Magnetic behavior of $Cr_{74}Fe_{26}$ alloy investigated by Mössbauer spectroscopy

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Two recently published comments [P. A. Beck, Phys. Rev. B **39**, 752 (1989) and D. Boumazouza *et al.*, Phys. Rev. B **39**, 749 (1989)] have drawn attention to the interesting magnetic behavior of reentrant spin-glass systems. We present a temperature-dependent Mössbauer-effect study on $Cr_{74}Fe_{26}$ alloy whose composition is close to and slightly above the percolation threshold ($x_{Fe}=19.9$ at. %), to resolve the controversy that has arisen out of these discussions. Our results suggest that the nature of magnetic ordering at the paramagnetic-to-ferromagnetic transition in this system also does not correspond to a true cooperative transition, as pointed out by Beck for $Au_{82.5}Fe_{17.5}$ alloy.

The nature of magnetic ordering at the paramagnetic-to-ferromagnetic (PM-FM) transition temperature (T_c) and ferromagnet-to-spin-glass transition (T_G) in the so-called reentrant spin-glass systems (RSG) has been discussed recently.[1-5] Rakers and Beck[1] reported low-field ac-susceptibility measurements on a $Au_{82.5}Fe_{17.5}$ alloy with an external field perpendicular to their disc-shaped samples and also parallel to the specimen plane. In the first case, the demagnetization factor ($D_\perp$) is large and a "knee" is observed at a temperature of 140 K, a demagnetization-limited susceptibility ($\chi_e \simeq 1/D_\perp$) between 140 and 60 K, followed by a decrease in χ_e at lower temperatures. This has hitherto been taken as evidence for RSG behavior; the "knee" temperature is associated with PM-FM transition and the susceptibility change at the lower temperature with spin-glass transition. However, in the low-demagnetization factor parallel field geometry ($D_\parallel$ small), the susceptibility did not show a plateau or knee but a well-defined maximum at 100 K in the 9.3-Oe field which shifted to lower temperatures in higher fields. Magnetothermal history effects and remenance behavior typical of spin glasses (SG) were also observed up to 110 K. This observation together with the evidence of ^{57}Fe Mössbauer studies, which give hyperfine splitting only at temperatures (T_M) lower than the knee temperature (T_c) associated with ferromagnetic order, was taken to imply that there is no true long-range ferromagnetic state between T_c and T_G and there exists only a Langevin-type quasisuperparamagnetic behavior in this temperature range.

On the other hand, Boumazouza *et al.*[2] present evidence from low-field magnetization in parallel and perpendicular geometries, Mössbauer spectroscopy, and small-angle neutron-scattering (SANS) data on amorphous $(Fe_xCr_{1-x})_{75}P_{15}C_{10}$ alloys (x above and near the critical concentration $x_c \simeq 0.60$) to show that there is a true FM-PM transition in these systems followed by a SG behavior at low temperature. While agreeing with the observations of Rakers and Beck regarding the absence of a knee at the PM-FM transition temperature in the parallel geometry, they also find that SANS data show a sharp peak at the FM-PM transition temperature (T_c) and below T_c the transverse correlation length diverges but the longitudinal correlation length is finite as given by mean-field theories.[4]

The Mössbauer spectrum above T_c is a pure quadrupole doublet and the width of the doublet (as seen by a plot of FWHM of the components of the doublet) increases "sharply" below the same T_c observed in SANS and perpendicular geometry susceptibility data, which they attribute to the onset of hyperfine field. The same T_c observed in all measurements were taken to strongly suggest a true cooperative PM-FM phase transition.

However, as pointed out by Beck[3] in reply to the comment of Boumazouza *et al.*, the transition in Mössbauer spectrum is not really sharp but extends over a temperature range of more than 30% of the T_c in contrast to a sharp transition expected for a ferromagnet. Beck has suggested that this temperature variation can result from superparamagnetic fluctuations as calculated by Rancourt and Daniels.[5] Further, since there is no evidence for a magnetic specific-heat anomaly at T_c (Ref. 6), but the SANS data give a sharp peak at T_c, it has been suggested by Beck that there is some cooperative interaction between the fluctuating superparamagnetic moments but it is not sufficiently strong to change the paramagnet into a ferromagnet.

