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Mössbauer-effect study of the reentrant transition in $Cr_{75}Fe_{25}$ alloy

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In-field ^{57}Fe Mössbauer-effect measurements reveal that a $Cr_{75}Fe_{25}$ alloy undergoes a transition from a "ferromagnetic" to a spin-glass state. The influence of an external magnetic field on the transition is also studied.

One of the most interesting phenomena which has been recently studied is the so-called reentrant transition from a ferro- (antiferro-) magnetic to a spin-glass (SG) phase. The transition is driven by temperature T and has been predicted theoretically[1-3] as well as observed experimentally for various systems including $Cr_{1-y}Fe_y$.[4-10]

Concerning this system its phase diagram has not yet been completely elaborated. The main confusion concerns the border of the spin-glass phase. According to Ref. 9 the SG does not exist for $y \geq 19$ at.% which is in conflict with measurements reported in Ref. 10 where it is claimed that the SG was observed for $0.05 \leq y \leq 0.22$. In our recent Mössbauer-effect (ME) study of an $Cr_{75}Fe_{25}$ alloy[11] we observed at $T \simeq 35$ K a steep increase of the average ^{57}Fe-site hyperfine (hf) field on lowering T which may indicate a transition into the SG phase. In this Rapid Communication we report on our further studies of this sample by means of ME measurements in an external magnetic field, H_{ext}. The idea underlying this method to study the FM $\rightarrow$ SG transition is based on the fact that if the propagation of the γ rays is parallel to the local magnetization vector **M** then the nuclear transitions for $\Delta m = 0$ vanish and consequently the second and fifth lines in the ^{57}Fe site Mössbauer spectrum disappear. If, however, the γ rays propagate perpendicularly to **M** the intensities of the two lines $I_{2,5}$ are four times larger than those of the third and fourth lines $I_{3,4}$. For the intermediate case $0 < x < 4$ is valid ($x = I_{2,5}/I_{3,4}$). In order to detect in that way a FM $\rightarrow$ SG transition one saturates the magnetic moments in the FM phase using an external field H_{ext} and measures the spectra at different decreasing temperatures T. Entering the SG state at $T = T_f$ the magnetic moments will be no longer aligned and, consequently, the intensity ratio becomes $x \neq 0$. This method has already been applied to study the FM $\rightarrow$ SG transition in the Au-Fe alloys.[12-14] However, the results obtained by the two groups are not quite compatible. In particular, the authors of Ref. 12 observed for a 17 at.% Fe-Au alloy a sharp transition, in agreement with the model of Gabay and Toulouse,[15] while in Ref. 13 the transition observed for a 19 at.% Fe-Au alloy was weak, i.e., T_f was not well defined experimentally. However, in the two experiments rather different values of H_{ext} were used (20 and 6 kOe, respectively) which may be, at least partly, responsible for the observed discrepancy. Therefore, the second aim of our present investigation was to study the influence of H_{ext} on T_f in more detail.

