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The evolution of spin glass state in AuFe re-entrant spin glass by Cr substitution: a magnetoresistance study

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Abstract. A detailed study of magnetoresistance ($\Delta\rho/\rho$) has been carried out on a series of $Au_{82}(Fe_{1-x}Cr_x)_{18}$ alloys with $0 \leqslant x \leqslant 0.6$ in the temperature range of 4.2–300 K and in magnetic fields up to 45 kOe. The AC susceptibility measurements carried out earlier showed that the system undergoes a transition from a re-entrant spin-glass phase for Fe-rich alloys to spin-glass phase for Cr-rich alloys. The magnetoresistance measurements show a progressive increase of magnetic disorder below the re-entrant spin glass transition. For $0.15 \leqslant x \leqslant 0.4$, $\Delta\rho/\rho$ shows a behaviour similar to that of cluster glasses. For $x = 0.6$, $\Delta\rho/\rho$ exhibits a spin-glass-type behaviour. The magnetic field (H) dependence of $\Delta\rho/\rho$ shows that there exists a unique power-law dependence of $H^{2/3}$ below the Curie temperature in Cr-containing re-entrant spin glasses.

1. Introduction

The magnetism of disordered alloys has been a subject of interest for several decades. Many systems have been discovered to date showing varieties of magnetic order [1]. One such type of magnetic order is spin glass where the spins are randomly frozen below a characteristic temperature (T_g). The spin-glass ordering was first discovered in noble-metal–transition-metal alloys such as **Au**Fe, **Cu**Mn, **Au**Mn, **Ag**Mn and **Au**Cr having a dilute concentration of magnetic impurity [2]. The magnetic phase diagrams of **Au**Fe and **Au**Cr derived from experimental data show a percolation limit at 15 at.% of magnetic impurity [3, 4]. The first-nearest-neighbour interaction is positive in **Au**Fe while it is negative in **Au**Cr. In **Au**Fe, there is a strong tendency to form Fe clusters even in alloys containing 3–4 at.% Fe. For alloys having 15–20 at.% Fe, a re-entrant spin-glass (RSG) behaviour has been observed where there is a transition from a paramagnet to a ferromagnet at a temperature T_c and a second transition to a spin-glass state at a lower temperature T_g. In **Au**Cr alloys, there is apparently some signature of RSG behaviour in 12 and 13 at.% Cr-containing alloys [4]. Since both the above systems have a percolation limit of 15 at.% but with opposite near neighbour exchange interaction, it would be interesting to look into the mixed magnetic phase of the two systems. We therefore carried out measurements of magnetic and electrical transport properties on the ternary **Au**FeCr system where the total magnetic impurity concentration has been kept at 18 at.% which is above but close to percolation concentration. Our earlier measurements of AC susceptibility on $Au_{82}(Fe_{1-x}Cr_x)_{18}$ alloys with $0 \leqslant x \leqslant 0.4$ showed that the system slowly evolves from an RSG phase to a spin-glass (or cluster-glass) phase with increasing x [5]. The magnetic phase diagram of $Au_{1-y}(Fe_{1-x}Cr_x)_y$ determined by Nakai *et al* through their magnetic susceptibility and neutron diffraction measurements shows that for $x = 0.6$, the spin-glass state is stable up to $y = 0.28$ which is well above the percolation limit of both **Au**Fe and **Au**Cr alloys [6].

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Magnetoresistance ($\Delta\rho/\rho$) measurements in canonical spin glasses have been reported by Nigam and Majumdar [7] as well as by Senoussi [8]. They observed a quadratic dependence of $\Delta\rho/\rho$ on magnetic field (H). Magnetoresistance in concentrated alloys has not been widely studied as compared to the dilute alloys. There have been a few studies by Rakoto *et al* [9] and Hamzic and Campbell [10] at high fields and by Barnard [11] at very low fields in a few concentrated **Au**Fe alloys. Magnetoresistance measurements on an RSG alloy, $Au_{82}Fe_{18}$ [12], and cluster glass (CG) $Au_{87}Fe_{13}$ [13] show characteristic features in their temperature dependence. It was found that in case of **Au**Cr spin glasses, the magnetoresistance was positive and very small [14] and could not be detected for alloys with Cr concentration above 5 at.%.

