Magnetic Double Transition in Au-Fe near the Percolation Threshold

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The ^{57}Fe Mössbauer effect on 16.8-at.% Fe-Au displays the magnetic double transition from paramagnetism to ferromagnetism at T_c = 165 and from ferromagnetism to spin-glass-like behavior at $T_f \sim 45$ K. Below T_f the magnetic state is characterized by freezing of Fe spin components which are transverse with respect to a strong applied dc field.

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One of the most interesting phenomena of *Au*Fe alloys is the suggested magnetic double transition for alloys near the percolation concentration (~16 at.% Fe): a transition from paramagnetism to ferromagnetism at the Curie temperature T_c followed by a transition at some lower temperature T_f which has been interpreted as a disappearance of ferromagnetism and the appearance of a spin-glass-like state.[1-4] This suggestion is based principally on the anomalous temperature behavior of the low-field ac and dc susceptibilities. However, magnetic measurements in these alloys are difficult to interpret.[4] Thus the magnetic double transition in Au-Fe alloys has been questioned recently.[5] Moreover it has been found in a neutron diffraction study[6] that ferromagnetism persists even down to the lowest temperatures.

In this Letter we report ^{57}Fe Mössbauer-effect results on *Au*Fe which give new information on Fe spin correlations and hyperfine interactions in this alloy. Our 16.8-at.% Fe-Au samples were foils of ~ 6 μm thickness, solution-annealed at 820 °C in vacuum for 49 h, and rapidly quenched in water.

The transition from paramagnetism to ferromagnetism on decrease of temperature is indicated by a sudden spectral broadening near T_c due to the development of magnetic hyperfine splitting (Fig. 1). The paramagnetic spectra obtained above 165 K are in agreement with earlier Mössbauer investigations.[7,8] Taking the onset of spectral line broadening for the transition yields T_c = 165 ± 5 K (see inset in Fig. 1). At 77 K our alloy shows typical *ferromagnetic* Fe spin alignment in an applied (longitudinal) field H_{ext} of 20 kOe. The corresponding spectrum (Fig. 2, top) indicates that essentially all of the Fe spins at 77 K were aligned parallel to H_{ext}, since the absorption lines No. 2 and No. 5 (i.e., the $\Delta m = 0$ nuclear transitions) disappear as compared to the zero-field spectrum at 77 K (Fig. 1).

The spectra shown in Fig. 2 were obtained by field cooling the sample from 77 to 4.2 K in a (longitudinal) field of H_{ext} = 20 kOe. The striking effect is the reappearance of the $\Delta m = 0$ lines at ~ 45 K and the gradual increase in their relative intensity as the temperature is lowered to 4.2 K. This means that the Fe spins (starting at ~ 45 K) gradually rotate away from the applied field direction as the temperature decreases, the ensemble of Fe spins now forming an average angle $\theta \neq 0$ relative to the applied field. Thus below 45 K the magnetic state is characterized by a freezing of Fe spin components which are *transverse* with respect to the applied field and which increase in magnitude if the temperature is lowered. When

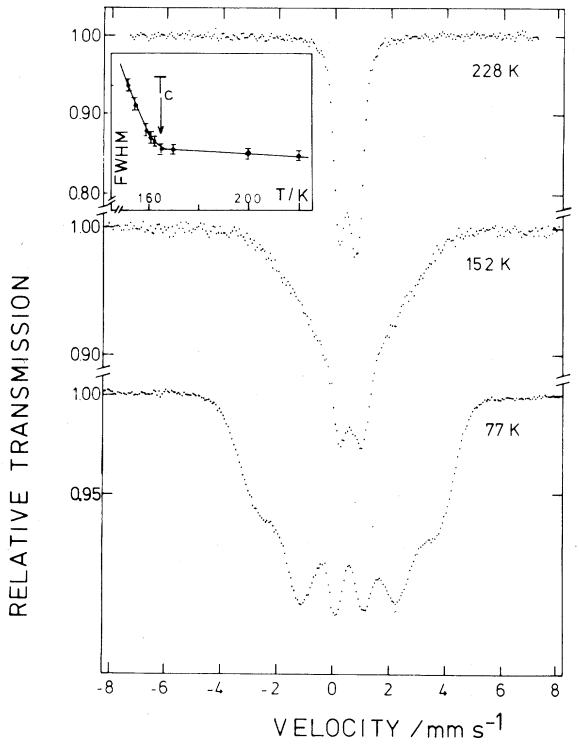

FIG. 1. Mössbauer spectra at 228, 152, and 77 K, $H_{ext} = 0$.