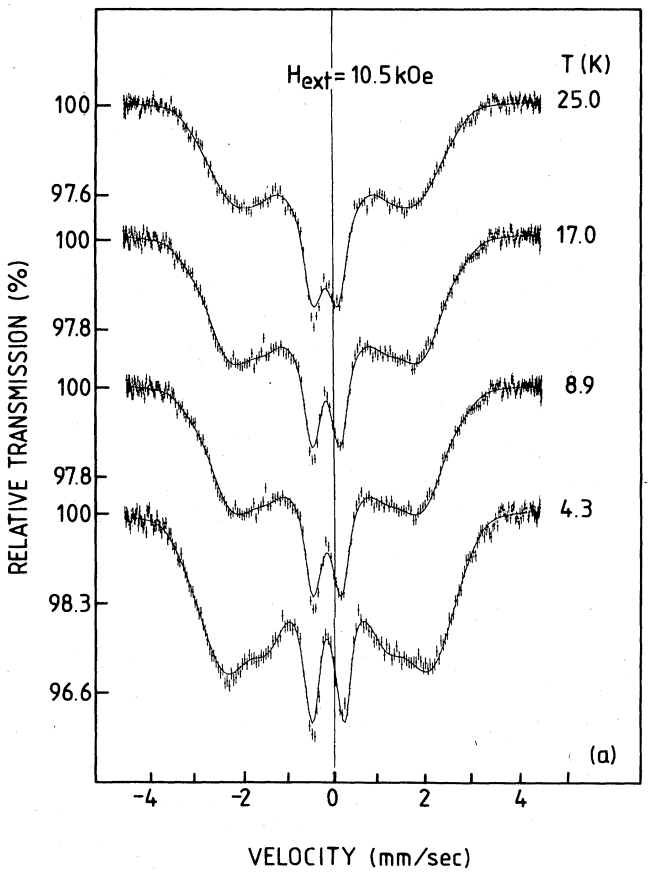
We collected the ^{57}Fe Mössbauer spectra using the same sample and setup as for the zero-field measurements.[11] As demonstrated in Ref. 11 the Fe/Cr chemical distribution within this sample can be regarded as random to a high degree. We started the measurements performed at different decreasing temperatures with cooling the sample from $\simeq 70$ K in a longitudinal magnetic field $H_{ext} = 10.5$ kOe. Each spectrum was collected within a 5–6 day run. After the 4.3-K spectrum was completed we started measurements with increasing T. Having completed this cycle of measurements, the sample was again slowly (within ~ 3 h) cooled in $H_{ext} = 10.5$ kOe down to 4.3 K and a spectrum was collected again. Afterwards T was kept constant and we measured spectra at different fields $H_{ext} = 20$, 5, and 0.3 kOe. Then the sample was heated up to room temperature and afterwards again cooled down to 4.3 K (from ~ 70 K in $H_{ext} = 10.5$ kOe), but this time rather fast (within ~ 20 min). After reaching 4.3 K a spectrum was collected. Then we again heated the sample up to ~ 70 K and started a new cycle by cooling it down in $H_{ext} = 20$ kOe taking spectra with 5–6 day running time. When this series of measurements was completed, the sample was heated up to ~ 70 K and then cooled fast in $H_{ext} = 20$ kOe down to 4.3 K taking a spectrum at this temperature. Figure 1(a) shows some spectra as obtained at $H_{ext} = 10.5$ kOe for different T, and Fig. 1(b) presents the spectra as measured at $T = 4.3$ K for different H_{ext}.

Although the resolution in this case is not as good as it was for the Au-Fe system,[12,13] one can readily see that on lowering T the intensities of the lines 2 and 5 increase. For a quantitative evaluation we fitted the spectra based on the field-distribution method outlined by Window[16] (the spectra are fairly symmetric, so the method is applicable). To choose the best value of the x parameter we used the following criterion: The standard deviation χ^2 should be minimum by changing the x value as a free parameter by steps and taking into account for the fit only field distributions $P(H)$ without negative amplitudes [$P(H)$ = distribution probability of the hf field H]. It has turned out that the field distribution was affected very sensitively by the x values: the typical error of x was ± 0.05. Figure 2 illustrates the influence of x on $P(H)$. Using this method we evaluated the x values for all the spectra measured. They are presented in Fig. 3 from which we are able to draw the following conclusions.

(1) An Fe-Cr alloy containing 25 at.% Fe exhibits a reentrant behavior which is rather sharp, i.e., similar to that observed for an 17 at.% Fe-Au alloy.[12]

(2) The transition does not show any hysteresis (within the accuracy of the present experiment).

