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Magnetic-moment distribution and environmental effects in dilute iron-based alloy with V, Cr, and Mn impurities

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The measurements of polarized neutron elastic, diffuse scattering on dilute iron-based alloys with about 1 and 2 at.% of Cr, V, and Mn impurities are reported. The measurements have been performed at room temperature using a polarized neutron beam with wavelength of 4.25 Å. The data indicate existence of a large magnetic moment on impurity atoms at about 1 at.% concentration, coupled antiparallel to the bulk magnetization. The observed values of average impurity moment agree very well with theoretical calculations done by Campbell and Gomés on the basis of Friedel's model of virtual bound-state formation in these alloys and support the idea of localized impurity states in the dilute limit. The value of the average impurity moment decreases rapidly with increasing alloy concentration, whereas the iron moment is nearly constant with a slight tendency to increase. The environmental effects are mostly confined to the first shell of atoms and depend on the kind of impurity and its concentration. The data are compared with coherent potential-approximation calculations for these alloys.

I. INTRODUCTION

Recent measurements of polarized neutron elastic, diffuse scattering on disordered bcc iron-rich Fe-Ti alloys[1] yield a large average magnetic moment on impurity sites of about $-2.1\mu_B$ in dilute limit ($C_{\mathrm{Ti}}=0.84$ at.%), decreasing rapidly in absolute value with increasing titanium concentration. The observed value of impurity moment agrees very well with theoretical calculations of Campbell and Gomés[2] done on the basis of Friedel's model[3] within Hartree-Fock approximation for Coulomb correlation energy. This observation stimulated us to study other dilute iron-based alloys for which the calculations of Campbell and Gomés[2] predict also a large magnetic moment on impurity atoms.

In this paper we report some results of similar measurements, as in an earlier study,[1] on dilute Fe-V alloys with 0.7 and 1.65 at.% of vanadium, Fe-Cr alloys with 1.04 and 1.46 at.% of chromium and Fe-Mn alloys with 0.79 and 1.85 at.% of manganese.

A general review of magnetic moment distribution in low concentrated iron-based alloys has been done by Collins and Low[4] by means of unpolarized neutron elastic diffuse scattering measurements. In the alloys containing about 1–2 at.% impurity these authors found a moment of about $-0.7\mu_B$ on titanium and chromium in Fe-Ti and Fe-Cr alloys, respectively, $-0.4\mu_B$ on vanadium in Fe-V alloys, and a negligible moment on manganese in Fe-Mn alloys. Collins and Low determined the impurity moment from the magnetic diffuse scattering cross sections at the scattering vector $k \simeq 1.35$ Å^{-1} ($k=4\pi\sin\theta/\lambda$), assuming that the environmental effects do not contribute to the magnetic scattering at this value of k (Shull-Wilkinson model[5]).

Campbell[6] reexamined the data of Collins and Low including environmental effects and obtained larger impurity moments of about $-1.1\mu_B$ with an error of $0.4\mu_B$ for titanium, $-0.9\mu_B$ with an error of $0.2\mu_B$ for vanadium and chromium and a negligible moment on manganese in Fe-Ti, Fe-V, Fe-Cr, and Fe-Mn alloys, respectively. Nearly the same value of chromium moment $(-1.16\pm0.10)\mu_B$ was reported recently by Aldred *et al.*[7] from the analysis of unpolarized neutron diffuse scattering measurements of Collins and Low[4] on an alloy with 2 at.% of Cr.

This technique of polarized neutron diffuse scattering is a useful tool in order to study magnetic disturbances in disordered systems. As it will be discussed later, this technique gives higher precision in the case of our alloys than that with unpolarized neutrons. Also several difficulties which appear when one uses unpolarized neutrons can be avoided and at dilute limit the results are model independent.

II. THEORY

The theory of neutron diffuse scattering from binary, disordered magnetic systems has been elaborated and described by Marshall[8] in the framework of a linear superposition model of magnetic disturbances induced by single impurities. More recently Gautier[9] proposed a general formulation, valid also for more concentrated alloys. Within Gautier's formalism the polarized neutron diffuse scattering cross section from a binary $A_{1-c}B_c$ disordered alloy is given by

$$\left(\frac{d\sigma}{d\Omega}(k)\right)_{\pm}=\left(\frac{d\sigma}{d\Omega}\right)_{\text{incoh}}+c(1-c)\left[(\bar{b}_B-\bar{b}_A)^2S(k)\pm 2\left(\frac{\gamma e^2}{2mc^2}\right)(\bar{b}_B-\bar{b}_A)f(k)\Delta\mu(k)+\left(\frac{\gamma e^2}{2mc^2}f(k)\Delta\mu(k)\right)^2\right] , \quad (1)$$

where $(d\sigma/d\Omega)_{\text{incoh}}$ is the incoherent scattering cross section, $\bar{b}_B$ and $\bar{b}_A$ are average coherent scattering lengths, $\gamma e^2/2mc^2$ is a constant equal to 0.2695×10^{-12} cm/μ_B, $f(k)$ is the magnetic form factor of the alloy, and ± signs stand for neutron spin parallel or antiparallel to the sample magnetization.

The function $S(k)$ describes the nuclear short-range order and for a polycrystalline sample

$$S(k)=1+\sum_i N_i\alpha_i\frac{\sin kR_i}{kR_i} , \quad (2)$$

where α_1's are the nuclear short-range-order parameters defined by Cowley[10] which measure a deviation from a perfect disorder in the alloy and N_i is the number of nearest neighbors distant by R_i from a central atom.

The second function in Eq. (1), $\Delta\mu(k)$ describes the magnetic moment disturbances in the alloy. Within Gautier's formalism this function for a polycrystalline alloy is expressed as follows

$$\Delta\mu(k)=(\bar{\mu}_B-\bar{\mu}_A)S(k)+\sum_i(1-\alpha_i)N_i(\delta\mu^i_{AB}-\delta\mu^i_{BA})\frac{\sin kR_i}{kR_i} , \quad (3)$$

where $\bar{\mu}_B$ and $\bar{\mu}_A$ are the individual average magnetic moments and the parameters $\delta\mu^i_{AB}$ describes the average modification of the magnetic moment of an atom A due to the presence of another one of type B in the ith shell of atoms.