The polycrystalline alloy system $Cr_{1-x}Fe_x$ is another similar system on which we present our careful Mössbauer-effect studies to examine the nature of transition at T_c. The critical concentration for the onset of ferromagnetic order in this system is $x_c = 19.9$ at. % (Ref. 7) and we have chosen the composition $Cr_{74}Fe_{26}$ ($x > x_c$), which shows reentrant spin-glass behavior.[8] Similar to the two systems mentioned earlier, inelastic-neutron-scattering studies[8] show a well-defined peak at the ferromagnet-paramagnetic transition temperature ($T_c \simeq 180$ K) followed by a significant decrease in spin-wave stiffness at lower temperatures suggesting a spin-glass phase with no well-defined excitations below about 50 K.

Figure 1 shows the temperature dependence of Mössbauer spectra recorded in transmission geometry on a spark-eroded 40-μm foil of $Cr_{74}Fe_{26}$ alloy sample. Experimental details are mentioned elsewhere.[9] One can notice from the spectra that there are sidewings in the spectra indicative of a nonzero hyperfine field even up to 173 K besides paramagnetic intense absorption near zero velocity. The magnetic hyperfine field distributions were

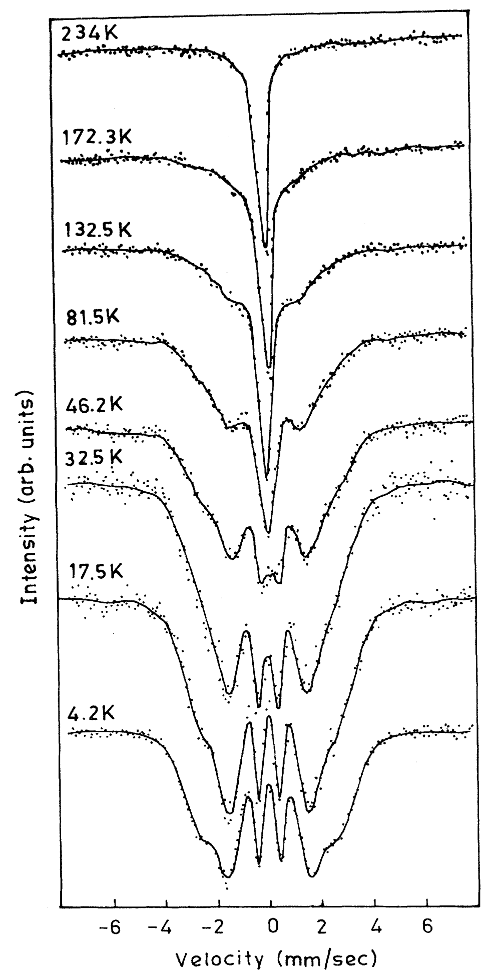

FIG. 1. ^{57}Fe Mössbauer spectra of the $Cr_{74}Fe_{26}$ alloy at different temperatures recorded in the transmission geometry. The solid lines through data are obtained by a model-independent field-distribution fit.

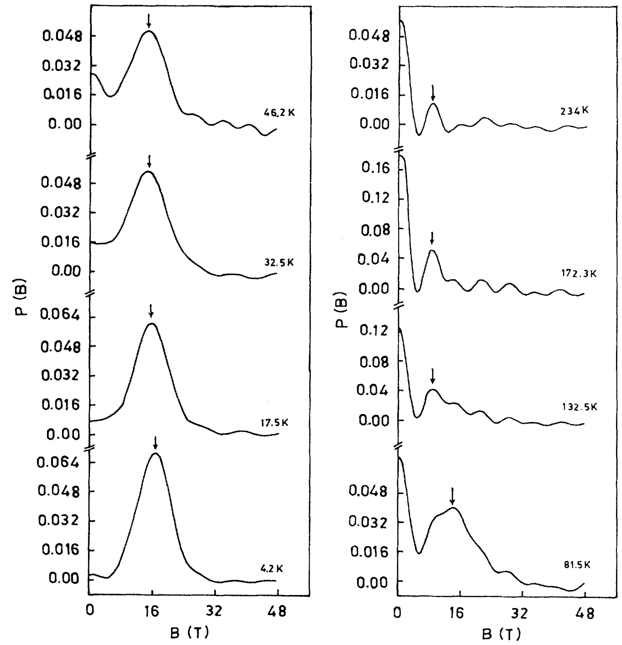

FIG. 2. The probability, $P(B)$, of the hyperfine field (B) obtained at different temperatures for the Mössbauer spectra of Fig. 1.

