INFLUENCE OF LOCAL ORDERING ON THE VALUE OF THE SPIN WAVE STIFFNESS CONSTANT IN FERROMAGNETIC TRANSITION METAL ALLOYS.
II. EFFECT OF SHORT-RANGE ORDER*

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We present numerical results from the study of the short-range order effect on the value of the spin wave stiffness constant.

1. Introduction

In the last few years there has been an increasing interest in the local environment effects in transition metal binary alloys. A large number of experiments show the importance of the local environment on the magnetic and electronic properties of alloys. In this paper we study the magnon energy of binary alloys using a method described in a previous paper (Jezierski [1]), hereafter referred to as I, and Falicov et al. [2]. Our aim is to include the short-range order in the expression for the spin wave stiffness constant (see I Eq. (16)). In paper I we introduced a formula for the local two-particle Green function. Then using the exact expression for the stiffness constant D and applying the coherent potential approximation (CPA) we made the numerical calculations for NiFe and NiCo alloys. In Section 3 (I) it was shown, basing on a simple cluster model, that the stiffness constant depended on the distribution of impurity atoms in the matrix. In this paper we present the results of numerical calculations for NiFe alloy (Section 4a) and for NiPt, NiPd and NiCu alloys (Section 4b).

2. The short-range order parameters

We characterized the alloy by the short-range order parameters p_A and p_B. The parameter $p_A(p_B)$ gives the probability that when choosing a nearest-neighbour pair such that one of the atoms is of class A (B), the other atom is of the same class. Then we can

define similarly as Falicov et al. [2] the parameters $\lambda_A(\lambda_B)$ and $q_A(q_B)$ by the relations

$$\lambda_A = p_A - q_A = 2p_A - 1, \tag{1}$$

$$\lambda_B = p_B - q_B = 2p_B - 1, \tag{2}$$

$\lambda_A = 1$ corresponds to complete segregation — all atoms of class A surrounded by atoms of class A, $\lambda_A = -1$ corresponds to a perfect binary compound. Similar definition is for λ_B. The parameter $q_A(q_B)$ gives the probability of finding a nearest-neighbour pair such that if one of the atoms is of class A (B) the other atom is of class B (A). It is easy to show that $p_A + q_A = 1$. The values of q_A and q_B are limited by the concentration c_A and c_B and by the requirement of conservation of the total number of particles [2]

$$c_A q_A = c_B q_B, \tag{3}$$

or from (1) and (2)

$$c_A \lambda_A - c_B \lambda_B = c_A - c_B, \tag{4}$$

where $c_A = 1 - c$ and $c_B = c$.

Sometimes the other short-range order parameters are used. Reck [3] and Mishra [4] defined α_1 parameter which may be written as

$$\alpha_1 = 1 - \frac{(1 - \lambda_B)}{2(1 - c)}. \tag{5}$$

Stern et al. [5] determined the probability p and q by β parameter and the concentration. We may write

$$p_A = 1 - \beta c, \tag{6}$$

$$q_A = \beta(1 - c), \tag{7}$$

$$p_B = 1 - \beta(1 - c), \tag{8}$$

$$q_B = \beta c, \tag{9}$$

where β is a parameter which may be different from unity, which describes the degree of local order. If $\beta = 1$ then the alloy is random, when $0 \leqslant \beta < 1$ then is tendency to segregation of A and B atoms into clusters of similar atoms. If $\beta > 1$ then atoms of one type tend to be surrounded by atoms of the other type (anticlustering). It is easy to show that the β parameter is connected with λ by the relation

$$\beta = \frac{1}{2c}(1 - \lambda_A), \tag{10}$$

and

$$\beta = \frac{1}{2(1 - c)}(1 - \lambda_B). \tag{11}$$

From (10) and (11) we get

$$(1-c)\lambda_A - c\lambda_B = (1-c) - c,$$ (12)

which is equivalent to (4).