(3) The transition temperature T_f^* depends clearly on H_{ext} in such a way that an increase of H_{ext} decreases T_f^*. This remains at least in a qualitative agreement with theoretical models,[15,17] and potentially it permits one to test the models



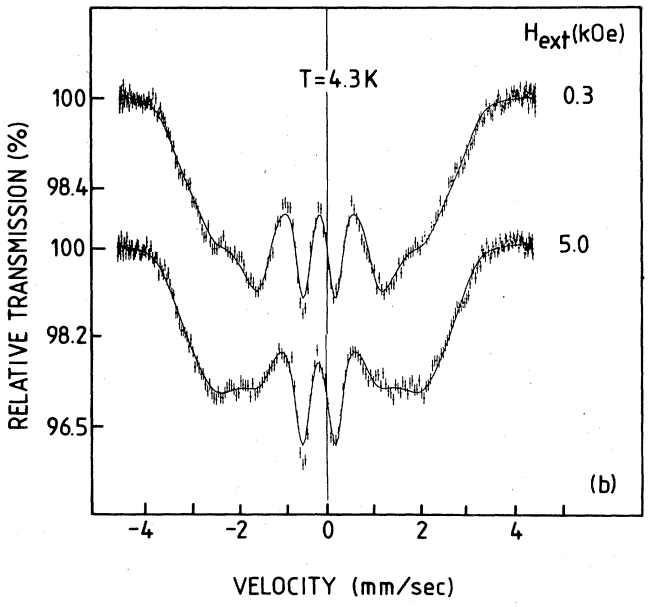


FIG. 1. Mössbauer spectra (a) as obtained in $H_{ext} = 10.5$ kOe for various temperatures T, (b) as obtained at $T = 4.3$ K after cooling in $H_{ext} = 10.5$ kOe and measured in various H_{ext} as shown. The solid lines are the best fits to the data.

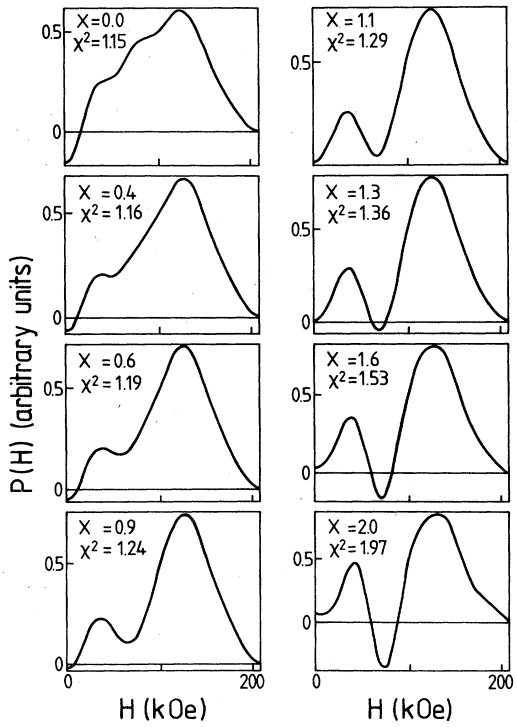


FIG. 2. hf field distributions as obtained for the studied sample from the Mössbauer spectra measured at $T = 4.3$ K and in $H_{ext} = 20$ kOe. The field distribution corresponding to the best fit was obtained with $x = 1.1$.

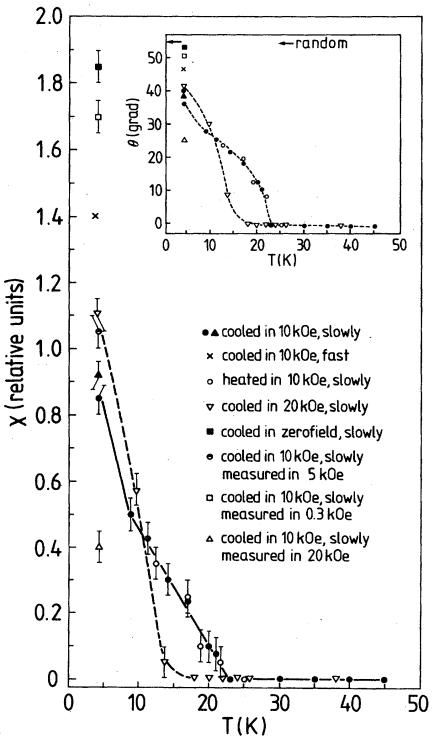


FIG. 3. Relative intensity of the second and fifth line, x vs temperature T. The inset shows the angle θ related to x by $\sin^2\theta = 2x/(4+x)$; the arrow indicates the value of θ for a random distribution of spins. The lines are drawn to guide the eye only.

by studying T_f^x vs H_{ext}.

