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The paramagnetic–ferromagnetic transition in re-entrant spin-glasses—a magnetoresistance study in ultra-low magnetic fields

R D Barnard
Department of Pure and Applied Physics, University of Salford, Salford M5 4WT, UK

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Abstract. The longitudinal magnetoresistance has been measured in the spin-glass and re-entrant spin-glass states of Au–14at.%Fe and amorphous $(Fe_{1-x}Ni_x)_{77}Si_{10}B_{13}$ alloys by a new technique which involves applied fields of less than two gauss. The measurements have concentrated on the paramagnetic–ferromagnetic transition where in both crystalline and amorphous systems a curious negative magnetoresistance has been found over a small temperature range, which rapidly changes sign as the ferromagnetic state becomes established with decrease of temperature. From measurements of the field dependence of the magnetoresistance near the Curie temperature, the negative contribution, as well as the positive contribution, is found to continue to develop into the ferromagnetic state. The model used by Beck in 1985, modified with the addition of a spin-glass type background matrix, is proposed from this study for the ferromagnetic state of these alloys.

1. Introduction

The re-entrant spin-glasses are characterised by an unusual magnetic transition from ferromagnetism to a spin-glass state as the temperature is lowered, usually at temperatures below 100 K. Whilst this type of alloy shows a typical Curie transition from paramagnetic to ferromagnetic behaviour as the temperature is reduced, the ferromagnetic state is unstable against further lowering of temperature and re-enters into a spin-glass state in which there is a drop in the AC susceptibility and other magnetic effects occur such as the onset of magnetic viscosity and history dependent effects. The nature of this transition has been the subject of much discussion over the years (Gabay and Toulouse 1981, Beck 1985) but much less attention has been paid to the transition from paramagnetism to ferromagnetism (PM–FM) which takes place at higher temperatures, it being generally assumed that this is of the Curie type typical of conventional ferromagnetic materials. However, as a result of some magnetoresistance (MR) measurements on both crystalline and amorphous re-entrant spin-glasses, we believe that this transition is rather more complicated and contains features which occur in conventional spin-glasses prior to the freezing process as the temperature is reduced.

In a recent paper on the magneto-transport properties of ordinary spin-glasses (Barnard and Ul-Haq 1988), we presented evidence for the existence of a second magnetic transition at temperatures in the region of $5T_f$ where T_f is the freezing temperature. This transition takes place when individual moments first couple under the

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RKKY interaction; these entities then behave collectively with respect to magnetic fields, their cluster size increasing as the temperature is lowered towards T_f where the blocking of the clusters and the establishment of the spin-glass state takes place. This lower temperature paramagnetic region between T_f and $5T_f$ is characterised by subtle changes in the low field Hall effect and deviations from the Curie–Weiss law. In this paper we present evidence that a similar phenomenon takes place prior to the establishment of the ferromagnetic phase (or, according to Beck (1985) of very large ferromagnetic clusters) but its temperature range is generally limited to only a few degrees. The evidence is based on MR measurements performed using a new technique (which will be described elsewhere) which has enabled us to measure the MR coefficient in re-entrant and conventional spin-glasses in fields comparable with that of the Earth, or comparable with those used in AC susceptibility determinations. This is an extremely important aspect of measuring the intrinsic properties of spin-glasses and the re-entrant variety for, as we have expounded in a number of papers (Barnard 1984, Barnard and Ul-Haq 1986, 1988), the presence of large magnetic fields severely disrupts the rather weak magnetic coupling which exists in these materials. In this paper we describe measurements performed on a sample of crystalline Au–14at.%Fe which, when quenched from 950 °C possesses predominantly spin-glass characteristics, but when aged at room temperature for a few days then possesses cluster-glass/re-entrant spin-glass characteristics (Sarkissian 1981) due to the atomic short range order which sets in. We also report measurements on some amorphous $(Fe_{1-x}Ni_x)_{77}Si_{10}B_{13}$ alloys, kindly supplied by Dr Miyazaki of Tohoku University, with values of x ranging from 0.95 to 0.80 which traverses pure spin-glass to re-entrant spin-glass behaviour as x decreases. In both crystalline and amorphous systems similar behaviour occurs in the MR with both the normal and re-entrant spin-glasses.

2. Experimental details

The sample of Au–14at.%Fe was prepared in an RF furnace from accurately weighed constituents and annealed at 950 °C for several days to ensure homogenisation before being quenched into water. After rolling and spark machining, a narrow strip 70 mm long, 2 mm wide and approximately 0.25 mm thick was produced which was further annealed at 950 °C for half an hour before quenching into water. This was followed immediately by installation in the MR apparatus for measurement at low temperatures. Further measurements were taken some 12 and 92 days later after the sample had been aged at room temperature. The preparation of the amorphous samples has been described by Miyazaki *et al* (1988); this paper also contains the temperature variation of the AC susceptibility showing clearly the re-entrant nature of the samples with $x < 0.93$ and normal spin-glass behaviour for $x > 0.93$.