In Marshall's model the corresponding function is separated into two terms

$$\Delta\mu(k)=S(k)[\bar{\mu}_B-\bar{\mu}_A+(1-c)G(k)+cH(k)] , \quad (4)$$

where $G(k)$ and $H(k)$ are Fourier transforms of magnetic moment disturbances at, respectively, host and impurity atom sites induced by impurity. Thus the main difference between Marshall's model and Gautier's formulation consists of the description of magnetic perturbations. These are separated in Marshall's model at two parts: one connected with impurity site, and the second one with the host atom. Such separation is not evident in Gautier's formalism where both terms interfere with the same weight. However, in dilute limit ($c\rightarrow 0$, $\alpha_i\equiv 0$) both formalisms are equivalent and in this case Eqs. (3) and (4) yield

$$\Delta\mu(k)=\bar{\mu}_B-\bar{\mu}_A+\sum_i\beta_iN_i\frac{\sin kR_i}{kR_i} . \quad (5)$$

From Eq. (1) it is seen that the magnetic disturbance function is given directly by the difference of spin-up (+) and spin-down (−) diffuse scattering cross sections

$$\Delta\frac{d\sigma}{d\Omega}=\left(\frac{d\sigma}{d\Omega}\right)_+-\left(\frac{d\sigma}{d\Omega}\right)_-=4\left(\frac{\gamma e^2}{2mc^2}\right)c(1-c)(\bar{b}_B-\bar{b}_A)\Delta\mu(k)f(k) . \quad (6)$$

This allows us to determine the difference of individual magnetic moments $\Delta\mu=\bar{\mu}_B-\bar{\mu}_A$ and the parameters β_i [Eq. (5)].

Within the formalism of Gautier[9] the parameters β_i are related to nuclear short-range order and magnetic-moment disturbance parameters through the following equation:

$$\beta_i=\alpha_i(\bar{\mu}_B-\bar{\mu}_A)+(1-\alpha_i)\delta\mu^i , \quad (7)$$

where

$$\delta\mu^i=\delta\mu^i_{AB}-\delta\mu^i_{BA} .$$

Thus really in the absence of nuclear short-range order $\beta_i=\delta\mu^i$ and similarly as in Marshall's model the disturbance is purely magnetic. When the nuclear short-range order is taken into account, Marshall's formalism gives the same results as Gautier's only in the case where the magnetic perturbations induced by impurity are confined to the impurity itself ($\delta\mu^i\equiv 0$) or very short ranged. It seems reasonable to assume that in our dilute alloys the nuclear short-range order is absent and the determined parameters β_i describe the magnetic-moment disturbances.

In some cases (cf. discussion by Parette and Kajzar[11]), if the value of the magnetic term $[(\gamma e^2/2mc^2)\Delta\mu]^2$ is sufficiently small compared to the square of difference of nuclear coherent-scattering length $(\bar{b}_B-\bar{b}_A)^2$, the short-range-order parameters can be obtained form the half sum of spin-dependent cross sections. In our alloys the dominating term is the magnetic one and such analysis cannot be applied.

Marshall[8] has shown that in the $k=0$ limit the magnetic diffuse scattering cross section should be proportional to the square of concentration variation of bulk moment.

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{mag.diff.}}(k=0)=c(1-c)\left(\frac{\gamma e^2}{2mc^2}\frac{d\bar{\mu}}{dc}\right)^2 . \quad (8)$$

In the presence of nuclear short-range order and in the case of polarized neutron diffuse scattering the

TABLE I. Physical constants used in data conversion.

Element	$\bar{b}$ (10^{-12} cm)	σ_{incoh} (b)	σ_{abs} (b)	$2B$ (Å^2)
Cr	0.353	1.76	7.1	0.56
Fe	0.951	0.4	6.0	0.71
Mn	−0.387	0.4	29.9	0.73
V	−0.052	5.07	12.2	1.14

corresponding relation is (Medina and Cable,[12] Medina and Garland[13]),

$$\Delta\frac{d\sigma}{d\Omega}(k=0)=4\frac{\gamma e^2}{2mc^2}c(1-c)(\bar{b}_B-\bar{b}_A)\frac{d\mu}{dc}S(0) \;. \tag{9}$$

For the alloy without nuclear short-range order $[S(0)=1]$ this relation implies that the loss of magnetization due to the impurity should be found in the magnetic diffuse scattering cross section in the forward direction.

III. EXPERIMENT

The diffuse scattering measurements have been performed on the polarized neutron spectrometer installed at the end of the cold-source neutron guide Belenus II of reactor EL 3 in Saclay. The neutron beam polarized and monochromatized by a Heusler Cu_2MnAl single crystal, kept in a permanent magnetic field of 2.4 kOe, and flipped in a longitudinal radio frequency coil. The chosen neutron wavelength of 4.25 Å, lying well above the cutoff for iron, assured small multiple-scattering effects. The half-wavelength contamination, which is small because of a favorable neutron guide spectrum, was removed by a pyrolytic graphite with large mosaicity, oriented with (002) plane in reflection for $\frac{1}{2}\lambda$. The scattered neutrons were counted in the horizontal plane by a linear multidetector composed from 400 cells and covering a 2θ range of about 80°.

The investigated alloys were prepared by Perrier de la Bathie from Crystaltech Grenoble from 99.997 pure elements by levitation technique (horizontal and vertical) in inductance furnace under argon atmosphere and their concentration was determined by chemical analysis. The samples were reduced from cylinders with diameter of about 10 mm into 8 mm. Cylindrical specimens were mounted vertically in the gap of electromagnet with magnetic field of 13 kOe perpendicular to the scattering vector and sufficient to saturate them. The scattered intensities, corrected for background, multiple-scattering effects, transmission, incomplete beam polarization and flipping efficiency, absorption, thermal and magnetovibrational effects, were put on absolute scale by comparison with a standard vanadium scatterer of the same dimensions. The magnetovibrational effects have been corrected for by subtracting corresponding terms (cross sections) determined for pure iron (cf. Kajzar and Parette[1]). The physical constants used in data conversion are listed in Table I. More details about apparatus, experimental procedure, and corrections can be found in earlier papers.[1,11]

IV. RESULTS AND ANALYSIS

The measured difference of differential cross sections $\Delta(d\sigma/d\Omega)$ for Fe-V, Fe-Cr, and Fe-Mn alloys are plotted in Figs. 1, 2, and 3, respectively. The experimental points represent the averages of counts from eight cells of multidetector and the bars denote the statistical errors only.

