

INFLUENCE OF LOCAL ORDERING ON THE VALUE OF SPIN WAVE STIFFNESS CONSTANT IN FERROMAGNETIC TRANSITION METAL ALLOYS*

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An expression for the spin wave stiffness constant D of ferromagnetic alloys is derived without using the Hartree-Fock Approximation. It is shown that D is very sensitive to distribution of impurity atoms in crystal. Using the coherent potential approximation and cluster model the numerical calculations are made for NiFe and NiCo alloys.

1. Introduction

The energy of a long wavelength magnon is given by the simple formula $E_q = Dq^2$, where D is the spin wave stiffness constant. Early calculations of magnon energy in the ferromagnetic transition metal alloys were based on the one-electron Green functions which were calculated within the coherent potential approximation (see: Fukuyama [1], Hill and Edwards [2], Riedinger and Nauciel-Bloch [3]).

An expression for D in terms of the one-electron Green functions is very useful to calculate the configurational average in CPA. However, this procedure has some shortcomings. One of them is that the Hartree-Fock Approximation has to be used. Another one is the difficulty of taking into consideration the change of the hopping integral with concentration. In this paper we present another method to calculate the value of the two-particle Green function without using the Hartree-Fock Approximation.

2. Outline of method

We consider the ferromagnetic binary alloys $A_{1-c}B_c$, where c is a concentration of the component B . The Hamiltonian for this system is the following:

$$H = \sum_{ij\sigma} t_{ij} a_{i\sigma}^+ a_{j\sigma} + \sum_{i\sigma} \varepsilon_i n_{i\sigma} + \sum_i I_i n_{i+} n_{i-}. \quad (1)$$

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Here ε_l is the atomic potential, and it takes the values ε_A and ε_B depending on whether site l is occupied by atom A or B , t_{ij} is the hopping integral which takes the values t_{ij}^{AA} , t_{ij}^{AB} , t_{ij}^{BA} , or t_{ij}^{BB} . Similarly, I_l the intraatomic Coulomb interaction takes the values I_A or I_B . The operators $n_{l\sigma}$, $a_{l\sigma}^+$ and $a_{l\sigma}$ are the occupation numbers, the creation and annihilation operators for electron in the Wannier states at the lattice site R_l with spin σ , respectively. Since the Hamiltonian (1) has a rotational symmetry with regard to the total spin $[H, S_0^-] = 0$ we may use, similarly as Hill and Edwards [2], an exact formula for the spin wave stiffness constant D derived by Edwards and Fischer [4].

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

where n_+ , n_- are the total numbers of + and - spin electrons and $\langle \langle a_{k-}^+ a_{k+}; a_{k'+}^+ a_{k'-} \rangle \rangle_{\omega}$ is the two-particle Green function. In the early papers the expression for D in the case of alloys was obtained by using the Hartree-Fock Approximation for the evaluation of the two-particle Green function. Now we calculate the two-particle Green function directly from equation of motion. Our aim is to find the local two-particle Green function. The calculations are made in the Wannier representation. We transform $\langle \langle a_{k-}^+ a_{k+}; a_{k'+}^+ a_{k'-} \rangle \rangle_{\omega}$ into the Wannier representation by the transformation

$$\begin{aligned} & \langle \langle a_{k-}^+ a_{k+}; a_{k'+}^+ a_{k'-} \rangle \rangle_{\omega} \\ &= \frac{1}{N^2} \sum_{\substack{l,j,m,p \\ \alpha,\beta,\gamma,\delta=A,B}} e^{i[k(l-j) - k'(p-m)]} \langle \langle a_{l-}^+ a_{j+}; a_{m+}^+ a_{p-} \rangle \rangle_{\omega}^{\alpha\beta\gamma\delta} * c_l^{\alpha} c_j^{\beta} c_m^{\gamma} c_p^{\delta}. \end{aligned} \quad (3)$$

Here c_l^{α} is zero-one operator (formula (7)).