From the theoretical viewpoint, magnetoresistance has been less dealt with than any other experimental measurements. Mookerjee [15] formulated a theory for magnetoresistance in dilute alloys based on the EA model for spin glasses [16], and its validity has been proved for canonical systems [17] and also some ternary alloy systems [18]. There has not been any calculation of magnetoresistance in re-entrant spin glasses since the magnetic exchange interactions in these systems are not clearly identified. There have been a few theoretical calculations of magnetoresistance due to s-d interaction in ferromagnetic and antiferromagnetic metallic systems [19]. Balberg has calculated the magnetoresistance of ferromagnets due to electron scattering by spin fluctuations in the framework of mean-field theory [20]. Yamada and Takada have also calculated the magnetoresistance of ferromagnetic metals due to electron scattering by the localized spins in the first Born approximation using the Nakano–Kubo–Mori approximate formula [21]. They have calculated the temperature and field dependences of magnetoresistance both below and above the Curie temperature. In the molecular-field approximation, they have obtained an $H^{2/3}$ dependence of $\Delta\rho/\rho$ around the Curie temperature while at lower temperatures a linear dependence of $\Delta\rho/\rho$ on H has been obtained.

In the present paper, we report magnetoresistance measurements on $Au_{82}(Fe_{1-x}Cr_x)_{18}$ alloys with $0 \leqslant x \leqslant 0.6$ in the temperature range of 4.2–300 K and in magnetic fields up to 45 kOe. The temperature and field dependences of $\Delta\rho/\rho$ have been examined over the entire range of temperatures and fields which are discussed in the subsequent sections.

2. Experimental details

The samples were prepared by arc melting the high-purity constituents in an argon atmosphere and homogenized by annealing the alloy buttons for 1 week at 850 °C. They were cold rolled into foils of 100 μm thickness from which rectangular strips ($\simeq$ 10mm×3mm) were cut out for measurement. These strips were annealed for 24 h at 850 °C, quenched in water and stored in liquid nitrogen until measurement. The samples were studied through x-ray diffraction and were found to have a single phase. Compositional analysis on a few samples via electron-probe micro-analysis showed that the analysed concentration did not deviate much from the nominal ones.

Longitudinal magnetoresistance measurements were carried out on $Au_{82}(Fe_{1-x}Cr_x)_{18}$ ($0.05 \leqslant x \leqslant 0.8$) alloys in fields up to 45 kOe and a temperature range of 4.2–300 K in a home-built superconducting magnet cryostat employing the standard four-probe DC technique. The sample temperature in the presence of a magnetic field was monitored with a Lakeshore carbon glass sensor below 70 K while a Lakeshore silicon diode sensor was used above 70 K. All measurements were automated using an IBM PC to which the measuring instruments were coupled via an IEEE-488 interface. The stability in measurements was better than 50 ppm.

3. Results and discussion

The AC susceptibility studies [5] showed that the alloys with $x = 0.05$ and 0.1 are RSGs with T_c around 127 K and 109 K respectively and T_g of 70 K for both the alloys. Alloys with $x > 0.1$ were found to exhibit a single peak in χ_{AC} similar to that of a CG while $x = 0.4$ showed a spin-glass-like cusp in χ_{AC} at 57.5 K. For alloys with $x \geqslant 0.6$, the susceptibility is too small and hence could not be measured on our χ_{AC} set-up. Recent measurements of DC magnetization carried out on a SQUID magnetometer on the $x = 0.6$ sample, in both zero-field-cooled and field-cooled states, show a deviation at around 70 K signifying a spin glass behaviour below this temperature. This agrees with the value reported by Nakai *et al* [6]. Detailed results of this measurement will be reported elsewhere. It is found that the spin glass transition temperature, T_g, for all the alloys is in the temperature range of 60–70 K indicating a weak Cr concentration dependence. This is presumably due to the compensating effects of damping of the mean free path which reduces T_g and of reduction in the mean ferromagnetic exchange interaction tending to increase T_g [5].

