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ac susceptibility of *Au*Fe near the percolation limit

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A detailed study of the ac susceptibility of a Au−15 at.% Fe sample in both a quenched and aged state is reported as a function of dc biasing field over a wide range of temperature. Both the quenched and aged samples exhibit considerable structure in their susceptibility-versus-temperature curves which might be taken as indicating various "transitions." However, unlike the situation reported recently for a variety of systems, we find that the predictions of scaling behavior are *not* followed at either transition; instead these data exhibit features which might be qualitatively interpreted in terms of a recently proposed triple-transition process in this system. The quantitative aspects of this latter model have yet to be worked out and tested, and so any definitive assignments still cannot be made.

In this report we are concerned with the behavior of the *Au*Fe system in the vicinity of that concentration regime where near-neighbor and indirect Ruderman-Kittel-Kasuya-Yosida interactions are in competition as the system evolves from a spin-glass state into a dilute ferromagnet. There has been considerable recent interest in this concentration regime following suggestions, based on model calculations,[1] of the possibility of reentrant behavior in the form of a paramagnetic to ferromagnetic transition at some Curie temperature T_c, followed at some lower temperature T_F by a second transition into a spin-glass state.[2,3] The principal aim of the present study was to ascertain whether the scaling-law predictions were followed in this system in the vicinity of both T_c and T_F, in a manner similar to that recently reported[4–6] for a number of "reentrant systems," or whether a more complicated situation existed. A Au + 15 at.% Fe alloy was prepared by diluting part of a 22 at.% Fe sample. Its physical dimensions and quenching treatment were similar to those previously reported,[7] and its ac susceptibility was measured at 2.4 kHz with an ac driving field of 0.46 Oe rms in a previously described magnetometer.[7]

The ac susceptibility $\chi(H,T)$ of the zero-field cooled quenched sample (Q), recorded while warming in zero dc field and small applied dc fields, is similar to that previously reported[2,3]; and here we concentrate on the behavior in moderate applied fields as summarized in Fig. 1. Previous investigators[2,3] have suggested that the high-temperature peak evident in Fig. 1 signals the onset of ferromagnetic ordering, and hence we have attempted to fit these data to the scaling-law predictions,[8] viz., the usual static scaling equation of state predicts that the susceptibility $\chi(h,t) = \partial m/\partial h$ should exhibit a peak at temperature T_M which increases above T_c with increasing field according to

$$t_m = (T_M - T_c)/T_c \propto h^{(\gamma+\beta)^{-1}} , \tag{1}$$

while the peak susceptibility $\chi(h,t_m)$ varies with field in an identical manner to that at T_c, viz.,

$$\chi(h,t_m) \propto h^{1/\delta-1} , \tag{2}$$

and further,

$$\chi(h=0,t) \propto t^{-\gamma}, \quad T > T_c . \tag{3}$$

The behavior summarized in Eqs. (1)−(3) has now been observed in a number of systems[4–8] and is applied below to the *Au*Fe system. Certainly the upper peak in Fig. 1 exhibits a behavior which is in qualitative agreement with that expected of a ferromagnet as summarized in Eqs. (1)−(3), viz., its height is re-

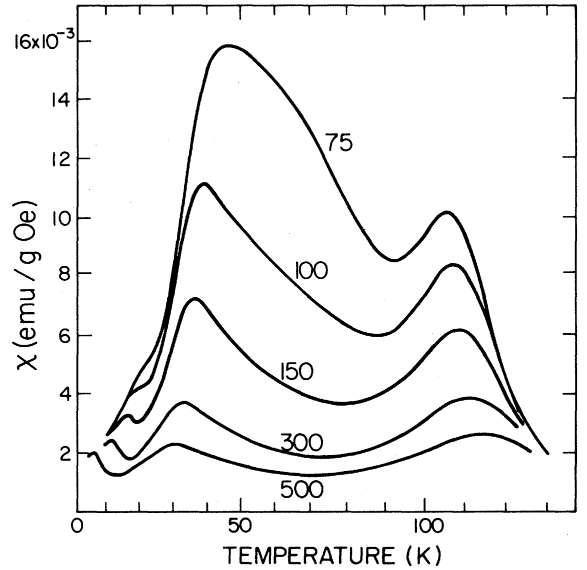

FIG. 1. ac susceptibility χ(emu/g Oe), measured in various static biasing fields, plotted against temperature (in kelvins). The static biasing field (in oersteds) is marked against each curve.

duced and the peak temperature increases with increasing field. A quantitative analysis, however, leads to a different conclusion.

