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Mössbauer-effect study of a nearly homogeneous $Cr_{75}Fe_{25}$ alloy doped with 0.6 at. % ^{119}Sn

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Using the Mössbauer effect at ^{57}Fe and ^{119}Sn nuclei we study a $Cr_{75}Fe_{25}$ alloy doped with 0.6 at. % ^{119}Sn in the temperature range of 4.2–331 K. We find that the sample, being chemically very homogeneous ($>90\%$), is magnetically very heterogenous, i.e., it cannot be described with only one characteristic magnetic temperature contrary to expectation from the existing phase diagram. On a microscale it consists of a magnetic (M) and a nonmagnetic phase, the latter persisting down to $T\sim9$ K. The contribution of the M phase increases discretely on lowering T. This permits the definition of characteristic temperatures T_N^c at which Fe atoms having another N Fe atoms in the first-neighbor shell become magnetic. At $T\sim20$ K the average hyperfine field shows a steeper change with T which may be indicative of a spin-freezing process expected to occur in that region.

I. INTRODUCTION

$Cr_{1-x}Fe_x$ alloys are among the most often studied magnetic systems. The most intensive investigations have been carried out for Fe concentrations $0.1<x<0.3$. This region is of particular interest because with decreasing x there is a change in the long-range magnetic order from ferromagnetic (F) to antiferromagnetic (A). This has stimulated numerous investigations aiming, first of all, at determining the phase diagram of the system in the T-x plane. In the literature one can find three versions of the phase diagram.

(a) At $T=0$ K there is one critical concentration, $x_A=x_F$, i.e., the F and A regimes meet.

(b) At $T=0$ K there is a gap between the F and A regimes, i.e., $x_A<x_F$.

(c) At $T=0$ K the F and A regimes overlap, i.e., $x_A>x_F$.

Although numerous experiments using different experimental techniques have been carried out, our knowledge of the Cr-Fe system is unsatisfactory and its final phase diagram has not yet been worked out. Most authors favor phase diagram (c), but they do not agree on the common area. Some of them argue that in this region both ferromagnetic and antiferromagnetic long-range order coexist.[1–5] Others, however, do not share this point of view and claim that a spin glass exists in this region.[6,7] We note here that the latter picture is also supported by a recent theory by Jo.[8]

A different phase diagram has recently been suggested by Burke *et al.*,[9] based on neutron scattering and low-field magnetization measurements. Its important features are (i) the A and F regimes do not overlap and (ii) spin-glass behavior occurs only between the critical concentrations $x_A=16$ at. % and $x_F=19$ at. %.

Other uncertainties concerning the magnetic behavior of the system are the values of the critical temperatures T_N (Néel) and T_C (Curie). In fact, the differences in T_C may be as high as 100 K, for a given concentration. It is often suggested (e.g., Loegel *et al.*[10]) that the main source of these differences is related to various experimental techniques involved in measurements. On the other hand, other authors (see, for example, Nemanich *et al.*[11]) show that the T_C values obtained with different methods are in accord. We think that the main reason for the discrepancy of T_C values reported in the literature is not due to different experimental methods but rather due to differences in the samples themselves. We have in mind here the degree of randomness, which may even be different for samples originating from the same ingot and undergoing the same heat treatment (e.g., annealing followed by quenching), but having different final shapes. For example, consider the results by Loegel *et al.*[10] The authors investigate a sample of $Cr_{76}Fe_{24}$ with two methods.

(a) The Mössbauer effect, for which they ground their heat-treated sample.

(b) Low-field permeability, for which they also use their heat-treated samples, as in (a), but as a bulk.

They account for the difference in T_C (110 and 140 K, respectively) by the different time scales involved in the two methods. This explanation is, in our opinion, not correct. Firstly, the authors of Ref. 11, using similar methods, did not detect any difference in T_C. Secondly, Carnegie *et al.*[12] showed for an Ni-Cu alloy that after plastic deformation, the T_C of the sample dropped from 2.5 to 1.2 K. Therefore, we think that Loegel *et al.*[10] obtained a smaller T_C from the Mössbauer-effect measurement because the sample for this experiment had been ground. It is known that a plastic deformation makes the atomic distribution more random. Probably, although both samples had the same nominal concentration, their randomness was different.