The experimental data were analyzed within Eq. (6) with $\Delta\mu(k)$ given by Eq. (5). As we mentioned before, we assume that in our dilute alloys there is no nuclear short-range order and the parameters β_i describe the magnetic-moment disturbance [see Eq. (7) with $\alpha_i=0$]. In the analysis we assumed the alloy magnetic form factor of the form $f(k)=1-0.061k^2$ close to that of pure iron (the same was used by Aldred *et al.*[7] for Fe-Cr alloys). In the least-squares fit of Eq. (6) to experimental data we introduced the successive shells of atoms surrounding the central one looking at the variation of χ^2 value. A satisfactory fit has been obtained including into computation only first shell of atoms for all studied alloys. The χ^2 values were slightly smaller when the second shell of atoms was taken into account, but both parameters β_1 and β_2 become important and have opposite signs, without significant change in the value of $\Delta\mu$. In fact, because of limited k-vector range (0.2– 2 Å^{-1}) a precise determination of β parameters is spurious. Thus in Tables II–IV, reassembling the results of least-squares

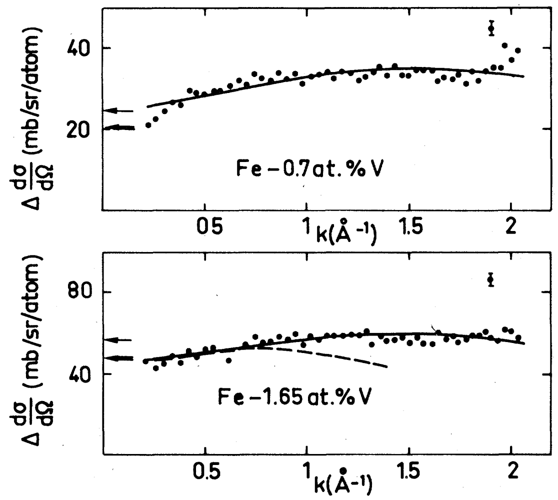

FIG. 1. Difference of differential cross sections in Fe-V alloys. Experimental points are averages of counts from eight cells of multidetector and the bars denote statistical errors only. Solid lines represent a fit as described in text. The calculated values of $\Delta(d\sigma/d\Omega)(k=0)$ from concentration variation of bulk moment are shown by single arrow [$d\bar{\mu}/dc=-3.286\mu_B$/atom (Nevitt and Aldred, Ref. 29)] and double arrow [$d\bar{\mu}/dc=-2.72\mu_B$/atom (Arrott and Noakes, Ref. 16) and $d\bar{\mu}/dc=-2.676\mu_B$/atom (Aldred, Ref. 30)]. Broken line represents the corresponding difference cross section measured by Collins and Low (Ref. 4) and converted to polarized neutrons (see text).

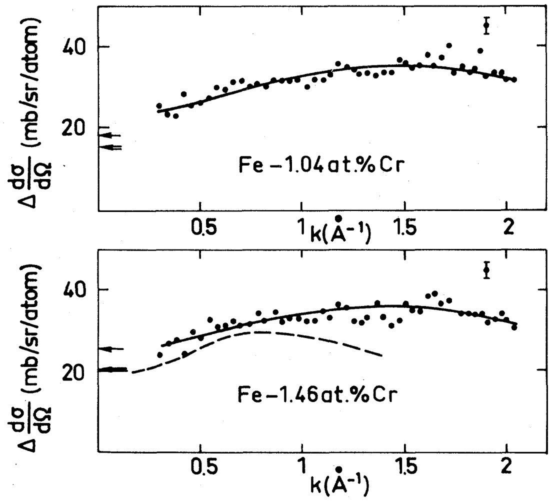

FIG. 2. Difference of differential cross sections in Fe-Cr alloys. The details are as in Fig. 1. Single arrow corresponds to $d\bar{\mu}/dc=-2.71\mu_B$/atom (derived from room-temperature data of Fallot, Ref. 31), double arrow to $d\bar{\mu}/dc=-2.30\mu_B$/atom (Arrott and Noakes, Ref. 16), $d\bar{\mu}/dc=-2.36\mu_B$/atom (room-temperature data of Dubiel *et al.* Ref. 32), and $d\bar{\mu}/dc=-2.4\mu_B$/atom (Aldred, Ref. 33). Broken line corresponds to the data of Collins and Low (Ref. 4).

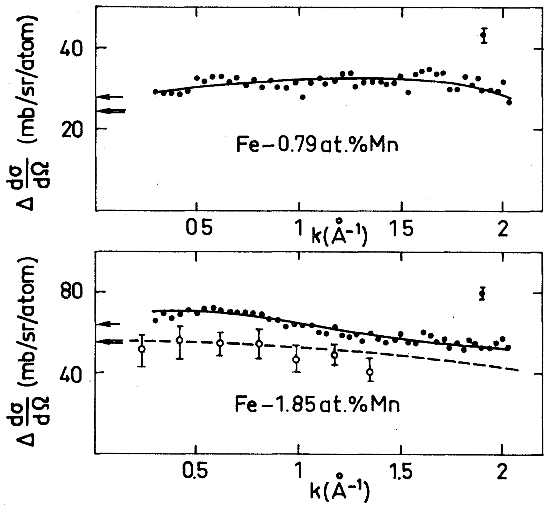

FIG. 3. Difference of differential cross sections in Fe-Mn alloys. The details are as in Fig. 1. Single arrow corresponds to $d\bar{\mu}/dc=-2.44\mu_B$/atom [derived from room-temperature measurements of Sadron (Ref. 34) with assumed iron moment $\mu_{\mathrm{Fe}}=2.177\mu_B$] and double arrow to $d\bar{\mu}/dc=-2.1\mu_B$/atom (Arrott and Noakes, Ref. 16). Broken line and open circles correspond to the data of Collins and Low (Ref. 4).

analysis, only those with first shell of atoms are given. The statistical errors of $\Delta\mu$ values are increased by an uncertainty in the concentration assumed to be of $\Delta c=0.05$ at.%; this being an upper limit in the chemical analysis accuracy. In dilute alloys the precision in concentration determination is very important, since the accuracy is directly proportional to it [see Eq. (6)]. The calculated values of $\Delta(d\sigma/d\Omega)$ are plotted in Figs. 1–3 (solid lines) for corresponding alloys.

The individual average magnetic moments are given by the following equations:

$$\bar{\mu}_A=\bar{\mu}-c\,\Delta\mu-\mu_0\ ,$$
$$\bar{\mu}_B=\bar{\mu}+(1-c)\Delta\mu-\mu_0\ , \tag{10}$$

where $\bar{\mu}$ is the bulk magnetic moment, $\Delta\mu$ is the difference between average individual magnetic moments as determined by polarized neutron diffuse scattering measurements, and μ_0 is the diffuse moment seen in polarized neutron Bragg-scattering measurements and for iron is equal to $-0.21\mu_B$.[14] We assume that in our dilute alloys the value of μ_0 is not affected and is equal to that in pure iron. In fact in Fe-V alloys at 5 at.% of vanadium the value of μ_0 is close to that of iron.[15]

The values of bulk moments have been determined using $d\bar{\mu}/dc$ reported in literature and are given in

TABLE II. Magnetic-moment distribution and magnetic disturbance parameters in Fe-V alloys. All quantities are in Bohr magnetons (μ_B).

