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ABSTRACT

The effect of aluminium addition on the magnetic moments of the constituent atoms of binary bcc Fe-Co, Fe-Cr and fcc Ni-Fe alloys was investigated by comparing the results of Mössbauer and average magnetization measurements. It was found that the Al neighbours do not perturb the moment of Fe atoms in the Fe-Co and Ni-Fe alloys whereas there is a decrease in the Co and Ni moments. This different behaviour of Fe, Co and Ni towards the Al diluent is explained in terms of a simple local model.

АННОТАЦИЯ

С помощью сравнения результатов измерения эффекта Мессбауэра и средней намагниченности было исследовано влияние, оказываемое алюминием на магнитный момент составляющих атомов двухкомпонентных сплавов объемно-центрированных Fe-Co, Fe-Cr и плоско-центрированного кубического сплава Ni-Fe. Было обнаружено, что соседние атомы алюминия не изменяют моментов атомов железа в сплавах Fe-Co и Ni-Fe, в то время как снижают моменты Co и Ni. Объяснение такого различного поведения, показанного моментами Fe, Co и Ni при добавлении к ним Al, мы проводим в рамках простой локальной модели.

KIVONAT

Aluminiumnak a kétalkotós tck Fe-Co, Fe-Cr és fck Ni-Fe ötvözetek alkotó atomjainak mágneses momentumára gyakorolt hatását vizsgáltuk a Mössbauer és átlagmágnesezettség mérések eredményeinek összehasonlításával. Azt találtuk, hogy az Al szomszédok nem változtatják meg a Fe atomok momentumát a Fe-Co és Ni-Fe ötvözetekben, mig a Co és Ni momentumokat csökkentik. A Fe, Co és Ni momentumok Al hozzáadására mutatott ezen különböző viselkedését egy egyszerü lokális modell keretein belül magyarázzuk.

1. INTRODUCTION

The study of ternary alloys is of great value not only in itself but in the understanding of binary alloys. One of the most investigated problems is the effect of Al substitution in different ferromagnetic matrices. It is well known, for example, that the Al impurities in an iron host cause a simple dilution of the average magnetization by $\frac{d\mu}{dc} = -2.2\,\mu_B$/Al atom whereas in a nickel host they result in a decrease much larger than simple dilution: $\frac{d\mu}{dc} = -2.8\,\mu_B$Al atom (Parsons et al. 1958, Crangle and Martin 1965). In agreement with this, diffuse neutron scattering measurements show that the Ni moments are appreciably decreased in a broad distance range around the Al impurities and there is no such moment perturbation in the Fe host (Comly et al. 1968, Holden et al. 1967). These quite different effects of the Al diluent towards the Fe and Ni matrices are not yet theoretically well understood. For example, attempts are made to attribute these effects to the different band structures of the Fe and Ni hosts (according to their different crystal structure)resulting in a different type of screening of the Al excess charge (by s- and d-like conduction electrons, respectively) (e.g. Mott 1964, Besnus and Meyer 1970).

Recent combined magnetization and Mössbauer measurements on bcc Fe-Ni alloys diluted by Al seem to rule out this above-mentioned possibility (Bardos et al. 1969, Vincze 1973). The average magnetization of these alloys shows an even larger decrease because of the Al diluent than does pure Ni, whereas the Mössbauer investigation showed that the Al neighbours do not cause any change in the magnetic moment of Fe atoms as in pure Fe. Thus the large decrease in the average magnetization (the saturation value is $\frac{d\mu}{dc}{}^{\text{Fe-Ni}}_{\text{Al}} = -4.0\,\mu_B$/Al atom) suggests a significant decrease in the magnetic moments of Ni in the environment of Al atoms similarly to that in pure Ni.

It seems that a reasonable account of the fundamental difference in the behaviour of Fe and Ni atoms towards Al diluent could be given in terms of a simple local model (Vincze 1973) based on one suggested by Marshall and published by Mott (1964). The model supposes an energy level arrangement similar to that of the free atom for the Fe and Ni atoms. If the $3s^2$ electrons of Al have a sharp localized level with an evergy above the level corresponding to the Ni 3d electrons yet below the level of the Fe 3d electrons, then these two Al 3s electrons can "overflow" only into the Ni atoms. At low Ni concentrations this results in a nearly linear variation of $\frac{d\mu_{Fe-Ni}}{dc_{Al}}$ with Ni concentration as the Ni neighbours of Al are too few to accommodate both 3s electrons. At higher Ni concentrations this leads to a saturation effect, corresponding to the accommodation of both excess Al-electrons by the Ni atoms. This behaviour was found by Bardos et al. (1969) in the total change of the average magnetization of bcc Fe-Ni alloys due to a single Al atom as a function of Ni concentration.