Many investigators, being aware of this problem, randomize their samples by annealing at elevated temperature and quenching (usually into water). The difference in T_C values obtained even for samples treated in this way reflect, in our opinion, different degrees of randomness. In other words, the above-described process of randomizing is obviously unsatisfactory. It is clear, from experiments by Carnegie *et al.*,[12] that annealing and quenching is not sufficient to obtain a random sample.

The annealing temperature, the sample volume, and the quenching rate all play important roles. We conclude from the above observations that the degree of randomness seems to be a dominant factor in determining the magnetic behavior of Cr-Fe alloys. It follows that microscopic methods are best for approaching the problem. Among them, Mössbauer-effect (ME) spectroscopy should be very useful as the hyperfine parameters are very sensitive to the local environment.

Hesse and Schossow[13] and Hesse and Rübartsch[14] were among the first to apply ME to Fe_xCr_{1-x} alloys for $0.24 \leq x \leq 0.31$. The results they obtained are of particular interest as they explicitly show how the heat treatment drastically influences T_C. In particular, for $Cr_{71}Fe_{29}$, $T_C = 295\,^\circ C$ or $395\,^\circ C$ for a quenched or slowly cooled sample, respectively. Based on the linear correlation "T_C versus degree of randomness" they obtained $T_C = 120$ K, by extrapolating to a completely random alloy. This shows how ill random their best real sample was. It follows that T_C can be taken as a good measure of the randomness of the Cr-Fe alloy.

In this paper we present results of our Mössbauer-effect measurements carried out on a very homogeneous (i.e., very randomly distributed) sample of $Cr_{75}Fe_{25}$ alloy doped with 0.6 at. % ^{119}Sn for a temperature range of 4.2—331 K. We discuss them in the light of the existing phase diagrams of this system.

II. EXPERIMENTAL DETAILS

A. Sample preparation

As master material we used 310 mg of $Cr_{75}Fe_{25}$ alloys (nominal composition) donated by S. K. Burke. The description of its preparation can be found elsewhere.[15] Here, we only want to emphasize that it was homogenized by annealing at 1050 °C for a few days followed by water quenching. The authors of Ref. 15 also claim that the concentration of Fe deviates from the average value by not more than $\pm 5\%$ across the sample. We melted the master alloy together with 7 mg of tin enriched to $\sim 91\%$ with ^{119}Sn in two steps. First, the sample was heated in an arc furnace under an atmosphere of pure argon. Here, the weight loss was ~ 2 mg. Second, the sample was heated in an induction furnace, again in an argon atmosphere. The sample lost ~ 1 mg in weight here.

Finally, we gave the sample a homogenizing treatment by annealing it in vacuum ($p < 10^{-4}$ mbar) at $T = 1000\,^\circ C$ for 24 h. Afterwards, the sample was quenched in oil. As a result of the last heat treatment the sample lost ~ 1 mg of weight.

Chemical analysis carried out on part of the sample after the final homogenization yielded the following composition: $x_{Fe} = 25.063$ at. %, $x_{Cr} = 74.353$ at. %, and $x_{Sn} = 0.583$ at. %. Assuming the nominal composition was the real starting composition we must conclude that (a) the mass loss was 70% tin and (b) tin replaced mainly Cr.

B. Mössbauer-effect measurements

For the Mössbauer-effect measurements the sample was filed, giving an average particle size of 60 μm. The density of the probe ^{119}Sn nuclei was 1 mg/cm^2. The sample was placed in a helium flow cryostat and its temperature was kept constant between 4.2 and 331 K with an accuracy of 0.1—1 K depending on the temperature range.

We carried out the measurements with a standard spectrometer having a 512-channel analyzer and we collected spectra using the Mössbauer effect both at ^{57}Fe and ^{119}Sn nuclei. For the former, 14.4-keV γ rays were emitted by a source of ^{57}Co in rhodium, while for the latter $CaSnO_3$ was used as a source of 23.8-keV γ rays. Spectra with good statistics could be collected within 24—36 h for ^{57}Fe and within 48 h for ^{119}Sn.