(i) The maximum ac susceptibility measured in zero field is less than 20% of the limit set by the dimensions of the sample.

(ii) A plot of $\chi(h,t_m)$ (corrected for background and demagnetizing effects) against the internal field H_i on a double-logarithmic plot yields a straight line as shown in Fig. 2. This simple power-law dependence is in agreement with the predictions of Eq. (2), but the value for δ' ($=3.4\pm0.15$) obtained from it is considerably less than that usually quoted for ferromagnets. However, this estimate for δ agrees with that recently deduced[9] ($\delta=3.5\pm0.2$) from static measurements along the critical isotherm for an alloy of comparable concentration.

(iii) Equation (1) predicts that T_M should increase with field above T_c as $\sim H^{1/2}$ (for typical choices of γ and β). Figure 3 shows a plot of such data taken in fields between 50 and 750 Oe and which (within experimental error) behave approximately in the manner indicated. The intercept of this plot at $H=0$ yields an estimate for T_c of 101 K.

(iv) With this value for T_c, an attempt was made to verify Eq. (3) by plotting $\chi(0,T>T_c)$ (corrected for background and demagnetizing effects) against t on a double-logarithmic plot. This plot showed considerable curvature throughout the region immediately above T_c which could not be removed by adjustments in the value of T_c by a few degrees in either direction. From this we conclude that the power-law dependence predicted by Eq. (3) from the static scaling-law equation of state is not obeyed in this system.

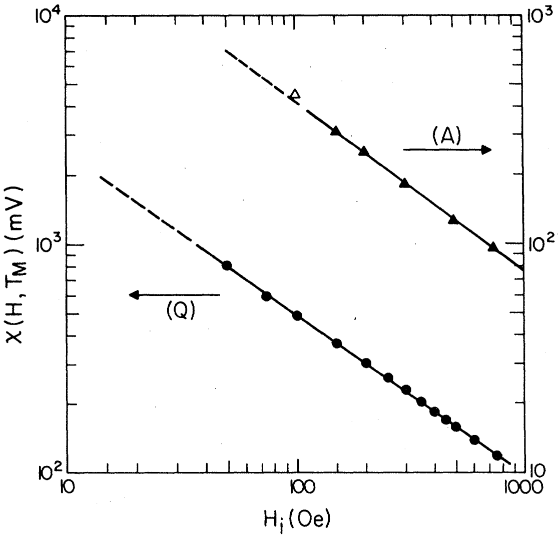

FIG. 2. Peak ac susceptibility $\chi(H,T_M)$, plotted against the internal field H_i (in oersteds), on a double-logarithmic plot: (●)—quenched sample (Q); (Δ)—aged sample (A). (Here 1 mV is equivalent to 8.6×10^{-7} emu/Oe.)

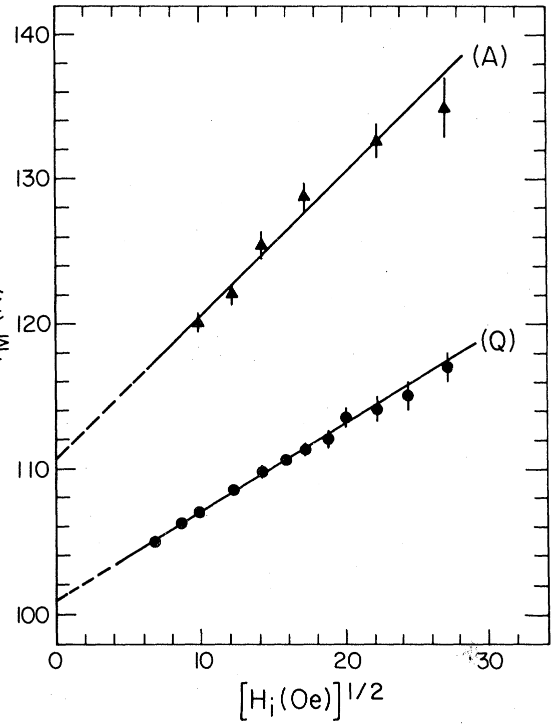

FIG. 3. Peak temperature T_M (in kelvins) plotted against the square root of the internal field H_i (in oersteds): (●)—quenched sample (Q); (Δ)—aged sample (A).