So far we have characterized the alloy by means of the parameters λ_A and λ_B, which can vary only between -1 and $+1$. Falicov et al. [2] considered three cases:

(a) Segregation sequence

The alloy has a tendency to segregate into two separate regions, the A and B regions, respectively. For this sequence

$$\lambda_A = \lambda_B = 1 \quad \text{for any } c_A.$$ (13)

(b) Random sequence

In this sequence, there is no short-range correlation between the atoms. This sequence corresponds to the coherent potential approximation.
In this case

$$\lambda_A = c_A - c_B \quad \text{and} \quad \lambda_B = c_B - c_A.$$ (14)

(c) Binary-compound sequence

All the atoms of class A prefer to be surrounded by atoms of class B and vice versa. In this sequence, we have

$$\lambda_A = -1 \quad \lambda_B = 1 - 2c_A/c_B \quad \text{if } c_A \leqslant c_B,$$ (15)

$$\lambda_B = -1 \quad \lambda_A = 1 - 2c_B/c_A \quad \text{if } c_A \geqslant c_B.$$ (16)

3. Spin wave stiffness constant and the short-range order properties

Our aim is to calculate the value of the stiffness constant. According to (I–2) we have

$$D = \frac{1}{3(n_+ - n_-)} \left[\frac{1}{2} \sum_k \langle n_{k+} + n_{k-} \rangle \nabla_k^2 \varepsilon_k - \sum_k \sum_{k'} \langle\langle a_{k-}^+ a_{k+}; a_{k'+}^+ a_{k'-} \rangle\rangle_{\omega=0} \nabla_k \varepsilon_k \cdot \nabla_{k'} \varepsilon_{k'} \right]$$ (17)

the local two-particle Green function $\langle\langle a_{k-}^+ a_{k+}; a_{k'+}^+ a_{k'-} \rangle\rangle_\omega$ is determined by the relation (I–8). In all formulae we used the same notations as in part I and $\langle\langle a_{k-}^+ a_{k+}; a_{k'+}^+ a_{k'-} \rangle\rangle_{\omega=0} \equiv G(k-k')$
where

$$G(k-k') = \frac{1}{Nz} \sum_{\substack{l,jl \neq j \\ \alpha,\beta}} G_0(l^\alpha j^\beta; j^\beta l^\alpha) e^{i(k-k')(l-j)}$$

$$= \frac{1}{Nz} \sum_{\substack{l,j\, l \neq j \\ \alpha, \beta}} e^{i(k-k')(l-j)} [W_1 + c_l^B W_2 + c_j^B W_3 + c_l^B c_j^B W_4$$

$$+ \sum_{r \neq l \neq j} (W_5 c_r^B + W_6 c_l^B c_r^B + W_7 c_j^B c_r^B + W_8 c_l^B c_j^B c_r^B)]. \tag{18}$$

The two particle Green function is taken in this paper in the pair approximation. We consider only the interaction between the central atom and its nearest-neighbour in the first shell. In this approximation the sum over r vanished because it includes the interactions between sites which do not belong to the first shell. Therefore we may write the configurational average of the two-particle Green function as

$$\overline{G(k-k')} = G(c, \lambda_B)\delta_{kk'} = [W_1 + \langle c_l^B \rangle W_2 + \langle c_j^B \rangle W_3 + \langle c_l^B c_j^B \rangle W_4]\delta_{kk'}. \tag{19}$$

It is easily found using the definition of p_B and λ_B that $\langle c_l^B \rangle = \langle c_j^B \rangle = c$ and $\langle c_l^B c_j^B \rangle = cp_B = 0.5\,c\,(1+\lambda_B)$. Then we can rewrite (19) as

$$\overline{G(k-k')} = \delta_{kk'}[W_1 + c\{W_2 + W_3 + \tfrac{1}{2}\, W_4(1+\lambda_B)\}]. \tag{19a}$$

Substituting (19a) to (17) we get

$$D(c, \lambda_B) = \frac{1}{3(n_+ - n_-)} \left[\tfrac{1}{2} \sum_k \langle n_{k+} + n_{k-} \rangle \nabla_k^2 \varepsilon_k - \sum_{\substack{k \\ \varepsilon(k) \leqslant \varepsilon_F}} G(c, \lambda_B) |\nabla_k \varepsilon_k|^2 \right]. \tag{20}$$

The electron energy ε_k was taken in the tight binding approximation (I–13), and the numerical values of $\nabla_k^2 \varepsilon_k$ and $|\nabla_k \varepsilon_k|^2$ were calculated in I (Section 3). The configurational average of the two-particle Green function $G(c, \lambda_B)$ can be written as

$$G(c, \lambda_B) = G_{CPA} + \tfrac{1}{2}\, W_4[\lambda_B - \bar{\lambda}_B]c, \tag{21}$$

where $G_{CPA} = W_1 + c[W_2 + W_3] + c^2\, W_4$ is calculated in the CPA and $\bar{\lambda}_B = 2c - 1$.

