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Competing Ferromagnetic and Spin-Glass Order in a *Au*Fe Alloy

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Magnetic and Mössbauer measurements on a *Au*–19-at.%-Fe alloy are reported. After ferromagnetic ordering at $T_c \cong 170$ K, spin-glass-like transverse ordering sets in below $T \cong 60$ K; strong irreversibility in the magnetization process only occurs below $T \cong 15$ K. This behavior is consistent with the predictions of a Heisenberg mean-field model, in which the transverse ordering transition is distinct from a crossover to strong irreversibility.

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A number of magnetic systems are spin-glasses over a certain concentration range but acquire a degree of ferromagnetic order when the concentration of the magnetic component is increased. For *Au*Fe and other systems,[1-7] phase diagrams have been proposed which show, for samples just above the critical concentration, a transition from ferromagnetic to "reentrant spin-glass" at a temperature below T_c. On the other hand, Heisenberg mean-field model calculations[8-12] suggest that there should be two distinct changes of behavior below T_c. We report magnetic and Mössbauer measurements on *Au*–19-at.%-Fe samples; the results show two transitions of quite different types having the essential qualitative features predicted by the model. This has important implications for the phase diagrams of other systems of this type, and by similarity[9] for the behavior of true spin-glasses in an applied field.

*Au*Fe alloys below about 15 at.% Fe are spin-glasses; above this concentration the alloys have a spontaneous magnetization at low temperatures. For alloys on the ferromagnetic side of the critical concentration the ac susceptibility falls off from the ferromagnetic limiting value at some temperature well below T_c; this falloff has been taken to indicate the onset of a "spin-glass-like" ordering.[5-7] However, the low-temperature spin-glass-like state is not a conventional spin-glass with randomly oriented local moments, because the zero-field magnetic-disorder resistivity decreases regularly as T decreases to $T = 0$.[13-15]

We have made magnetic and Mössbauer measurements on samples of nominal concentration *Au* + 19 at.% Fe. Magnetization measurements were made using a Faraday balance up to 9 kG, and hysteresis loops were taken at fixed temperatures with a home-built vibrating-sample magnetometer; the field was swept from +5 to −5 kG in about 5 min and the magnetization was continuously recorded. In both cases the sample was needle shaped, with a small demagnetization factor. We estimate the demagnetization field to be about 5 G at the lowest temperatures.

Down to about 10 K, identical hysteresis loops were obtained when the sample was in the zero-field-cooled state or when it had been cooled under 25 kG applied field. At lower temperatures the value of the cooling field began to influence the shape of the hysteresis loop, so that the loops became magnetic-history dependent. The results in the present paper refer to the zero-field-cooled state only, and the magnetic history effects will be discussed elsewhere.

The magnetization measurements at each temperature can be characterized by a spontaneous magnetization and a coercive field H_c. The spontaneous magnetization per Fe atom $\langle m_z(T)\rangle$ was evaluated by extrapolating $m(H,T)$ back to $H = 0$, using linear extrapolation at low temperatures and an Arrott plot procedure for the temperatures near T_c. The magnetization evolves with

decreasing temperature in much the same way as in a standard ferromagnet, increasing only very little below $T_c/2$ (see also Ref. 16). However, the zero-temperature value of the spontaneous magnetization $\langle m_z(0)\rangle$ is only $1.8\mu_B$ per Fe atom while the true saturation magnetization m_{sat} obtained by aligning all the Fe moments in a very strong applied field is about $2.6\mu_B$ per Fe atom at this concentration as found by Rakoto and Ousset[17] who carried out magnetic measurements up to 320 kG.

Hysteresis loops and the value of the coercive field H_c as a function of temperature are shown in Fig. 1. The results show a low value of H_c until around 15 K; then H_c increases dramatically before saturating around 4 K. We are observing the onset of strong irreversible behavior in the magnetization process of the alloy from 15 K.[18] It is well known that in standard ferromagnets high coercive fields are a sign of domain-wall blocking; this may well be true here also, but with an intrinsic blocking mechanism related to local frustration effects.

Mössbauer experiments at the same *Au*Fe concentration have been described in an earlier article,[19] where we focused attention on the polarization of the Mössbauer spectrum in an applied field sufficient to produce technical saturation.

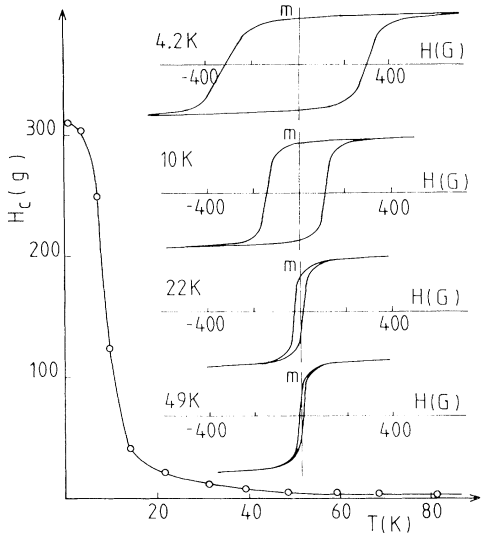

FIG. 1. Insets: Hysteresis loops $M(H)$ on a *Au*–19-at.%-Fe needle-shaped sample at fixed temperatures. The spontaneous magnetization extrapolated back to $H=0$ from higher fields corresponds to about $1.8\mu_B$ per Fe atom in the low-temperature range. Main figure: The coercive field H_c as a function of temperature. T_c for this sample is about 170 K.

