Disappearance of Ferromagnetic Order in Au-Fe

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Very-low-field magnetization measurements on specially prepared Au-Fe alloys are reported. It was possible to follow the spontaneous magnetization through a double magnetic transition. The spontaneous magnetization appears below the Curie temperature and disappears again at a second critical temperature.

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In recent years, considerable effort has been directed toward understanding the magnetic phase diagram of alloys with concentrations near the percolation threshold.[1] One of the most interesting phenomena studied is the disappearance at low temperature of long-range ferromagnetic order. It has been suggested that, due to frustration caused by random fields, some alloys, after becoming ferromagnetic at their Curie temperature, undergo a second phase transition at a lower temperature, supposedly into a spin-glass state.[1-6] This would mean that the ferromagnetic order parameter, the spontaneous magnetization, after gradually increasing below the Curie temperature, has to disappear again with decreasing temperature at a second critical temperature. However, due to hysteresis developing below the Curie temperature, the magnetization measurements in these alloys are difficult to interpret.[7] It has not been possible to directly measure the spontaneous magnetization in these alloys so far, and much controversy exists about this second phase transition.[8-11]

In a recent paper[12] we have demonstrated that by carefully controlling the atomic clustering of Fe atoms in Au-Fe alloys we can tune the Curie temperature of certain samples arbitrarily close to the multicritical point where the ferromagnetic and spin-glass phase lines intersect. We thus can obtain a state where the two transition temperatures are so close together in temperature that hysteresis should be negligibly small at the lower transition. Our method of changing the degree of atomic clustering, rather than the chemical concentration, makes this experiment possible.

Figure 1 displays low-field magnetization data in the form of Arrott plots, σ^2 vs H/σ, for a 14-at.%-Fe sample. The sample preparation has been described earlier.[12] All isotherms are completely reversible. The maximum field of the data displayed is 20 Oe. The data are corrected for demagnetization effects. The demagnetization factor has been determined experimentally from the magnetization data. The intercept of the isotherms with the H/σ axis yields the inverse initial

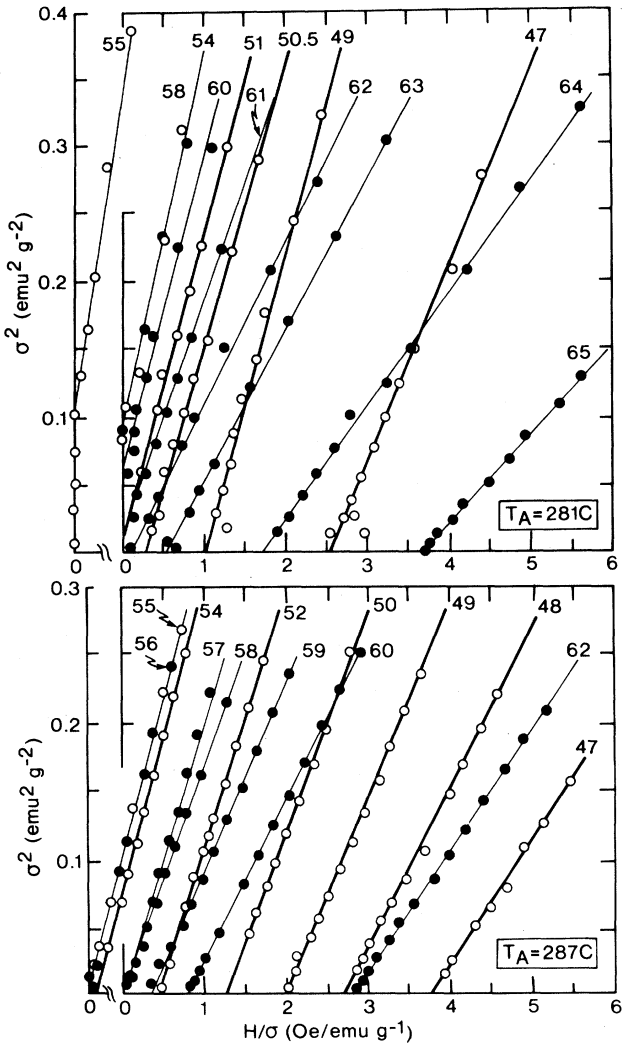

FIG. 1. Arrott plots of a Au:14-at.%-Fe sample. Upper part, after annealing at $T_A = 281$ C; lower part, $T_A = 287$ C. Numbers are temperatures in degrees kelvin. Lines through solid circles are isotherms from 65 to 56 K; lines through open circles are from 56 to 47 K.

susceptibility $1/\chi$, and the intercept with the σ^2 axis yields the spontaneous magnetization.

In the state quenched from 28 C (upper part of Fig. 1) the sample is ferromagnetic in the temperature range from 61.5 to 51 K. Above and below this range, the isotherms intercept the H/σ axis at positive values, yielding finite values of χ. The low-temperature ($T<51$ K) "quasiparamagnetic" state has considerable magnetic viscosity. However, the magnetic relaxation times down to 47 K are considerably smaller than the dc measurement time (~30 sec), i.e., at each value of the magnetic field the sample is allowed to reach its thermodynamic equilibrium magnetization and no coercivity is observed. The spread of the very-low-field data points of the 47-K isotherm is due to the onset of coercivity. Below 47 K, the effect of the hysteresis increases rapidly and the intercept with the H/σ axis can no longer be determined unambiguously.