(at.%) V	$\bar{\mu}$	$\bar{\mu}_{\rm Fe}$	$\bar{\mu}_{\rm V}$	$\Delta\mu$	β_1
0.7	2.158	2.404 ± 0.003	−2.78 ± 0.40	−5.18 ± 0.40	0.238 ± 0.009
1.65	2.132	2.405 ± 0.002	−1.40 ± 0.13	−3.80 ± 0.13	0.146 ± 0.004

Tables II–IV for corresponding alloys. In view of some differences in $d\bar{\mu}/dc$ determination by different authors, we have used those given by Arrott and Noakes[16] from magnetization measurements on dilute alloys (up to about 3 at.% of impurity concentration). In fact in our low-concentration alloys, the imprecision in $d\bar{\mu}/dc$ values does not alter significantly the values of bulk moment. For pure iron we have used the room-temperature value of $2.177\mu_B$ given by Crangle and Goodman.[17]

The results for individual average magnetic moments given in Tables II–IV indicate a moment enhancement on impurity sites at dilute limit in all alloys studied. The average impurity moment is coupled antiparallel to the net magnetization and takes the values of $(-2.78 \pm 0.40)\mu_B$, $(-3.46 \pm 0.33)\mu_B$, and $(-0.82 \pm 0.23)\mu_B$ at 0.7 at.% of V, 1.04 at.% of Cr, and 1.85 at.% of Mn in iron, respectively. The value of impurity moment decreases rapidly with increasing alloy concentration and at 1.65 at.% of V, 1.46 at.% of Cr, and 1.85 at.% of Mn is equal, respectively, to $(-1.40 \pm 0.13)\mu_B$, $(-1.88 \pm 0.18)\mu_B$, and $(-0.23 \pm 0.9)\mu_B$. The errors given in brackets correspond only to those in $\Delta\mu$ determination. We have not accounted for eventual errors in $\bar{\mu}$ and μ_0 values.

The magnetic-moment disturbance is most pronounced in Fe-Cr alloys and in all alloys decreases with increasing alloy concentration. In the case of Fe-Mn alloys the moment disturbance changes sign between 0.79 and 1.85 at.% of Mn. This is in agreement with other studies on Fe-Mn alloys by Child and Cable,[18] Mezei,[19] and Radhakrishna and Livet[20] who found a negative moment disturbance in more concentrated alloys.

In Figs. 4–6 we have plotted the measured values of half sums of spin-dependent cross sections for alloys with lowest impurity concentration. Similar as before, experimental points are average of counts from eight cells of multidetector and the bars denote the statistical errors only. The solid lines in these figures represent the following different contributions to $\frac{1}{2}\sum d\sigma/d\Omega$ [cf. Eq. (1)]

$$\frac{1}{2}\sum\frac{d\sigma}{d\Omega}=\left[\frac{d\sigma}{d\Omega}\right]_{\rm incoh}+\left[\frac{d\sigma}{d\Omega}\right]_{N-N}+\left[\frac{d\sigma}{d\Omega}\right]_{M-M}, \tag{11}$$

where

$$\left[\frac{d\sigma}{d\Omega}\right]_{\rm incoh}=(1-c)\left[\frac{d\sigma}{d\Omega}\right]_{{\rm incoh},A}+c\left[\frac{d\sigma}{d\Omega}\right]_{{\rm incoh},B} \tag{12}$$

and

$$\left[\frac{d\sigma}{d\Omega}\right]_{N-N}=c(1-c)(\bar{b}_A-\bar{b}_B)^2 \tag{13}$$

are calculated using the incoherent scattering cross

TABLE III. Magnetic-moment distribution and magnetic disturbance parameters in Fe-Cr alloys. All quantities are in Bohr magnetons.

(at.%) Cr	$\bar{\mu}$	$\bar{\mu}_{\rm Fe}$	$\bar{\mu}_{\rm Cr}$	$\Delta\mu$	β_1
1.04	2.153	2.424 ± 0.003	−3.46 ± 0.33	−5.88 ± 0.33	0.298 ± 0.012
1.46	2.143	2.416 ± 0.002	−1.88 ± 0.18	−4.30 ± 0.18	0.195 ± 0.009

TABLE IV. Magnetic-moment distribution and magnetic disturbance parameters in Fe-Mn alloys. All quantities are in Bohr magnetons.

(at.%) Mn	μ	$\bar{\mu}_{\mathrm{Fe}}$	$\bar{\mu}_{\mathrm{Mn}}$	$\Delta\mu$	β_1
0.79	2.160	2.395 ± 0.002	-0.82 ± 0.23	-3.21 ± 0.23	0.087 ± 0.007
1.85	2.138	2.397 ± 0.002	-0.23 ± 0.09	-2.63 ± 0.09	-0.017 ± 0.004

sections and scattering lengths for alloy elements from Table I. The magnetic contributions were calculated approximately with the values for $\Delta\mu$ from Tables II–IV for corresponding alloys

$$\left(\frac{d\sigma}{d\Omega}\right)_{M-M} = c(1-c)\left(\frac{\gamma e^2}{2mc^2}\Delta\mu\right)^2 . \qquad (14)$$

It is seen, that except for the Fe-Mn alloy, the calculated values of $\frac{1}{2}\sum d\sigma/d\Omega$ agree well with measured ones. In the case of Fe-Mn alloy the experimental values are slightly higher than the calculated ones. The difference may be due to the omission of the interference term in Eq. (11) which vanishes only in perfect experimental conditions (100% of polarization and flipping efficiency) and is most important (because of largest value of Δb) in the case of Fe-Mn alloys (our estimation gives a value of about 2 mb for this term). On the other hand, because of possible systematic errors, the precision in the absolute values determination of the half sum of spin-dependent cross section is smaller than in the case of difference cross section, where most of corrections cancel themselves. Thus the good agreement between calculated and observed values of $\frac{1}{2}\sum d\sigma/d\Omega$ confirms the correctness of experimental procedure and data treatment as well as the large magnetic diffuse scattering in this alloy.

