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The magnetoresistance behaviour of $Au_{87}Fe_{13}$ cluster glass

A K Nigam, S Radha and Girish Chandra
Tata Institute of Fundamental Research, Homi Bhabha Road, Bombay 400 005, India

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Abstract. Magnetoresistance ($\Delta\rho/\rho$) measurements for $Au_{87}Fe_{13}$ are reported in the temperature range 4.2–200 K and for magnetic fields (H) up to 45 kOe. The remarkable feature of the experimental observations is the rapid decrease of $|\Delta\rho/\rho|$ below 15 K at low fields. This behaviour is associated with the breaking up of large clusters into smaller ones as a result of the dominating RKKY interaction in the system at low temperatures. The observed behaviour corroborates the neutron depolarization studies carried out earlier.

1. Introduction

There have been extensive studies of spin glasses and related systems during the past two decades. All these studies have led to a considerable debate over the nature of magnetic interactions in various identified phases, such as spin glass, re-entrant spin glass and ferromagnetic phases, which occur in these systems in different compositions and temperature ranges. The magnetic phase diagram of AuFe shows the existence of all the above magnetic phases depending on the concentration of Fe atoms [1]. It shows a spin glass behaviour below about 10 at.% Fe, while above 15 at.% Fe a re-entrant spin glass behaviour is observed. In the concentration range of 10–15 at.% Fe, a cluster glass behaviour is found. There have been extensive studies of AuFe alloys in the spin glass and the re-entrant spin glass regime, but relatively few studies exist in the cluster glass regime. The AC magnetic susceptibility of AuFe in the cluster glass regime has been studied by Cannella and Mydosh [2] as well as by Sarkissian [3]; they found a sharp peak at a characteristic temperature T_{cg} similar to that found in spin glasses, but the magnitude of the AC susceptibility is much larger. In this concentration regime the magnetic properties are strongly sensitive to the thermal history of the samples, such as the annealing temperature, the quenching rate, and so on. For example, Crane and Claus [4] have studied the magnetization and AC susceptibility of AuFe with 14 and 15 at.% Fe with different annealing temperatures and found that the 15 at.% Fe alloy is more sensitive to the annealing temperature than the 14 at.% Fe alloy. This indicates that the alloys near the percolation limit are rather sensitive to their thermal history. Mössbauer absorption spectra of Fe^{57} in AuFe have scarcely been studied in the cluster glass region. Ridout [5] has studied Au–12.8 at.% Fe alloy, which shows a hyperfine field splitting below 26 K. Murani [6] has carried out small-angle neutron scattering measurements on AuFe alloys containing 10 and 13 at.% Fe, and found that there exists a series of freezing temperatures corresponding to relaxation times of finite magnetic clusters. Sarkissian [7] has recently reported neutron-spin depolarization studies on AuFe alloys in the concentration range between 15–19 at.% Fe. While the alloys with concentrations above the percolation limit

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(17–19 at.% Fe) continue to remain ferromagnetic at all temperatures below the Curie temperature T_c, the 15 at.% Fe alloy reveals an anomaly at two distinct temperatures. The first corresponds to the temperature of formation of ferromagnetic clusters (around 90 K). The lower temperature (around 50 K) is associated with the breaking of these clusters into smaller ones, which subsequently freeze into a spin glass state. The results of these measurements suggest that the spin system in the cluster glass alloys is localized into finite magnetic clusters.

The magnetoresistance studies in a few cluster glass alloys have been carried out by Rakoto and co-workers [8] at very high fields but only at 4.2 K, and by Senoussi [9] at very low fields. Barnard has also studied magnetoresistance in a few spin glasses in fields of a few gauss [10, 11]. In $Au_{86}Fe_{14}$ alloy he observed a negative magnetoresistance in the quenched alloy with a maximum at T_{cg}, while room-temperature aging of the same alloy for a few days resulted in a positive magnetoresistance that was highly enhanced in magnitude compared to the negative one in the quenched state [10]. In the present paper, a detailed study of magnetoresistance in the temperature range 4.2–200 K and for magnetic fields up to 50 kOe is reported for $Au_{87}Fe_{13}$.

