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Spin canting and ferromagnetism in a *Au*Fe alloy: Mössbauer and magnetic measurements

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We have made Mössbauer polarization and susceptibility measurements above technical saturation on Au–19 at.% Fe. Just below the magnetic ordering temperature the system behaves as a ferromagnet, but random local canting sets in at lower temperatures. A well-defined canting transition temperature cannot be established from the data, but canting begins above the temperature at which the low-field susceptibility shows a sharp falloff.

Among the open questions in the domain of spin-glasses is that of the transition of spin-glass to a ferromagnet which occurs in a number of systems when the concentration is varied, and that of the behavior of samples just on the ferromagnetic side of the critical concentration. One of the most intensively studied alloy series of this type is *Au*Fe. Principally from susceptibility measurements[1–4] the following pattern of behavior has been established: At low Fe concentration the alloys are spin-glasses, but beyond about 14 at.% Fe (the exact value is sensitive to annealing procedures[5]) the alloys acquire a nonzero spontaneous magnetic moment and a well-defined Curie temperature T_C. The initial susceptibility is high from T_C down to a less well-defined temperature T_m where it tends to drop off.[1–4] T_m has been identified with a "ferromagnetic–to–spin-glass transition."[2,4] For this type of system, a Heisenberg mean-field model has been proposed[6]; according to the model, at a given concentration above the critical concentration one should observe the successive transitions: para → ferro at T_C; ferro → mixed I at T'; mixed I → mixed II at T''.

Roughly, T' is a canting transition and T'' is a "replica symmetry-breaking" transition. Further work on the same model[15] showed that at the transverse ordering point T' weak irreversability sets in, while the T'' behavior represents only a gradual crossover to stronger irreversability.

We have made Mössbauer polarization measurements on a Au–19 at.% Fe sample in order to observe directly the degree of alignment of the Fe local spins as a function of temperature. We find that at temperatures below but close to T_C alignment is nearly ferromagnetic but at lower temperatures disalignment (canting) sets in and steadily increases. Susceptibility measurements made above technical saturation showed an extra low-temperature term associated with the canted state.

The Mössbauer measurements were performed on a 9-μm-thick foil sample containing enriched ^{57}Fe. The sample was annealed at 800 °C, quenched, and kept in liquid nitrogen except during experiments. The ferromagnetic ordering as monitored by the thermal scanning technique (constant velocity) occurred progressively in the range 175–165 K in good agreement with literature values at this concentration[4] (before heat treatment the temperature range for magnetic ordering was 175–150 K).

Full spectra were taken at different temperatures either in zero applied field or in a 0.6-T field applied parallel to the γ-ray direction. This field is enough to technically saturate the sample at all temperatures below T_C. Field-off spectra at 4.2 K (magnetically ordered) and at 300 K (paramagnetic) closely resemble equivalent spectra taken by Window[7] at a similar concentration and show rather broad lines with distributions of hyperfine field, quadrupole effect, and isomer shift. We will only discuss the polarization effects here. As is well known, for a pure magnetic spectrum the relative line intensities depend on the angle θ between the local hyperfine field and the γ-ray direction.

Such a situation is roughly that of *Au*Fe, since below ~150 K the quadrupole interaction is much weaker than the magnetic one. We can also assume that the local Fe hyperfine field is parallel to the local moment at each site. Therefore Mössbauer line intensity measurements in the technically polarized state give relevant information on the local canting angle θ: Actually the relative line intensities $3{:}x{:}1{:}1{:}x{:}3$ lead to $\langle \sin^2\theta \rangle = 2x/(4+x)$. The limits

for $\langle \sin^2\theta \rangle$ are 0 for a true ferromagnet and $\frac{2}{3}$ for a true spin-glass.

A set of typical field-on spectra is shown on Fig. 1. It is seen from the intensity of lines 2 and 5 (arrows) that spin alignment is roughly achieved above 70 K, while at lower temperatures spin canting occurs. Because of the overlap of the Mössbauer lines, accurate analysis of the line intensities ($3{:}x{:}1{:}1{:}x{:}3$) requires unusual methods such as a linear combination of "standard" Mössbauer spectra (i.e., of spectra whose line intensities are known[8]). By carefully comparing the field-off spectra recorded at 77 K either at normal incidence to the γ beam or at the magic angle[9] we ensured that the texture effect[10,11] was negligible. Consequently, the field-off spectra were used as standard spectra of line intensities 3:2:1:1:2:3. No other standard spectra could be obtained because full spin alignment was never completely achieved. We thus analyzed the spectra by matching the outer lines of the field-off and field-on spectra; typical examples are shown in Fig. 2. We measured the ratio of the absorption area of the field-off–field-on spectra: $R = 12/(8+x)$. Then $\langle \sin^2\theta \rangle$ was easily determined at each temperature. The statistical error on $\langle \sin^2\theta \rangle$, obtained by varying the limits of the fitted part of the spectrum was 0.02 in most cases. A detailed consideration of instrumental effects (such as thickness effects) and intrinsic difficulties [the tail of $P(H_{\text{eff}})$ towards high fields] shows that the measured $\langle \sin^2\theta \rangle$ might be slightly overestimated.