The MR was measured using a new technique involving a pulsed longitudinal field of only 2 Gauss magnitude at typically 38 Hz, the residual vertical component of the Earth's field being annulled by a separate solenoid carrying a direct current. With the sample and field configuration already described, the demagnetisation factor D was negligible and measurements could be performed between 4.2 and 300 K.

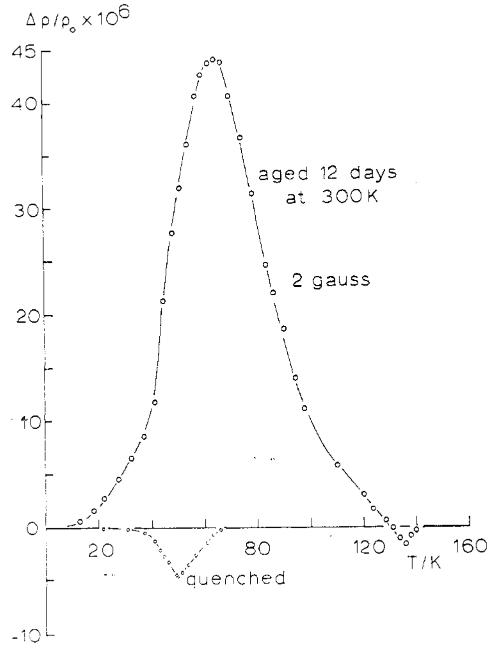

Figure 1. The magnetoresistance of Au–14at.%Fe in (*a*) the quenched state and (*b*) when aged for 12 days at 300 K.

3. Results

The AC susceptibility of the amorphous alloys studied in this work have been published by Hisatake *et al* (1986) and Miyazaki *et al* (1988). We have remeasured some of the re-entrant samples near the PM–FM transition in order to ascertain precise values of transition temperatures for comparison wiith the MR results. The general variations of the in-phase susceptibility obtained were similar to those given by Miyazaki *et al* except that in our case smaller and more rounded peaks were observed probably due to the larger magnetic field (~1 Gauss) used in our determinations. The AC susceptibility of Au–14at.%Fe in the quenched state was rather typical of that of a conventional spin-glass, peaking at a 'freezing temperature' of 50 K. On aging for 12 days a large increase in susceptibility occurred, the peak shifting upwards to 65 K and becoming more rounded.

In figure 1 is shown the MR results obtained on Au–14at.%Fe in both the quenched and aged conditions measured in a peak field of 2 Gauss with the vertical component of the Earth's field annulled. In the quenched state the MR was entirely negative, peaking, like the susceptibility, at 50 K. This existence of negative MR appears to be a rather general feature of spin-glasses for it has been reported in Au–8at.%Mn and Cu–4.6at.%Mn also (Barnard 1984). However, on aging for 12 days at 300 K, a complete sign change occurred coupled with a considerable increase in magnitude but with a curious negative MR re-establishing itself beyond 130 K before decaying to zero at higher temperatures.

In view of the unusual sign change in the region of 130 K, a field dependence study was undertaken over the range 0–30 Gauss. Measurements were taken after the sample had been further aged at room temperature for 80 days by which time the cross-over temperature had increased slightly from 130 K to 138.5 K due to further short range order setting in. The temperature 137.9 K was chosen for the field study so that initially

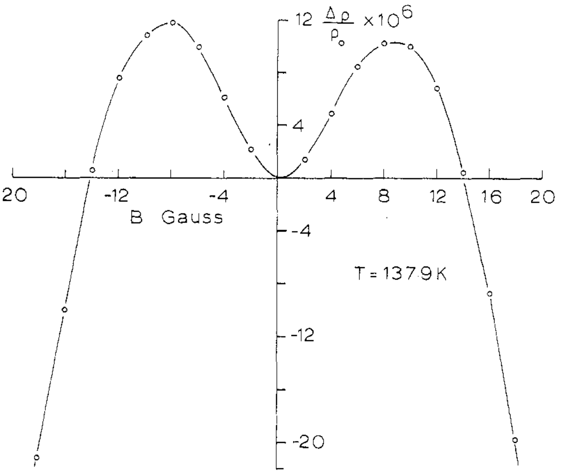

Figure 2. The field dependence of Au–14at.%Fe (aged 92 days at 300 K) at 137.9 K for both positive and negative values of the applied field.