The measured magnetoresistance ($\Delta\rho/\rho$) defined as

$$\Delta\rho/\rho = \frac{(R(H) - R(0))}{R(0)}$$

where $R(H)$ is the resistance in the presence of field H and $R(0)$ is the resistance in zero field, is plotted as a function of magnetic field H at 10 K for all the samples in figure 1. All the alloys exhibit negative magnetoresistance in the range of temperatures measured. With increasing Cr concentration, $|\Delta\rho/\rho|$ decreases. In the $x = 0.6$ alloy, the magnetoresistance is very small and could only be detected at a field of 40 kOe while in the $x = 0.8$ alloy, no magnetoresistance could be detected within the accuracy of our set-up.

3.1. Temperature dependence of magnetoresistance

3.1.1. Re-entrant spin glass alloys behaviour. The temperature variation of $|\Delta\rho/\rho|$ for the RSG alloys ($x = 0$, 0.05, 0.1) from 4.4 K to 300 K is shown in figure 2 in fields of 5 and 40 kOe. It shows the existence of two distinct phases. At low fields (5 kOe), $|\Delta\rho/\rho|$ initially decreases with increasing temperature up to around T_g and then increases up to a temperature which is close to T_c (determined from χ_{AC} measurements) for the $x = 0$ alloy but is higher than T_c for the $x = 0.05$ and 0.1 alloys. For example, the peak in $|\Delta\rho/\rho|$ for the $x = 0.1$ alloy occurs at 135 K while T_c is 109 K. This increase of $|\Delta\rho/\rho|$ beyond T_c indicates that the ferromagnetic clusters exist at temperatures well above T_c. Below T_g and at low fields, $|\Delta\rho/\rho|$ is nearly the same for $x = 0.05$ and 0.1 alloys but significantly lower than the $x = 0$ value. At higher fields (40 kOe), $|\Delta\rho/\rho|$ decreases as x increases from 0 to 0.1, the drop being more significant as x changes from 0 to 0.05. This difference in $|\Delta\rho/\rho|$, at higher fields, between the two Cr-containing alloys narrows down to almost zero at T_g and remains so until T_c. Further, $|\Delta\rho/\rho|$ is independent of temperature between T_g and T_c. The temperature dependence of $\Delta\rho/\rho$ may be explained qualitatively in the following way:

(i) Below T_c and above technical saturation, the magnetoresistance is primarily due to scattering of electrons from spin wave excitations. As a result of reduction in electron–magnon scattering with decreasing temperature, this contribution to magnetoresistance decreases as the temperature is lowered. This behaviour is similar to that observed in normal ferromagnets [22].

(ii) The observed increase of $|\Delta\rho/\rho|$ below T_g could be explained if spin disordering takes place below this temperature. The increased spin disorder will lead to a higher

scattering cross-section thereby increasing the negative magnetoresistance. This has been found to occur as a result of RKKY interaction dominating over short-range ferromagnetic interaction. This would cause canting of spins as envisaged in various theoretical models [23]. This canting transition has been seen through Mössbauer polarization measurements by Varret *et al* [24]. Murani [25] from his neutron scattering measurements and Gangopadhyay *et al* [26] from their spontaneous magnetization measurements have observed that not all the spins are integrated into the infinite cluster formed at T_c. These uncorrelated spins do not contribute to the ferromagnetism of the alloy. At lower temperatures, these spins would be correlated to form small magnetic clusters. The conduction electron scattering from these finite clusters having short-range ferromagnetic correlations will progressively reduce as the temperature drops below T_g. This would also lead to an increase of negative magnetoresistance. It is therefore possible that the increase of $|\Delta\rho/\rho|$ below T_g is due to both contributions.

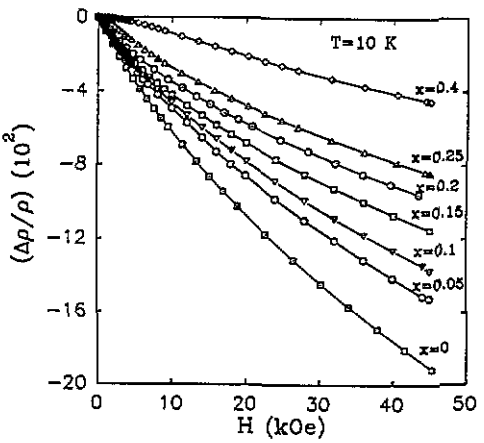

Figure 1. The magnetic field (H) dependence of $\Delta\rho/\rho$ of the $Au_{82}(Fe_{1-x}Cr_x)_{18}$ ($0 \leqslant x \leqslant 0.4$) alloys at 10 K. Lines joining the points are a guide to the eye.

