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H. Ebert, *University of Bristol*
H. Winter
B. Gyorffy, *University of Bristol*
Duane D. Johnson
F. J. Pinski, *University of Cincinnati*

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THEORETICAL STUDY OF THE ENVIRONMENTAL EFFECTS ON THE HYPERFINE FIELDS OF Ni AND Fe IN $Ni_{0.75}Fe_{0.25}$

H. Ebert[+], H. Winter[o], B. Gyorffy[+], D.D. Johnson[++], and F.J. Pinski[oo]

[+]H.H. Wills Phys. Lab., Univ. Bristol BS8 1TL, UK

[o]Kernforschungszentrum Karlsruhe, Institut für Technische Physik, P.O.B. 3640, D-7500 Karlsruhe, FRG

[++]Naval Research Lab., Washington, D.C. 20375, USA

[oo]Department of Physics, University of Cincinnati, Cincinnati, Ohio 45221, USA

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The dependence of the hyperfine fields B_{hf} of Ni and Fe in $Ni_{0.75}Fe_{0.25}$ on the surrounding atomic configuration has been studied by performing charge selfconsistent 6 shell-cluster Korringa-Kohn-Rostoker Coherent Potential Approximation (KKR-CPA) bandstructure calculations. By replacing the CPA-scatterers in the various shells around the central atom by Ni- or Fe-atoms, respectively, it could be shown that the hyperfine fields vary linearly with the number of Fe-atoms within a given shell and that the changes of B_{hf} due to simultaneous changes of the atomic configurations of different shells are additive. The changes of the hyperfine fields upon ordering of $Ni_{0.75}Fe_{0.25}$ solid solution as deduced from our calculations agrees reasonably well with experiments.

Mößbauer spectroscopy and ferromagnetic nuclear resonance (FMR) have been extensively used to investigate the short range order in Ni_xFe_{1-x}. The permalloy and invar region of concentration (e.g. Ref. 1-5) have been the subject of particularly keen interest. In contrast to the classical methods of X-ray diffraction and neutron scattering these resonance techniques probe only a relatively small region around the resonating nucleus. However they do not suffer from the difficulty of X-rays and neutron scattering for which Ni- and Fe-atoms are very similar (see e.g. references given in Ref. 1). Unfortunately, for the interpretation of the experimental Mößbauer and NMR-data a number of simplifying assumptions has to be made. Most workers assumed that there is a linear relation between the hyperfine field B_α (α = Fe, Ni) and the number of Fe atoms in a surrounding atomic shell and that at most the configuration of the first two neighbouring shells determine B_α: Namely, it is assumed that

$$B_\alpha = B_\alpha^o + \Delta B_\alpha^1 \cdot n^1 + \Delta B_\alpha^2 \cdot n^2 \qquad (1)$$

where B_α^o is the hyperfine field for an α-atom with only Ni on its first two neighbouring shells and the hyperfine field coefficients ΔB_α^i give the change of B_α due to an occupation of the sites on shell i by n^i Fe-atoms.

The primary aim of the present work is to investigate how well the above assumptions are justified. For this purpose charge self-consistent cluster KKR-CPA [6] calculations have been performed for 6-shell disordered $Ni_{0.75}Fe_{0.25}$ clusters. To investigate the dependence of the electronic structure of a (central) Ni- or Fe-atom on its atomic surrounding we have calculated charge self-consistently (for the central site), its electronic structure assuming that all atoms of one of the surrounding atomic shells are occupied by Ni- or Fe-atoms respectively, while all other shells are occupied by CPA-scatterers. Because our programs were designed to make full use of the cubic symmetry of the central site, we studied only those extreme configurations for which all shells were occupied by one type of scatterer. The resulting local density of states curves for Ni with a nearest neighbor (NN) shell of Ni- or Fe-atoms are compared in Fig. 1 to the all CPA results. Because the d-phase shifts for the spin-up (majority) electrons of Fe and Ni in Ni_xFe_{1-x} nearly coincide the spin-up subsystem behaves almost virtual crystal like. For this reason, replacing the CPA scatterers on the NN shell by Ni or Fe-atoms affects the density of states curves only moderately. In contrast to this behaviour, one finds drastic changes for the spin-down bands. As one would expect, the changes are more pronounced when we set Fe on the

[+]Present address: Siemens AG, ZFE TPH1, Paul-Gossen-Str. 100, D-8520 Erlangen, FRG