Putting the expression (21) to (20) and dividing over D_{Ni} (the value of stiffness constant for a pure nickel) we obtain

$$\frac{D(c, \lambda_B)}{D_{Ni}} = \frac{D(c, \bar{\lambda}_B)}{D_{Ni}} - \frac{cW_4(\lambda_B - \bar{\lambda}_B)}{6NnD_{Ni}} \sum_{\substack{k \\ \varepsilon(k) \leqslant \varepsilon_F}} |\nabla_k \varepsilon_k|^2, \tag{22}$$

or using the definition of P(I–15a)

$$\frac{D(c, \lambda_B)}{D_{Ni}} = \frac{D(c, \bar{\lambda}_B)}{D_{Ni}} - \frac{P\left(\dfrac{4\varepsilon_F}{W}\right) a^2 t^2 c W_4(\lambda_B - \bar{\lambda}_B)}{6nD_{Ni}} \tag{23}$$

here $n = n_+$ is the total number of $\uparrow$ spin electrons. (We consider the case when $n_- = 0$), and $W = 16t$.

In our calculations we assume that the hopping integral t changes linearly with concentration. We take t in a form (I–10)

$$t = t_{AA} + 2cH,\tag{24}$$

where $H = 0.5\,(t_{BB} - t_{AA})$. The formula (24) is obtained from the expression $t = (1-c)^2\,t_{AA} + 2c(1-c)\,t_{AB} + c^2\,t_{BB}$.

4. Numerical results

(a) NiFe alloys

In part I we calculated D using the coherent potential approximation. Now we apply formula (23) in order to get the values of the stiffness constant $D(c, \lambda_B)/D_{Ni}$.

First we will consider NiFe alloy. Magnetic disorder-scattering indicates that the short-range order may exist over much of the concentration range of the NiFe alloys.
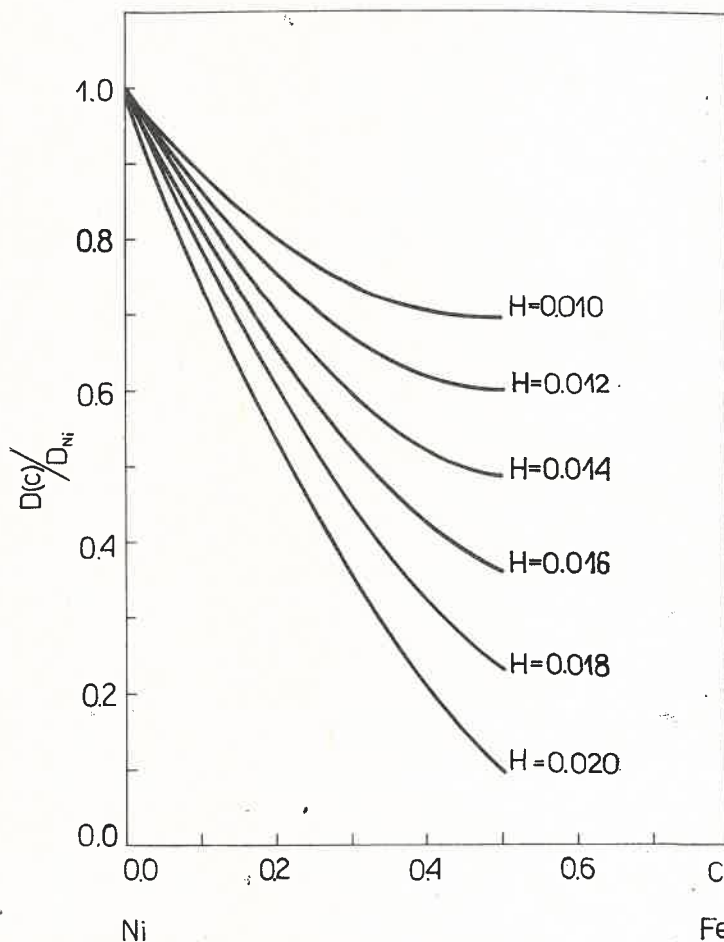

Fig. 1. Values of $D(c)/D_{Ni}$ as a function of impurity concentration c for NiFe alloy