The two-particle Green function $\langle \langle a_{l-}^+ a_{j+}; a_{m+}^+ a_{p-} \rangle \rangle_{\omega}^{\alpha\beta\gamma\delta}$ satisfies the equation

$$\begin{aligned} G_0(l^{\alpha} j^{\beta}; m^{\gamma} p^{\delta}) &= [\omega - \varepsilon_j^{\beta} + \varepsilon_l^{\alpha}]^{-1} \langle \langle [a_{l-}^+ a_{j+}; a_{m+}^+ a_{p-}] \rangle \rangle_{\alpha\beta\gamma\delta} \\ &+ \sum_{r \neq l \neq j} \sum_{\varepsilon=A,B} [t_{jr}^{\beta\varepsilon} G_0(l^{\alpha} r^{\varepsilon}; m^{\gamma} p^{\delta}) - t_{rl}^{\varepsilon\alpha} G_0(r^{\varepsilon} j^{\beta}; m^{\gamma} p^{\delta})] \\ &+ t_{lj}^{\alpha\beta} [G_0(l^{\alpha} l^{\alpha}; m^{\gamma} p^{\delta}) - G_0(j^{\beta} j^{\beta}; m^{\gamma} p^{\delta})] - I_l^{\alpha} G_0(l^{\alpha} l^{\alpha}; m^{\gamma} p^{\delta}) \delta_{lj} \delta_{\alpha\beta} \\ &+ I_j^{\beta} G_2(j^{\beta} j^{\beta} l^{\alpha} j^{\beta}; m^{\gamma} p^{\delta}) - I_l^{\alpha} G_1(l^{\alpha} l^{\alpha} l^{\alpha} j^{\beta}; m^{\gamma} p^{\delta}), \end{aligned} \quad (4)$$

where

$$\begin{aligned} G_0(l^{\alpha} j^{\beta}; m^{\gamma} p^{\delta}) &= \langle \langle a_{l-}^+ a_{j+}; a_{m+}^+ a_{p-} \rangle \rangle_{\omega}^{\alpha\beta\gamma\delta}, \\ G_1(l^{\alpha} l^{\alpha} l^{\alpha} j^{\beta}; m^{\gamma} p^{\delta}) &= \langle \langle n_{l+} a_{l-}^+ a_{j+}; a_{m+}^+ a_{p-} \rangle \rangle_{\omega}^{\alpha\alpha\alpha\beta\gamma\delta}, \\ G_2(j^{\beta} j^{\beta} l^{\alpha} j^{\beta}; m^{\gamma} p^{\delta}) &= \langle \langle n_{j-} a_{l-}^+ a_{j+}; a_{m+}^+ a_{p-} \rangle \rangle_{\omega}^{\beta\beta\alpha\beta\gamma\delta}. \end{aligned} \quad (5)$$

Here Greek superscripts indicate the type of atoms (A or B) at the given site. In order to solve equation (4) we apply the Random Phase Approximation to the functions G_1 and G_2 .

$$(G_1^{\text{RPA}} \simeq \langle n_{l+} \rangle (1 - \delta_{lj} \delta_{\alpha\beta}) \cdot G_0(l^\alpha j^\beta; m^\gamma p^\delta) \text{ and } G_2^{\text{RPA}} = 0.)$$

In this way we get the following equation for the function G_0

$$\begin{aligned} G_0(l^\alpha j^\beta; m^\gamma p^\delta) \simeq & [\omega - \varepsilon_j^\beta + \varepsilon_l^\alpha + I_l^\alpha \langle n_{l+} \rangle (1 - \delta_{lj} \delta_{\alpha\beta}) + I_l^\alpha \delta_{lj} \delta_{\alpha\beta}]^{-1} \\ & * [\langle [a_{l+}^+ - a_{j+}^+; a_{m+}^+ a_{p-}^+] \rangle_{\alpha\beta\gamma\delta} + t_{lj}^{\alpha\beta} \{G_0(l^\alpha l^\alpha; m^\gamma p^\delta) \\ - G_0(j^\beta j^\beta; m^\gamma p^\delta)\} + & \sum_{r \neq l \neq j} \sum_{\varepsilon=A,B} \{t_{jr}^{\beta\varepsilon} G_0(l^\alpha r^\varepsilon; m^\gamma p^\delta) - t_{rl}^{\varepsilon\alpha} G_0(r^\varepsilon j^\beta; m^\gamma p^\delta)\}]. \end{aligned} \quad (6)$$

