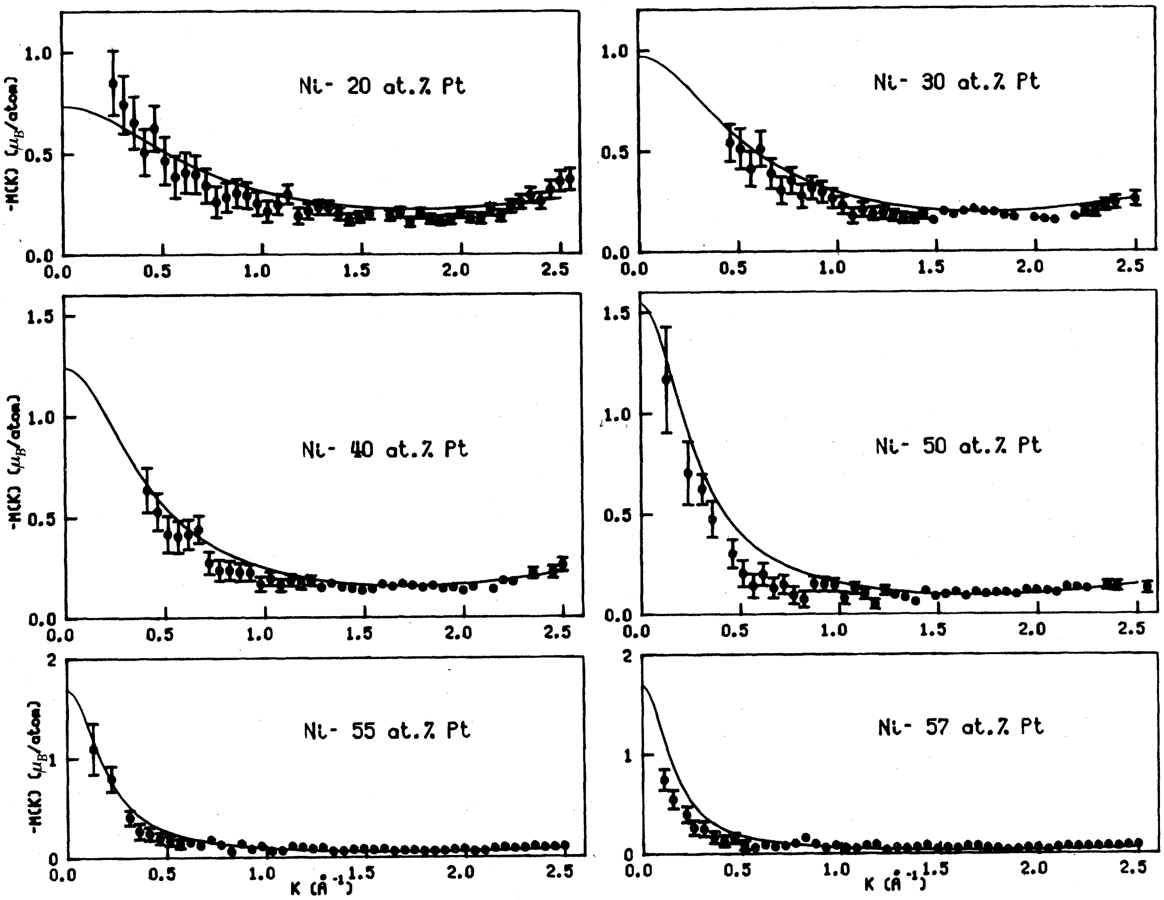

FIG. 8. Moment disturbances of random Ni-Pt alloys from Ref. 12. The continuous curve corresponds to the values obtained with the model.

data of Ni-Pd (Ref. 11) were not corrected for chemical short-range order (SRO), as they were for Ni-Pt. SRO effects disappear as the concentration approaches the extreme values 0 and 1; this explains the good agreement obtained near the critical concentration (~ 98 at.% Pd). The importance of SRO for the Ni-Pd data is evident when one compares the concentration derivative of the average moment (0.72 for the 70 at.% Pd alloy) with the $M(0)$ value obtained by the extrapolation of the data[11] at the same concentration (0.32) (see Fig. 6). The two values ought to coincide for random alloys.[14] On the other hand, the calculated value of $M(0)$ is 0.76.