As calibration standards we used a metallic iron foil and $BaSnO_3$ powder.

III. RESULTS AND DISCUSSION

A. ^{57}Fe-site spectra

Figure 1 shows a set of selected ^{57}Fe-site Mössbauer spectra for various temperatures T. They resemble in shape those reported earlier[10,11,13,14] in that (a) they are, at first glance, symmetric, and (b) they consist of a single-line subspectrum and a broadened subspectrum. The latter obviously increases its contribution and its splitting with falling temperature becoming predominant at lowest temperatures. The main difference compared with the previous spectra of other authors is that the broad subspectrum can be detected in our case only for $T \sim 100$ K, while Nemanich *et al.*[11] observed it already around 160 K and the authors of Refs. 13 and 14 observed it at higher T. The most frequently given Curie temperature for this composition is $T_C \sim 160$ K, i.e., at $T \sim 100$ K the sample should already be quite ferromagnetic.

The question is, why we do not see any indication of the phase transition at $T \sim 160$ K. Our spectra remain single-line ones with constant width down to $T \sim 100$ K where something occurs. At this point two questions arise: (1) Why such a big discrepancy in behavior can be observed with the same experimental technique for samples having a similar nominal composition, and (2) does the appearance of the broadened subspectrum indicate the phase transition (paramagnetic-ferromagnetic), i.e., can the temperature at which this occurs be attributed to T_C? We think that the answer for the first question is rather simple. It should be the difference in the actual distribution of Fe atoms in the Cr matrix which causes such differences in the shape of Mössbauer spectra [the hyperfine (hf) parameters and especially the hf field are very sensitive to the local environment]. Hesse *et al.*[13,14] have already showed that a more homogeneous distribution leads to a lower value of T_C. We also think that the answer to the second question is not difficult. Using the Mössbauer effect, one investigates a system on a microscale, i.e., the existence of a split spectrum does not necessarily mean that the whole system is magnetically ordered. However, if the whole system becomes magnetic, i.e., if there exists long-range order, then the total Mössbauer spectrum will split in all cases.

This is obviously not the case here, as the spectrum does not broaden as a whole, but at all T down to ~ 50 K it is dominated by the single-line subspectrum. This, in

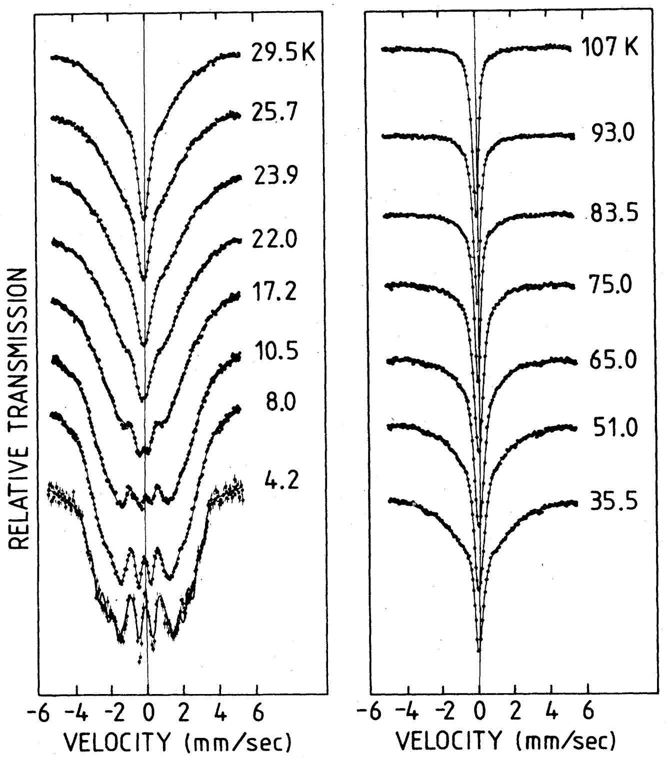

FIG. 1. ^{57}Fe-site Mössbauer spectra of $Cr_{75}Fe_{25}$ for different temperatures.