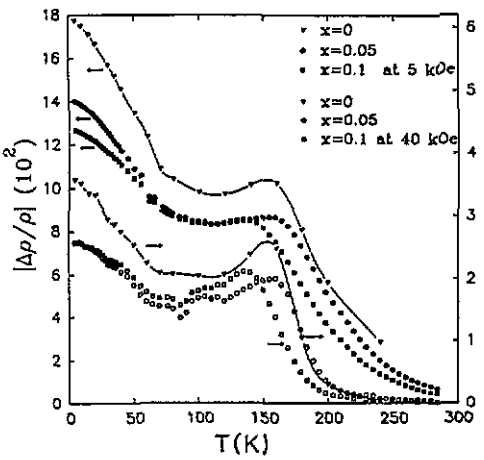

Figure 2. The temperature (T) dependence of $|\Delta\rho/\rho|$ at 5 and 40 kOe for the re-entrant spin glass alloys ($0 \leqslant x \leqslant 0.1$).

In the RSG alloys with $x = 0.05$ and 0.1, transverse magnetoresistance was also measured in the temperature range of 4.4–180 K and no anisotropy could be detected between the longitudinal and transverse magnetoresistance.

3.1.2. Cluster glass and spin glass behaviour. The temperature variations of $|\Delta\rho/\rho|$ for the $x = 0.15$–0.4 alloys at 5 kOe and 40 kOe are shown in figures 3 and 4. Some of the salient features are the following.

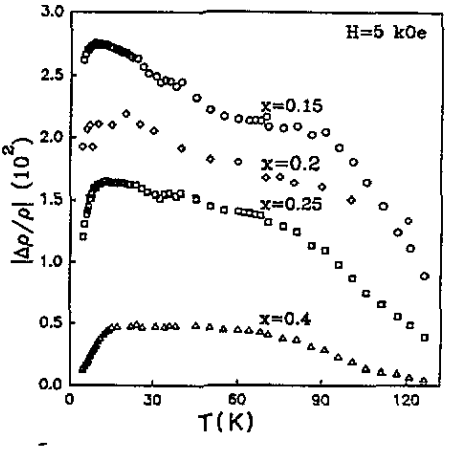

Figure 3. The temperature (T) dependence of $|\Delta\rho/\rho|$ at 5 kOe for alloys with $0.15 \leqslant x \leqslant 0.4$.

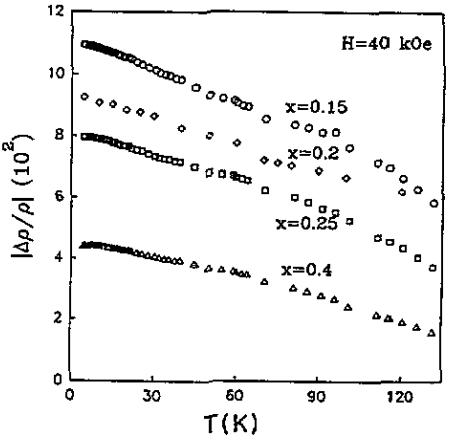

Figure 4. Variation of $|\Delta\rho/\rho|$ with T at high field (40 kOe) for alloys with $0.15 \leqslant x \leqslant 0.4$.

At low fields (5 kOe), $|\Delta\rho/\rho|$ initially increases with temperature and shows a maximum in the temperature range of 10–20 K for various alloys. This maximum becomes more pronounced with increasing x. A similar effect has been seen in $Au_{87}Fe_{13}$ cluster glass [13]. The maximum in $|\Delta\rho/\rho|$ occurs at a temperature (T_m) which is well below T_g of the alloy. In $Au_{85}Fe_{15}$, neutron depolarization measurements show two magnetic anomalies, one below 90 K due to formation of large but finite clusters and the other at 25 K due to spin-glass-like freezing of small clusters formed as a result of breaking up of large clusters which had formed at 90 K [27]. The breaking up of large clusters into smaller ones occurs due to frustration caused by the long-range RKKY interactions within the spin system. The initial increase of $|\Delta\rho/\rho|$ mentioned above could be accounted for by this model. However, in AuFeCr alloys, frustration within the spin system could result from both the RKKY interaction and the competing ferromagnetic and antiferromagnetic interactions between the magnetic

atoms. The increase of Cr concentration in the reported alloys is expected to cause more frustration which would in turn lead to breaking of finite clusters. This cluster-breaking temperature would increase with increasing Cr concentration. This effect is observed in the present study since the temperature at which $|\Delta\rho/\rho|$ is a maximum varies from 9 K to 15 K as x increases from 0.15 to 0.4.