TABLE I

	I (eV)	t (eV)	ε (eV)
Ni	0.95	0.075	0.000
Fe	0.97	0.089	−0.135

Reck [3], from a comparison of the measured and calculated effective magnetomechanical ratio g' values, estimated the value of the short-range order parameter α_1 as a function of the atomic fraction of iron. The values of α_1 estimated by Reck were small in absolute
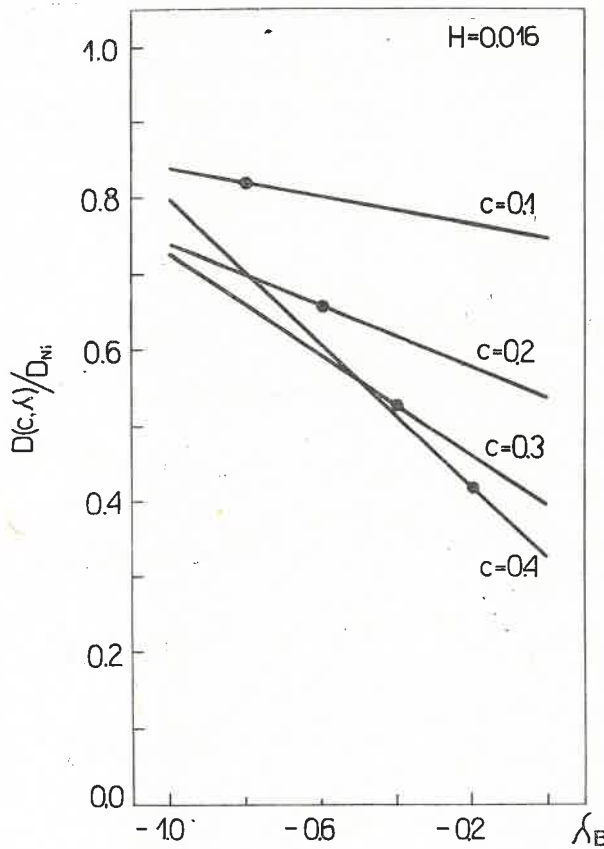

Fig. 2. Plot of $D(c, \lambda_B)/D_{Ni}$ versus λ_B for the different concentration Fe. The points denote the CPA results

magnitude, and corresponded to random sequence. For $Ni_{0.75} Fe_{0.25}$ alloy the value of the stiffness constant $D(c, \alpha_1 = -0.003)$ is greater than CPA result by only about 3%. In our calculation we assume the values of hopping integral t, intraatomic Coulomb interaction I, and atomic potential ε for NiFe alloy as in Table I. We take $n_+^{Fe} = n_+^{Ni} = 0.6$.
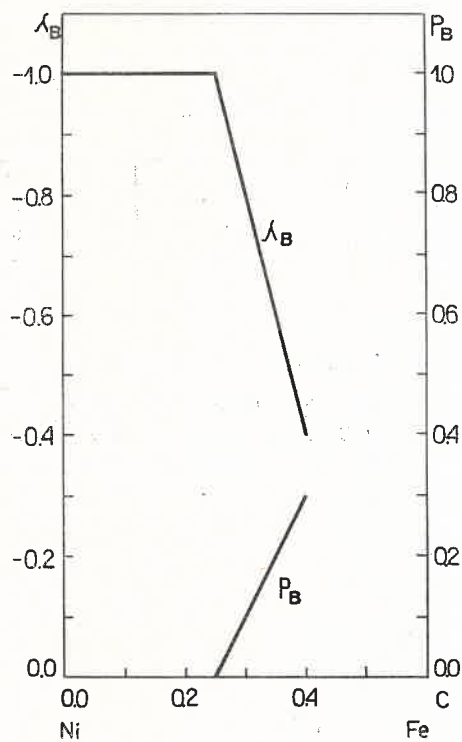
Fig. 3. Estimated dependence of the short-range parameter λ_B and probability p_B on the concentration c for ordering NiFe alloy
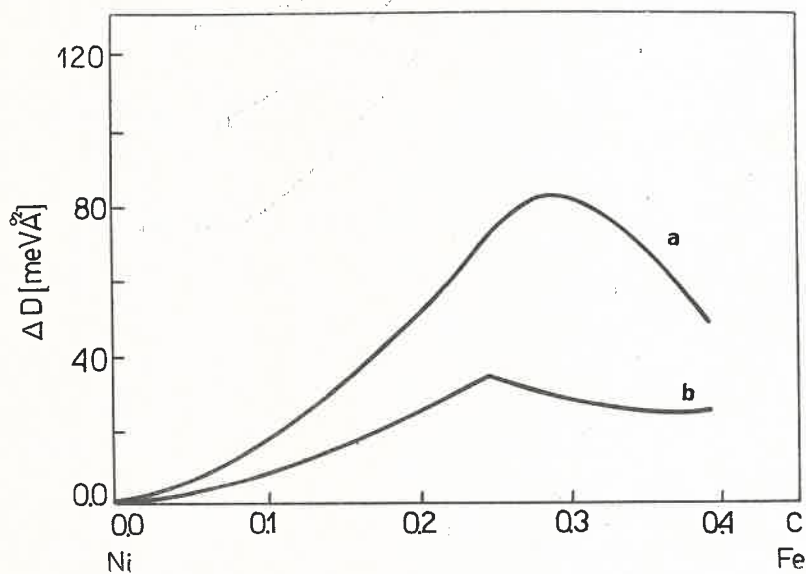