the MR was positive, but as is clear from figure 2, further increasing the field resulted in a maximum followed by strong negative MR being established. It should be pointed out that this unusual type of variation of MR can be observed in conventionally behaved materials in which a finite magnetisation exists prior to the application of the MR field. The situation is clarified in figure 3, where a conventional $\Delta\rho/\rho_0 \propto M^2$ variation is shown but with the initial starting point at, say, the point A. The application of the MR field, (B_{MR}) in the direction which decreases the remanent magnetisation, results in a measured $\Delta\rho/\rho_0$ which initially *increases* and then turns over to become negative. This apparent anomalous variation may be easily recognised by taking measurements with the MR field reversed which will then only give negative values of $\Delta\rho/\rho_0$. It is clear that in the case of Au–14at.%Fe (figure 2), such a situation does *not* obtain, as exactly the same variation occurs with field reversal and thus this anomalous behaviour must be an intrinsic property of this material.

In figures 4 and 5 are shown the MR versus temperature variations for three of the amorphous FeNi alloys. For $x = 0.95$, which is a typical conventional spin-glass, the MR is entirely negative and thus conforms to the behaviour discussed above of other spin-glasses, the MR peaking at the freezing temperature. Here the MR was so small that an MR field of 16 Gauss was necessary for the measurements. In the case of $x = 0.92$, changeover to re-entrant behaviour is apparent; the MR is now predominently positive with a temperature variation superficially similar to that of the AC susceptibility. However, as with Au–14at.%Fe, a just discernible sign change of MR occurs at 2 G at the tail end of the decay of the AC susceptibility which is revealed more strongly by the field dependence study shown in figure 6 where the variations are similar to that of Au–14at.%Fe shown in figure 2. However, this phenomenon is displayed even more prominently in the case of $x = 0.875$ (figure 5) where we also show details of the AC susceptibility, measured in 1 Gauss at 161 Hz and the MR in the vicinity of the Curie temperature. In figure 6 is shown the evolution of the field dependence of the MR as a function of T across the positive to negative transition in the case of $x = 0.92$. It is clear from figure 6 that this transition is field dependent; the higher the field the lower the

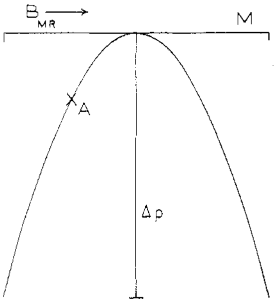

Figure 3. A schematic magnetoresistance versus magnetisation variation for a conventional ferromagnet in which the starting point is at A; a positive and then a negative magnetoresistance is observed for an applied field in the direction indicated.

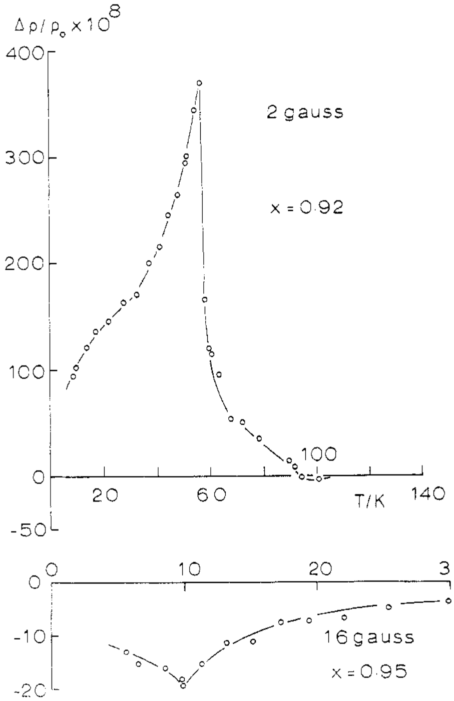

Figure 4. Magnetoresistance versus temperature for amorphous $(Fe_{1-x}Ni_x)_{77}Si_{10}B_{13}$ for the values of x indicated.

temperature of the transition. Below 95 K and at very low fields, a clear positive MR is first observed with what appears to be a B^n variation with $n > 1$, but at higher fields, an increasing negative contribution becomes established which ultimately drives the MR negative. Only above 95 K is the MR wholly negative with an apparent B^m variation with again $m > 1$.