The temperature dependence of $|\Delta\rho/\rho|$ between T_m and T_g in $x = 0.15$ alloy is somewhat similar to that of the alloys with $x \leqslant 0.1$. In addition, the drop in $|\Delta\rho/\rho|$ is analagous to that observed in cluster glasses, as has been discussed above. Above T_m, $|\Delta\rho/\rho|$ decreases with temperature as shown in figure 3. These observations indicate that the $x = 0.15$ alloy lies near the boundary of RSG and cluster-glass phases.

For $x = 0.2$–0.4 alloys, $|\Delta\rho/\rho|$ is very weakly dependent on temperature between T_m and T_g at low fields. At the higher field (40 kOe), $|\Delta\rho/\rho|$ decreases monotonically with temperature as against a nearly temperature independent region in RSG alloys.

For the $x = 0.6$ alloy, $|\Delta\rho/\rho|$ is quite small as shown in figure 5, where the field dependence of $\Delta\rho/\rho$ is plotted at different temperatures. $|\Delta\rho/\rho|$ has a maximum value of 0.8% in 40 kOe in this alloy as against more than 4% in the $x = 0.4$ alloy. The inset in figure 5 shows the temperature dependence of $|\Delta\rho/\rho|$ in 40 kOe which exhibits a peak around 25 K. Our recent measurement of DC magnetic susceptibility on a SQUID magnetometer shows a field-dependent double magnetic transition in this alloy. The higher-temperature transition (70 K) is field independent while the other transition moves to lower temperatures with increasing fields. For example, at 15 kOe, the lower transition occurs at 40 K. It is possible that the observed peak in $|\Delta\rho/\rho|$ in 40 kOe would correspond to the lower magnetic transition. At present, we are not very clear about the nature of the lower magnetic transition and it does not appear to be a simple spin-glass state as claimed by Nakai *et al* [6].

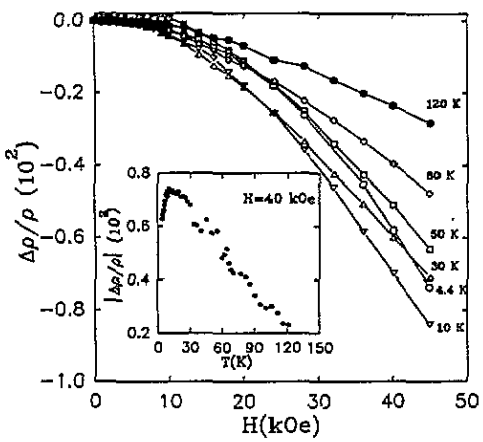

Figure 5. The magnetic field (H) dependence of $\Delta\rho/\rho$ at different temperatures for the $x = 0.6$ alloy. Inset shows the temperature dependence of $|\Delta\rho/\rho|$ at 40 kOe.

Figure 6 shows the magnetoresistance as a function of Cr concentration x at 10 K in fields of 5 and 40 kOe. At 5 kOe, $|\Delta\rho/\rho|$ drops sharply as x varies from 0 to 0.05 and then decreases slowly up to $x = 0.15$ followed by a rapid drop until $x = 0.4$. The initial sharp drop of $|\Delta\rho/\rho|$ indicates that the presence of a small amount of Cr strongly reduces the canting of spins. The latter drop of $|\Delta\rho/\rho|$ indicates that there is a breaking up of long-range ferromagnetic order into a cluster glass state as a result of strong competing ferromagnetic and antiferromagnetic exchange interactions. The magnetoresistance of these