2. Experimental details

The sample (provided by B R Coles almost a decade ago) was prepared by induction melting the high-purity constituent metals and was drawn into wire form and homogenized at 800 °C for 36 h. Prior to measurement, it was annealed at 850 °C for 24 h and quenched in water.

Longitudinal magnetoresistance was measured using the standard four-probe DC technique in the temperature range 4.2–200 K and for magnetic fields up to 45 kOe generated by a home-built superconducting magnet. In the presence of magnetic fields the specimen temperature below 70 K was monitored with a Lake Shore Carbon Glass Sensor; above 70 K a Lake Shore Silicon Diode Sensor was used. The zero-field electrical resistivity measurements were carried out in the temperature range 1.8–300 K. The electrical contacts to the specimen were spot-welded. All measurements were automated using an IBM-PC via an IEEE-488 interface with the measuring instruments. The stability in measurements was better than 50 PPM.

3. Results and discussion

In their AC susceptibility studies on $Au_{87}Fe_{13}$, Cannella and Mydosh [2] found the cluster glass transition temperature, T_{cg}, to be 38 K; Sarkissian [3] found it to be 45 K. We measured the AC susceptibility of the alloy reported here and found a similar behaviour, but T_{cg} was 40 K. These variations in T_{cg} could be due to the slightly different annealing temperatures used by the authors prior to quenching. The magnetoresistance ($\Delta\rho/\rho$) defined as

$$\Delta\rho/\rho = [R(H) - R(0)]/R(0)$$

where $R(H)$ is the resistance of the sample in a magnetic field H, is plotted as a function of H at various temperatures in figure 1. It is found that $\Delta\rho/\rho$ is negative at all temperatures and fields, increasing in magnitude with the field. There seems to be a large increase in $|\Delta\rho/\rho|$ as the concentration of Fe increases beyond 7 at.%. This can be qualitatively explained if one assumes a model where the ferromagnetic clusters are coupled via an s–d exchange type of interaction, identical to that of a spin glass in which the magnetic spins play the role of these clusters. The magnetoresistance, being a microscopic measurement, will have two contributions, one due to the ferromagnetic interaction among the spins inside a cluster, and the other due to the spin glass type of interaction among these clusters. This latter interaction seems justified because, in this concentration range, the magnetic impurity concentration is such that these clusters stay far apart so that no direct magnetic interaction may take place between them. Due to the additive nature of the two contributions, the negative magnetoresistance is expected to be significantly higher than what is expected from either of the above two types of interaction. Mirza and Loram [12] have explained their specific heat data on **AuFe** on the basis of a similar model. This anomalous increase in magnetoresistance when going from the spin glass to the mictomagnetic regime is evident from the measurements made on some of the alloys in that concentration range [8, 13].

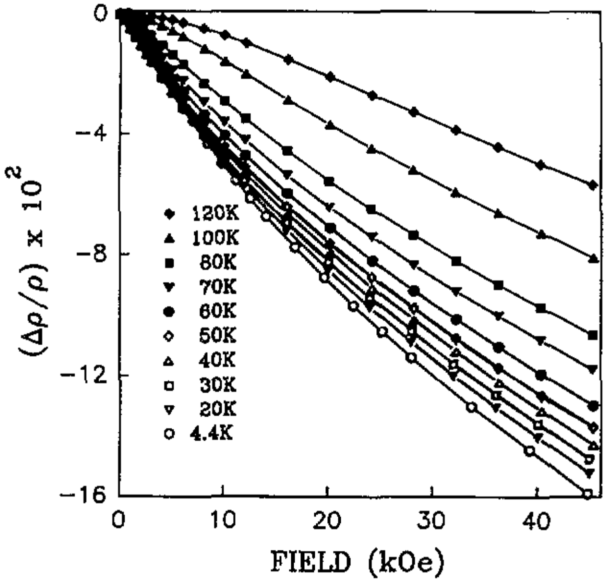

Figure 1. The magnetic field (H) dependence of $\Delta\rho/\rho$ at various temperatures.