The aim of the present work is to study by Mössbauer measurement the effect of Al on the magnetic behaviour of iron atoms in ternary alloys containing Al and to complete the earlier magnetization measurements of Bardos et al. (1967) and Besnus Meyer (1970). In the cases of bcc Fe-Co and Fe-Cr, further valuable information is expected from these experiments for the interaction between the Al and the 3d constituents of the alloys, whereas in the case of fcc Ni-Fe alloys the non-dependence of this interaction on the crystal structure was investigated.

2. EXPERIMENTAL DETAILS AND RESULTS

The same alloys prepared for earlier magnetization measurements (Besnus and Meyer 1970) and completed with some new series were used. The Mössbauer measurements were performed at room temperature on powder specimens (grain size less than 50 μm) with a conventional constant-acceleration spectrometer using a 20 mCi ^{57}Co in Cr source. Data were stored in a 1024-channel analyzer. Each spectrum was taken with 300-500 x 10^3 counts per channel. The depth of the outer lines in the spectra is generally about 40-60 x 10^3 counts per channel. Specimens for saturation magnetization measurements were fabricated into prolate ellipsoids with dimensions 8.0 x x 4.0 mm. The experimental method and the apparatus employed have been described previously (Herr 1970). The saturation magnetizations were obtained by $\sigma(H^{-2})$ extrapolations and are considered to be accurate within ±0.2 %.

The Mössbauer spectra of the different alloys series required different evaluation techniques. In the following we detail the results for the individual binary alloy systems.

a/ bcc (Fe-Co)-Al

Figure 1 shows the results of the average magnetization measurements.

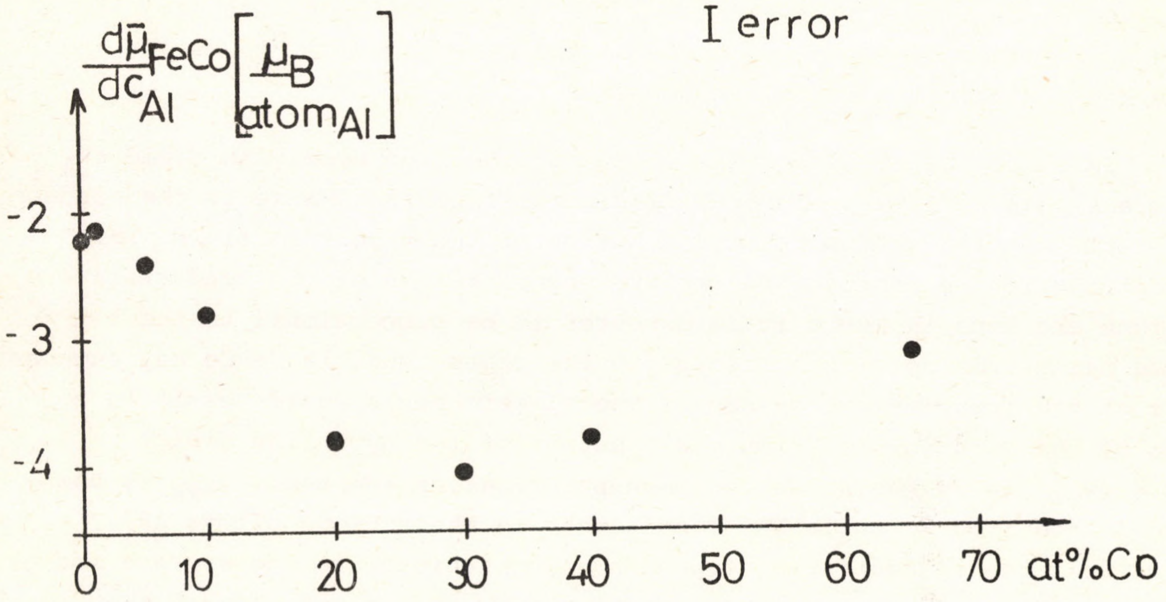

Fig. 1 *Total change of the average magnetizations of bcc Fe-Co alloys caused by a single Al atom as a function of Co concentration*

For the Mössbauer experiments four series of bcc alloys were chosen; in each the iron- to -cobalt ratio was increased. The four series were based on binary Fe-Co alloys containing 5, 10, 20 and 30 at% Co and diluted by 4 and 8 at% Al (or in part by 5 and 10 at% Al).