V. DISCUSSION

We have performed polarized neutron elastic diffuse scattering measurements on dilute binary iron-based alloys with V, Cr, and Mn impurities. The data indicate existence of a magnetic-moment enhancement on impurity sites at dilute limit. The largest moment is found on chromium $(-3.46 \pm 0.23)\mu_B$ and the smallest one on manganese $(-0.82 \pm 0.23)\mu_B$. The value of impurity moment decreases rapidly with increasing alloy concentration. Similar phenomenon has been also observed in dilute Fe-Ti alloys.[1] The observed values of impurity moment at its lowest concentration agree surprisingly well with theoretical calculations done by

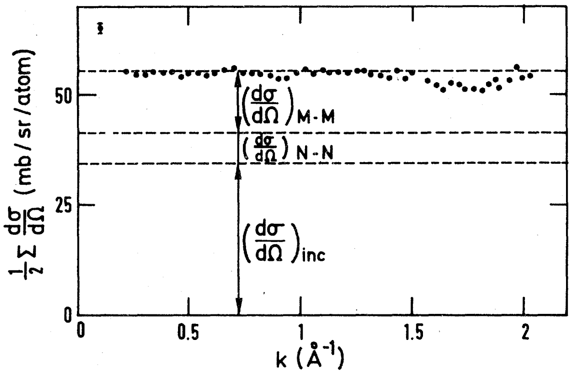

FIG. 4. Half sum of differential cross sections for an Fe-V alloy with 0.79 at.% of V. Broken lines represent calculated contributions of incoherent, nuclear diffuse (N-N) and magnetic (M-M) scattering cross sections. Other details as in Fig. 1.

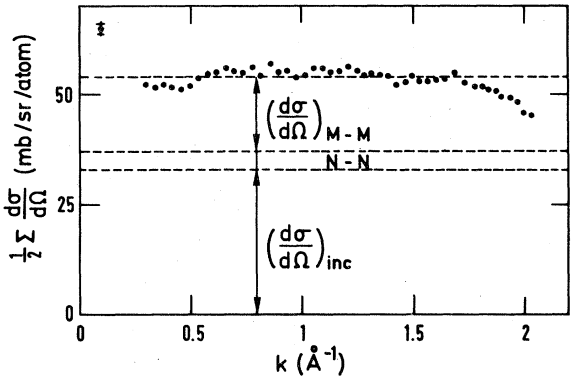

FIG. 5. Half sum of differential cross sections for an Fe-Cr alloy with 1.04 at.% of Cr. For details see Fig. 4.

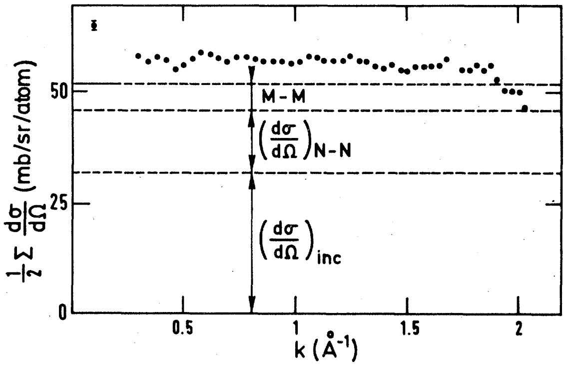

FIG. 6. Half sum of differential cross sections for an Fe-Mn alloy with 0.7 at.% of Mn. For details see Fig. 4.

Campbell and Gomés (see Fig. 7) on the basis of Friedel's model.[3] This well supports the idea of existence of isolated impurity states in dilute alloys with virtual bound-state formation. We note here, that similar magnetic-moment enhancement, with strong concentration dependence, has been also observed in Ni-Rh alloys.[21] In the last case, the impurity moment was found to be coupled parallel to the net magnetization.

Our results at lower impurity concentration differ significantly from those reported by Collins and Low[4] for corresponding alloys. As Collins and Low studied alloys with impurity concentration between 1 and 2 at.% it seems more adequate to compare their cross sections with ours for more concentrated alloys. Such comparison is displayed in Figs. 1–3 where the values are plotted of $\Delta(d\sigma/d\Omega)$ (broken lines) extracted from unpolarized neutrons data of Collins and Low assuming our impurity concentrations (1.65 at.% V, 1.46 at.% Cr, and 1.85 at.% Mn). For Fe-V and Fe-Cr alloys our difference cross sections $\Delta(d\sigma/d\Omega)$ agree with those deduced from the data of Collins and Low up to $k=1$ Å^{-1}. At higher values of k the difference cross sections corresponding to the data of Collins and Low fall rapidly, whereas they are still increasing in the present study. For Fe-Mn alloy the values of $\Delta(d\sigma/d\Omega)$ derived from the data of Collins and Low lie systematically below ours with, however, a similar k dependence. A main difficulty of the present work consists on elimination of magnetovibrational contribution to the diffuse scattering (Collins and Low used time-of-flight method which eliminates inelastic part in diffuse scattering). This was taken into account by subtracting of corresponding terms for pure iron, under an assumption that in our dilute alloys the magnetovibrational effects are of the same magnitude.[1] This assumption holds quite well for the half sum of cross sections $\frac{1}{2}\sum d\sigma/d\Omega$ where these effects give a greater contribution (of about 30 mb at $k=2$ Å^{-1}) than for the difference cross sections (about 13 mb). This fact and also similar k dependence of $\Delta(d\sigma/d\Omega)$ for Fe-Mn alloys allows us to believe that the used procedure for magnetovibrational correction is not bad. If, however, our assumption on the magnetovibrational corrections does not hold and these are more important for alloys than for pure matrix this should *a priori* affect the observed strong concentration dependence of impurity moment (effect of such corrections will be certainly more important at lower impurity concentration where the measured difference cross section takes smaller values). In order to check it out, the following reasoning is possible. Let us suppose that the measured difference cross sections still contain a term $\phi(k)$ due to not entirely eliminated magnetovibrational effects, then Eq. (6) will be replaced by the following expression:

$$\Delta\frac{d\sigma}{d\Omega}=4\left(\frac{\gamma e^2}{2mc^2}\right)c(1-c)(b_B-b_A)\Delta\mu(k)f(k)+\phi(k)\ . \tag{15}$$

For pure iron the contribution due to magnetovibrational effects is nearly given by $Ak^2f(k)$ (cf. Ref. 1) where $A=4.6$ mb Å^2. It is reasonable to assume a similar k dependence of this contribution in alloys. Moreover, due to a small difference in concentration of our alloys (less or of about 1 at.%), one can also assume that this contribution should be nearly the same at both alloy concentrations.