An interesting effect has been found by Gillespie and Schindler[27] concerning the Ni-Pt system, namely, that the ordered Ni-54 at.% Pt and Ni-50 at.% Pt are paramagnetic while for the same concentrations the disordered alloys are ferromagnetic. From the values of the parameters for the Ni-Pt disordered alloys, we can estimate the critical concentration of the ordered alloys. The number of nearest Pt neighbors of a Ni atom in an ordered alloy is eight, therefore the field acting upon a Ni atom is

$$h_{\mathrm{Ni}}=(\tfrac{1}{3}-\delta)\mu_{\mathrm{Ni}}+(\tfrac{2}{3}+\delta)\alpha\mu_{\mathrm{Pt}}\ , \tag{3.9}$$

while the one acting upon a Pt atom is

$$h_{\mathrm{Pt}}=(\tfrac{1}{3}+\delta)\mu_{\mathrm{Pt}}+(\tfrac{2}{3}-\delta)\beta\mu_{\mathrm{Ni}}\ , \tag{3.10}$$

where δ is the excess Pt concentration over the stoichiometric composition. Replacing these field expressions into Eqs. (3.1) and (3.2), one obtains the following condition for the appearance of ferromagnetism:

$$A\exp[Q_1(8+12\delta)]\left(\frac{[(\frac{2}{3})^2-\delta^2]\Gamma_i\alpha\beta}{1-(\frac{1}{3}+\delta)\Gamma_i}+(\tfrac{1}{3}-\delta)\right)>1 \tag{3.11}$$

This yields a critical concentration of $c'=0.504 \pm 0.005$ which is consistent with the data of Gillespie and Schindler.

IV. Pd AND Pt WITH ISOLATED MAGNETIC IMPURITIES

The high values of Γ for Pd and Pt are responsible for the giant moment polarization clouds that appear when magnetic impurities such as Fe are introduced in these metals. For an isolated magnetic impurity that polarizes its neighbors and considering a linear-response function as in Eq. (3.1) one obtains, with no other approximations, the following expression for $M(K)$:

$$M(K)=\mu_0+\frac{\Gamma g_1}{B(\Gamma)-1}\left(\frac{1}{1-\Gamma F_1(\vec{\mathrm{K}})}-B(\Gamma)\right)\ . \tag{4.1}$$

Here, μ_0 is the impurity moment and g_1 is the moment of first-neighbor host atoms. Neutron data[30,31] have been analyzed assuming a Lorentzian form for $M(K)$, which is valid for small-K values. In this limit $F_1(K)=1-(aK)^2/12$ and one gets the following relationship between Γ and the range parameter of the Lorentzian:

$$k_1=a^{-1}[12(1-\Gamma)/\Gamma]^{1/2}\ . \tag{4.2}$$

Using the proper values of the lattice parameters "a" one obtains the value $k_1=0.21$ Å^{-1} for Fe-Pd, which should be compared with the reported value[27] of 0.2 Å^{-1}. For Fe-Pt we predict a value of $k_1=0.33$ Å^{-1}.

V. CONCLUSIONS

A local magnetic environment model developed for Ni-Cu alloys, and in which the Ni moments are a function of the moments of the neighboring atoms, has been extended to the cases of Ni-Pd and Ni-Pt where the impurity atoms are also magnetic. The model describes well the diffuse scattering neutron measurements of these alloys once the parameters of the theory have been obtained from fitting the model to the moment versus concentration data. With these parameters one can also calculate the range parameter of the moment disturbances in the giant moment systems Fe-Pd and Fe-Pt.

Some comments, however, must be made about the approximations used in this model. The saturating function and the chemical effect functions used are arbitrary to the extent that they are the simplest available and that the parameters obtained give a qualitative measurement of the effects. For the same reasons, we have also disregarded any chemical environment dependence of the parameters Γ_i, α, and β. Although contributions to the exchange field due to neighbors beyond the first ones cannot be excluded, the agreement of the calculated $M(K)$ with the neutron data shows that only first-neighbor interactions are necessary to explain the observations. The moment perturbations produced by first neighbors are propagated to first neighbors of those neighbors and so on. This explains the long-range-moment disturbances measured with neutrons.

Since the model presented here is phenomenological, a more fundamental theory is needed. However, the model describes, with a few parameters, the magnetic behavior of Ni-based alloys; therefore the task of a first-principles theory may be reduced to an explanation of those parameters. The success of the magnetic environment model, which is based on a local moment picture, suggests that a theory like the quasilocalized spin theory[32–37] should be adequate for explaining the magnetism of nickel and other elements of the same group.