The evaluation of the Mössbauer spectra closely followed the same procedure used for the bcc (Fe-Ni)-Al alloys (Vincze 1973). The Mössbauer spectra of binary bcc Fe-Co alloys showed only a broadening without resolvable satellites (Johnson et al. 1963). Thus in the evaluation of the Mössbauer spectra of the bcc (Fe-Co)-Al alloys it was assumed that lines corresponding to the iron atoms with 0, 1, 2, ... first Al neighbours; that is, we determined merely the change of the hyperfine field and isomer shift at the iron atoms due to the Al neighbours. Moreover it was assumed that the probability of the occurrence of each configuration is given by a binomial distribution because of the disordered state and that the contributions are additive and independent of the actual arrangement of Al neighbours. The satisfactory description of the spectra and the consistent values of the parameters obtained from the fit support these suppositions. Figure 2 shows the change of the iron hyperfine field ΔH_1 due to a single Al first neighbour (the average is shown when two Al compositions were used for the evaluation of ΔH_1). There is a small increase in the absolute value of ΔH_1 with respect to the value obtained for binary Fe-Al alloys (Vincze and Cser 1972), which seems to be proportional to the average magnetization of the bcc Fe-Co alloys. The change in the isomer shift of iron atom Δi_1 caused by a single Al first neighbour is the same 0.020(3) $\frac{\text{mm}}{\text{sec}}$ as for binary Fe-Al within the experimental error.

In binary Fe-Al alloys the change in the iron hyperfine field ΔH_1 due to a single Al first neighbour originates from the change in the conduction electron polarization CEP contribution of the hyperfine field. This CEP contribution is proportional to the magnetization of the conduction electrons and thus in Fe-Co it is expected to be proportional to the average magnetization of the alloys. On the other hand, in Fe-Co any supposed change of the iron moments caused by the Al neighbours would result in a change of the core-polarization contribution of the hyperfine field: $H_{CP} = a\,\Delta\mu_{Fe}$ ($a \simeq 70$ kOe/μ_B is the core-polarization constant, $\Delta\mu_{Fe}$ is the change in the iron moment). The result that in these Fe-Co alloys ΔH_1 follows well the concentration dependence proportional to the average magnetization (Fig. 2) suggests that the Al neighbours do not perturb the moment of Fe atoms, similarly to the Fe-Al case.