(4) The nature of the canting which starts at T_f^x is not that of the usual anisotropy, since it depends strongly on the particular temperature and field cycling procedure, as shown in detail in Fig. 3. For example, one should note that cooling in a larger H_{ext} produces larger x.

(5) Dynamical aspects of the canting process can be seen through different values of x as revealed for different rates of cooling.

(6) The sample is magnetically heterogeneous as it can be described in terms of two phases reflected by the two prominent peaks in the hf distributions; their mutual contribution changes with T [Fig. 4(a)] and H_{ext} [Fig. 4(b)] in a different way.

The insert of Fig. 3 shows the average angle θ between $\mathbf{M}$ and H_{ext}. It is interesting to note that although for $H_{ext}=20$ kOe the process of canting sets in at lower T_f^x (H_{ext} stabilizes the FM phase) than for $H_{ext}=10.5$ kOe, it proceeds faster, and below $T\simeq 10$ K the difference between the two phases is more pronounced, i.e., $\theta(20\text{ kOe}) < \theta(10.5\text{ kOe})$.

In Ref. 14 it was shown that T_f as deduced from the x values measured in external fields corresponds with T_f determined from the anomaly of the average hf field, $\overline{H}$, as obtained from zero-field measurements. However, it did not agree with T_f obtained from the anomaly of $\overline{H}$ as obtained from the in-field measurements. This inconsistency although neglected by the authors could be important as it may have serious consequences as far as the validity of the method itself is concerned. We have plotted in Fig. 5 $\overline{H}$ vs T for measurements at $H_{ext}=0$, 10.5, and 20 kOe, in order to see in our case (i) if and how H_{ext} influences $\overline{H}$, (ii) if $\overline{H}$ exhibits any anomaly at all, and (iii) how such an anomaly would compare to the T_f^x data. In addition, we displayed also $H_{max}(1\text{ T})$, i.e., the peak value of H in the field distributions for the measurements at $H_{ext}=10.5$ kOe. The results presented in Fig. 5 can be summarized as follows.

(a) Both $\overline{H}$ and H_{max} (1 T) exhibit anomalies at certain temperatures which can be regarded as spin-freezing temperatures $T_f^{\overline{H}}$ [the anomaly of $\overline{H}(0)$ is very weak and it could be only vizualized by plotting $d\overline{H}(0)/dT$ vs T, see Ref. 11].