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APPENDIX

The coefficients of Eqs. (2.32) and (2.33) are

$$
\begin{aligned}
d = {} & c(1-c)(1-\alpha\beta)(B_0+B_1)^2 - B_0B_1 \\
& + c\Gamma_i\{(1-c)(1-\alpha\beta)B_1[(1-c)B_1 - cB_0)] - (1-\alpha\beta)c(1-c)B_2B_1 + B_0B_2[\alpha\beta + c^2(1-\alpha\beta)]\} \\
& + (1-c)\Gamma_h\{c(1-\alpha\beta)B_1[cB_1-(1-c)B_0] - c(1-c)(1-\alpha\beta)B_2B_1 + [\alpha\beta+(1-\alpha\beta)(1-c)^2]B_0B_2\} \\
& + (1-\alpha\beta)c(1-c)\Gamma_i\Gamma_h[c(1-c)(1-\alpha\beta)(B_1+B_2)^2 - B_1B_2] \ , \\
a_1d = {} & B_1[-B_1+(1-\alpha\beta)c(B_0+B_1)] + (1-c)\Gamma_hB_1\{[(1-c)B_2-cB_1](1-\alpha\beta)+B_2\alpha\beta\} \\
& + c\Gamma_iB_2\{\alpha\beta B_1 - (1-\alpha\beta)[cB_0-(1-c)B_1]\} \\
& - c(1-c)\Gamma_i\Gamma_h(1-\alpha\beta)B_2\{[(1-c)B_2-cB_1](1-\alpha\beta)+\alpha\beta B_2\} \ , \\
a_2d = {} & B_1^2 - (1-c)\Gamma_h[(1-\alpha\beta)B_1(B_2-cB_1) + (1-\alpha\beta)cB_0B_2 + B_1B_2\alpha\beta] - c\Gamma_iB_1B_2 + c(1-c)\Gamma_i\Gamma_h(1-\alpha\beta)B_2^2 \ , \\
a_3d = {} & (-cB_0 + (1-c)B_1 + (1-c)\Gamma_h\{(1-\alpha\beta)[cB_1-(1-c)B_2] - \alpha\beta B_2\})(B_1 - c\Gamma_iB_2) \\
& + \alpha\beta c(1-c)\Gamma_hB_2(B_0+B_1-\Gamma_iB_2) \ , \\
a_4d = {} & (-cB_0 + (1-c)B_1 + (1-c)\Gamma_h\{(1-\alpha\beta)[cB_1-(1-c)B_2] - \alpha\beta B_2\})c\Gamma_iB_2 \\
& + c(B_0+B_1-\Gamma_iB_2)[B_1-(1-c)\Gamma_hB_2] \ .
\end{aligned}
$$

Exchanging c with $(1-c)$ and Γ_i with Γ_h one obtains the coefficients b_i.

The functions B_n are defined as

$$B_n = \frac{1}{V_c^*}\int \frac{[F_1(\vec{K})]^n d^3K}{1-\Gamma F_1(\vec{K}) + DF_1(\vec{K})^2} \ ,$$

where Γ and D are given in Eqs. (2.36) and (2.37). These functions can be reduced to the function $B(\Gamma)$ defined as

$$B(\Gamma) = \frac{1}{V_c^*}\int \frac{d^3K}{1-\Gamma F_1(\vec{K})} \ ,$$

which has an analytic expression[38] for the fcc lattice.

For example, the functions B_n that appear in the model are

$$
\begin{aligned}
B_0 &= \tfrac{1}{2}[B(\Gamma_1)+B(\Gamma_2)] + \tfrac{1}{2}\Gamma B_1 \ , \\
B_1 &= [B(\Gamma_2)-B(\Gamma_1)]/(\Gamma^2-4D)^{1/2} \ , \\
B_2 &= D^{-1} + D^{-1}(B_1-B_0) \ ,
\end{aligned}
$$

where

$$
\begin{aligned}
\Gamma_1 &= \tfrac{1}{2}[\Gamma - (\Gamma^2-4D)^{1/2}] \ , \\
\Gamma_2 &= \tfrac{1}{2}[\Gamma + (\Gamma^2-4D)^{1/2}] \ .
\end{aligned}
$$

The parameters of $M(\vec{K})$ in Eq. (2.35) are given by the following expressions:

$$
\begin{aligned}
U = {} & -(1-c)g_1[(1-c)\Gamma_h + \beta c\Gamma_i] - ch_1[c\Gamma_i + \alpha(1-c)\Gamma_h] \ , \\
V = {} & -\bar{\mu}_h[(1-c)\Gamma_h + \beta c\Gamma_i] + \bar{\mu}_i[c\Gamma_i + \alpha(1-c)\rho h] + c(1-c)g_1[\Gamma_h - \beta\Gamma_i + (1-\alpha\beta)(1-c)\Gamma_h\Gamma_i] \\
& + c(1-c)h_1[\Gamma_i - \alpha\Gamma_h + (1-\alpha\beta)c\Gamma_i\Gamma_h] + (1-c)\Gamma_hZ_1\rho_h + c\Gamma_iZ_1\rho_i \ , \\
W = {} & -c(1-c)\Gamma_i\Gamma_h\{(1-\alpha\beta)(\bar{\mu}_i-\bar{\mu}_h) + g_1[c-\beta+(1-c)\alpha\beta] \\
& + h_1(1-c-\alpha+\alpha\beta c) + Z_1\rho_h(1-\beta) + Z_1\rho_i(1-\alpha)\} \ .
\end{aligned}
$$

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