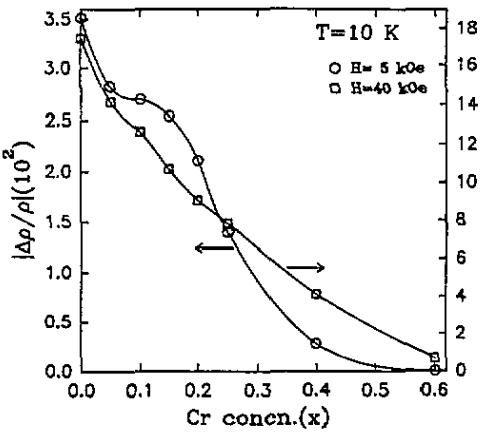

Figure 6. The variation of $|\Delta\rho/\rho|$ with Cr concentration (x) at 10 K and in fields of 5 and 40 kOe. Lines joining the points serve as guides to the eye.

alloys at 40 kOe monotonically decreases with increasing x since large external fields inhibit the breaking up of the clusters due to increased spin–spin interaction strength.

3.2. Field dependence of magnetoresistance

The field dependence of magnetoresistance ($\Delta\rho/\rho$) in the field range 3–45 kOe was fitted using a least squares fit program, to a functional form

$$\Delta\rho/\rho = aH^n \tag{1}$$

where a is a proportionality constant. The uncertainty in the determination of the fitted parameters is of the order of 1%. The values of n at different temperatures for various alloys are given in table 1. The following behaviour has been observed for alloys in the different concentration regimes:

Table 1. Results of fits to $\Delta\rho/\rho = aH^n$. x is the Cr concentration in $Au_{82}(Fe_{1-x}Cr_x)_{18}$. T_1 indicates the range of fit. Freezing temperature T_g and Curie temperature T_c (in the case of a double transition) are from [5].

x	T_g, T_c (K)	n (±0.02)	T_1 (K)
0	65, 151	0.74–0.82	4.4–120
		0.68	140–160
0.05	72, 127	0.67	4.4–200
0.1	69, 109	0.67	4.4–200
0.15	66.6	0.67–0.71	10–100
0.2	73.3	0.67–0.70	10–100
0.25	56.6	0.73–0.77	10–80
0.4	57.5	0.97–1.00	10–80
0.6	70.0	1.90–2.10	10–120

(i) In RSG alloys ($x = 0.05$ and 0.1), n is found to be 0.67 or $\simeq \frac{2}{3}$ in the temperature range 4.2–200 K for fields between 3 and 45 kOe. This $H^{2/3}$ dependence of $\Delta\rho/\rho$ for the $x = 0.05$ alloy at a few temperatures is shown in figure 7. A similar value of n has been reported for an $Fe_{57}Ni_{23}Cr_{20}$ alloy exhibiting RSG behaviour in the field range 2–16 kOe

[28]. It is interesting to note that in all these alloys the range of $H^{2/3}$ dependence of $\Delta\rho/\rho$ extends beyond the Curie temperature. In our study on $Au_{82}Fe_{18}$ re-entrant spin glass alloy, n is found to be 0.67 only between 140 to 160 K which is close to T_c (150.7 K). Below 140 K, n increases with decreasing temperature to a value of 0.80 below T_g. Our earlier study on the same alloy showed that $n = 0.85$ below T_g in low fields (below 5 kOe) [12]. The functional field dependence of magnetoresistance has not been checked in most of the studies carried out on re-entrant spin glasses. The present study as well as that carried out on $Fe_{57}Ni_{23}Cr_{20}$ [28] show $H^{2/3}$ dependence of $\Delta\rho/\rho$ to be a characteristic feature of re-entrant spin glasses in fields above technical saturation. Senoussi has derived a linear dependence of $\Delta\rho/\rho$ on H based on the quadratic dependence of $\Delta\rho/\rho$ on magnetization [8]. This is not truly revealed by his experimental data on $Ni_{79}Mn_{21}$ where one clearly sees that n is less than unity [29]. According to Balberg [20], the field dependence of magnetoresistance at high fields is given by