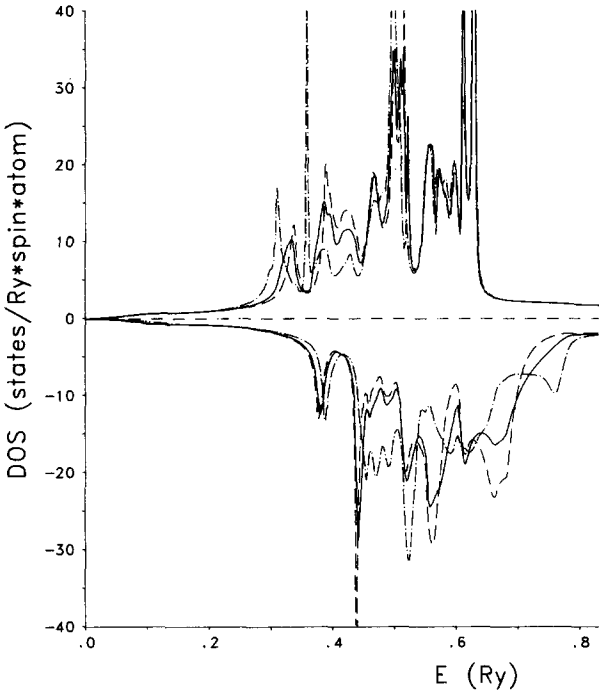

Fig. 1

Partial density of states for Ni in $Ni_{0.75}Fe_{0.25}$ embedded in the CPA-medium (——) and surrounded in addition by a nearest neighbour shell of Ni (---) or Fe (-·-·) atoms respectively. The upper part is for spin up, the lower one for spin down.

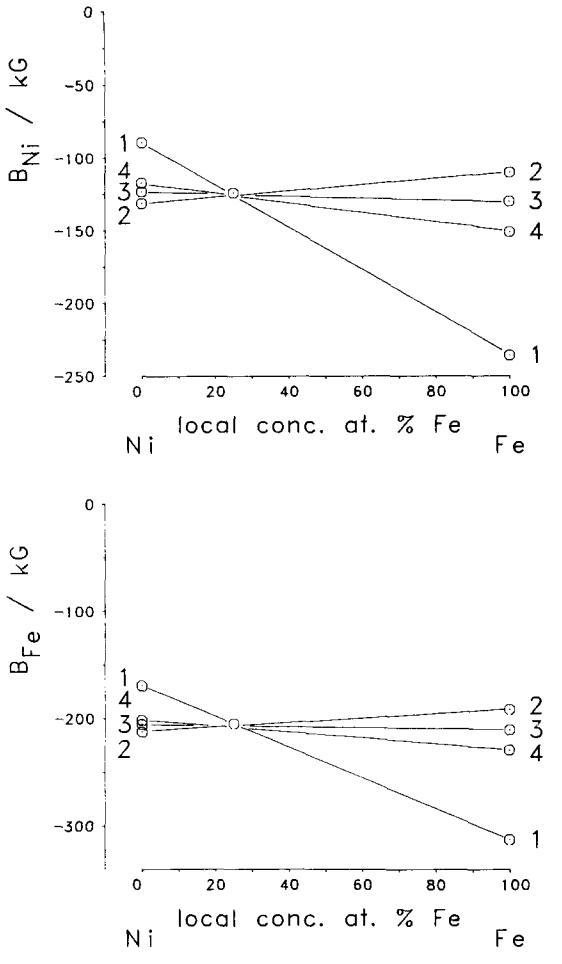

Fig. 2

Variation of the hyperfine fields of Ni (a) and Fe (b) with the configuration within one of the surrounding atomic shells. The numbers label the shells according to table 1, where also the number of atoms per shell are given. Results for shell 5 have been omitted for the sake of clarity.

NN shell than Ni, because the Ni-content in $Ni_{0.75}Fe_{0.25}$ is three times higher than that of Fe. In the case of Ni NN's the primary effect on the curves is to sharpen the peaks.

The hyperfine fields for Ni and Fe, that emerged from our calculations are displayed in Figs. 2a) and 2b). The most striking result is that for all configurations studied, the CPA-result lies more or less exactly on the straight line connecting the hyperfine fields for the two extreme configurations of a given shell (only Ni- or Fe-atoms). This behaviour has also been found for all other concentrations of Ni_xFe_{1-x} that we considered [7]. These findings lead us to the conclusion, that the hyperfine fields indeed depend linearly on the number of Fe-atoms in a shell and therefore justify one of the assumptions on which eq. (1) is based. Note that the above result is in some variance with the work of Kanashiro and Kunitomi [8] who claim to have found from their Mößbauer spectra of Fe in Ni_xFe_{1-x} alloys, with x around 0.75 a slight nonlinear dependence of B_{Fe} on n^1. However, in fitting their experimental spectra, these workers took only 6 atomic configurations into account and made the severe assumption that ΔB^i_{Fe} is identical zero. This later assumption is however in striking contrast to the experimental results of others [1,2,5] and also to our hyperfine field coefficients, which are given in Tab. 1. Beside the close similarity of the coefficients for Ni and Fe, the most interesting result is that in this alloy B_α is not only determined by the two next neighboring shells but also by the outer ones. In Fig. 3 ΔB^i_{Fe} has been plotted against the distance of shell i from the central site. The tentatively drawn connecting line obviously shows Friedel-like oscillations and is very similar to the curve which describes the satellite Knight shift distribution of (e.g.) Cu-atoms around a Mn-impurity [9]. Interestingly,