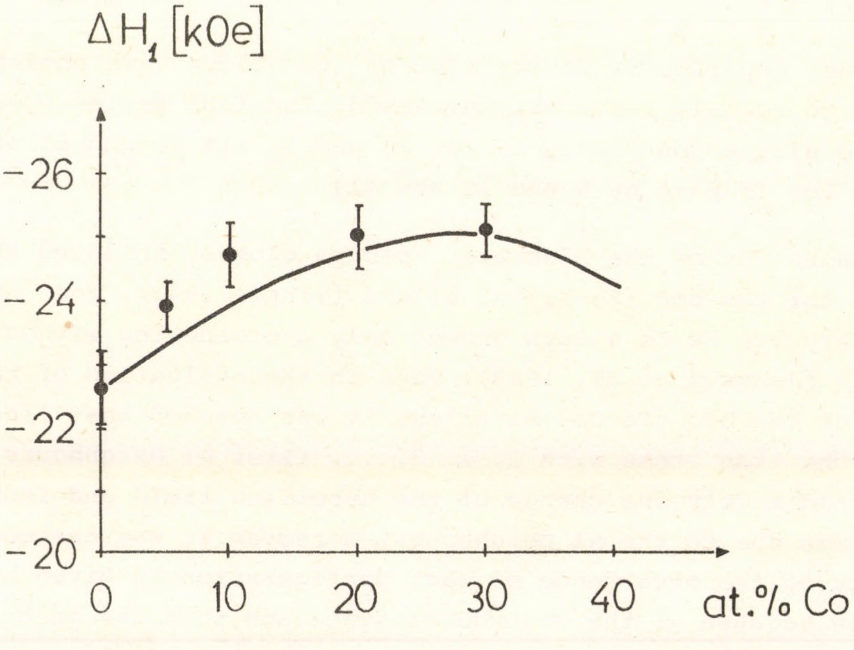

Fig. 2 *Change in the hyperfine field of iron atoms caused by a single first Al neighbour ΔH_1 as a function of Co concentration. The full line shows the concentration dependence proportional to the average magnetization of bcc Fe-Co alloys*

b/ fcc (Ni-Fe)-Al

Two series of fcc alloys were investigated, in which the Ni-to-Fe ratio was kept constant (namely 50 and 15 at% Fe) as the Al concentration was increased. Because of the weak resolution of the spectra in the case of the Ni-Fe (15 at%) series (diluted by Al from 2 to 10 at%) we could determine only the values of the average iron hyperfine field shown in Fig.3. The substitution of Al into this system causes a decrease in the iron hyperfine fields: $\frac{d\bar{H}_{Fe}}{dc_{Al}} = -(183 \pm 7)$ kOe.

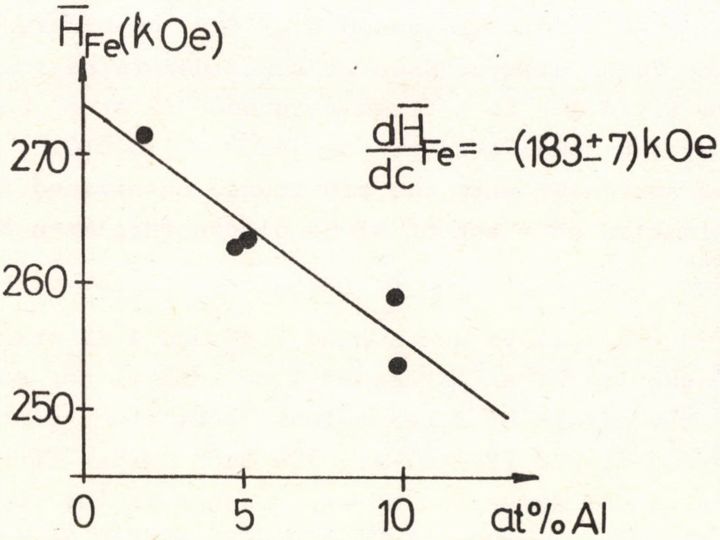

Fig. 3 *Room temperature values of the average iron hyperfine field in Ni-Fe (15 at%)-Al as a function of Al content*

From the average iron hyperfine fields we can estimate the decrease in the average magnetic moment of iron due to Al neighbours as follows. The average iron hyperfine field in the Ni-Fe alloys can be well represented by the frequently used phenomenological expression:

$$\bar{H}_{Fe} = a\mu_{Fe} + b\mu_{Ni-Fe} \qquad (1)$$

with the values a $\approx$ 90 kOe/ μ_B and b$\approx$ 60 kOe/ μ_B (Erich et al. 1969). The analogous expression for the total change in the average iron hyperfine field for the Al substitution is

$$\frac{d\bar{H}_{Fe}}{dc_{Al}} = a \frac{d\mu_{Fe}}{dc_{Al}} + b \frac{d\mu_{Ni-Fe}}{dc_{Al}} , \qquad (2)$$

where the measured value of the decrease in the mean magnetic moment of the Ni-Fe (15 at%) alloy is $\frac{d\bar{\mu}_{Ni-Fe}}{dc_{Al}} = -2.83\mu_B$ (Besnus and Meyer 1970). Thus the estimation for the second term in eq.2 is -170 kOe which agrees with the above-mentioned measured value of $\frac{d\bar{H}_{Fe}}{dc_{Al}}$ within the experimental error. Consequently $\frac{d\mu_{Fe}}{dc_{Al}} \approx 0$ from the zero estimation of the first term in eq.2, that is: magnetic moments of iron are not perturbed by the presence of Al.