It is observed that $|\Delta\rho/\rho|$ decreases with increasing temperature except at low fields, as can be seen from figure 1. Figure 2 shows that at low fields (below 4 kOe), $|\Delta\rho/\rho|$ varies faster than H, whereas at higher fields $|\Delta\rho/\rho|$ varies more slowly than H, which is similar to what has been observed in re-entrant spin glasses, such as $Au_{82}Fe_{18}$ [14] and amorphous $(Fe_{0.2}Ni_{0.8})_{75}P_{16}B_6Al_3$ [15] below their Curie temperatures. The field dependence of $\Delta\rho/\rho$ has been fitted to an expression

$$\Delta\rho/\rho = \alpha H^n$$

in the field range 4–45 kOe; the value of n is found to be 0.75±0.02 in the temperature range 4.2–50 K. A similar magnetic field dependence of $\Delta\rho/\rho$ has been observed in $Fe_{59}Ni_{21}Cr_{20}$, which has been claimed to be a spin glass [16]. However, it is most likely a cluster glass since the composition of the alloy is very close to that of a re-entrant spin glass studied by the authors. Above 50 K, n increases continuously with temperature to a value of 1.5 at 120 K. This indicates that short-range spin correlations exist even at 120 K, since in a completely paramagnetic state n is expected to be 2 [14].

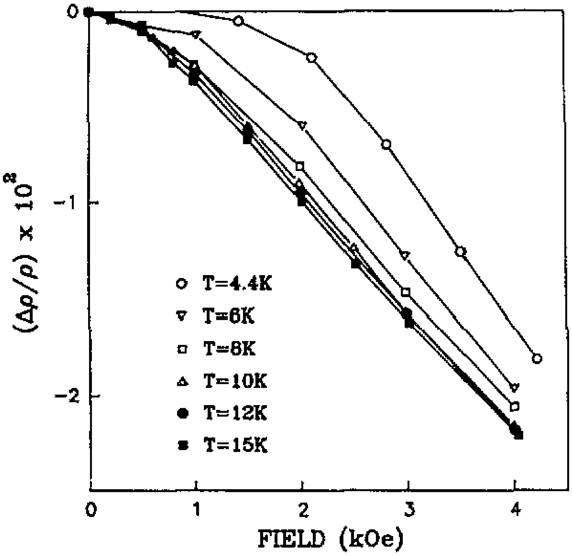

Figure 2. The variation of $\Delta\rho/\rho$ with H at low fields (up to 4 kOe) and low temperatures (up to 15 K).

The temperature dependence of the magnetoresistance in different magnetic fields is interesting. This is seen from figures 3 and 4, where $|\Delta\rho/\rho|$ has been plotted as a function of temperature at several magnetic fields in the range 1–45 kOe. At low fields, below about 5 kOe, $|\Delta\rho/\rho|$ increases with temperature, initially up to about 15 K, and then tends to flatten out at higher temperatures up to around the cluster glass transition temperature, T_{cg}. Above 50 K, $|\Delta\rho/\rho|$ decreases continuously with temperature. The initial increase of $|\Delta\rho/\rho|$ with temperature is quite significant at low fields (below 2 kOe) as shown in the inset of figure 3. This is a remarkable feature, which has not been reported until now in cluster glasses at such fields. The impurity concentration of $Au_{87}Fe_{13}$ lies well below the percolation threshold. The measurements of low-field magnetization and AC susceptibility of Au–Fe alloys of 14 and 15 at.% Fe by Crane and Claus [4] do not show any RSG behaviour for samples quenched from above 400 °C. The annealing temperature used for our sample is much above this. Furthermore, comparing with our earlier magnetoresistance studies made on an RSG alloy, $Au_{82}Fe_{18}$ [14], the present measurements of $\Delta\rho/\rho$ at higher fields do not indicate any re-entrant phase, since $|\Delta\rho/\rho|$ decreases monotonically with temperature rather than showing any increase at an intermediate temperature range. All these observations confirm that $Au_{87}Fe_{13}$ is not a re-entrant spin glass.