evaluated from the spectra using a model-independent field-distribution program.[10] A χ^2 minimization procedure[11] was adopted to get best fits to the data by varying the intensity ratios of the lines of the component spectra. The parameter b (ratio of the intensities of the second and fifth lines to the third and fourth lines) varied between 2.1 and 4 to yield optimum fits to data. The field distributions, $P(B)$, are shown in Fig. 2 as a function of temperature. At 4.2 K there is a well-defined peak in the

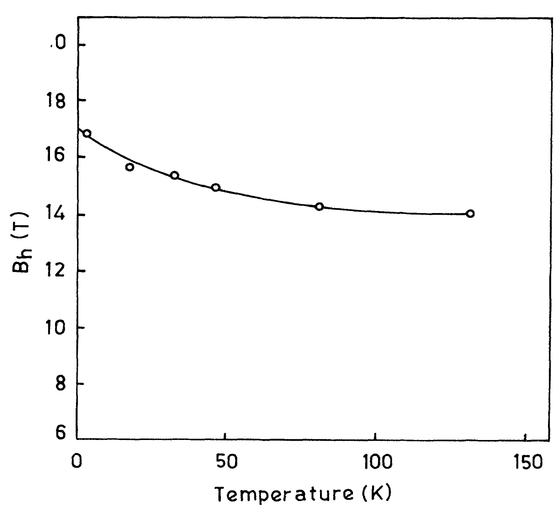

FIG. 3. The value of hyperfine field (B_h) corresponding to the high-field peak (indicated by arrows in Fig. 2) plotted as a function of temperature.

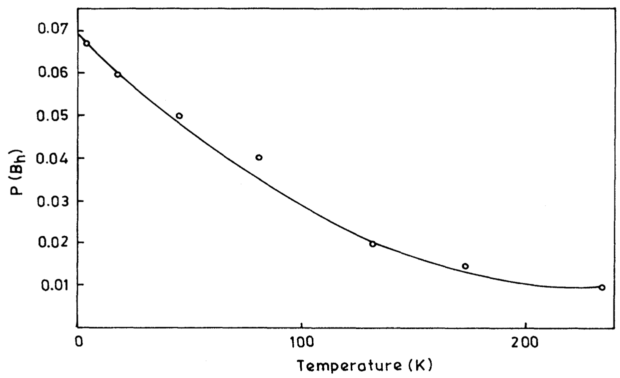

FIG. 4. The probability $P(B_h)$ for the high-field peak, B_h, of Fig. 3 as read from the field distributions of Fig. 2.

field distribution at 16.8 T together with a very small probability for low fields. As the temperature increases, the high-field peak shifts to slightly lower values but the field value does not go to zero even up to T_c, the ferromagnetic-to-paramagnetic transition temperature (Fig. 3). At higher temperatures ($\simeq 200$ K) there is uncertainty in the determination of peak hyperfine field value due to oscillations in $P(B)$. The probability for maximum field is also shown in Fig. 4 and it decreases upto 200 K before leveling off above this temperature. Figure 5 shows the probability for zero field as obtained from the field distributions. Up to 50 K the increase in the zero-field probability is slow and it increases rapidly thereafter. The Mössbauer spectrum at 4.2 K recorded in an external field of 5 T applied || to the plane of the foil is shown in Fig. 6 together with the field distribution. The shape of the distribution is not altered but it shifts to lower fields as expected for a negative sign of hyperfine field. The intensity ratio b defined earlier, which gives the best fit to the distribution, is $b=3.6$ and not $b=4$ which would be expected for a saturation of magnetization in the direction of the external field.