Table 1

shell i	number of atoms in shell	ΔB^i_{Ni}/kG	ΔB^i_{Fe}/kG
1	12	-12.4	-11.9
2	6	+4.4	+3.5
3	24	-0.22	-0.17
4	12	-2.5	-2.2
5	24	-0.08	-0.09

Hyperfine field coefficients ΔB^i_α for Ni and Fe for the first 5 surrounding atomic shells as deduced from 6-shell cluster calculations (see Fig. 2).

in a calculation of the spin polarization on the host Pd atoms by Fe impurities significant contributions were found by Oswald et al. [10] from a considerable number of nearest neighbour shells. Evidently, our results reflect the same long range correlation phenomena. Moreover, quite recently, the same group studied the hyperfine fields of impurities in Ni and their NN Ni-atoms. For Fe in Ni they find ΔB^1_{Ni} to be -19 kG. This is of same order of magnitude and sign as our result for $Ni_{0.75}Fe_{0.25}$ given in table 1. Because the first two shells give the most important contributions to B_α it is not completely senseless to compare the coefficients of table 1 to experimental values although these are based on eq. (1). Driver et al. [1] found from their extensive Mößbauer study on $Ni_{0.75}Fe_{0.25}$ alloys ΔB^1_{Fe} = -10.5 kG and ΔB^2_{Fe} = -3.3 kG. These results are very similar to those reported by Frackowiak [5] who gives ΔB^1_{Fe} = -10.5 kG and ΔB^2_{Fe} = -3.3 kG. Both values for ΔB^i_{Fe} agree reasonably well with that given in table 1. However, for ΔB^2_{Fe} the experiments differ in signs from the prediction of the theory. Unfortunately, the experimental values depend not only on the validity of eq. 1 but are also influenced by the fitting procedure by which they are deduced from experimental spectra. For example, in a recent paper, Cranshaw [2] finds for slightly Fe-rich ordered $Ni_{0.75}Fe_{0.25}$ ΔB^1_{Fe} = -8.3 kG and ΔB^2_{Fe} = +9.3 kG, where the last value is assumed to be less certain. To check the additivity of the contributions to B_α coming from different shells, we have also performed calculations for clusters, for which the first two shells were occupied by Ni- and/or Fe-atoms. The resulting hyperfine fields should then be given by

$$B_\alpha = B_\alpha^{CPA} + \sum_i \Delta B^i_\alpha (n^i - \bar{n}^i) \qquad (2)$$

Here B_α^{CPA} is the CPA result for B_α and $\bar{n}^i$ is the average Fe occupation number of shell i corresponding to the concentration x = 0.75. As can be seen from Table 2, where calculated hyperfine fields are compared to ones obtained using eq. (2), the additivity of the shell contributions is obviously nicely fulfilled.

Using the coefficients ΔB^i_α given in table 1 it is possible to estimate the change of B_α upon ordering of disordered $Ni_{0.75}Fe_{0.25}$. For Ni and Fe we find -10 kG and +30 kG which is to be compared to the experimental values of -20 kG [3,12,13] and +12 kG [14,15] respectively. For the change of B_{Fe} due to a positioning of Fe on a Ni-site instead on a Fe-site in ordered $Ni_{0.75}Fe_{0.25}$, we find -45 kG while Cranshaw [5] deduced a value of -46 kG from his experimental work.