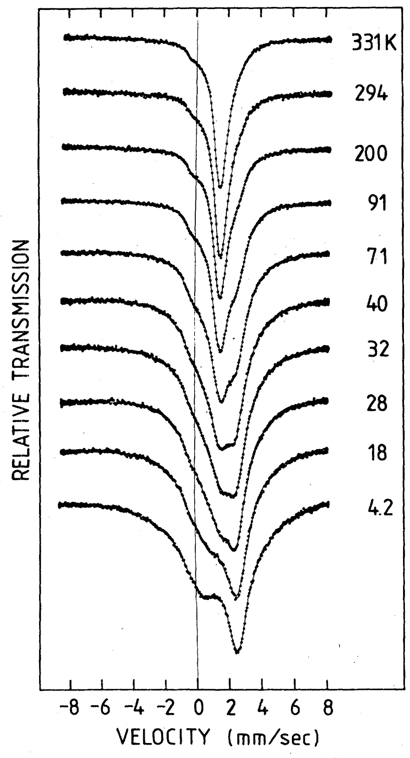

FIG. 2. ^{119}Sn-site Mössbauer spectra of $Cr_{75}Fe_{25}$ for different temperatures.

our opinion, must be taken as an argument that at $T \sim 100$ K there is no uniform phase transition from a paramagnetic (P) to a ferromagnetic state. Loegel *et al.*[10] suggested that superparamagnetism should be responsible for the actual shape of the Mössbauer spectra. If this was the case, the Mössbauer spectra would be symmetric. The analysis of our spectra showed, however, that the isomer shifts of the single-line subspectrum differ meaningfully from that of the broad subspectrum. In addition, ^{119}Sn-site spectra shown in Fig. 2 are completely asymmetric, although the corresponding time-scale involved in their measurement is similar to that characteristic of ^{57}Fe nuclei.

Due to these arguments, we assume a static model in the following analysis of the spectra. They were analyzed in two independent ways, I and II. In the analysis I it was assumed that each spectrum consists of two parts: a nonmagnetic one associated with the single-line subspectrum and related to the nonmagnetic phase in the sample and a magnetic (M) one associated with the broadened subspectrum and related to the magnetic phase of the sample. From the fitting procedure we obtained the relative abundance of the magnetic phase, A_M:

$$A_M(\%) = \frac{S_M}{S_T} 100 , \tag{1}$$

where S_M stands for the area of the magnetic subspectrum and S_T is the area of the total spectrum. It is now interesting to plot A_M versus T to see how the magnetic phase develops with decreasing T.

Figure 3 provides this plot. Its main feature is that A_M increases in a steplike manner. In fact, there are six steps, just as many different atomic configurations as are expected assuming the Fe atoms are distributed at random and including only the first neighbor shell (NN) (probabilities of configurations with six and more Fe atoms in NN are so small that they could not be detected with ME).

Therefore we associate the steps in A_M with these configurations which seem to behave like independent mag-

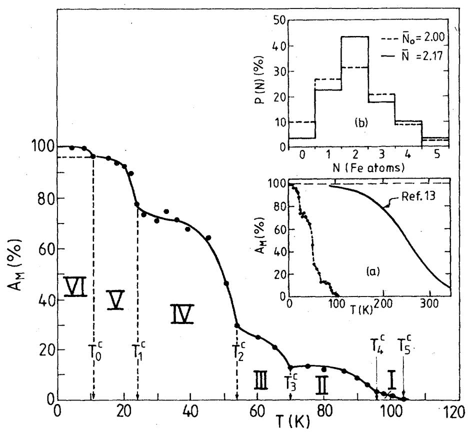

FIG. 3. Magnetic phase contribution A_M vs temperature T. Inset (a) shows a comparison of presently determined A_M (dots) with that given in Ref. 13, and inset (b) presents the probability histogram as obtained (——) and as expected for the random distribution (— — —).

netic units as they become magnetic at rather well-defined temperatures, $T_N^c(N=0,1,2,3,4,5)$ characteristic of a given configuration. If we plot T_N^c versus N (see Fig. 4) we obtain a nice linear correlation which permits extrapolation to $T_8^c=166$ K, i.e., the characteristic microscopic temperature at which an Fe atom having its maximum number of Fe neighbors becomes magnetic. T_8^c coincides with the temperature which is most frequently claimed as the Curie temperature of this system. It is also interesting to note [see Fig. 3, inset (b)], how much our A_{M} differs from the corresponding quantity given in Ref. 13 for a very heterogenous sample.