(b) $T_f^{\overline{H}}$ values like T_f^x values are reduced by H_{ext} in

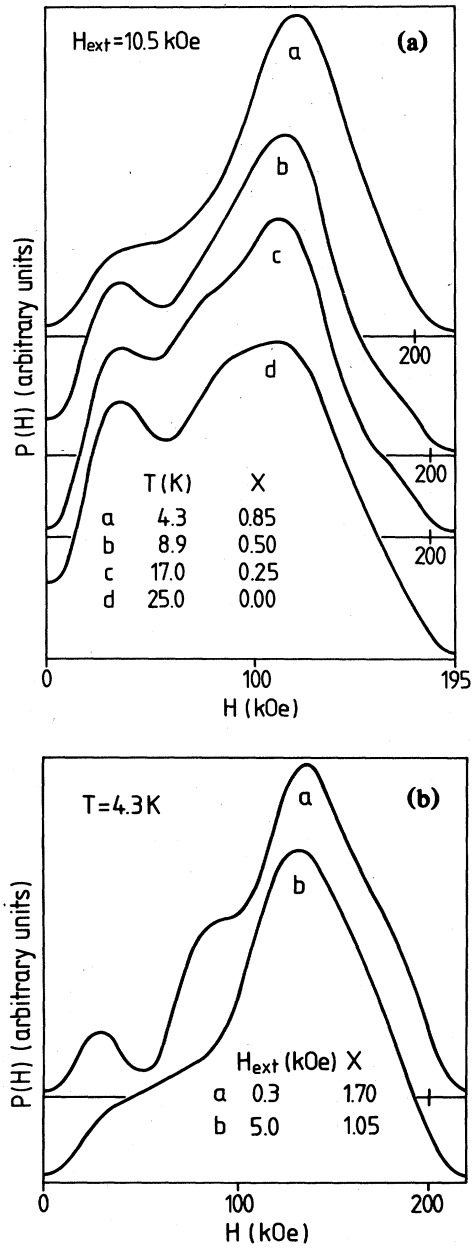


FIG. 4. hf field distributions corresponding to the best fits of the spectra shown (a) in Fig. 1(a) and (b) in Fig. 2(b). Note the change of shapes with T (a) and with H_{ext} (b).

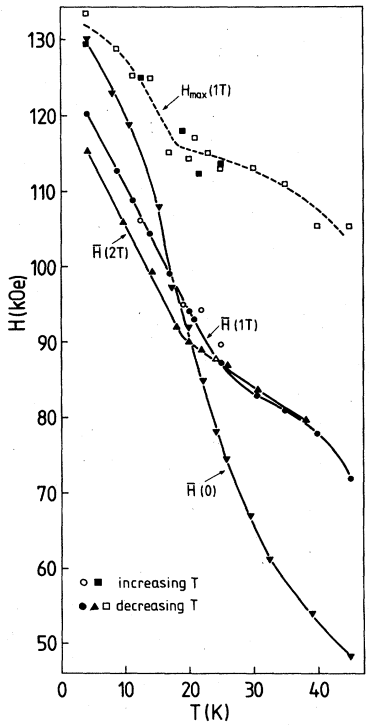


FIG. 5. Average hf field $\overline{H}$ and the most probable field H_{max} vs T, as obtained from the field distributions. The lines are drawn to guide the eye only.

a similar way: $\Delta T_f^{\bar{H}} = T_f^{\bar{H}}(2\ \mathrm{T}) - T_f^{\bar{H}}(1\ \mathrm{T}) \simeq -7$ K and $\Delta T_f^{\chi} = T_f^{\chi}(2\ \mathrm{T}) - T_f^{\chi}(1\ \mathrm{T}) \simeq -8$ K. They do not coincide with each other as $T_f^{\chi}(1\ \mathrm{T}) \simeq 23$ K, $T_f^{\bar{H}}(1\ \mathrm{T}) \simeq 26$ K, $T_f^{\chi}(2\ \mathrm{T}) \simeq 15$ K, $T_f^{\bar{H}}(2\ \mathrm{T}) \simeq 19$ K.

(c) $T_f^{H^{\max}}(1\ \mathrm{T}) \simeq 19$ K agrees neither with $T_f^{\bar{H}}(1\ \mathrm{T})$ nor with $T_f^{\chi}(1\ \mathrm{T})$.

The three temperatures fulfill the relation $T_f^{H^{\max}} < T_f^{\chi} < T_f^{\bar{H}}$.

In conclusion, we see that, although in-field ME measurements permit the detection of a FM → SG transition, the open question remains which quantity must be taken as an adequate measure of the transition temperature T_f. Possibly, the present results may indicate that there is no unique T_f. The idea of the existence of a series of freezing temperatures was already introduced by Murani.[18] The difference between $T_f^{\bar{H}}(1\ \mathrm{T})$ and $T_f^{H^{\max}}(1\ \mathrm{T})$ as revealed in the present study is compatible with this idea: we see that for all temperatures $H^{\max}(1\ \mathrm{T}) > \bar{H}(1\ \mathrm{T})$ and $T_f^{H^{\max}}(1\ \mathrm{T}) < T_f^{\bar{H}}(1\ \mathrm{T})$ in agreement with the expected[15,17] influence of H_{ext} on T_f, which has been demonstrated now experimentally in this work.

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