4. Discussion

The MR results presented in this paper under very low field conditions reveal that as the temperature is lowered in the paramagnetic region, the re-entrant alloys (whether crystalline or amorphous) develop a negative MR over a small temperature range before this changes sign and the ferromagnetic state becomes established. In view of the fact that negative MR is characteristic of all the ordinary spin-glasses we have studied including those presented in this paper, it is tempting to conclude that the magnetic coupling which first becomes established in the re-entrant alloys must be similar to that which first marks the development of the conditions necessary for an ordinary spin-glass to occur. However, for the re-entrant spin-glasses, out of this initial coupled state the so-called ferromagnetic state develops leading to a sign change in MR. But it is also clear that the spin-glass (SG) type coupling continues to develop even when the alloys have entered

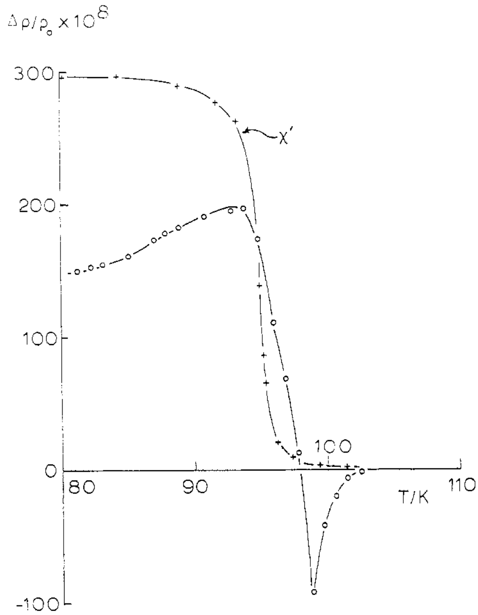

Figure 5. Magnetoresistance versus temperature in the vicinity of the Curie temperature for $(Fe_{1-x}Ni_x)_{77}Si_{10}B_{13}$ $x = 0.875$ in a field of 2 G. The corresponding AC susceptibility (arbitrary units) is also shown.

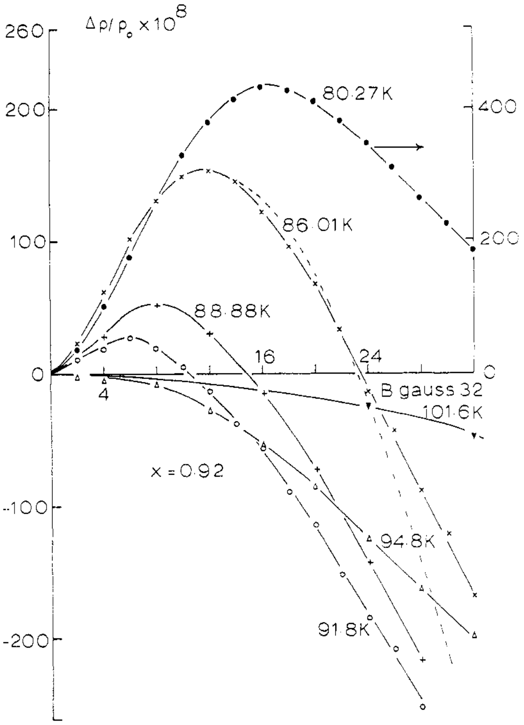

Figure 6. The field variation of $(Fe_{1-x}Ni_x)_{77}Si_{10}B_{13}$ for $x = 0.92$ for different temperatures near the Curie temperature.

into the ferromagnetic state since negative MR is still present as our measurements show in the higher applied fields. It thus appears that the ferromagnetic state is characterised, at least initially, by a mixed phase, probably with ferromagnetic clusters growing out of the precursor of the ordinary spin-glass state which itself is growing as more moments couple with reduction of temperature. Indeed, judging by MR studies at higher fields by other authors (e.g. NiMn at 4.2 K, Senoussi and Oner, 1984) it is highly likely that this mixed system still obtains at very much lower temperatures within the ferromagnetic state although by then growth of ferromagnetic clusters and the SG coupled state is likely to have ceased. The model which is suggested by these measurements is thus one of nearest neighbour moments coupled ferromagnetically and these constitute the clusters, and a weaker coupled system including frustration constituting the precursor of the SG-like state. Generally, the two states should not be regarded as independent as the cluster growth occurs at the expense of the SG precursor which itself grows with reduction of temperature. However cessation of growth at some temperature between the PM–FM and FM–SG transitions should occur; the two phases then act to some extent independent of one another.