Fig. 4. Plot of the computed difference $\Delta D = D(c, \lambda_B) - D(c, \bar{\lambda}_B)$ for ordering and for the disordered NiFe alloy

TABLE II

c	λ_B	$D(c, \lambda_B)/D_{Ni}$	$D(c, \bar{\lambda}_B)/D_{Ni}$ [CPA]	ΔD [meV Å²]
0.10	−1.0	0.84	0.82	10
0.20	−1.0	0.74	0.66	43
0.25	−1.0	0.72	0.59	72
0.30	−0.8	0.66	0.53	83
0.40	−0.4	0.51	0.42	46

The values of the spin wave stiffness constant calculated in the coherent potential approximation are in agreement with the experimental values of Mikke et al. [4].

In the paper I we have shown the dependence of $D(c)$ on the change of the intra-atomic Coulomb interaction. Now we calculated in the CPA the values of $D(c)/D_{Ni}$ for
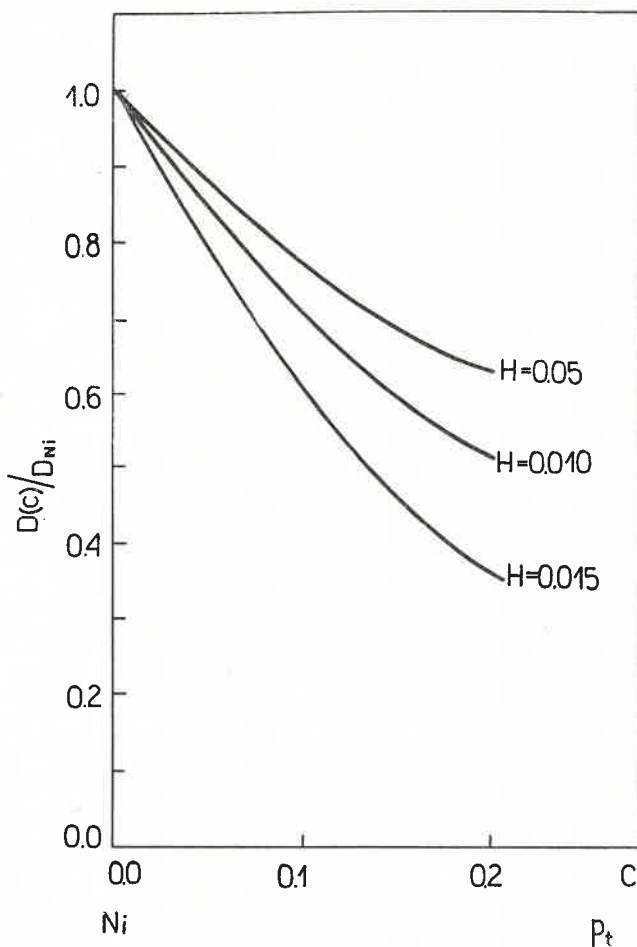