The function G_0 depends on the distribution of impurity atoms in crystal. We may introduce zero-one operators c_l^α to equation (6). Our operators c_l^α will be expressed by standard c_l operators (see Edwards, Jones [5]) by the relation

$$c_l^\alpha = 1 - \delta_{B\alpha} + c(2\delta_{B\alpha} - 1). \quad (7)$$

Equation (6) may be solved by iteration. We assume that the ratio of t_{lj}/I_l is small for the ferromagnetic transition metals. In the following calculations we will consider only the case of strong ferromagnet where in the ground state all the electrons spins are aligned in the same direction. We make the assumption that $\langle n_{l\sigma} \rangle$ is the same for all A and B sites. After some manipulation we obtain the following expression for the two-particle Green function, neglecting the terms of higher order than $(t_{lj}/I_l)^3$.

$$c_l^\alpha c_j^\beta c_m^\gamma c_p^\delta \cdot G_0(l^\alpha j^\beta; m^\gamma p^\delta) \approx G_0(l^\alpha j^\beta; j^\beta l^\alpha) \delta_{lp} \delta_{jm},$$

where

$$\begin{aligned} G_0(l^\alpha j^\beta; j^\beta l^\alpha) = & W_1 + c_l^B W_2 + c_j^B W_3 + c_l^B c_j^B W_4 + W_5 \sum_{r \neq l \neq j} c_r^B \\ & + W_6 c_l^B \sum_{r \neq l \neq j} c_r^B + W_7 c_j^B \sum_{r \neq l \neq j} c_r^B + W_8 c_l^B c_j^B \sum_{r \neq l \neq j} c_r^B, \end{aligned} \quad (8)$$

where

$$W_1 = x(A, A) + (z-1) \cdot y(A, A),$$

$$W_2 = p(B, A) + (z-1) \cdot s(B, A) - W_1,$$

$$W_3 = -W_1 + p(A, B) + (z-1) \cdot u(A, B),$$

$$W_4 = -W_1 - W_2 - W_3 + x(B, B) + (z-1) \cdot r(A, B),$$

$$W_5 = -r(B, A),$$

$$W_6 = -W_5 - s(B, A) + u(B, A),$$

$$W_7 = -W_5 - u(A, B) + s(A, B),$$

$$W_8 = -W_5 - W_6 - W_7 - r(A, B) + y(B, B),$$

$$\begin{aligned}
u(\alpha\beta) &= b(\alpha\beta)d(\alpha\beta) [t^2(\beta\beta)d(\alpha\beta) + t^2(\alpha\beta)d(\beta\beta) + 2t^2(\alpha\beta)t(\beta\beta)[f(\alpha)d(\alpha\beta) - f(\beta)d(\beta\beta)]], \\
s(\alpha\beta) &= b(\alpha\beta)d(\alpha\beta) [t^2(\beta\beta)d(\alpha\beta) + t^2(\alpha\beta)d(\beta\beta) + 2t^2(\alpha\beta)t(\beta\beta) [f(\alpha)d(\alpha\beta) - f(\beta)d(\beta\beta)]], \\
p(\alpha\beta) &= b(\alpha\beta) [1 + 2t^2(\alpha\beta)d(\alpha\beta) \{f(\alpha) + f(\beta)\}], \\
x(\alpha\beta) &= b(\alpha\beta) [1 + 4t^2(\alpha\alpha)d(\alpha\alpha)f(\alpha)], \\
y(\alpha\alpha) &= b(\alpha\alpha)2t^2(\alpha\alpha)d^2(\alpha\alpha), \\
r(\alpha\beta) &= b(\beta\beta)d(\beta\beta) \{t^2(\alpha\beta) [d(\beta\alpha) + d(\alpha\beta)] + 2t(\beta\beta)t^2(\alpha\beta)f(\beta) [d(\beta\alpha) - d(\alpha\beta)]\}, \\
d(\alpha\beta) &= [\omega - \varepsilon_\beta + \varepsilon_\alpha + A_\alpha]^{-1}, \\
b(\alpha\beta) &= d(\alpha\beta) \langle a_{j+}^+ a_{j+} \rangle_{\beta\beta}, \\
f(\alpha) &= [\omega + I_\alpha]^{-1}, \\
t(\alpha\beta) &= t^{\alpha\beta}, \\
A_\alpha &= \langle n_\alpha^\alpha \rangle I_\alpha.
\end{aligned} \tag{9}$$