Senoussi [9] has observed an initial rapid increase of $|\Delta\rho/\rho|$ with temperature in an AuFe(14.5 at.%) alloy at 100 Oe, which exhibited two peaks, one at 35 K and the other at

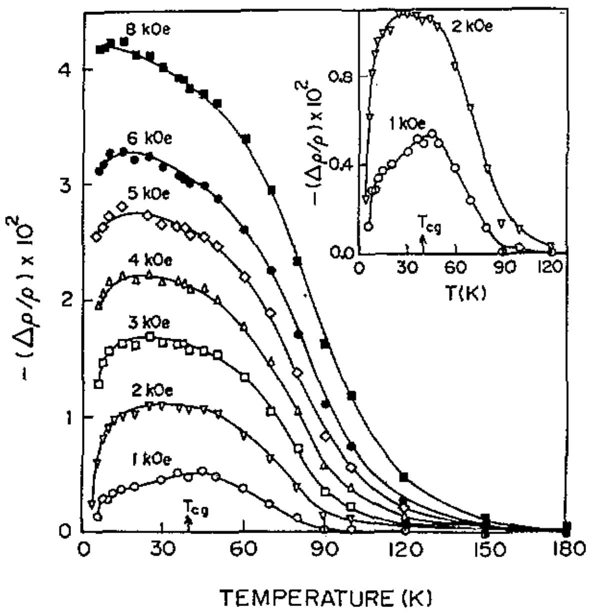

Figure 3. The temperature (T) dependence of $\Delta\rho/\rho$ at different fields (H). The inset shows $\Delta\rho/\rho$ at low fields (1–2 kOe).

75 K. Senoussi claimed the rapid increase up to 35 K to be due to a re-entrant spin glass (RSG) phase. This is contrary to what has been observed in one of our earlier studies of magnetoresistance in the RSG alloy $Au_{82}Fe_{18}$ [14], where below the RSG transition $|\Delta\rho/\rho|$ was found to increase with decreasing temperature. It was argued in the above paper that the observation of a rapid decrease of $|\Delta\rho/\rho|$ in AuFe(14.5 at.%) below 35 K could be due to the fact that the low field (100 Oe) used by Senoussi was small enough not to disrupt the spin glass phase, as compared to that of ours (around 3 kOe). But the observation of a rapid decrease of $|\Delta\rho/\rho|$ at lower temperatures in the present study goes against the above argument, since here this behaviour is observed even up to 3–4 kOe. Thus it seems that the drop in $|\Delta\rho/\rho|$ below T_{cg} is not due to a re-entrant spin glass phase, but is probably be due to the reason described below.

As the temperature decreases, the clusters having their relaxation times longer than the measuring probe time ($\simeq 10^{-15}$ s in the present case) appear frozen. The decrease of $|\Delta\rho/\rho|$ below about T_{cg} and at a low field (1 kOe) as observed in the present study is possible if the number of clusters with relaxation times comparable to the measuring probe time increases. This process can occur only if the large clusters are broken into smaller ones which would have shorter relaxation times. This has indeed been found in the neutron depolarization studies by Sarkissian [7] on an $Au_{85}Fe_{15}$ alloy. In his measurement, it was found that below about 50 K the large clusters start breaking into smaller ones which are frozen below 25 K. This breaking up of large clusters occurs because of the dominance of the RKKY interaction over the direct d–d interaction between spins within the cluster. This temperature for freezing depends on the inter-cluster interaction, and hence would change with a change in Fe concentration. This temperature could be lower for $Au_{87}Fe_{13}$, since the broken cluster may be comparatively smaller in size leading to a weaker inter-cluster interaction. This is

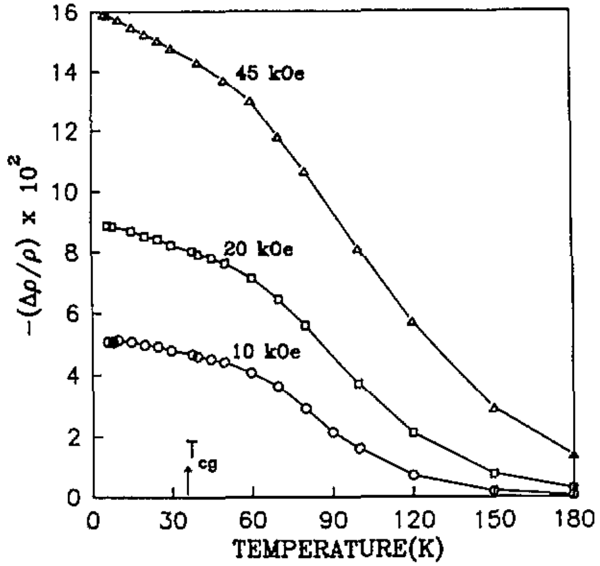

Figure 4. The variation of $\Delta\rho/\rho$ with T at higher fields (10–45 kOe).