Figure 3 shows the behavior of $\langle \sin^2\theta \rangle$. Just below T_C the system is close to being fully ferromagnetic; $\langle \sin^2\theta \rangle$ is not quite zero, which may be an artifact due to the baseline error we have mentioned or, more probably, a real effect. Below about 70 K there is a change of regime with the degree of canting increasing sharply. The system is becoming progressively less ferromagnetic as the temperature drops (even thought the average moment in the z direction continues to increase[11]). The residual deviations from true ferromagnetic alignment in the higher-temperature region can be ascribed to random crystal-field effects; the Fe local moments are not pure S-like in this alloy[12] and Window[7] has already pointed out evidence for local easy-magnetization axes. However, the significant observation is that the degree of canting begins to increase steeply below ~70 K and increases continuously as T drops to zero.

This behavior is much as would be expected below

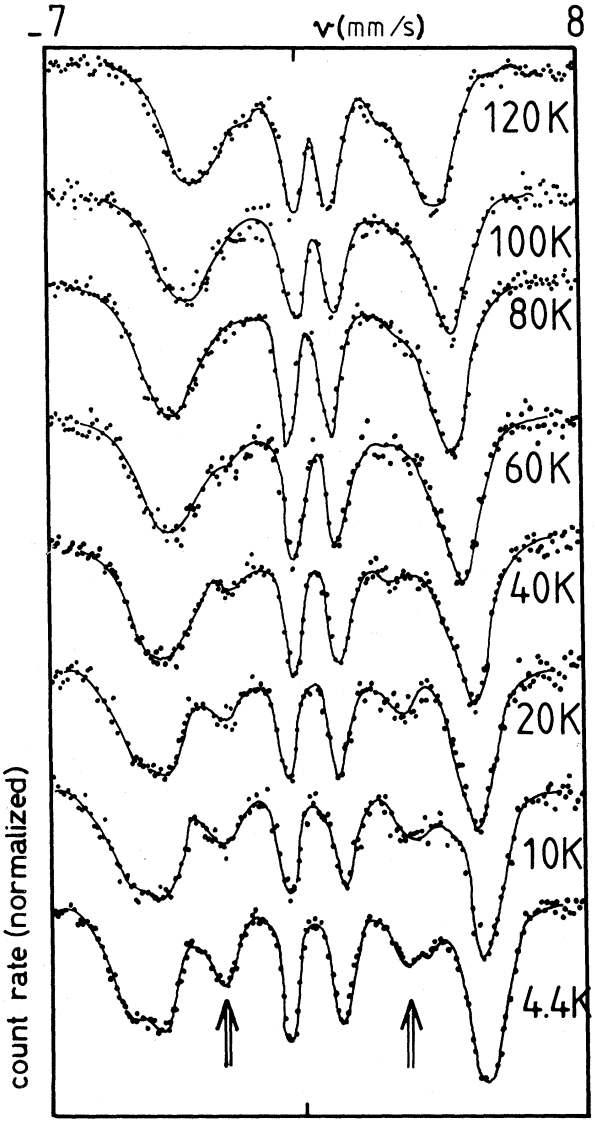

FIG. 1. Mösbauer spectra of Au–19 at.% Fe in a magnetic field of 0.6 T parallel to the γ beam (arrows indicate lines 2 and 5, sensitive to spin canting).

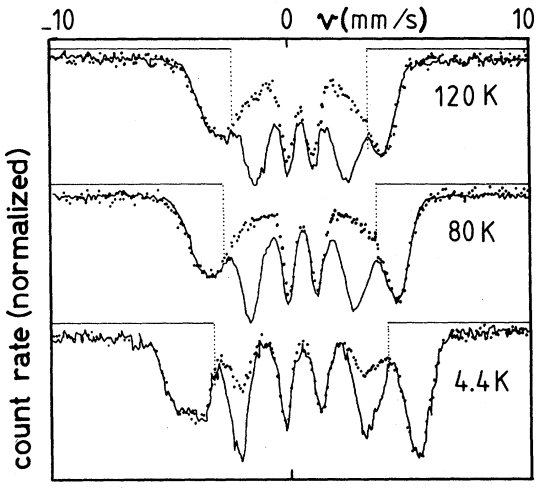

FIG. 2. Linear fitting of experimental spectra; the "standard" spectrum is shown as a full line. The horizontal bars indicate the channels involved in the fit.

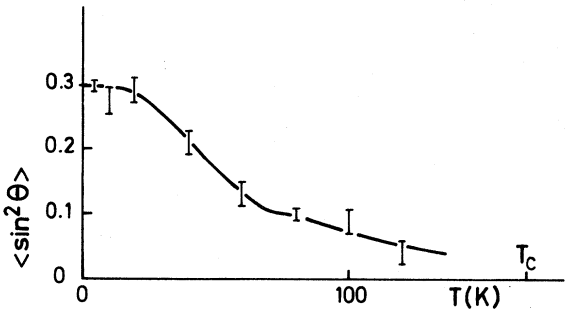

FIG. 3. Measured $\langle \sin^2\theta \rangle$, where θ is the local canting angle from the Mössbauer analysis.

the ferromagnetic to mixed-I transition in the Heisenberg mean-field model[6] except that the present results cannot be taken as showing a sharply defined canting transition temperature.