Let us consider following difference function

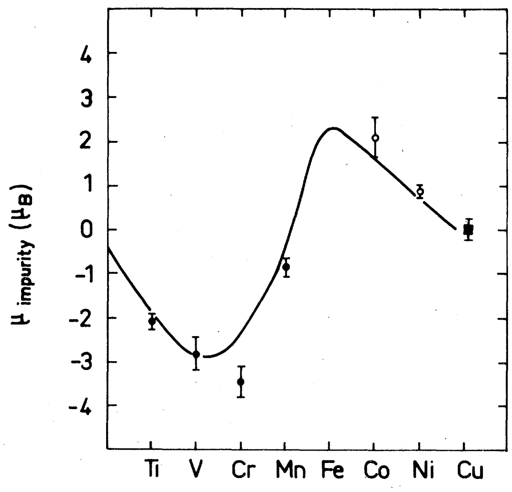

FIG. 7. Impurity moments in iron solid circles and solid square represent present study and Refs. 1 and 24 (Fe-Ti and Fe-Cu alloys, respectively), empty circles represent Collins and Low (Ref. 4).

$$\Delta(k) = \Delta\mu_2'(k) - \Delta\mu_1'(k) = \Delta\mu_2(k) - \Delta\mu_1(k) + \frac{\phi(k)}{Wf(k)}\left[\frac{1}{c_2(1-c_2)} - \frac{1}{c_1(1-c_1)}\right]$$

$$= \Delta\mu_2(k) - \Delta\mu_1(k) + \frac{A}{W}\left[\frac{1}{c_2(1-c_2)} - \frac{1}{c_1(1-c_1)}\right]k^2 , \qquad (16)$$

where

$$\Delta\mu_i'(k) = \frac{[\Delta(d\sigma/d\Omega)]_i}{Wc_i(1-c_i)f(k)} \quad (i=1,2) \qquad (17)$$

is the experimentally determined magnetic disturbance function, supposed here to be not entirely free of magnetovibrational contribution [otherwise $\Delta\mu_i'(k)=\Delta\mu_i(k)$] and $W=4(\gamma e^2/2mc^2)\,(b_B-b_A)$. The subscripts (1,2) refer to the lower and higher impurity concentration of a given alloy, respectively. It is seen that not entirely eliminated magnetovibrational contributions give a quadratic term in k in the right side of Eq. (16). At large values of k where the contribution due to magnetic environmental effects is negligible the difference of magnetic disturbance functions $[\Delta\mu_2(k)-\Delta\mu_1(k)]$ is nearly constant and equal to the difference of impurity moment at two alloy concentrations $[\bar{\mu}_B(c_2)-\bar{\mu}_B(c_1)]$. Thus the k dependence of $\Delta(k)$ at larger values of k will be a test whether or not the magnetovibrational effects have been correctly taken in account and the impurity moment depends on concentration. The measured values of the quantity $\Delta(k)$ are plotted in Fig. 8 for corresponding alloys. It is seen that for Fe-V and Fe-Cr alloys this quantity increases with increasing k at small values of k ($k < 1$ Å^{-1}) and is nearly constant within experimental errors at large values of k ($k > 1.4$ Å^{-1}). For Fe-Mn alloys this quantity is nearly constant (close to zero) up to $k=1$ Å^{-1}, after increases up to $k=1.7$ Å^{-1} and thereafter slightly decreases. These behaviors of the quantity $\Delta(k)$ for all alloys are different of what it would be expected if the contribution due to magnetovibrational effects was not correctly eliminated in the difference cross sections $\Delta(d\sigma/d\Omega)$. In fact, assuming that the difference between our difference cross sections $\Delta(d\sigma/d\Omega)$ and those derived from the data of Collins and Low of about 10 mb at $k=1.35$ Å^{-1}, for FeV and FeCr alloys is due to not entirely eliminated magnetovibrational effects, we obtain for the parameter A in Eq. (16) a value of 6.2 mb Å^2. The calculated values of $\Delta(k)$ through Eq. (17) assuming a constant impurity moment $[\Delta\mu_1(k)=\Delta\mu_2(k)]$ are plotted in Fig. 8 (solid lines). It appears that calculated in this way $\Delta(k)$ show quite different k behavior than the observed values for Fe-V and Fe-Cr alloys where the concentration dependence of impurity moment is the most pronounced one. This is also true for Fe-Mn alloys where at large k the experimental values of $\Delta(k)$ are nearly constant with a slight ten-

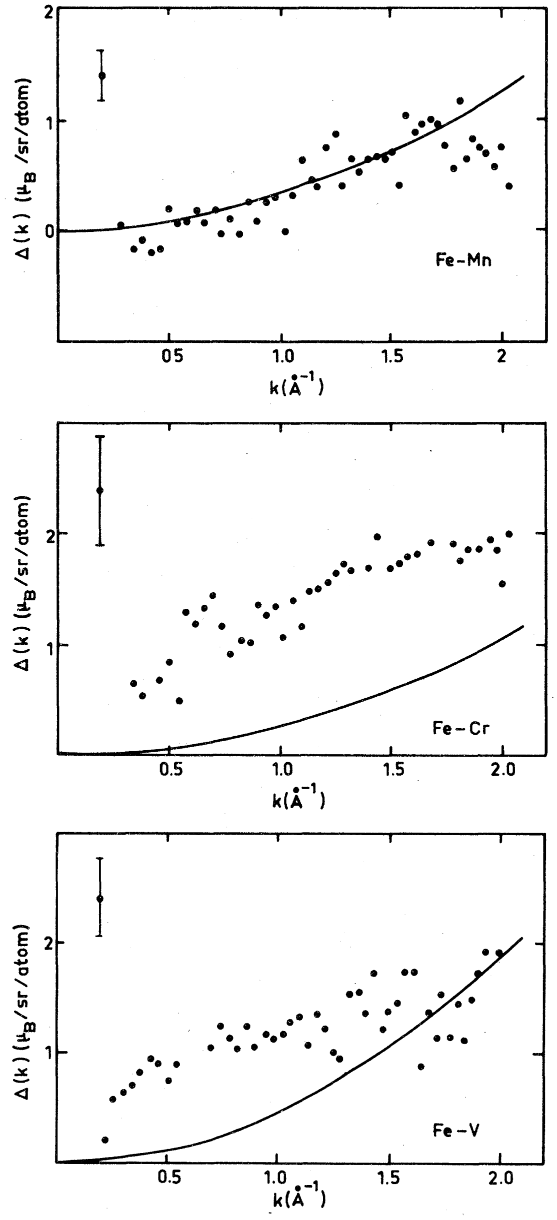

FIG. 8. The difference $\Delta(k)=\Delta\mu_2'(k)-\Delta\mu_1'(k)$ for studied alloys. Points represent experimental values and the solid lines the calculated values if there is no concentration dependence of impurity moments and the magnetovibrational effects are more important for alloys than for pure matrix (see text).

dency to decrease whereas the calculated ones increases as k^2. Thus it seems to us that magnetovibrational contribution to diffuse scattering is correctly evaluated in the present study and the observed strong concentration dependence of impurity moment in dilute alloys is an objective fact.