presumably the reason why we observe a sharp drop in $|\Delta\rho/\rho|$ around 15 K, indicating a spin glass type of freezing. The small-angle neutron scattering measurements on AuFe(13 at.%) alloy by Murani [6] showed a series of q-dependent freezing temperatures, which suggests a large distribution of cluster sizes. The characteristic size of these clusters determined through his study ranges from 40–100 Å (in the temperature range 25–35 K). As the field is increased the correlation among clusters increases, causing them to grow in size. This would result in a higher relaxation time of clusters but reduce the range of their distribution, thereby leading to a weak temperature dependence of $|\Delta\rho/\rho|$, as is seen for magnetic fields in the range of 4–6 kOe. At still higher fields the clusters grow further in size, leading to a marked reduction in their number. Consequently, the conduction electron scattering due to the inter-cluster interaction decreases, while that due to the intra-cluster interaction caused by the direct d–d interaction increases and becomes predominant. This would give rise to a monotonically decreasing magnetoresistance with increasing temperatures since the temperature would try to disrupt the spin–spin correlations.

The variation of the magnetic contribution to resistivity ρ_m (obtained by subtracting the contribution of the pure-metal host from the total resistivity) and its first derivative, $d\rho_m/dT$, as a function of temperature is shown in figure 5. The value of the resistivity is quite large ($\simeq 68\,\mu\Omega$ cm at 2 K). It has been fitted to the relation

$$\rho_m = \rho_0 + AT^{3/2}$$

and it is found to hold up to the ordering temperature (40 K). This indicates that the $T^{3/2}$ dependence of ρ_m is due to excitations in clusters of ferromagnetically ordered spins, as discussed by Fischer [17], and is not due to spin-diffusion modes. According to the Rivier–Adkins theory of spin-diffusion modes [18], the range of the $T^{3/2}$ dependence of ρ_m is well below the spin glass freezing temperature, which is not the case in the present alloy. A similar behaviour was found by Mydosh and co-workers [19] in Au–12 at.% Fe alloys.

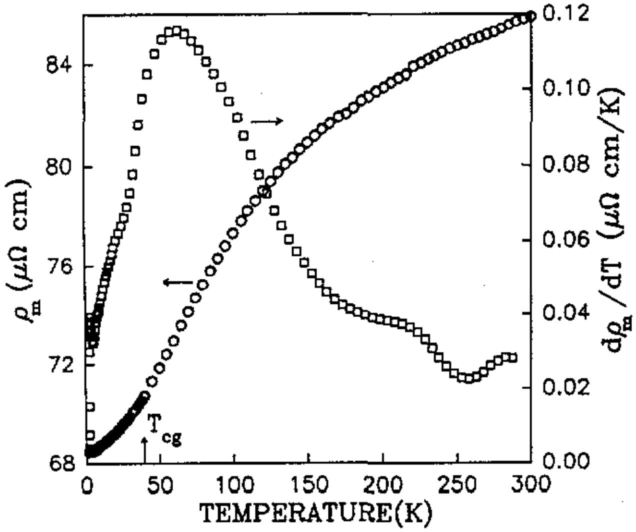

Figure 5. The magnetic contribution to resistivity ρ_m and its first derivative $d\rho_m/dT$ plotted as a function of temperature.

The first derivative, $d\rho_m/dT$, shows a maximum at 60 K and probably indicates that cluster formation starts around this temperature.

4. Conclusion

To summarize, a detailed study of magnetoresistance has been reported in $Au_{87}Fe_{13}$ cluster glass over a wide temperature range at several magnetic fields up to 45 kOe. The anomaly at low temperature (below 15 K) seems to occur as a result of the breaking of large clusters into smaller ones with relaxation times comparable to that of the measuring probe. The present study corroborates the results of neutron scattering and neutron-spin depolarization measurements on a similar alloy [6, 7].

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