In formulae (8) and (9) there are three different hopping integrals t^{AA} , t^{BB} , and $t^{AB} = t^{BA}$. In our calculations t^{AB} is set equal to the average of $t^{AA} + t^{BB}$. We consider only the change induced when A atom is replaced by B atom. Therefore we may rewrite

$$t_{AB} = t_{AA} + h, \tag{10}$$

and

$$t_{BB} = t_{AA} + 2h,$$

where

$$h = \frac{1}{2}(t_{BB} - t_{AA}). \tag{11}$$

In formula (8) we assumed that the hopping integral is different from zero only for nearest neighbours.

Let us consider a simple two-dimensional square lattice. We take a cluster consisting in a central atom and its nearest neighbours. We divide all crystals into such clusters. In figure 1 we denoted the sites which were included in the summation in formula (8). In our averaging procedure we consider any central atom and all of the correlations from nearest neighbours. The summation is over all atoms in the crystal (formula (8)). By solving equation (6) we obtain an expression for two-particle Green functions, whose values depend on the local value of ε_l , t_{lj} and I_l . In formula (2), which determines the spin wave stiffness constant, there are sums over all l and j sites in the crystal. This sums we calculate for two cases:

1. we take an appropriate average of equation (8) over possible distributions of impurity atoms;
2. we assume a specified distribution of impurity atoms, in matrix (cluster model).

In the first case we are using the Edwards methods [5] to obtain the configurational average of two-particle Green functions in CPA

$$G_0(lj; j'l) = W_1 + c[W_2 + W_3 + (z-1)W_5] + c^2[W_4 + (z-1)W_6 + (z-1)W_7] + c^3(z-1)W_8. \quad (12)$$

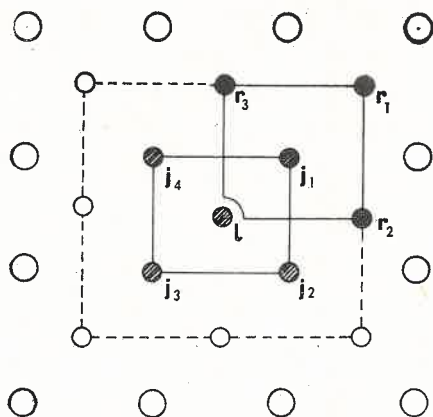


Fig. 1. Schematic diagram illustrating the two dimensional cluster consisting of a central atom and the nearest neighbours. The central atoms are chosen at l, j_1, j_2, j_3 and j_4 sites

Substituting (12) into (3) and (2) we get the final formula for the spin wave stiffness constant for disordered alloys. Numerical results are described in Section 3. In the second case we assume some ordering in alloys. The whole crystal is divided into small clusters consist-