To summarize points (i) to (iv) above, the state evolving below $T_c^*=101$ K in this system, while being strongly magnetic, cannot be classified as ferromagnetic in the sense of being *fully* described by the scaling-law equation of state with unique (and conventional) values of the critical indices. The lack of true critical behavior has also been recently reported[10] for this system based, however, on arguments related to dynamical behavior; here we concentrate on the failure of *static* scaling laws which have been more widely applied.

The second peak in $\chi(H,T)$ around 40 K, evident in Fig. 1, has been taken to signify[3] the onset of a spin-glass transition below some characteristic temperature T_F. In other reentrant spin-glass systems it has recently been suggested[4–6] that an analogous form of the scaling law applies around T_F. Such assumption would lead to an analogous set of equations for the behavior of $\chi(\tilde{h},\tilde{t})$ near $\tilde{t}=(T_F-T)$. $T_F^{-1}\simeq0$, and our data have been analyzed on such a basis:

(i) A plot of the susceptibility at the temperature of the *second* peak (at T_p), $\chi(H,T_p)$ against H on a double-logarithmic plot does *not* yield the straight-line behavior analogous to Fig. 2, and reported in other systems.[4–6] Thus the power-law dependence of $\chi(H,T_p)$ on H with a unique index $(\tilde{\delta})$ does *not* exist here.

(ii) A plot of T_p vs $H^{(\tilde{\gamma}+\tilde{\beta})^{-1}}$ should yield a straight line whose intercept gives T_F. While no *a priori* values for $\tilde{\gamma}$ and $\tilde{\beta}$ exist, the data reported above suggests that $\tilde{\gamma}+\tilde{\beta}$ would have to be $\geqslant 10$ because T_p *initially* decreases rapidly with increasing field. We reject such a choice because it yields an estimate for T_F well in excess of T_c^*, which is clearly unphysical.

(iii) With no reliable estimate for T_F, no check on the relationship $\chi(0,\tilde{t})$ vs $(\tilde{t})^{-\tilde{\gamma}}$ could be performed.

In summary, the above data do not support the assertion that a scalinglike relationship applies near T_F in this system, in marked contrast to various other systems.[4–6]

As yet, no mention has been made of the third peak evident at low temperatures in fields greater than 150 Oe. Even in systems exhibiting two peaks in their $\chi(H,T)$ vs T curves, which can be reproduced qualitatively by numerical calculation based on the Sherrington-Kirkpatrick (SK) model,[1] the underlying physical processes are not clear. Certainly, the strongly magnetic state evolving below T_c^* appears to result from the ordering of Fe neighbors connected predominantly by direct d-d overlap in the percolation chain (PC); however, the subsequent evolution of this system into a zero-moment state at lower temperatures could be brought about by either (a) the PC and the remaining "loose spins" existing separately immediately below T_c^*, with the loose spins freezing out and locking onto the PC at T_F, or (b) a gradual pick up of loose spins by the PC on cooling below T_c^*, mostly involving antiferromagnetic coupling, leading to a diminishing net moment which becomes zero at T_F—now the effect compensation temperature, or (c) a realignment of the spins in the PC at T_F so that they become part of a zero-moment spin structure.