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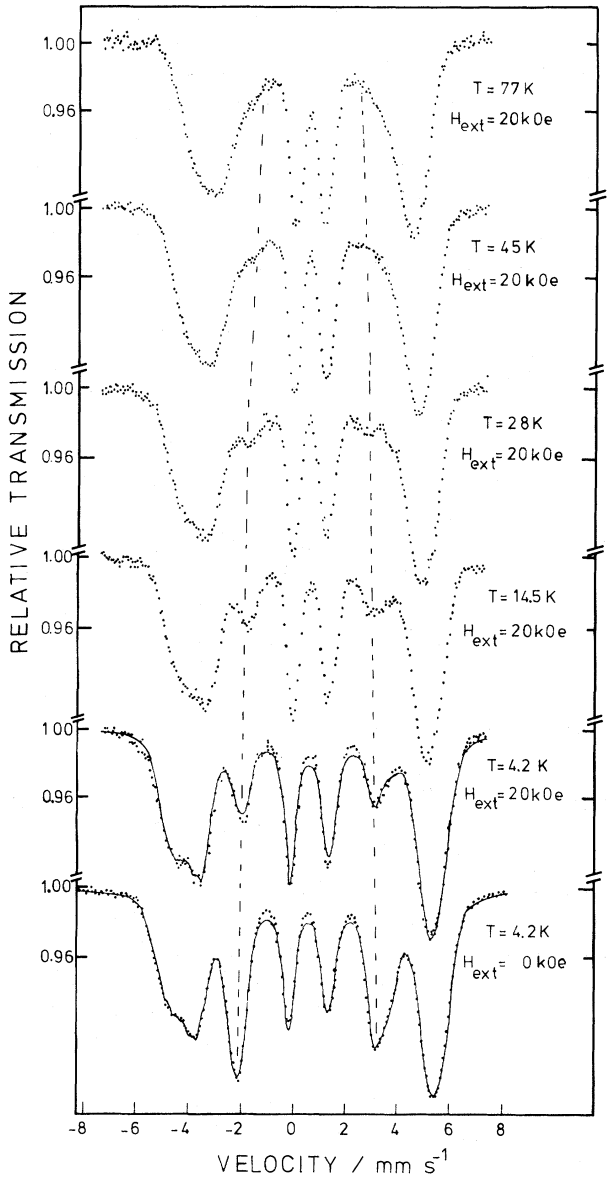

FIG. 2. Mössbauer spectra at $H_{ext} = 20$ kOe and at various temperatures; bottom: at 4.2 K, $H_{ext} = 0$. Solid curves: Fitted curves corresponding to $P(H)$ distributions of Fig. 4.

the field is decreased to zero at 4.2 K after field cooling the sample to 4.2 K, the spectrum taken is as shown in Fig. 2 (bottom). The strong $\Delta m = 0$ line intensities indicate that the Fe spins were randomly reoriented after H_{ext} had been removed.

The onset of a gradual spin rotation below ~ 45 K is more clearly demonstrated in Fig. 3 (bottom) where we have plotted the peak intensity ratio $[N_2(77\text{ K}) - N_2(T)]/N_3(T)$. $[N_2(77\text{ K})$ and $N_2(T)$ are the normalized numbers of counts for the absorption peak of line No. 2 at 77 K and at temperature

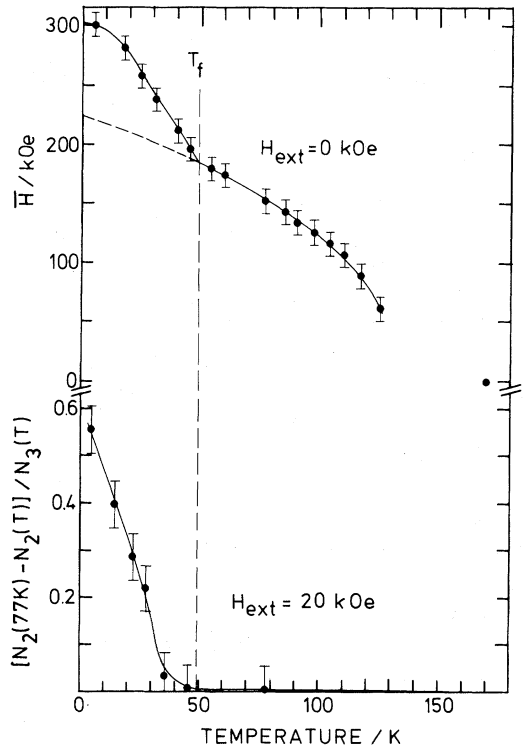

FIG. 3. Top: average hyperfine field $\bar{H}(T)$; bottom: peak intensity ratio vs temperature (see text).