The straight line behavior of the isotherms in Fig. 1 is somewhat misleading. It does not indicate mean-field behavior. At higher fields (>20 Oe) definite curvature develops at all temperatures (the critical isotherm has an exponent of $\delta = 3.5 \pm 0.2$). We have restricted the display to the lowest fields to demonstrate the well-defined intercepts. The isotherm at 55 K, displaced to the left, demonstrates how well the vertical rise is defined, allowing the experimental determination of both the demagnetization factor and the spontaneous magnetization.[13]

The lower part of Fig. 1 shows isotherms of the same sample annealed at 287 C. The ferromagnetic range is from 58 to 52 K. The spontaneous magnetization is now too small to be determined.

Annealing at a temperature lower than 281 C increases both the Curie temperature and the spontaneous magnetization σ_s, but hysteresis makes it impossible to follow σ_s through the second phase transition.

Arrott plot intercepts are summarized in Fig. 2 for states corresponding to four different annealing temperatures. In all four states, $1/\chi$ is strongly curved, indicating complex magnetic short-range order (the critical exponent γ is about 2). In the state quenched from 281 C the susceptibility diverges at $T_{c1} = 61.5$ K. Below T_{c1} spontaneous magnetization develops (solid squares) first increasing and then decreasing again. It disappears at a second critical temperature $T_{c2} = 51$ K. Below T_{c2} the susceptibility becomes finite again. The scatter in the spontaneous magnetization data is due mainly to small temperature drifts during an isotherm measurement. Thus the functional form of $\sigma_s(T)$ cannot be accurately determined. For the state quenched from 287 C, $1/\chi$ becomes zero at $T_{c1} = 58$ K. Below T_{c1}, $1/\chi$ remains zero and no measurable spontaneous magnetization develops. Below $T_{c2} = 54$ K, $1/\chi$ becomes finite again.

After annealing at 300 and 311 C the susceptibility remains finite at all temperatures. The broad minimum in $1/\chi$ probably corresponds to the direct transition from paramagnetic to spin-glass state observed at lower Fe concentrations. However, the low-temperature state here is quite different from the usual spin-glass state. The

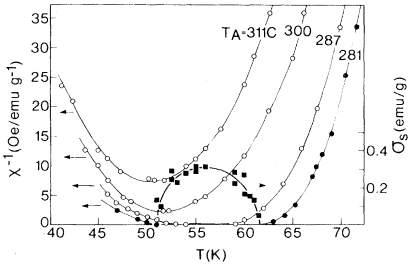

FIG. 2. Solid circles, open circles inverse susceptibility, scale on left. Solid squares, spontaneous magnetization, scale on right. For sample in four different annealing states.

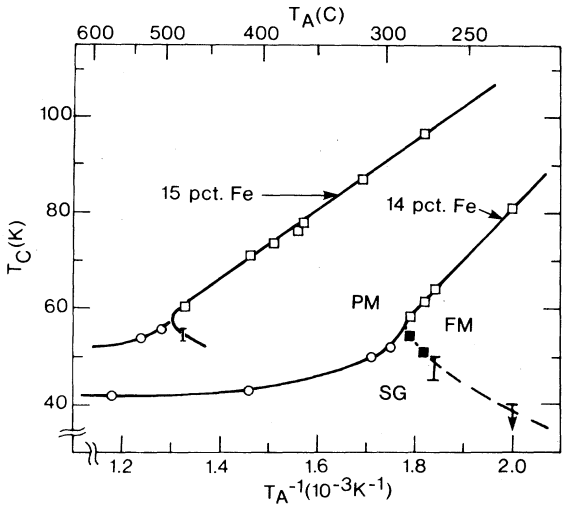

FIG. 3. Magnetic phase diagram: T_C vs reciprocal annealing temperature, T_A (in degrees K). Open squares paramagnetic to ferromagnetic transition; open circles paramagnetic to spin-glass; solid squares ferromagnetic to spin-glass; data points with error bars, estimated from ac susceptibility.

data in Fig. 2 all represent thermodynamic equilibrium values of the susceptibility, determined from hysteresis-free magnetic isotherms. The decrease in χ at low temperature is therefore a static effect and not due to a dynamic freezing of the spins.

Figure 3 shows the magnetic phase diagram for Au-Fe as determined from our measurements. We display the transition temperature versus the reciprocal annealing temperature. For similar alloys it has been shown that T_A^{-1} is proportional to the degree of atomic clustering,[14] a parameter comparable to the concentration. The data for the 15-at.% alloy and also some data for the 14-at.% alloy are taken from earlier ac susceptibility measurements.[12] The two solid squares for the 14-at.% alloy represent ferromagnetic to spin-glass transitions. Because of the rapid onset of hysteresis, this latter phase line cannot be followed to lower temperatures, at least not by bulk measurements.

The phase diagram near the multicritical point as shown in Fig. 3 may not be complete. There is a qualitative difference between the low-temperature states to the left and to the right of the multicritical point. To the right of this point, the magnetic isotherms in the spin-glass state are well behaved as seen in Fig. 1. However, to the left, the isotherms develop strong curvature at the lowest fields, similar to the behavior recently predicted.[15]

In conclusion, we were able to measure the spontaneous magnetization of a weakly ferromagnetic alloy and to demonstrate clearly the existence of a second phase transition out of the ferromagnetic state. Because of hysteresis below the Curie temperature, this can be done only very close to the multicritical point.

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