We have also used a Faraday balance to make measurements of dM/dH on an alloy of the same nominal concentration in applied fields of 0.5 to 1.0 T, i.e., in the region above technical saturation. The results are shown in Fig. 4. In agreement with conclusions drawn from magnetoresistance measurements,[13] this single-domain susceptibility has a maximum at T_C as in all ferromagnetics, but then goes through a minimum before reaching a flat plateau at low temperature. In a conventional local moment ferromagnet, dM/dH above technical saturation would drop continuously to zero at $T=0$. When we compare the data with the Mössbauer canting-angle results, it can be seen that the "extra" low-temperature susceptibility is associated with the increase in canting; we can put it down to the magnetic

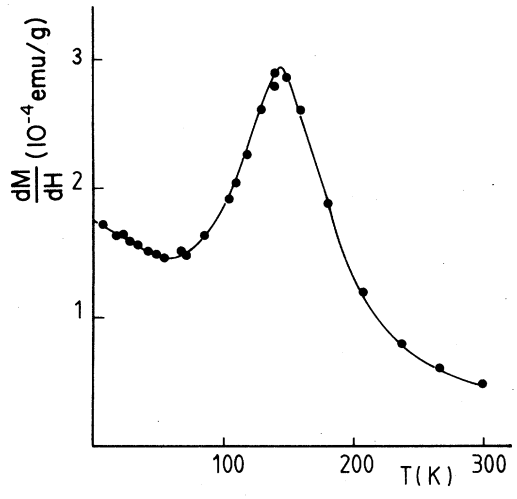

FIG. 4. The susceptibility dM/dH measured above technical saturation for Au–19 at.% Fe. dM/dH was measured between 0.5 and 1.0 T at each temperature.

field tilting the perpendicular moment components towards the z direction (as in the perpendicular susceptibility of an antiferromagnet).[13]

We can discuss briefly certain implications of these results. The fact that the alignment between T_C and the "canting transition" is close to being purely ferromagnetic does not appear to be compatible with a simple percolation description[4] where a distinction is made between spins participating in the infinite ferromagnetic cluster and the other spins which are randomly aligned. The Mössbauer polarization shows that near T_C almost complete ferromagnetic alignment is achieved for all spins so the percolation image seems inappropriate for AuFe.

There is a breakdown in ferromagnetic behavior at low temperatures in this type of alloy as is shown by an upturn in the neutron scattering intensity at fixed Q in the data of Murani *et al.* on AuFe[1] or as a similar effect accompanied by a collapse of the effective spin-wave stiffness at low temperature in CrFe.[14] The upturn in the results of Murani *et al.* for Au–19 at.% Fe occurs at about the same temperature as the rapid increase in canting angle observed here. This suggests that both this upturn, which is probably dominated by quasielastic moment direction fluctuations scattering, and the CrFe spin-wave collapse are due to increase in canting. It seems physically reasonable that spin waves no longer have well-defined wave vectors when there is a strong degree of random local canting.

The exact temperature T_m characteristic of the falloff in the low-field initial susceptibility is difficult to define experimentally because of demagnetizing factors, etc.[2,4,5] It seems that for Au–19 at.% Fe this falloff happens at a temperature ~45 K (Refs. 2 and 4) or lower[3] whereas a fair degree of local canting has already set in at higher temperatures. Even though both "transitions" are rather blurred experimentally, the onset of canting and the falloff in susceptibility seem to happen at distinctly different temperatures. One can speculate that T_m can be identified with the "lower-T" crossover in the vector mean-field model,[6,15] though this does not appear to be clear for the moment.

We can summarize the conclusions that we draw from the Mössbauer polarization, high-field dM/dH magnetoresistance[13] and neutron scattering[1] data on Au–19 at.% Fe. The Mössbauer experiments show that canting is weak from T_C down to about 70 K, and then develops progressively as T is lowered. The other experiments all show departures from conventional ferromagnetic behavior in the same temperature range. Above about 70 K the system behaves essentially as a ferromagnet; below, as a result of canting, there is an extra tilting susceptibility term in dM/dH and a gradual breakdown of the ferromagnetic structure. The vector mean-field model of Gabay and Toulouse[6] predicts an increase in canting as the

temperature is lowered; however, the model suggests a sharply defined canting transition temperature T' with pure ferromagnetic behavior between T_C and T', while experiment shows a more gradual rise in canting (though experimental error bars are such that a sharp change of regime somewhere near 70 K cannot be ruled out). This may either be because in the real system there are extrinsic effects not included in the model, such as random crystal fields, or it may be due to the fact that the model is a mean-field approximation.

After this paper was submitted, a very similar Mössbauer experiment on a Au–16.8 at.% Fe alloy has been reported.[16] General conclusions are similar to ours though the data are interpreted as showing a sharp canting transition at ~45 K, and pure ferromagnetic behavior above this temperature. The question of whether a sharp transition exists or not has obvious implications for the problem of spin-glasses in an applied field.[6,17]

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