The situation is rather different for the other investigated Ni-Fe series. The spectrum is much more broadened even for the binary Ni-Fe (50 at%) alloy than for the bcc Fe-Co alloys. Thus we could determine the increase in the iron hyperfine field due to a first neighbour Ni atom ΔH_1^{Ni} from the line broadening by the usual assumptions. $\Delta H_1^{Ni} = (10.0 \pm 0.4)$ kOe was obtained in very good agreement with the previously determined value from the Mössbauer investigation of a set of Ni-Fe alloys (Heilmann and Zinn 1967).

Two Ni-Fe (50 at%) alloys containing 1.96 and 4.92 at% Al were investigated. The assumption of an increased linewidth is not sufficient to take into account the effect of first neighbour Ni atoms contrary to the cases of bcc (Fe-Ni)-Al and (Fe-Co)-Al. The much larger linewidth enables the simultaneous determination of the changes in the iron hyperfine field caused by a first neighbour Ni, ΔH_1^{Ni} and that caused by first neighbour Al, ΔH_1^{Al}. The probability of the occurrence of a certain configuration is given by the superposition of the two binomial distributions. Within the experimental errors the same values were obtained for the two compositions, thus supporting the assumptions. The value of $\Delta H_1^{Ni} = (10.9 \pm 0.4)$ kOe agrees with that obtained on the binary Ni-Fe (50 at%) alloy, whereas $\Delta H_1^{Al} =$ $= (-19.7 \pm 1.2)$ kOe is a little smaller than that for the binary Fe-Al alloys.

Unfortunately, the detailed comparison of the hyperfine field changes caused by Al is difficult because the crystal structures are different in the pure Fe and the Ni-Fe (50 at%) cases. Furthermore, a smaller CEP contribution to ΔH_1^{Ni} is expected for the Ni-Fe (50 at%) alloy than for Fe if the radial dependence of the CEP contribution is nearly the same as in Fe, since the first neighbour distances are about identical, while the average magnetization of the alloy ($\bar{\mu} = 1.676\ \mu_B$) is smaller than that of Fe. Thus these data of the Ni-Fe (50 at%) alloy do not contradict the previous conclusion that the Al is a simple diluent for Fe in the fcc Ni-Fe alloys.

Figure 4 shows the concentration dependence of the total change in the average magnetization due to a single Al atom in these fcc Ni-Fe alloys and in those measured by Bardos, Aldred and Beck (1967).

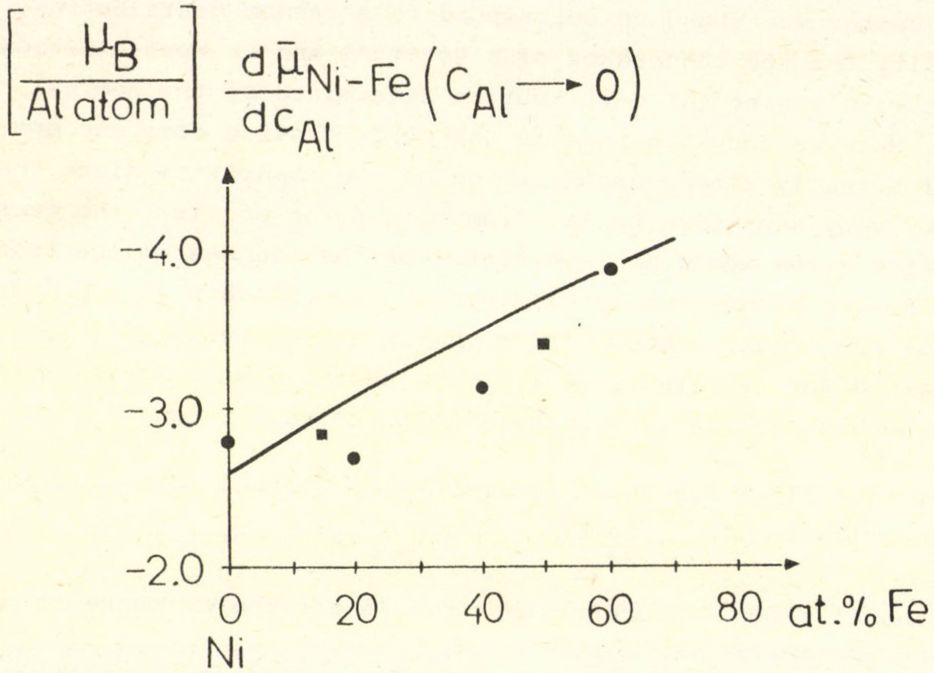

Fig. 4 *Total change of the average magnetization of fcc Ni-Fe alloys caused by a single Al atom as a function of Fe concentration. The squares show the results of Bardos et al. (1967). The full curve is given by eq. 3*