If our association of the steps observed in A_{M} with the atomic configurations is correct, then Fig. 3 permits one to evaluate the abundances of these configurations (as differences between subsequent plateaus). The inset (a) in Fig. 3 shows a comparison between such values obtained this way (solid line) and those calculated from a random distribution (dashed line). We see that the actual histogram is quite similar to that expected from the random distribution. This reflects itself in the figures, namely, the average number of Fe atoms within the NN shell $\overline{N}=2.17$ which differs only by 8.5% from the corresponding value $\overline{N}_0$ expected for the random distribution. Therefore, we concluded that our sample is to a high degree ($>90\%$) chemically homogeneous. This also explains, in the light of arguments given in Ref. 13, why the first indication of micromagnetic order occurs only at $T\sim 100$ K. The behavior of A_{M} also shows, that from a magnetical point of view, the sample is very heterogeneous.

The magnetic inhomogeneity of the Fe-Cr system for this or a similar composition is already in the literature as it has been observed by various investigators using different experimental techniques. However, it has been shown, to our knowledge for the first time, that on a microscale, there is not one temperature which would mark a phase transition. Instead, there is a discrete spectrum of such temperatures T_N^c which are strictly determined by the number of Fe atoms in the corresponding neighbor configuration. This picture seems to have support from theoretical calculations by Maksymowicz[16] based on the configurational model of magnetic moments.

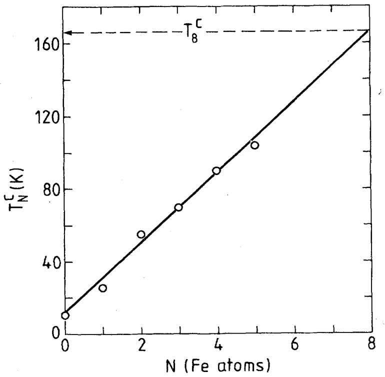

FIG. 4. Characteristic temperatures of micromagnetic order T_N^c vs number of Fe atoms in the NN shell N.

In the second way (II) of spectra analysis we used the method by Window[17] which permitted us to obtain field distributions, $P(H)$, from the Mössbauer spectra. Those $P(H)$ corresponding to the spectra shown in Fig. 1 are presented in Fig. 5. They reflect quite well the magnetic heterogeneity of the sample. In particular, the nonmagnetic phase persists down to $T\sim 10$ K and the magnetic phase is characterized by a broad field distribution. These two features agree well with the results obtained by the first analysis of the spectra and thereby support it.

By integrating $P(H)$ over H we also evaluated the average hf field $\overline{H}$ which is presented in Fig. 6 as a function of T. We note two features. First, the behavior of $\overline{H}$ around the temperature (~ 110 K) where $\overline{H}$ starts to increase (the broad subspectrum appears) is irreversible. This fragment of the $\overline{H}$-T plot is shown again as inset (a) in Fig. 6 where the irreversibility is easily seen. The understanding of the onset of micromagnetic order which obviously occurs here seems to be further complicated by the fact that, before the magnetic subspectrum appears, the width of the single-line spectrum (keeping its Lorentzian shape) slightly increases as T decreases from ~ 120 to ~ 110 K. The width then experiences a small, sharp increase [see inset (b) in Fig. 6] corresponding to an increase of $\overline{H}$ by ~ 0.5 kOe. Although it is a rather small effect it cannot be neglected, as this type of feature can be observed for all T.