T, respectively; $N_3(T)$ is the normalized number of counts for peak No. 3]. The sharp rise in this intensity ratio below $T_f \sim 45$ K is a clear manifestation of the freezing process below T_f where Fe spins are canted relative to H_{ext}.

The magnetic transition at $T_f \sim 45$ K has also been observed as an anomaly in the temperature behavior of the average hyperfine field, $\bar{H}$, obtained from Mössbauer spectra in zero applied field (Fig. 3, top). $\bar{H}(T)$ has been determined from the distribution $P(H)$ of hyperfine fields in these spectra (see below). The hyperfine field is usually proportional to $\langle S \rangle$ for a local Fe spin S. This anomaly either is related to a difference in the temperature behavior of the order parameter for the two phases, or it means that $\langle S^2 \rangle$ is not equal to $\langle S \rangle^2$. $\langle S^2 \rangle$ is the (spin-glass) Edwards-Anderson order parameter; in the ferromagnetic state the order parameter is $\langle S \rangle$. Extrapolation to $T = 0$ of the ferromagnetic part of the $\bar{H}(T)$ curve shows that the average saturation hyperfine field of the ferromagnetic state is roughly 75 kOe smaller than that of the spin-glass state. The reason might be either a different average Fe spin or a change in the conduction-electron spin polarization for both magnetic states.

Our spectra at 4.2 K have been evaluated using a histogram type of hyperfine-field distribution analysis. For this purpose nine six-line subspectra with Lorentzian lines of equal width (15.8 kOe full width at half maximum) at equally spaced values of the magnetic splitting were least-squares fitted to the experimental spectra. A hyperfine-field analysis is complicated by the fact that unknown distributions of quadrupole interactions and of isomer shifts exist[8] which are correlated with different components of the hyperfine-field distribution. In order to analyze our measured spectra the Fourier series method[9] of Window was also successfully used for $T \gtrsim 20$ K. However, this method failed at 4.2 K where the spectra are better resolved and more asymmetric. The obtained distributions $P(H)$ (Fig. 4) at 4.2 K are rather broad and clearly show a main (low-field) peak (at ~ 270 kOe for $H_{ext}=0$) and a (high-field) shoulder (at ~ 325 kOe for $H_{ext}=0$). We attribute the high-field part in the $P(H)$ curve to Fe-rich clusters, while the low-field part in $P(H)$ is associated with Au-rich clusters where an Fe atom has fewer Fe atomic neighbors. This interpretation is based mainly on the fact that the high-field shoulder in the $P(H)$ curve increases relative to the low-field peak with increasing Fe concentration.[10] The width and shape of the distribution $P(H)$ is *not* very much affected by the applied field, the main effect being a rigid shift of $P(H)$ towards lower effective fields since for ^{57}Fe the internal field is opposite to the Fe moment. Our analysis suggests that the alignment in a given external field is similar for all Fe spins irrespective of their local neighbor configuration. This can be concluded from the correlation between subspectra hyperfine field and line-intensity ratio I_2/I_1 (line No. 2 to line No. 1) (Fig. 4, bottom). For $H_{ext}=0$ the ratio I_2/I_1 has a constant value of 0.66 implying random Fe spin orientation for all hyperfine fields (and for all the corresponding Fe neighbor configurations). For $H_{ext}=20$ and 60 kOe the values for I_2/I_1 are 0.4 and 0.2, respectively, both ratios being independent of the subspectra hyperfine field (and neighbor configuration) as well. Since the relation between the line-intensity ratio I_2/I_1 and the angle θ is given by[11] $\cos^2\theta = (4-3I_2/I_1)/(4+3I_2I_1)$ we are able to calculate an average angle of $\theta = 43°$ and 31° for $H_{ext}=20$ and 60 kOe, respectively (assuming that the spin distribution is almost all "forward"). The magnitude of the shift of the distribution $P(H)$ for $H_{ext}\neq 0$ is consistent with these angles. For example, by using simple vector addition of H_{ext} and H ($H<0$), we would expect for $H_{ext}=60$ kOe (and $\theta=31°$) a value of ~ 221 kOe for the low-field peak and ~ 275 kOe for the high-field shoulder, which is in reasonable agreement with the corresponding values in Fig. 4 (~ 230 and ~ 280 kOe, respectively). Therefore our results confirm quantitatively conclusions drawn from earlier Mössbauer studies on *Au*Fe alloys[12]: Complete alignment ($\theta=0$) of a substantial fraction of Fe moments with the remaining fraction preserving its initial random orientation can be excluded. Thus we do *not* observe the coexistence of spatially segregated ferromagnetic and spin-glass regions in this alloy, but instead the local magnetic behavior is homogeneous.