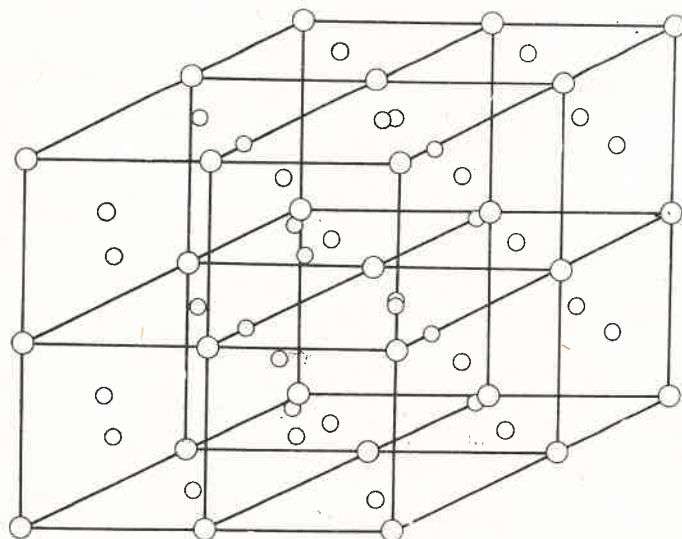


Fig. 2. Cluster model used in the numerical calculation

ing of 63 atoms (figure 2). Every cluster is formed by a central atom surrounded by its z nearest neighbours, by z_1 second neighbours, z_3 third, and z_4 fourth neighbours. Then we included into our cluster eight corner atoms so that we get cluster presented in figure 2. Early experiments have pointed out that NiFe alloys have a strong tendency to order. Mikke et al. [6] have shown experimentally and theoretically, basing on the Heisenberg model, that spin wave stiffness constant have the anomalous behaviour near 25% Fe. Then in our calculations we investigate NiFe alloy with 20.6% Fe (i. e. 13 atoms in cluster) The numerical results are described in Section 3.

3. Model calculation for nickel alloys. Numerical results

We consider the strong ferromagnetic alloys. Our calculations are made in the tight binding approximation for electron energy. We take

$$\varepsilon_k = 4t[3 - \cos(\frac{1}{2} ak_x) \cos(\frac{1}{2} ak_y) - \cos(\frac{1}{2} ak_x) \cos(\frac{1}{2} ak_z) - \cos(\frac{1}{2} ak_y) \cos(\frac{1}{2} ak_z)], \quad (13)$$

where $t = W/16$, W is the bandwidth and a the lattice parameter. The values of the sums $\frac{1}{N} \sum_k n_{k,+} f(k)$, where $f(k) = |\nabla_k \varepsilon_k|^2$ or $\nabla_k^2 \varepsilon_k$ were obtained by integration over $1/8$ at the first Brillouine Zone for the f. c. c. lattice. The mesh of 125000 points was used. The Fermi energy ε_F is determined from the equation

$$\frac{1}{N} \sum_{k\sigma} n_{k\sigma} = n. \quad (14)$$

Here n is the number of holes.

In the following calculations we used the notation

$$\frac{1}{N} \sum_k n_{k,+} |\nabla_k \varepsilon_k|^2 = W^2 a^2 \frac{P(A)}{16^2} \quad (15a)$$

$$\frac{1}{N} \sum_k n_{k,+} \nabla_k^2 \varepsilon_k = W a^2 \frac{L(A)}{16} \quad (15b)$$

$$A = \frac{4\varepsilon_F}{W}. \quad (15c)$$

The numerical values of functions L , P and n are presented in Figures 3 and 4.

Now we rewrite the formula (2) in the form

$$D(c) = \frac{W a^2}{96(n_+ - n_-)} [L(A) - \frac{1}{8} W \cdot G \cdot P(A)], \quad (16)$$