c/ bcc (Fe-Cr)-Al

The following alloys have been investigated: Fe-Cr (1.0 and 10.0 at%) diluted by 4.8 and 10.0 at% Al, respectively; and Fe-Cr (20.0 at%)containing 4.9 and 9.8 at% Al.

The Fe-Cr alloys differ from the previously studied cases since in their Mössbauer spectra there are well resolvable satellites corresponding to the first and second neighbour Cr atoms. Only the average change in the iron hyperfine field due to a first or second neighbour Cr was determined as $\Delta H_{1,2}^{Cr}$ = -26.9 kOe, because the contributions from the two shells were not separable on the basis of Mössbauer measurements (Vincze and Campbell 1973). On the other hand, a single first neighbour Al casuses a ΔH_1^{Al} = -22.6 kOe decrease of the iron hyperfine field in the binary Fe-Al alloys. The superposition of these satellite systems results in very broad, almost structureless lines of the Mössbauer spectra of the high Cr-concentration (10 and 20 at%) (Fe-Cr)-Al samples. For this reason we could not unambiguously evaluate these spectra.

However, in the case of the $(Fe_{0.99}Cr_{0.01})_{0.952}$-$Al_{0.048}$ alloy, because of the smaller number of satellites, we could determine both the hyperfine field changes caused separately by Al and Cr neighbours and the relative amplitudes of the satellites, i.e. the distribution of Al and Cr atoms. This latter was found to correspond to a random distribution, that is the probability for the occurrence of a given Al and Cr configuration was in agreement with a binomial distribution determined by the nominal concentrations. Here we should emphasize that this finding does not prove the existence of a really random distribution of the impurities since the iron sites are not very sensitive to it. From this point of view, the measurement at the impurity sites would be important. For the changes of the iron hyperfine the following values were obtained: $\Delta H^{Cr}_{1,2} = (-29.8 \pm 0.7)$ kOe and $\Delta H^{Al}_{1} = (-20.1 \pm 0.5)$ kOe. Again, though these values are a little different from those determined for the binary alloys, the deviations do not seem to be significant because of the strong correlation between them.

For this alloy the total change of the average magnetization caused by the Al substitution is $\frac{d\mu_{Fe-Cr}}{dc_{Al}} = -2.23\mu_B$, which corresponds to a simple dilution - the Al atoms disturb neither the Fe nor the Cr magnetic moments.

The initial rate of the change in the average magnetization of the Fe-Cr alloys for Al as a function of Cr concentration is shown in Fig.5.