The lower values of magnetic diffuse scattering cross section for Fe-Mn alloys observed by Collins and Low, as well as a rapid decrease of this one at $k > 1$ Å^{-1} for Fe-Cr and Fe-V alloys, are probably due to a poor accuracy of unpolarized neutron diffuse scattering techniques applied to dilute alloys. For Fe-Mn alloy with 1.85 at.% Mn the signal is of about 8.4 mb at $k = 1$ Å^{-1} measured on an incoherent scattering background of 32 mb (in fact 64 mb because of two independent measurements needed to its determination). This signal is more important for Fe-Cr and Fe-V alloys because of larger values of $\Delta\mu$, however it decreases rapidly at large values of k due to the decrease of the square of the magnetic form factor $f(k)$.

The polarized neutrons, as it can be seen from Eq. (6), give a better precision in magnetic diffuse scattering term determination than the unpolarized ones (except small Δb). The ratio of this term in both methods is roughly equal to

$$\frac{\text{polarized neutrons}}{\text{unpolarized neutrons}} \simeq \frac{\Delta b}{0.27\Delta\mu(k)f(k)} \tag{18}$$

and at $k = 1.8$ Å^{-1} is equal to 3.6, 1.9, and 7.7 for lower impurity concentration in the case of Fe-V, Fe-Cr, and Fe-Mn alloys, respectively. As $\Delta\mu$ decreases with increasing alloy concentration, this ratio also increases.

Besides the magnitude of measured signal in unpolarized neutron diffuse scattering, there exists also another difficulty connected with existence of nonlinear and nonlocal terms (see discussion by Cable and Medina[22]) arising from vertex corrections (Sacchetti[23]). These probably are negligible in dilute alloys but can become important for more concentrated alloys. These terms are absent in the difference of differential cross sections [Eq. (6)].

In Figs. 1–3 we denote by arrows the values of $\Delta(d\sigma/d\Omega)$ calculated using Eq. (9) with $S(0) = 1$ and $d\bar{\mu}/dc$ values as determined by different authors. Generally the measured values of $\Delta(d\sigma/d\Omega)$ extrapolate well to the calculated ones.

The magnetic-moment disturbance is almost confined to the first shell of atoms and is positive for the alloys with lower impurity concentration, indicating an increase of iron moment in the first shell of atoms surrounding an impurity site. However, as is seen from Figs. 1–3, the moment disturbance is more extended, especially in the case of Fe-V and Fe-Cr alloys at lower impurity concentration. As we mentioned before, due to the numerical difficulties (appearance of oscillating signs in β parameters), limited k-vector range of measurements, we have fitted our data with only one parameter β_1. This allows a comparative observation of the variation of magnetic-moment disturbance with increasing impurity charge Z. Such variation can be seen from Fig. 9, where we have plotted the values of β_1, $\Delta\mu = \bar{\mu}_{\text{Fe}} - \bar{\mu}_{\text{imp}}$, and $d\bar{\mu}/dc$ for different $3d$ impurities in iron (the values of $\Delta\mu$ and β_1 for Ti, V, Cr, and Mn correspond to the lowest impurity concentration). It is seen that the magnetic-moment disturbance for the elements on the left side of iron varies similarly with Z as $\Delta\mu$. Large values of $\Delta\mu$ are connected with large values of β_1. This explains why the variation of $d\bar{\mu}/dc$ is different than that of $\Delta\mu$. Contrariwise the difference $(8\beta_1 - \Delta\mu)$ is close to the values of $d\mu/dc$. There is no such information for the elements lying on the right or iron. However, the measurements of Collins and Low[4] for Fe-Co and Fe-Ni alloys and Kajzar and Parette[24] for Fe-Cu alloys indicate a negative moment disturbance around impurity sites.

With increasing alloy concentration the moment disturbance decreases and in the case of Fe-Mn alloys changes also its sign. The case of Fe-Mn alloys is probably more complicated and the value of impurity

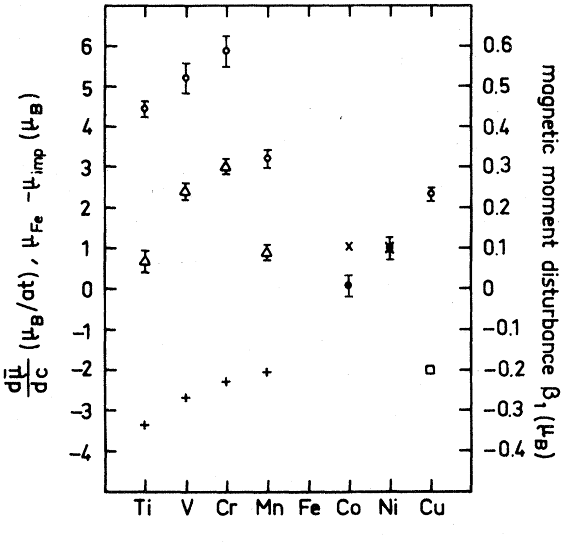

FIG. 9. Magnetic-moment disturbance parameter $\beta_1(\Delta)$, difference of average individual magnetic moments $(\bar{\mu}_{\text{Fe}} - \bar{\mu}_{\text{imp}})$(○, ●) and concentration derivative of bulk moment $d\bar{\mu}/dc$(+, ×, □) for different $3d$ impurities in iron. Δ, ○— present study and Refs. 1 and 24; ●— Collins and Low (Ref. 4); +— Arrot and Noakes (Ref. 16); ×— Stearns (Ref. 35); and □— Aldred (Ref. 36). For elements at the left of iron (Ti, V, Cr and Mn) the values of $\beta_1(\Delta)$ and $\bar{\mu}_{\text{Fe}} - \bar{\mu}_{\text{imp}}$(○) correspond to lower impurity concentration.

moment as well as the sign of magnetic-moment disturbance depend on temperature as was observed in more concentrated alloys by neutron diffuse scattering measurements[18, 19] and in abnormal temperature behavior of magnetic hyperfine field.[25] It seems to be interesting to do polarized neutron diffuse scattering measurements on dilute Fe-Mn alloys as a function of temperature.

The average host moment, compared to that of pure iron ($2.387\mu_B$), shows a slight tendency to increase. This increase is most pronounced in Fe-Cr alloys.