Within our study, we have also considered the influence of relativistic effects by calculating the hyperfine matrix elements completely relativistically. The change of the hyperfine fields compared to the non-relativistic ones for Ni and Fe that emerged are exactly the same as those found recently by Blügel et al. [11] for Ni and Fe dissolved in Ni. Be-

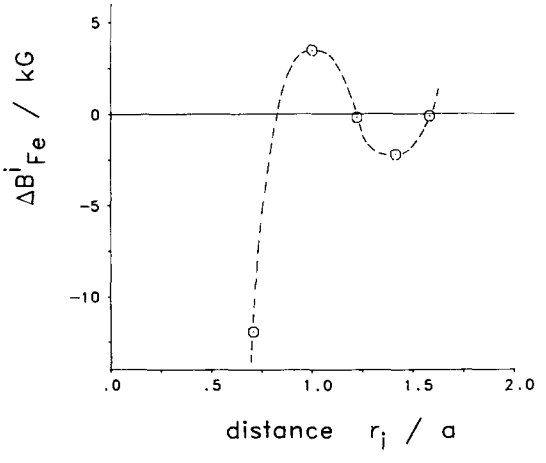

Fig. 3

Hyperfine field coefficients ΔB^i_{Fe} plotted against the distance of shell i from the central site.

Table 2

Configuration shell i (N=Ni F=Fe, C = CPA) 1 2 3 4 5	B_{Ni}/kG eq. (2)	B_{Ni}/kG band-structure	rel. diff. %	B_{Fe}/kG eq. (2)	B_{Fe}/kG band-structure	rel. diff. %
N N C C C	- 93.6	- 91.3	2.4	-173.9	-173.2	0.4
F N C C C	-242.4	-235.5	2.8	-316.8	-312.6	1.3
F F C C C	-216.0	-220.3	1.9	-295.8	-298.9	1.1
N F C C C	- 67.2	- 70.3	4.6	-152.9	-156.3	2.1
N C N C C	- 85.7	- 84.7	1.1	-167.7	-168.5	0.4
F C N C C	-234.5	-233.9	0.2	-310.5	-311.2	0.2
F C F C C	-239.8	-238.3	0.6	-314.6	-315.2	0.2
N C F C C	- 91.0	- 87.2	4.1	-171.8	-169.2	1.4
N C C N C	- 79.6	- 79.5	0.1	-162.3	-165.2	1.8
F C C N C	-228.4	-234.7	2.7	-305.0	-313.2	2.6
F C C F C	-258.0	-246.1	4.6	-330.9	-320.3	3.1
N C C F C	-109.2	-117.8	7.8	-188.0	-197.2	4.8

Hyperfine fields of Ni and Fe on the central site, surrounded by different atomic configurations according to eq. (2) and as obtained by bandstructure calculations.

cause these changes do not affect the conclusions drawn from our non-relativistic results (Table 1), the relativistic results will be given elsewhere [7].

In summary, one can say that our work strongly supports the fundamental assumption of most Mößbauer investigations of Ni_xFe_{1-x} that the contributions to the hyperfine field B_α from different neighbouring shells are additive and that each contribution depends linearly on the occupation number of the corresponding shell. Although the numerical results for the hyperfine field coefficients for the outer shells are presumably affected by the finite cluster size we used, they suggest that in many alloy systems the summation in eq. 1 cannot be restricted to the first two neighbouring shells.

Finally, it is illuminating to compare the above discussion of the hyperfine fields with investigations of the environmental effects on the local moments in alloys. Denoting the fluctuating magnetic moment on the i-th site occupied by an α type atom by

$$\mu_i^\alpha = \bar{\mu}_i^\alpha + \sum_j \gamma_{ij}^\alpha \, \delta C_j \qquad (3)$$

where $\bar{\mu}_i^\alpha$ is the averaged moment on an α type atom and δC_j is a fluctuation of the probability C_j that the j-th site is occupied by an α atom from its average value $\bar{C}$ we see that the coefficients γ_{ij} play the same role as ΔB_j^α in eq. 2. Interestingly γ_{ij}^α can be measured in a spin polarized quasi-elastic neutron scattering experiment and has been studied for many alloys. Moreover, a theory of γ_{ij}^α has been developed by Staunton et al. [16] on the basis of a spin-polarized self-consistent KKR-CPA theory. In this theory which in the few instances where it has been implemented appears to agree with experiments, γ_{ij}^α is identified with the derivative $\partial\mu_i^\alpha/\partial C_j$ and hence eq. 3 is viewed as a small δC_j expansion. If we assume that the local hyperfine field scales with the concentration fluctuation of environment in the same way as the local magnetic moment then the cluster calculations reported here can be interpreted as implying that eq. 3 can be fruitfully used even when δC_j is not small. Evidently, the linear dependence of the local magnetic moment on the compositional fluctuation of the environment is an important simplification whose range of validity is of considerable general interest. Our results suggest that this range is surprisingly large.

Having noted that the forms of eq. 2 and 3 are governed by closely related physics we suggest that directly comparable neutron scattering and Mößbauer studies would be most effective in making progress with understanding the interactions between magnetic and compositional fluctuations.

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