From zero-applied-field spectra taken in the same series of experiments we can estimate the average Fe hyperfine field. If $I(v)$ is the Mössbauer signal intensity as a function of the velocity v and v_0 is the center of the spectrum, then we evaluate as follows:

$$R = \int_{-\infty}^{\infty} |v - v_0| I(v)\, dv \Big/ \int_{-\infty}^{\infty} I(v)\, dv.$$

The zero-applied-field spectra showed no texture effects[19] and the spectra though broadened are dominated by the magnetic hyperfine interaction. Under these conditions, if we have a distribution of hyperfine fields together with weaker distributed quadrupole and isomer shift effects (correlated or not with the hyperfine field), then the ratio R should be proportional to the average hyperfine field to quite high precision.

The average hyperfine field $\langle H_{hf}\rangle$, Fig. 2, comes to an initial plateau in parallel with $\langle m_z(T)\rangle$ as the temperature is lowered, but then increases sharply again below about 60 K. This is just the temperature below which the polarization analysis[19] indicated a rapid increase in canting angle.[20]

The magnetization, polarization, and hyperfine-field data taken together give a consistent picture of behavior as the temperature is lowered: (1) Above 60 K, residual local canting of the Fe moments is low, so that $\langle m_z(T)\rangle$ and $\langle H_{hf}\rangle$ level off together with each $H_{hf}{}^i$ parallel to z. (2) Below 60 K, $\langle m_z(T)\rangle$ remains unchanged but static moment components $m_x{}^i$, $m_y{}^i$ appear accompanied by hyperfine-field components. The moduli of the local moment $|\vec{m}_i|$ and of the local hyperfine field $|\vec{H}_{hf}{}^i|$ increase and their direction is no longer parallel to z. The ratio of the $T=0$ hyper-

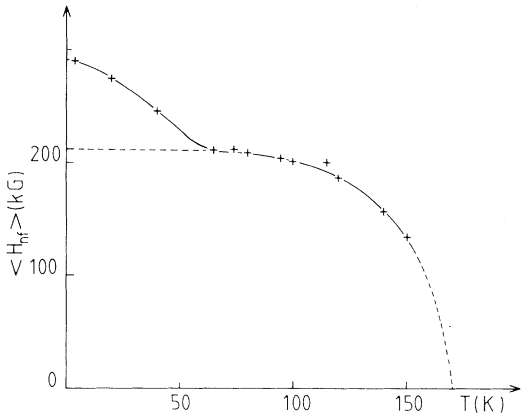

FIG. 2. Average hyperfine field as a function of temperature.

fine field to the plateau value is approximately equal to the ratio of the true Fe local moment (from the high-field data[17]) to the technical saturation moment $\langle m_z(0)\rangle$ as one would expect.

This is a canting transition, but of a very unusual type. Generally at a canting transition (see, e.g., Kouvel and Forsyth[21]) $|\vec{m}|$ is conserved but $\langle m_z\rangle$ decreases, whereas here a transverse spin-glass-like order sets in while the ferromagnetic order parameter is unaffected, so that $\langle m_z\rangle$ is conserved but $\langle|\vec{m}|\rangle$ increases.

We can now compare all these results with the Heisenberg mean-field model predictions.[8-12] According to the model, one should observe a series of transitions: First at T_c the system passes from paramagnetic to ferromagnetic. Then at a lower temperature T_{GT} (the Gabay-Toulouse transition) each spin should begin to acquire static transverse-moment components, without the ferromagnetic order parameter changing. At a yet lower temperature T_{AT} (the de Almeida-Thouless transition) a crossover from weakly irreversible to strongly irreversible behavior should occur.

For our alloy, $T_c \cong 170$ K. The canting with conservation of $\langle m_z\rangle$ that we have observed at $T \cong 60$ K can clearly be identified with the T_{GT} transition. The sharp onset of strongly irreversible behavior that we observe at $T \cong 15$ K indicates the T_{AT} crossover. The two aspects of spin-glass ordering (frozen-in random spin orientations and strong irreversibility) which occur together at T_g in a standard spin-glass appear here at distinct temperatures.

Many further points remain to be elucidated in these systems. The start of the falloff in the ac susceptibility[5-7] seems to occur between T_{GT} and T_{AT}, at an ill-defined temperature which varies considerably with the exact experimental conditions. For a spherical sample of $Au + 19$ at.% Fe, where the demagnetizing factor is relatively high, the falloff begins[6] close to the temperature where we estimate the magnetization behavior to become strongly irreversible. For a needle-shaped sample where the ac susceptibility is much more sensitive to all forms of blocking mechanisms, after a sharp peak at T_c the ac susceptibility drops to a plateau and then begins to fall off gradually somewhere below the canting transition temperature.[7] This may be because of the weak irreversibility which sets in at the canting transition according to the mean-field model.

Although the model[8-12] gives remarkably good predictions for the overall qualitative behavior of this system, we should be wary of assuming that it gives a perfect representation of all aspects of the true physical situation. It is not yet clear experimentally if the canting transition is "sharp" in the thermodynamic sense; the susceptibility above technical saturation shows a minimum near the canting temperature[14,19] which does not seem to be predicted by the model. A full microscopic picture of the transitions remains to be given—neutron-diffraction results on a number of alloys of this type (e.g., Refs. 4 and 22) could be interpreted in terms of a spontaneous breakup into microdomains starting near the canting temperature. The Bragg-peak intensity anomalies observed by Murani[23] do not seem to correspond to the transition temperatures we propose.

In conclusion, we observe in $Au + 19$ at.% Fe a series of transitions as predicted by the Heisenberg mean-field model.[8-12] It would be important to establish the existance of canting and strong irreversibility transitions in other systems of this type.

The Laboratoire de Physique des Solides is a laboratoire associé au Centre National de la Recherche Scientifique. The Laboratoire de Spectrometrie Mössbauer, Equipe de Recherche associée 682, is a laboratoire associé au Centre National de la Recherche Scientifique.

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