The linewidth of the single-line subspectrum oscillates irregularly with T. This could mean that some small cooperative (itinerant) mechanism, e.g., spin waves or polarization of conduction electrons exists for all T and causes the paramagnetic phase to show some remanent

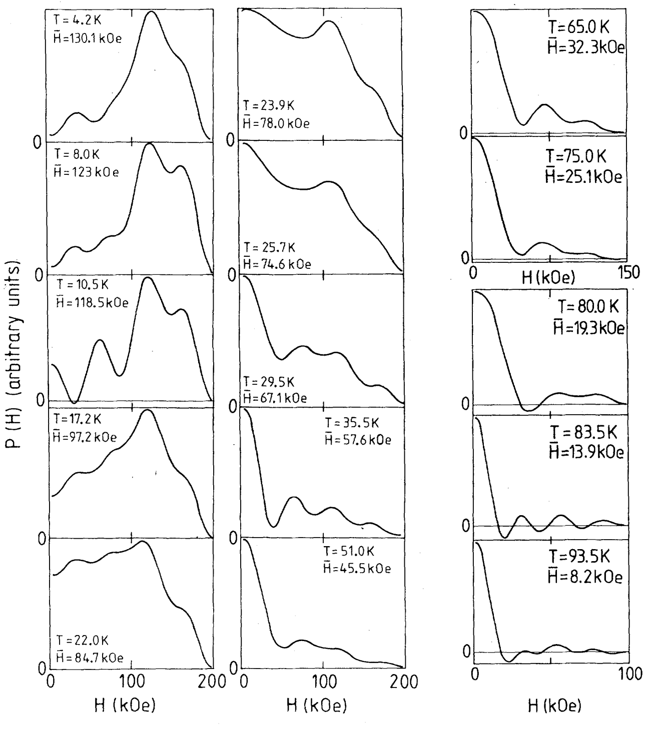

FIG. 5. Hyperfine field distributions as obtained from the spectra shown in Fig. 1.

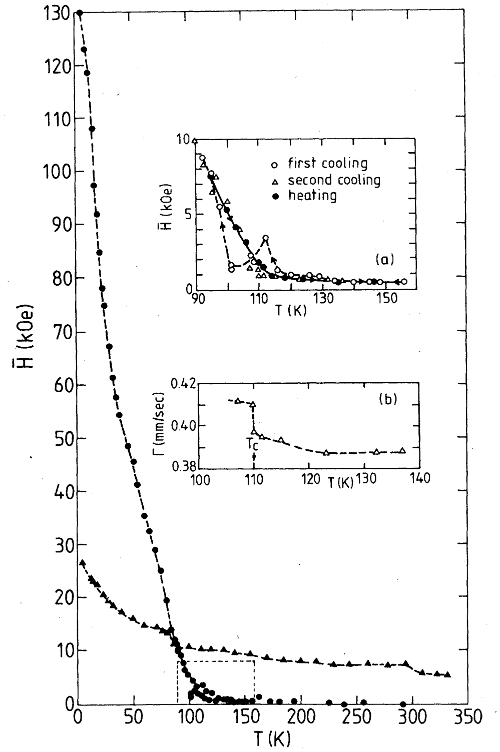

FIG. 6. The average hf field $\overline{H}$ as obtained from the ^{57}Fe-site spectra (●) and from the ^{119}Sn-site spectra (▲). Inset (a) shows a fragment of the $\overline{H}$-T plot in the vicinity of T where the onset of micromagnetic order appears, and inset (b) illustrates the dependence of the linewidth of a single-line spectrum on T in this region.

polarization. (It seems reasonable here to argue again that it is not relaxation which makes part of the single-line spectrum, otherwise its width should increase continuously with decreasing T.)

A second feature of the $\overline{H}$-T plot worth mentioning is that $\overline{H}$ does not change with T in a Brillouin-like fashion, but around $T \sim 20$ K it experiences a steeper increase. This feature shows up clearer when plotting $d\overline{H}/dT$ versus T (see Fig. 7). Such an additional increase of $\overline{H}$ could be related with a freezing process of Fe spins, characteristic of a spin-glass state. Measurements in external fields which may cast more light on this problem are in progress.