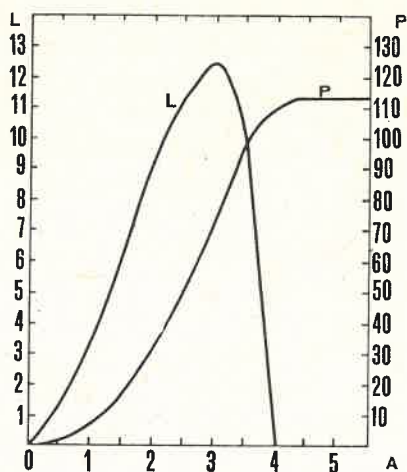


Fig. 3. Plots of the functions L and P versus $A = 4 \varepsilon_F/W$

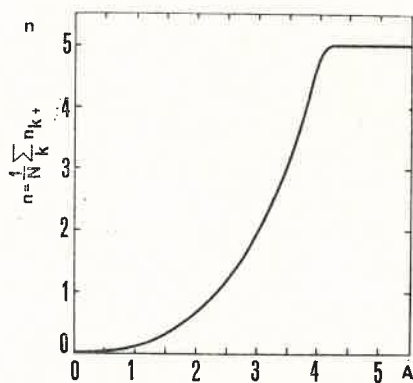


Fig. 4. Plot of the function n versus $A = 4 \varepsilon_F/W$

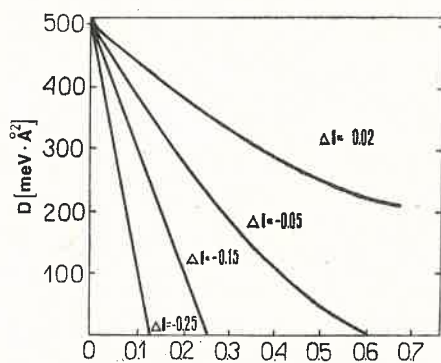


Fig. 5. Plot of exchange spin wave stiffness constant D versus concentration Fe for different ΔI

where $G = \frac{1}{Nz} \sum_{lj} G_o(lj;jl)$, and $G_o(lj;jl)$ is determined by Eq. (8). For pure nickel we choose the following values of the parameters: $n = 0.6$, $I = 0.95$ eV, $W = 1.2$ eV. The value of D for such parameters is 542 meV $\cdot \text{\AA}^2$ in agreement with the observed value at $T = 0^\circ\text{K}$ (Stringfellow [7]). The values of atomic potentials were taken from Hasegawa and Kanamori [8]. Then we took the values of bandwidth for Fe and Co as Cornwell et al. [9] and Batallan et al. [10], respectively. We examined the dependence of D for NiFe alloy on the value of $\Delta I = I_{\text{Fe}} - I_{\text{Ni}}$. The results are presented in Fig. 5. For the ΔI negative we obtained the critical concentration at which D vanishes.

Substituting (12) into (3) and (2) and using the coherent potential approximation we made numerical calculations for NiFe and NiCo alloys. We took similar values of intraatomic Coulomb interaction and bandwidth as Cornwell et al. [9] and Batallan et al.

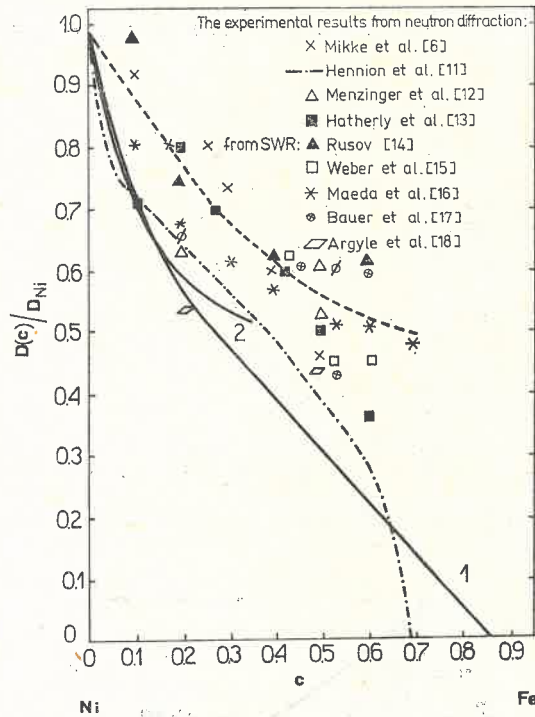


Fig. 6. Value of $D(c)/D_{\text{Ni}}$ as a function of impurity concentration c for NiFe alloy. The full curve 1 is reproduced from Riedinger et al. [3] the curve 2 presents Hill and Edwards [2] results and the broken curve corresponds to the method described in the text

[10]. On Fig. 6 we presented our theoretical results. Points denote experimental data, the solid lines are the theoretical results of other authors. Curve 1 presents Riedinger and Nauciel-Bloch [3] results and curve 2 those of Hill and Edwards [2]. The broken line across experimental points corresponds to our theoretical results. As can be seen from

Fig. 6 our model and method of calculations (without using the Hartree-Fock Approximation) gives reasonable agreement with experimental data for chosen parameters [8-10].