While (a) seems to be the most popular current interpretation of such data,[2] we suggest that consideration be given to (b) and (c), particularly if gradual PC spin realignment is incorporated into (b). Specifically, the long-range ordering of the PC immediately below T_c^* would not result in a fully collinear spin configuration because of existing random competing interaction which already cause an incipient spin-glass behavior. Further reduction in the temperature results in a gradual pick up of loose spins which leads to a zero-moment state below T_F due to the *gradual* randomizing of spin directions. Such a picture certainly agrees qualitatively with existing data—a reduced saturation moment per impurity,[11] a smeared transition near T_c^* in the ac susceptibility, and magnetoresistance results which demonstrate noncollinearity on a scale of the electronic mean free path.[12] Further, the instability of the intra-PC ferromagnetic (FM) groupings would occur only near the percolation limit because they would become less susceptible to the competing interactions with their surroundings as the FM volume increases, leading to a rapidly diverging T_c^* and T_F as the concentration increases. In addition, since the application of an external field would favor a collinear spin arrangement, one would expect the "compensation point" of the zero-moment state to be correspondingly depressed, leading to the large field sensitivity of the spin system near $T_F(H)$.

Now, to return to the third, low-temperature peak in $\chi(H,T)$. Various mean-field theories[13,14] have suggested the possibility of triple reentrant phase diagrams, with the latter hinting at a field dependence which is similar to that exhibited by the two lower peaks in $\chi(H,T)$, discussed above. As with the SK model, the underlying physical processes in these models are not clear; however, in terms of the discussion given above, a further anomaly might occur below T_F if, for example, the coarsely compensated zero-moment state occurring at T_F were to be replaced by a microscopically more random ground state so that the lower peak might signify a change in the scale of the randomness (brought about by a temperature-dependent range of interaction). Obviously, since the physical processes responsible for the SK-type behavior remain to be clarified, this third step is rather tentative—although the experimental situation is clear and a third peak does exist in $\chi(H,T)$.

Various interpretations of studies of aging effects following quenching in the *Au*Fe system[15] have sug-

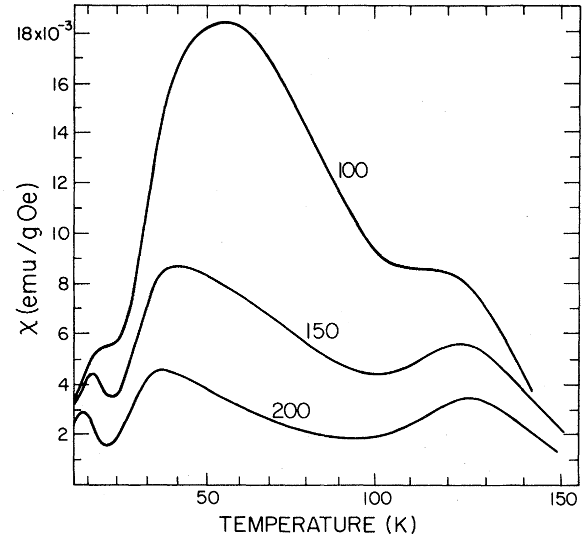

FIG. 4. ac susceptibility χ(emu/g Oe), measured in the various static biasing fields shown (in oersteds), plotted against temperature (in kelvins) for the aged sample. The data taken at 200 Oe has been shifted downwards by one scale unit (1×10^{-3} emu/g Oe) for clarity.

gested that it produces an increase in the number of nearest-neighbor Fe impurities. Following the measurements reported above, the Au–15 at.% Fe sample under investigation was aged at room temperature for about 190 h, after which the ac susceptibility was again measured in various fields over a wide range of temperature. In the aged sample the zero-field susceptibility is generally reduced in magnitude compared with the quenched specimen, but the principal maximum in $\chi(0,T)$ has moved *up* in temperature to around 101 K, whereas the low-temperature shoulder has moved *down* to about 50 K, in qualitative agreement with expectations based on the previous discussion. The effects of various applied fields can be seen in Fig. 4. The upper "pseudoferromagnetic" critical peaks require larger fields (100 compared with 50 Oe in the Q sample) to resolve them, indicating that the response at intermediate temperatures is more anisotropic, and hence the magnetization is more difficult to saturate. These peaks are considerably broader than their counterparts in the Q sample, and hence the temperatures T_M at which they occur are more difficult to determine accurately. These data have been analyzed in the same manner as for the Q sample and are included in Figs. 2 and 3. They lead to identical conclusions, viz., that the conventional, static scaling laws do *not* adequately describe the behavior of this system near its various "transition" points.

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