In Figs. 10, 11, and 12 we compare magnetic-moment distribution in Fe-V, Fe-Cr, and Fe-V with CPA (coherent potential approximation) calculations. Because the concentration variation of diffuse moment μ_0 is unknown, we show the sums of host and diffuse moments ($\bar{\mu}_h+\mu_0$) and impurity and diffuse moment ($\bar{\mu}_{\text{imp}}+\mu_0$) as determined by different authors. The theoretical results (solid lines) comes from the CPA calculations done by Kajzar[26] (Fe-V alloys), Frolani *et al.*[27] (Fe-Cr alloys), and Hasegawa and Kanamori[28] (Fe-Mn alloys). It is seen, that similarly as in Fe-Ti alloys, CPA gives a good description of concentration dependence of average individual moments in more concentrated alloys, but fails at dilute limit. From the departure of CPA results from experimental points one can estimate a limit of CPA applicability which is of about 2 at.% of impurity concentration.

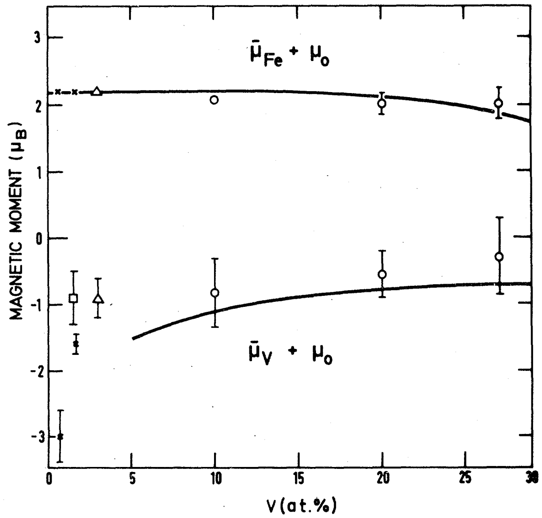

FIG. 10. Magnetic-moment distribution in Fe-V alloys. Solid lines represent the results of CPA calculations by Kajzar (Ref. 26); × —present study; ○—Yamashita *et al.* (Ref. 37); □—Campbell (Ref. 6) (determined from neutron diffuse scattering measurements of Collins and Low, Ref. 4); and Δ—Child and Cable (Ref. 18).

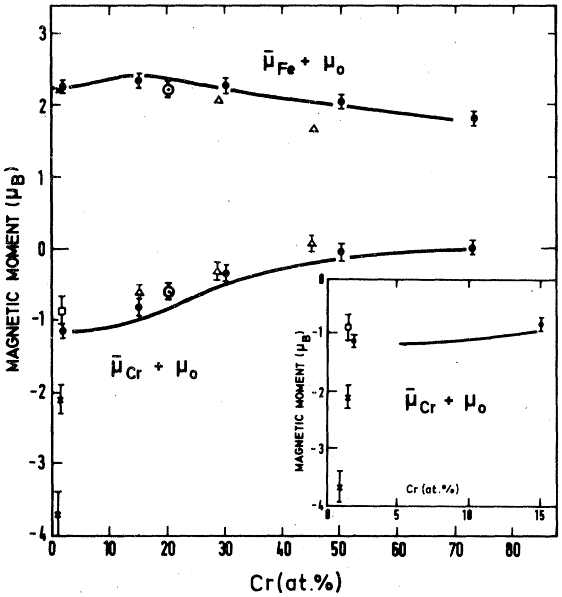

FIG. 11. Magnetic-moment distribution in Fe-Cr alloys. Solid lines represent results of CPA calculations by Frolani *et al.* (Ref. 27); ×—present study; ⊙— Lander and Heaton (Ref. 38); ● — Aldred *et al.* (Ref. 7) (4.2 K); □ — (Ref. 6) (determined from experimental data of Collins and Low, Ref. 4); and Δ— Shull and Wilkinson (Ref. 5) (room temperature). The insert shows impurity moment variation at low chromium concentration.

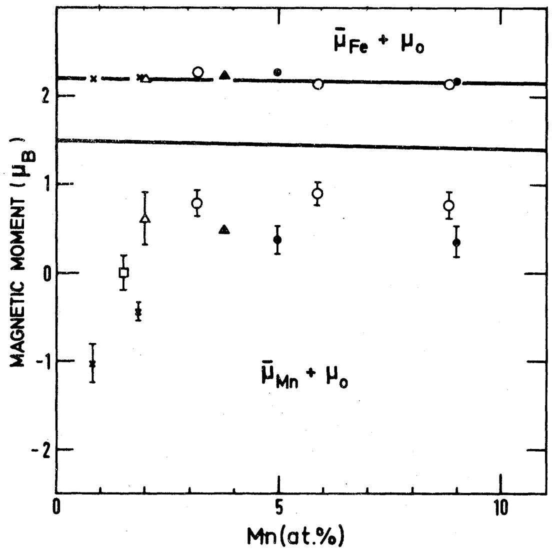

FIG. 12. Magnetic-moment distribution in Fe-Mn alloys. Solid lines represent results of CPA calculations by Hasegawa and Kanomori, (Ref. 28); × — present study; ● — Nakai and Kunitomi (Ref. 39); ○— Radhakrishna and Livet (Ref. 20); Δ—Child and Cable (Ref. 18); □—Campbell (Ref. 6) (determined from experimental data of Collins and Low, Ref. 4); and Δ—Mezei (Ref. 19). All experimental values correspond to room temperature.

The strong concentration dependence of average impurity moment indicates existence of a critical distance d_{crit} between impurities. If the two impurity atoms are in a distance smaller than this critical one, they lose magnetic moment. If they are, however, distant by a value greater than d_{crit}, they can be considered as isolated and carry a large magnetic moment. At dilute limit this critical distance d_{crit} can be evaluated from the following equation:

$$d_{\text{crit}} = (V/N_{\text{imp}})^{1/3} , \tag{19}$$

where V is the sample volume and N_{imp} the number of impurity atoms. For bcc structure we have

$$d_{\text{crit}} = a\,(1/2C_{\text{crit}})^{1/3} , \tag{20}$$

where a is the lattice parameter and C_{crit} is the critical impurity concentration. For $c < C_{\text{crit}}$ the impurity atoms are isolated and their moment does not depend on concentration. From our measurements this concentration can be estimated to be of about 1 at.%. Thus the critical distance will be of about $d_{\text{crit}} \simeq 10.5$ Å. Existence of such critical distance between impurity atoms indicate a long-range interaction between impurity atoms with wide spread (or with long tails) wave functions.

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