Then we apply our model to NiCo alloys. Numerical results will be obtained for the values of parameters assumed as Batallan et al. [10]. For such parameters we give the dependence of the spin wave stiffness constant on concentration presented in Fig. 7. Our results are close to the Yamauchi et al. [19] experimental data, but are different from

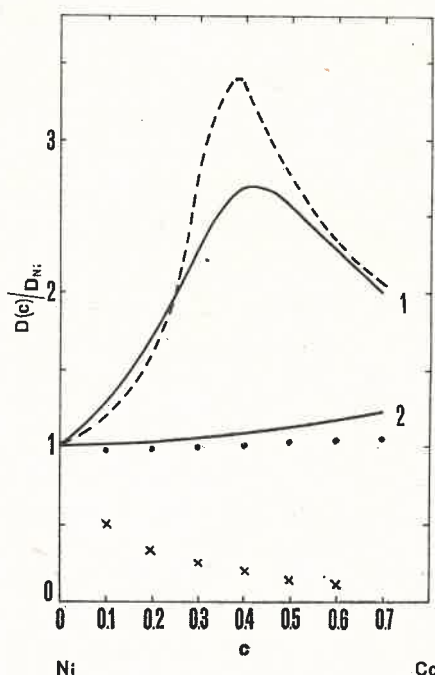


Fig. 7. Values of $D(c)/D_{Ni}$ as a function of impurity concentration c for NiCo alloy. The full curve 1 is the RPA-CPA results (Hill and Edwards [2]), the curve 2 presents our results for parameters described in the text. The broken curve is rigid band results (Wakoh [21]). Points denoted experimental data: \times Hinoul and Witters [20]; \bullet Yamauchi et al. [19]

the Hinoul et al. [20] experimental data and the Wakoh [21] theoretical results. The difference is that our model included change of hopping integral and we did not apply the Hartree-Fock Approximation in calculations of the two-particle Green function. Next the spin wave stiffness constant D was calculated for the cluster model described in Section 2. We made numerical calculations for three cases, corresponding to the concentration of impurity which was assumed as 20.6%:

- case 1 — the Fe atoms are in the center of cluster and the rest of atoms occupy the first shell,
- case 2 — the Fe atoms are in second, third and fourth shell,
- case 3 — seven atoms Fe occupy the first shell and six atoms of Fe are distributed in the second shell.

The values of D for these three different distributions of Fe atoms in cluster we listed in Table I. As it is evident from Table I the value of spin wave stiffness constant D depends on the distribution of impurity atoms in the matrix. Similar results were obtained by Mikke et al. [6].

TABLE I

Concentration	Distribution of Fe atoms	Stiffness constant D [meV Å ²]
20.6%	1	471
	2	390
	3	445
	CPA (disorder)	430

4. Conclusions

Hill and Edwards [2] discussed the problem of magnon energy in the ferromagnetic transition metal alloys using the Hartree-Fock Hamiltonian. In the limit of pure metal they obtained the RPA results for D (see Edwards [22]). The calculations we report in this paper were done within the Random Phase Approximation. We applied this approximation to the higher Green function in the equation of motion for the two-particle Green function. Our iterative method described in Section 2 is true if $t/\Delta < 1$. The numerical results showed that the contributions from the terms higher than $(t/\Delta)^3$ were very small. For pure metal our results give the RPA result and some contribution from the band structure. If we compare our method of calculation of the two particle Green function with Hill's and Edwards' one [2] we find that our method may give results beyond the RPA. All calculations described here may be applied to the weak ferromagnetic alloys. In recent paper Edwards and Hill [2] showed that the spin wave stiffness constant is very sensitive to the assumed exchange splitting in the pure nickel. In this paper we presented the influence of the change $\Delta I = I_{\text{Fe}} - I_{\text{Ni}}$ on the final value of D for NiFe alloys and made calculations assuming the different value of hopping integral t^{AA} , t^{AB} , and t^{BB} . The value of parameters was taken from Hasegawa and Kanamori [8], Cornwell et al. [9] and Batallan et al. [10], and our results are in agreement with the experimental values.

At last, assuming simple cluster model in which we consider interaction between all nearest neighbours, we calculated the dependence D of the distribution impurities atoms in cluster for three different cases. Our results are similar to those of Mikke et al. [6].

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