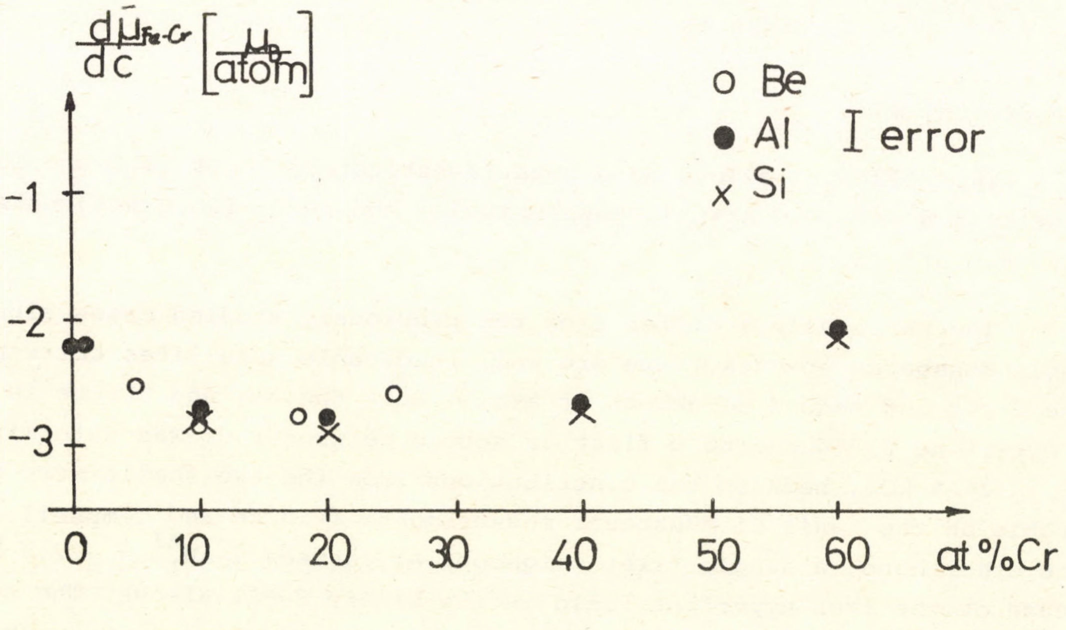

Fig.5 Total change of the average magnetization of bcc Fe-Cr alloys as a result of the addition of Be, Al or Si as a function of Cr concentration

The curve significantly deviates from a simple dilution behaviour, the possible origin of which will be discussed in the next section.

3. DISCUSSION

The experimental results show that the Al substitution does not change the magnetic moments of iron atoms in the bcc Fe-Ni, Fe-Co and fcc Ni-Fe alloys, while at the same time the magnetic moments of Ni and Co atoms are appreciably influenced by it. In the case of bcc Fe-Cr alloys the situation is not so unambiguous, the data allow - at least at high Cr-concentration - the disturbance of either Fe or Cr magnetic moments.

At first, the behaviour of the Fe-Ni and Fe-Co alloys will be discussed. According to the very simple, phenomenological, qualitative model sketched in the introduction, the basic difference between the Fe and Ni behaviour for Al substitution is that the latter can accept the two 3s electrons of Al while the Fe cannot. This is a strictly local property, it is independent of the crystal structure (at least for bcc and fcc structures). In the absence of theoretical band calculations which could be used simultaneously both for <u>Fe</u>Al and <u>Ni</u>Al, a hypothetical atomic energy level arrangement is assumed in order to be able to interpret the observed magnetic behaviours. It is well known of course, that the corresponding atomic electron states are considerably broadened in metals, thus this model is a simple working hypotheses which makes no attempt to explain the mechanism of charge transfer, but only to take into account in a somewhat consistent manner the fundamental experimental findings.

The basic assumption is that the Al $3s^2$ energy level lies above the Ni 3d level and below the Fe 3d level. This explains why the Al is a simple diluent for Fe and an electron donor for Ni atoms in the same alloys. Since the Ni atoms have about 0.6-0.8 3d holes in the whole concentration range of iron, they can gather both 3s electrons of Al if their number around the Al is sufficient for it. Here a further assumption has been used according to which the 3p electrons of Al form a collective conduction electron band with the 4s band of the host. The soft X-ray and Knight-shift measurement on high concentration Ni-Al alloys are in good agreement with this latter supposition (Wenger et al. 1971, Seitchik and Walmsley 1965).

On the basis of this model the total change of the average magnetization in fcc Ni-Fe due to a single Al atom will be the following:

$$\frac{d\mu_{Ni-Fe}}{dc_{Al}} (c_{Al} \rightarrow 0) = -\mu_{Ni-Fe} - 2\mu_B \qquad (3)$$

since here the number of Ni atoms is always enough to accommodate both 3s electrons of Al. The full curve of Fig. 4 calculated on the basis of eq.3 gives a reasonable fit to the experimental data without any adjustable parameter. The deviations may be attributed in part to the possible formation of short range order in these alloys.

Thus we can conclude that the effect of Al addition to the Ni-Fe alloys is independent of the crystal structure, it is purely local effect and it could be explained by the above model. The addition of Be of Si to these alloys causes a similar decrease in the average magnetization of the alloys (Besnus et al. 1971), the concentration dependence of which is similarly-shaped but its absolute value is larger for Si and less for Be than for Al because of their different outer electron structures - in agreement with our expectation.