Fig. 5. Calculated values of $D(c)/D_{Ni}$ for NiPt alloy for the different H
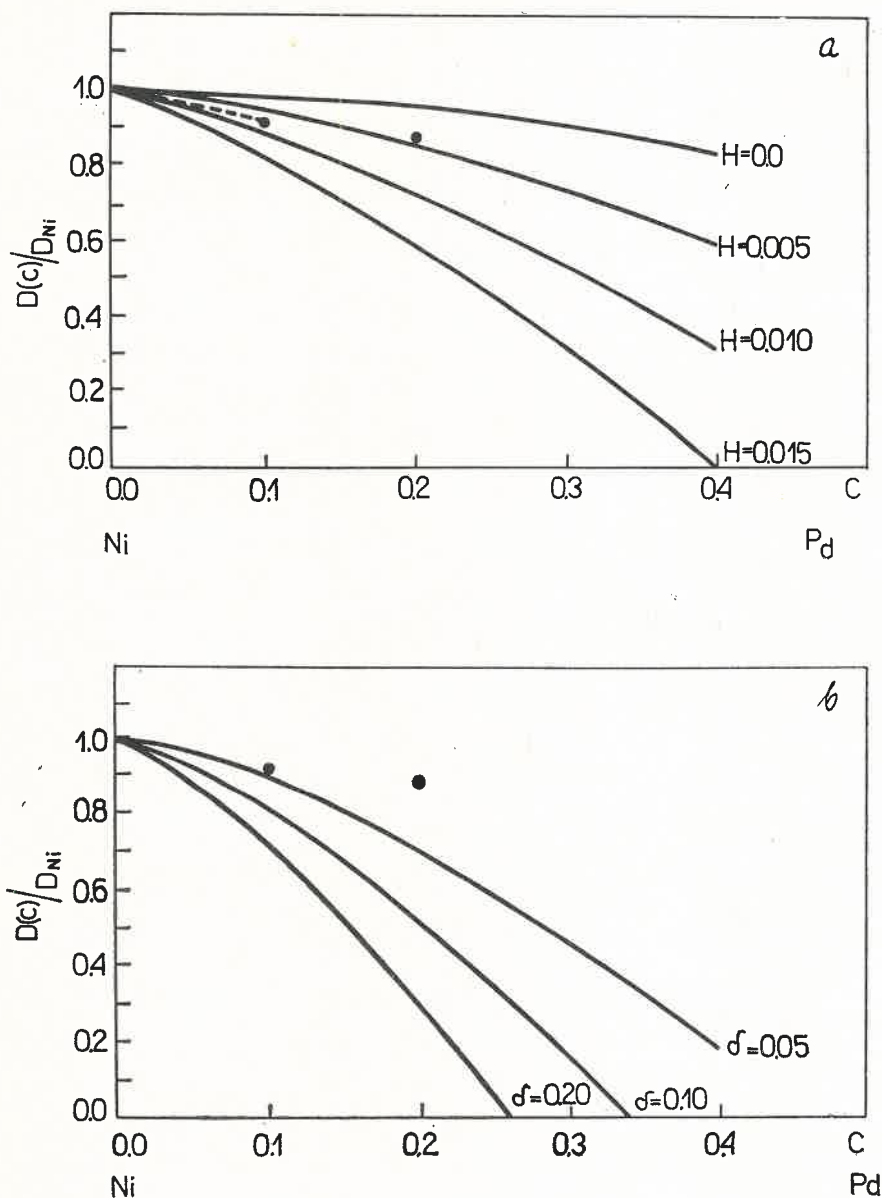
Fig. 6. Calculated results of $D(c)/D_{\text{Ni}}$ for NiPd alloy a — $I = 0.9$ eV, $\delta = 0$, $H = 0.0, 0.005, 0.01, 0.015$ eV b — $I = 0.72$ eV, $H = 0.01$ eV, $\delta = 0.05, 0.10, 0.20$ eV. Points denote the experimental values (Stringfellow [14]) The broken curve shows Morkowski's [12] result for $H = 0.005$

the different hopping integrals. These results are presented in figure 1. Then we examined the influence of a local ordering on the value of the stiffness constant. We used the expression (23) and calculated the $D(c, \lambda_B)/D_{\text{Ni}}$ for the different λ_B (see figure 2). As evident from figure 2 the value of $D(c, \lambda_B)/D_{\text{Ni}}$ may be greater than the CPA result.