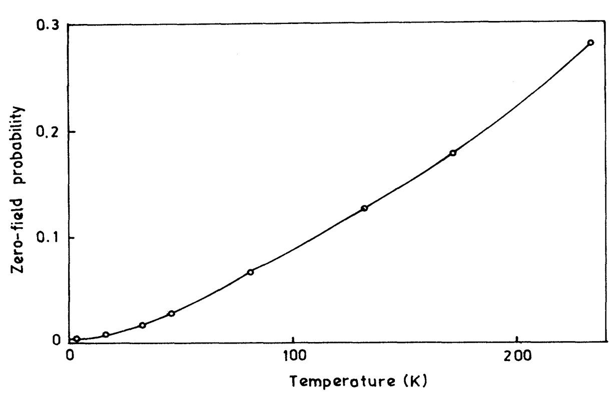

FIG. 5. The probability at $B=0$ (zero-field probability) as a function of temperature.

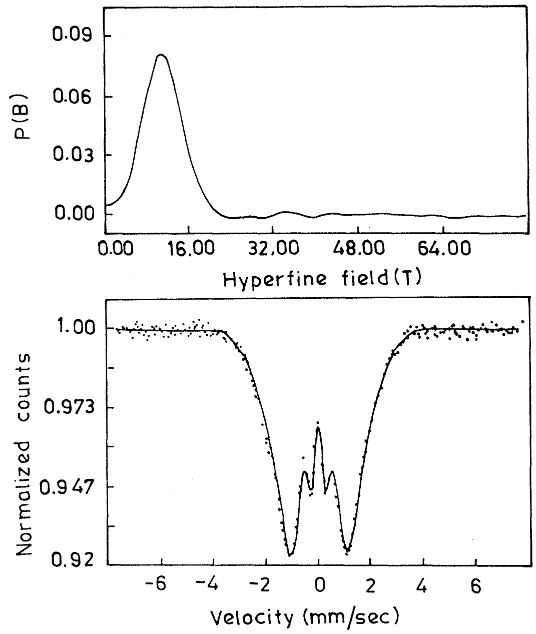

FIG. 6. The Mössbauer spectrum for $Cr_{74}Fe_{26}$ in an externally applied field of 5 T in a direction perpendicular to the direction of γ transmission, and the corresponding field distribution.

The observations presented above can be viewed within the framework of the model proposed by Burke and Rainford[12] and Kaul[13] to account for neutron scattering and other data in the concentration region close to and above the percolation threshold. The magnetic state of the system is assumed to be comprised of finite spin clusters embedded in an infinite percolation matrix. Similar to the observations of Boumazouza *et al.*, they[12] associate the peak in the SANS data at T_c with the onset of a

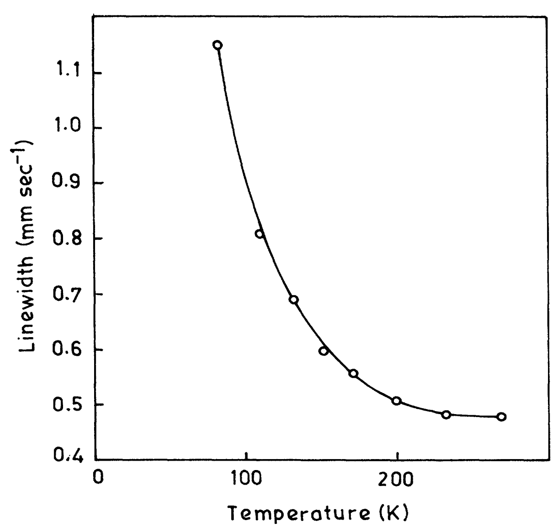

FIG. 7. The experimental linewidth of Mössbauer spectra plotted as a function of temperature.

paramagnet-ferromagnet transition of the infinite cluster and the quasielastic peak below 50 K with the spin-glass phase due to freezing of finite spin clusters. Our Mössbauer spectra, however, do not indicate any abrupt increase of the hyperfine field value below T_c (Fig. 3) but only an increase of the high-field probability (Fig. 4). To compare with the analysis of Boumazouza *et al.*, we have plotted in Fig. 7 the FWHM read directly from the Mössbauer spectra as a function of temperature without taking into account the detailed distribution of fields. This behavior tends to support the viewpoint adopted by Beck[3] that the transition is not sharp but extends over an appreciable temperature range. Another important aspect to be noted is that there exists appreciable zero-field probability down to 50 K (Fig. 5) indicating that the paramagnetic contributions dominate appreciably in the temperature range between T_c and T_G. This further lends support to the hypothesis of Beck that a Langevin-type superparamagnetic behavior predominantly exists in this temperature range.

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