In the bcc Fe-Co alloys the Fe is again not affected for the Al addition whereas the Co is disturbed similarly to the Ni atoms in Fe-Ni. However, there seems to be a substantial difference between the behaviours of Co and Ni towards Al in that the Co has holes in both spin-up and spin-down bands in contrast to the Ni where the spin-up band (or energy level) is full. Thus it is very probable that the "overflow" of the excess Al electrons will take place into both Co bands. The indication of this (in the absence of reliable $\frac{d\bar{\mu}}{dc}$ for CoAl) originates from the comparison of the $\frac{d\bar{\mu}}{dc}$ values obtained by Besnus et al. (1971) for CoBe and $(Fe_{0.5}Ni_{0.5})$-Be ($-3.10\mu_B$ and $-3.05\mu_B$, respectively), and for CoSi and $(Fe_{0.5}Ni_{0.5})$-Si ($-4.04\mu_B$ and $3.96\mu_B$, respectively). Thus if we take the value $\frac{d\mu}{dc} = -3.4\mu_B$ of $(Fe_{0.5}Ni_{0.5})$-Al to be a good approximation for the $\frac{d\bar{\mu}}{dc}$ value of CoAl, then the moment of the neighbouring Co atoms will decrease only by $1.6\mu_B$ from the transferred excess Al electrons.

Figure 1 shows the total change of the average magnetization in bcc Fe-Co due to a single Al atom. This curve has two characteristic features. First of all, the initial slope at small Co concentrations is about three times smaller then for the bcc Fe-Ni alloys and appreciably deviates from a linear concentration dependence. Secondly, it saturates at about $-4.0\mu_B$ in the region of 20 at% Co. This saturation is the same as for the bcc Fe-Ni alloys and it is because of the sufficiently large number of Co neighbours around the Al atoms. The saturation value is given by the sum of the previously mentioned $1.6\mu_B$ extra decrease of Co moments and the average magnetization of the alloys which is about $2.4\mu_B$ in this concentration range. The form of the initial part of $\frac{d\mu_{Fe\text{-}Co}}{dc_{Al}}$ could have different origins, however we cannot differentiate between them. One possibility is that the number of Co neighbours is less than that required by a random distribution at atoms. The tendency for short-range order in bcc Fe-Co alloys at low Co concentrations has already been observed by NMR measurements (Khoi et al. 1974) and was used for the explanation of the Co concentration dependence of the iron hyperfine field (Vincze et al. 1974). Another less likely possibility is a hypothetical Co concentration dependence of the Co moment change caused by the transferred Al electrons.

The addition of Be or Si to the high Co concentration bcc Fe-Co alloys has the same effect as in fcc Ni-Fe: the largest decrease in μ_{Fe-Co} is caused by Si, the smallest by Be - again in agreement with the model used.

The behaviour of bcc Fe-Cr alloys on addition of Al is somewhat puzzling. According to our simple model neither the Fe nor the Cr magnetic moments are expected to be changed by the Al presence because the Cr energy levels are above that of the Fe. In contrast to this, the total change of the average magnetization in these alloys as a result of a single Al indicates a significant deviation from the expected simple dilution behaviour. The analysis of the Mössbauer spectra can give no information whatsoever about the iron behaviour because of their very complex shapes. Therefore we cannot exclude the possibility of the Al-caused decrease in the moments of Fe of Cr, or in both. In any case it is very surprising that the substitution of Be or Si causes exactly the same moment perturbation independently of their outer electron number (Fig.5) contrary to the behaviour found in Ni-Fe and Fe-Co. Combining this observation with the Mn^{55} NMR measurement of Khoi et al. (1974) according to which the Al and Si impurities avoid forming pairs with Mn impurities in iron we suggest that the situation is similar in our ternary alloys: there is a repulsion between Cr and Be, Al, Si. The result of this supposed repulsion is a formation of short-range order and consequently we can attribute the larger decrease than simple dilution in the average magnetization of these bcc Fe-Cr alloys caused by the addition of Be, Al or Si to the decrease in the Fe and/or Cr moments as a result of the changes in the local concentrations. At present we are unable to give a quantitative estimation of this effect.

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