When we assume that λ_B or p_B changes with concentration as in figure 3 we get the values of $D(c, \lambda_B)/D_{Ni}$ similarly as Mikke et al. [6]. In figure 4 we show the computed difference $\Delta D = D(c, \lambda_B) - D(c, \bar{\lambda}_B)$ (curve a) calculated in this paper (Table II) for ordering and for disordered alloy and Mikke et al. [6] results calculated in terms of the Heisenberg model (curve b).

(b) Stiffness constant in NiPd, NiPt and NiCu alloys

We computed the values of the stiffness constant in the CPA for several nickel ferromagnetic alloys Ni T (where T = Pd, Pt, Cu). The intraatomic Coulomb integrals I and the values of $\delta = \varepsilon_B - \varepsilon_{Ni}$ for Pd, Pt and Cu were taken as in Table III. The ratio of
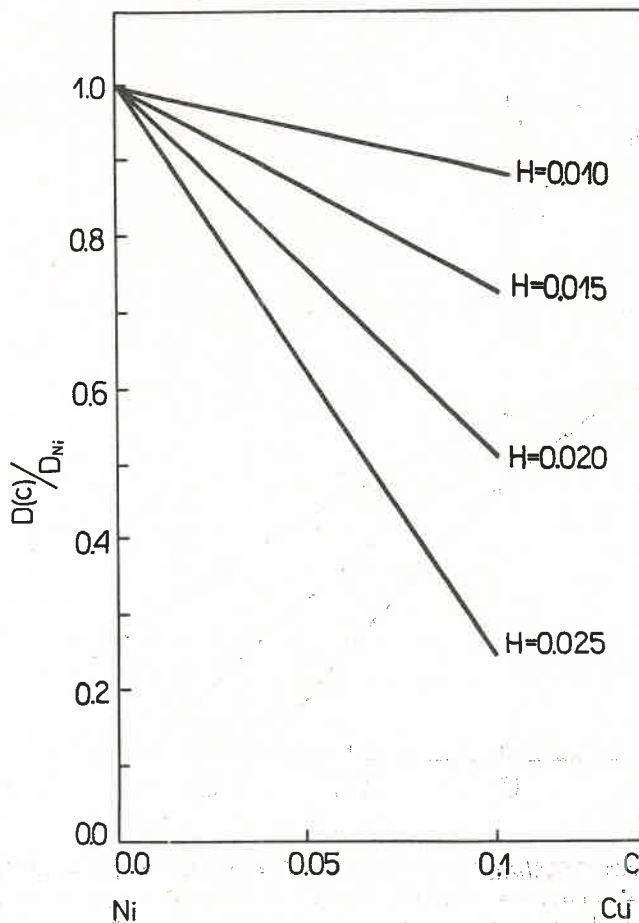

Fig. 7. Calculated values of $D(c)/D_{Ni}$ for NiCu alloy for the different H

I_T/I_{Ni} (T = Pd, Pt) we assumed similarly as Inoue et al. [9], and δ for Cu as Inoue et al. [11]. In this calculations the saturation moment of alloys (NiPt, NiPd, NiCu) changed with increasing inpurity concentration as reported by Mishra [4], Fischer et al. [7] and Fischer et al. [8]. The values of $D(c)/D_{Ni}$ for NiPt, NiPd and NiCu alloys are presented

TABLE III

	I (eV)	δ (eV)
Pd	0.9	0.0
	0.72	0.05—0.20
Pt	1.08	0.0
Cu	0.95	−2.0

in figures 5–7. In figure 6a the broken curve shows the Morkowski [12] results calculated for the ferromagnetic transition metal dilute alloys in the Random Phase Approximation using the method of effective magnon Hamiltonian.

5. Conclusions

In this paper we calculated the spin wave stiffness constant in the coherent potential approximation and then as a function of the short range order parameter λ_B. According to the definition λ_B for $\lambda_B = c_B - c_A$ there is no short-range correlation between the atoms. This case corresponds to CPA. The numerical results for $\lambda_B = 2c - 1 = \bar{\lambda}_B$ are the same as the CPA results which were obtained using the Edwards and Jones [15] method (see I). The effect of short-range order depended in our model on the value of $\lambda_B - \bar{\lambda}_B$. This result was obtained in the pair approximation, but when we took all the terms in the two-particle Green function into account we get more complicated expression for the spin wave stiffness constant. For NiPd, NiPt and NiCu alloys the stiffness constant decreases when the concentration of impurity increases. In the case NiPd alloy our results are similar to Stringfellow et al. [14] experimental values.

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