B. ^{119}Sn-site measurements

^{119}Sn-site measurements were carried out in the same temperature range as those at ^{57}Fe nuclei. As already mentioned, the resultant Mössbauer spectra (Fig. 2) are asymmetric, i.e., they do not have a relaxation character. Because of the asymmetry they could not be fitted with the Window method either. Instead we fitted them assuming they consisted of a number of subspectra, each corresponding to a probe ^{119}Sn nucleus with differing numbers of Fe (and Cr) atoms in the NN shell.

It was necessary to include seven such subspectra with

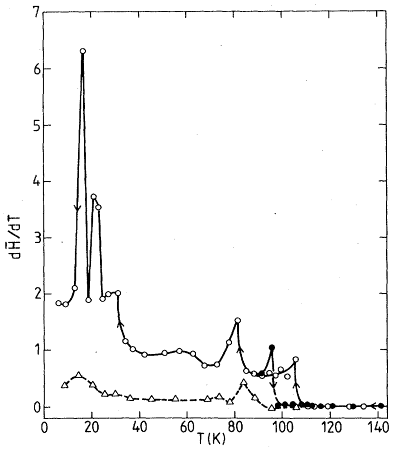

FIG. 7. Temperature derivation of the average hf field, $d\overline{H}/dT$ vs temperature T for ^{57}Fe site (○) and for ^{119}Sn-site results (△).

different isomer shifts to successfully and consistently fit all the spectra. The inset of Fig. 8 shows the actual histogram and the one expected from the random distribution. One can see that the former deviates from randomness (by 27%) and is shifted towards a larger number N of Fe atoms in the NN shells. This can be explained in part by the fact that Sn atoms have mainly substituted for Cr atoms. It also means that there is some attraction between Sn and Fe atoms.

The above analysis makes it possible to trace $H(N)$ versus T for each of the seven configurations, $N=0,1,2,3,4,5,6$. The behaviors obtained are presented in Fig. 8 [it was assumed that $H(0)<0$]. The characteris-

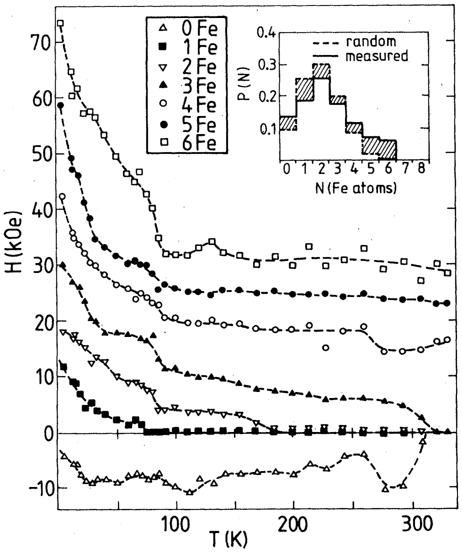

FIG. 8. Hyperfine field at ^{119}Sn sites having $N=0$–6 Fe atoms in the NN shell H vs temperature T. The inset shows the actual probability histogram (———) and the one expected from the random distribution (— — —).

tic and rather unexpected feature is that for all $N \geq 2$, $H(N) > 0$, even for T larger than ~ 100 K (i.e., the temperature at which the first indication of micromagnetic order in the ^{57}Fe-site spectrum appears). $H(2)$ vanishes at $T \sim 180$ K, $H(3)$ at $T \sim 320$ K, and the splitting associated with $N \geq 4$ could be detected even at $T = 331$ K (i.e., the highest temperature measured). However, the common feature of all $H(N)$, $N > 0$, is that the influence of T is mainly confined to the region below $T \sim 100$ K. Here, we can distinguish two steps: the first around 90 K, the second around 30 K.

It is also interesting to note that the $H(0)$ field is almost T independent and vanishes at $T \sim 310$ K which exactly agrees with the Néel temperature of pure chromium. It should be mentioned that Fe in a pure Cr environment shows at $T = 4.2$ K a small hf field of ~ 35 kOe only[20] due to a compensation of the Fe-core polarization field and the transferred hf fields. Therefore, it is not astonishing that the high characteristic temperatures detected by ^{119}Sn are not observed in the ^{57}Fe spectra.