An extension of these ideas would suggest that the re-entrant behaviour of these alloys into a SG state could be closely associated with the continued existence of the background state. If this is so, it is possible to qualitatively account for the characteristic

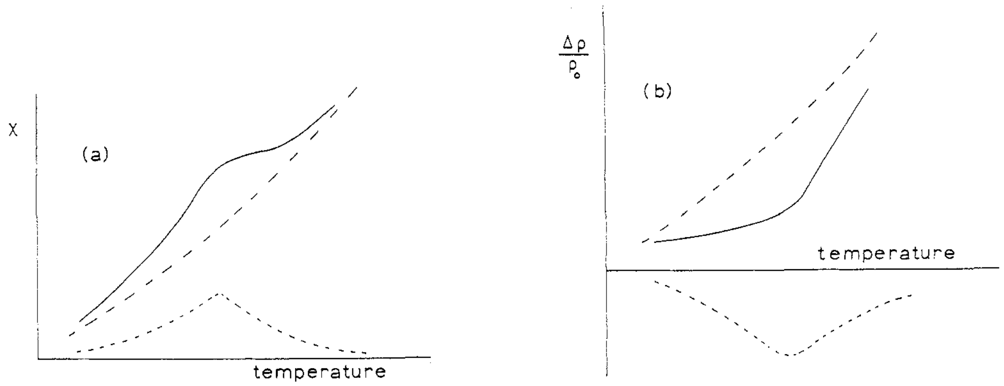

Figure 7. Schematic temperature variations for (*a*) the AC susceptibility and (*b*) the magnetoresistance (full lines) of a re-entrant spin-glass when two contributions exist, one from magnetic clusters (long dashed lines) and another from a spin-glass matrix (short dashed lines).

knee which occurs in the AC susceptibility of re-entrant spin-glasses at the FM–SG transition. In figure 7(*a*) we show the variation of the FM component as a monotonically increasing function together with an SG component exhibiting the characteristic cusp at the freezing temperature. The sum of the two contributions clearly reveals the knee due to the decreasing susceptibility of the SG component beyond its freezing temperature. A similar explanation for the inflection in the AC susceptibility curve has been given by Takahashi *et al* (1983) but our SG curve they regarded as a cluster curve and our cluster curve they regarded as a ferromagnetic matrix curve. In the case of the MR where the FM and SG components are of opposite signs, the net variation of the MR will contain somewhat different features. Figure 7(*b*) shows the type of variation which could occur but it is now clear that the freezing temperature is marked by a reasonably sharp increase in MR although its precise details will depend on the form and relative magnitudes of the two components. It is evident that the MR variation shown in figure 4 for $x = 0.92$ is not incompatible with this proposal in which a freezing temperature of about 30 K is suggested. However, a complete investigation of the FM—SG transition will be reported in a later paper.

In analysing the field dependence of FeNi ($x = 0.92$) shown in figure 6 we have examined an equation of the form

$$\Delta\rho/\rho_0 = XB^n - YB^m \qquad (1)$$

where X and Y are constants at a given temperature and B is the magnetic induction. Whilst reasonable fits to the data can be obtained with such an equation, strong deviations are in evidence at high fields, which suggests that simple power law variations are only applicable at very low fields. A typical fit in the case of $T = 86.01$ K is also shown in figure 6 by the dashed curve in which the m and n values are 2.0 and 1.2 respectively, although these values are by no means unique as the values 1.8 and 1.3 respectively can give a similar variation at low fields. However, despite the shortcomings of equation (1), the existence of two dominant contributions, one positive and the other negative would appear to be necessary to give the form of variation shown in figures 2 and 6. In addition, it is also clear that the parameters X and Y (or their equivalents in a more accurate equation) must increase as the temperature is lowered into the FM state thereby confirming the continued growth of the ordinary type of SG state. It will also be evident, in the light of the MR results presented here, that measurements of MR using high fields on these re-entrant spin-glasses will, in general, be difficult to interpret since erroneous transition temperatures will be indicated and the existence of, and the relative weighting of the two contributions will not be apparent. Clearly, the intrinsic behaviour of these materials is only revealed from measurements in vanishing magnetic fields in the vicinity of the Curie temperature.

Finally, the interpretation given here on the magnetic structure of re-entrant spin-glasses in the FM state gives, we believe, some support for the magnetic cluster model proposed by Beck (1985) and Rakers and Beck (1988) for Au–Fe in which no long range ferromagnetism is involved but rather superparamagnetism with random interaction between the cluster moments. Beck (1985) has given a comprehensive review of various measurements which support this viewpoint. The continued existence of the precursor to the ordinary SG phase in the so-called FM state suggests to us also that true ferromagnetism involving infinite clusters is precluded although the SG state is a new feature which we would add to his model. It differs also from the mixed phase model of Gabay and Toulouse (1981) in which transverse and longitudinal components of the moments are separated into essentially SG and FM characters, the former freezing in random directions. Furthermore our results also indicate that the crystal structure is not significant either, in view of the similarities in the MR of both crystalline and amorphous systems.

Acknowledgments

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