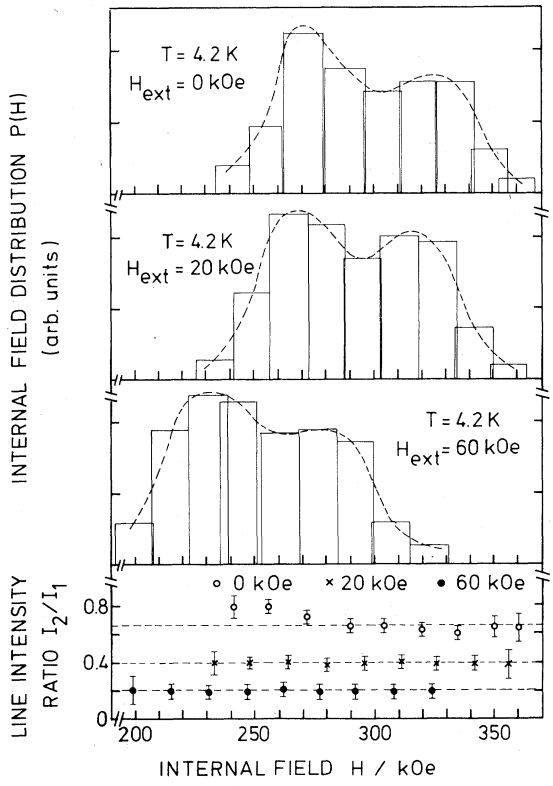

FIG. 4. Hyperfine-field distributions $P(H)$ at 4.2 K and $H_{ext}=0$, 20, and 60 kOe (from top). (The dashed curves are an aid for the eye). Bottom: line-intensity ratio I_2/I_1 for $H_{ext}=0$, 20, and 60 kOe.

It is interesting to notice that more dilute *Au*Fe spin-glasses ($c \lesssim 9$ at.% Fe) at 4.2 K do not show remarkable polarization effects in applied fields.[10,12] The different behavior found for our concentrated alloy suggests that (at least short-range) ferromagnetic spin correlation is maintained in the spin-glass-like state of 16.8 at.% Fe-Au, since

in this case a larger degree of alignment towards H_{ext} can be achieved by the interaction of (larger) Fe cluster moments and H_{ext}.

Our results are in qualitative agreement with a recent theory[13] which predicts the coexistence of spin-glass and ferromagnetic ordering. This "mixed phase" is characterized by the coexistence of a spontaneous (longitudinal) magnetization (ferromagnetic order) and a spin-glass ordering of the transverse components of the spins. Our observations in an applied field suggest this behavior, i.e., ferromagnetic spin correlation for $T > T_f$, and preferentially ferromagnetic spin correlation combined with the development of a transverse spin component due to long-range antiferromagnetic interaction for $T < T_f$.

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Pinning of a Vortex Line to a Small Defect in Superconductors

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It is shown that quasiparticle scattering by a defect in a superconductor leads to large elementary pinning energies of flux lines. In the case of a small radius (R) point defect the new mechanism outweighs the traditional volume effect by the factor ξ_0/R.

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It is a long-standing problem in the context of critical current densities of type-II superconductors that the measured volume pinning forces turn out larger than predicted by theory.[1] Type-II superconductors are resistanceless only as long as the magnetic-flux vortex lattice which penetrates them remains stationary. Current tries to push the vortex lattice into motion and the critical current is determined by the pinning force which holds the lattice back. This force is a volume effect arising from a large number of elementary contributions from individual pinning centers. The vortex lattice deforms elastically to accommodate itself to the presence of pinning centers. The problem of determining the energy barriers between different equilibrium vortex configurations for a random array of pinning centers is called the statistical summation problem. This statistical theory needs the elementary pinning potentials and the elastic properties of vortices

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