$$\Delta\rho \propto -H^{(1-\alpha)/\beta\delta} \tag{2}$$

provided $(\mu H/kT) \gg |1 - T/T_c|$ where μ is the magnetic moment of the ion, α is the specific heat exponent and β and δ are the critical exponents for the spontaneous magnetization and critical isotherm respectively. Studies on **Au**Fe RSG alloys near the critical temperature examined by magnetization [26] and non-linear susceptibility techniques [30] show that the critical exponents obtained are in close agreement with those predicted for 3D Heisenberg ferromagnets. If we substitute the theoretical values of critical exponents for the 3D Heisenberg system [31] ($\alpha = -0.115$, $\beta = 0.3645$ and $\delta = 4.80$), we obtain a value of $n(= (1-\alpha)/\beta\delta)$ equal to 0.64 which is very close to the value found experimentally in **Au**FeCr RSG alloys. According to the above condition, equation (2) is valid either at temperatures very close to T_c (within 0.5 K) or at very high fields ($\simeq$ 100 kOe). However in the present study, we find that equation (2) is valid in a much wider temperature range. The field dependence of $\Delta\rho$ given by equation (2) apparently arises due to existence of small q fluctuations. It is probable that in inhomogeneous ferromagnetic systems, such as **Au**Fe, the small q fluctuations may exist well below T_c leading to a similar field dependence of $\Delta\rho$. In **Au**FeCr alloys, the frustration caused as a result of competing exchange interactions could lead to the presence of small q fluctuations in a much wider temperature range which is probably the reason for observing a $H^{2/3}$ dependence of $\Delta\rho/\rho$ down to 4.4 K.

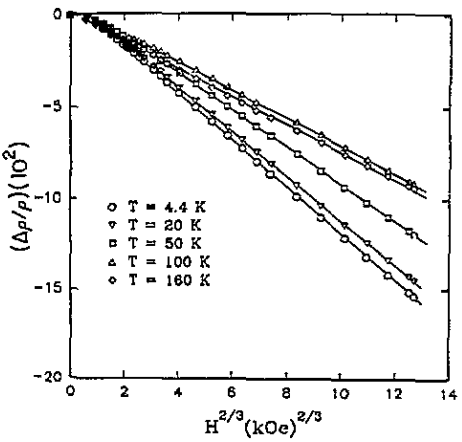

Figure 7. The $H^{2/3}$ dependence of $\Delta\rho/\rho$ for the $x = 0.05$ alloy in the field range 3–45 kOe, at temperatures indicated in the plot.

(ii) In cluster glass alloys, below T_g, n is found to be around 0.7 for alloys with $0.15 \leqslant x \leqslant 0.25$ and $n \simeq 1$ for the $x = 0.4$ alloy. The temperature dependence of susceptibility and magnetoresistance (as described above) of these alloys show a cluster-glass-like behaviour. Hence the magnetoresistance will have contributions due to both inter-cluster interaction which is spin-glass type and intra-cluster interaction which is ferromagnetic. The spin-glass type of interaction leads to a nearly quadratic field ($\simeq H^2$) dependence of $\Delta\rho/\rho$ and the ferromagnetic interaction leads to $H^{2/3}$ dependence as found above. One therefore expects a value of n between $\frac{2}{3}$ and two for the cluster-glass alloys depending upon the strength of the two interactions. Therefore the observed increase of n with x indicates that the spin-glass type of interaction becomes important ultimately dominating at higher x. For $x = 0.6$, n is found to be around two which is close to the value found for canonical spin glasses [7]. The electrical resistivity study of this alloy as a function of temperature exhibits a maximum, characteristic of spin glasses [32]. However the DC magnetization study shows that the alloy is not a simple spin glass as discussed in the preceding section.

4. Conclusion

In the present study we have reported detailed magnetoresistance measurements on AuFeCr alloys as a function of temperature and field. The study clearly shows that the system evolves from a re-entrant spin-glass phase on the Fe-rich side to a spin-glass phase on the Cr-rich side. The presence of Cr in these alloys causes weakening of ferromagnetic exchange interaction as a result of competing ferromagnetic and antiferromagnetic interactions. This leads to breaking up of ferromagnetic clusters. The temperature dependence of magnetoresistance shows certain features characteristic of re-entrant spin glasses as well as of cluster glasses below the transition temperatures. The re-entrant spin-glass alloys do not show any anisotropy in resistivity in either ferromagnetic or re-entrant spin-glass phases. The field dependence of magnetoresistance in Cr-containing re-entrant spin glasses appears to have a unique power-law dependence below the Curie temperature.

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