Knowing the probability of each $H(N)$, $P(N)$ we evaluated the average field $\bar{H}_{\mathrm{Sn}} = \sum H(N)P(N)$ and plotted it in Fig. 6 together with the Fe-site average field, $\bar{H}_{\mathrm{Fe}}$. Although its behavior versus T is rather different from that of $\bar{H}_{\mathrm{Fe}}$ ($\bar{H}_{\mathrm{Sn}}$ has only a transferred origin), its common feature which can be seen in Fig. 7 is that its temperature derivative $d\bar{H}/dT$ exhibits two characteristic peaks at $T \sim 20$ K and $T \sim 90$ K.

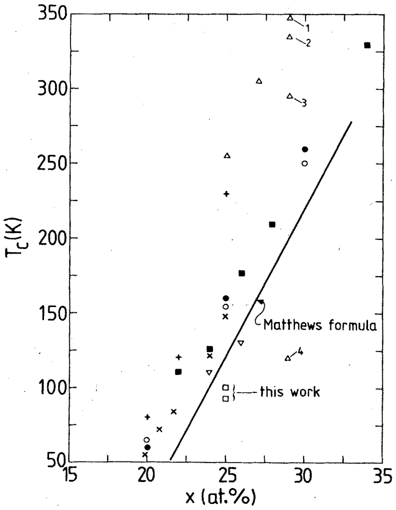

FIG. 9. Fragment of the phase diagram of the $\mathrm{Cr}_{1-x}\mathrm{Fe}_x$ system showing a compilation of literature data of the "Curie" temperature T_C vs the iron concentration x. The solid line has been shown according to the Matthews formula (Ref. 18). The meaning of other symbols is as follows: △ represents Ref. 14 (1 represents slowly cooled, 2 represents as obtained, 3 represents water quenched, and 4 represents homogeneous extrapolated value), ● represents Ref. 11, ■ represents Ref. 7, 0 represents Ref. 19, × represents Ref. 9, + represents Ref. 4, ∇ represents Ref. 10 and □ represents this work (for the ^{57}Fe-site result, the upper case, and for ^{119}Sn-site result, the lower case).

IV. CONCLUDING REMARKS

The results which we obtained in the present investigations can be summarized with the following conclusions.

(1) The $\mathrm{Cr}_{75}\mathrm{Fe}_{25}$ alloy sample studied was to a high degree ($>90\%$), chemically homogeneous.

(2) For the first time the detailed microscopic properties of such a homogeneous sample have been revealed.

(a) The sample is magnetically heterogeneous; it consists of a magnetic (*M*) and a nonmagnetic (*NM*) phase. Part of the latter (ascribed to Fe nuclei with no other Fe atoms in the NN shell) persists down to $T_0^c \sim 9$ K.

(b) A steplike increase of the M-phase abundance observed on lowering T enables determination of characteristic temperatures T_N^c, linearly correlated with the number of Fe atoms in the NN shell N.

(c) T_N^c-N correlation permits one to estimate $T_8^c \sim 166$ K, i.e., the maximum temperature at which the onset of micromagnetic order can occur in this sample.

(d) From the existence of these characteristic temperatures, attributed to discrete Fe configurations, we have to conclude that short-range magnetic interactions play a dominant role in the $\mathrm{Cr}_{1-x}\mathrm{Fe}_x$ system, at least for the Fe concentration $x = 0.25$.

(3) The average hf field can be taken to some extent as a measure of a bulk feature. There are two characteristic temperatures: $T_{\mathrm{I}} \sim 90$—100 K and $T_{\mathrm{II}} \sim 20$ K. The former corresponds to the onset of the micromagnetic order for $N = 5$, the latter may indicate a transition to a spin-glass state.

(4) The present investigation demonstrates the importance of chemically homogeneous samples in studying the magnetic properties of the $\mathrm{Cr}_{1-x}\mathrm{Fe}_x$ system, especially for $x \leq 0.35$. Fig. 9 shows a compilation of what has been called the Curie temperature versus x for $0.20 \leq x \leq 0.35$. One can readily see a wide spread in the data, indicative of a high sensitivity of magnetic properties of this system on the chemical order.

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