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MAGNETIC PROPERTIES
OF AuFe ALLOYS BELOW THE PERCOLATION LIMIT

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It is well known that solute spins even in very dilute Au-Fe alloys (\( \sim 0.1 \) at. \% Fe) [2] freeze magnetically at low temperatures in almost random orientations giving rise to a complex magnetic system which has been variously described as a Spin Glass [3] or a Mictomagnet [4]. The former term has been suggested to be rather appropriate [5], because of the similarity of some of the properties of such systems with real glasses, and may in general be applied to any binary solid solution alloy, consisting of moment bearing atoms dissolved in a non-magnetic metallic matrix, which is neither simple ferromagnetic nor antiferromagnetic but which exhibits properties which are the result of a mixture of both such types of interactions. In magnetisation measurements the spin glass behaviour manifests itself as a broad maximum in the static "susceptibility" and the observation of isothermal remanance and field cooling effects below the temperature of the maximum.

It is commonly believed that the spin glass magnetism found in the lower concentration Au-Fe alloys gives way to long range ferromagnetism when the Fe concentration is increased above about 12 at. \% [1]. The first observations of the transition to ferromagnetism in Au-Fe were made by Pan et al. [6], who have claimed that the alloys with more than 10 at. \% Fe were ferromagnetic, and were followed later by the work of Crangle and Scott [7] who measured the magnetisation of several Au-Fe alloys as a function of magnetic field and temperature and analysed their data with the help of Arrott plots of \( \frac{H}{M} \) vs. \( M^2 \) to obtain the ferromagnetic Curie temperatures \( T_c \). They found that an alloy containing 11.1 at. \% Fe was not ferromagnetic according to this criterion but another having 14.9 at. \% Fe had a Curie temperature of 110 K.

De Mayo [8] has reported magnetisation mea-
ments on Au-Fe alloys with 5.10 and 13.8 at. % Fe and shows that the magnetisation vs. temperature behaviour of the alloys in a constant field of 12.6 kOe is strongly dependent on cold working and annealing heat treatments given to the samples as also are the ordering temperatures obtained from Arrott plots for the two higher concentration samples. The author also observes strong super-paramagnetic behaviour which is, again, affected by the metallurgical treatment of the samples and concludes that the properties of the alloys are due to clusters dispersed in what the author calls the spin glass matrix. However, the strong dependence of the magnetic properties on metallurgical treatment is a general feature of most binary alloys. It has been shown in the case of concentrated Cu-Ni alloys, where the tendency towards atomic clustering is also pronounced, that high temperature annealing heat treatment can produce good random substitutional alloys whose properties are accountable in terms of purely statistical fluctuations of concentration [9]. As in all such systems the fact that non-random clustering can be induced does not mean that it cannot be avoided. We shall adopt this view point for the Au-Fe system. The present results show that the properties of the alloys, including the occurrence of giant superparamagnetic behaviour, have a simple physical interpretation which does not depend upon atomic clustering effects.

The transition to ferromagnetism claimed by the magnetisation results mentioned earlier is, however, not corroborated by the Mössbauer effect measurements. Borg [10] has found that the low temperature Mössbauer spectra of Au-Fe alloys with up to 18 at. % Fe are essentially similar and indicate spins frozen in random orientations which cannot be aligned parallel as a whole even with the application of magnetic fields of strengths up to 50 kOe. Borg and Kitchens [11] have measured magnetic properties of Au-Fe alloys with concentrations up to 12.5 at. % Fe. Although no bulk magnetisation measurements beyond this concentration are reported the authors suggest that «the magnetic behaviour of Au-Fe in the concentration region

$$0.05 \leq rFe \leq 17\,\%$$

conforms to the standard definition of micromagnetism», which conclusion is based mainly (especially for concentrations above 12.5 at. %) on their earlier Mössbauer results [10].

Another very important discrepancy between the magnetisation results and the Mössbauer effect measurements which has hitherto remained unexplained is the vast differences in the ordering temperatures obtained by the two types of measurements for Au-Fe alloys containing more than 12 at. % Fe (the concentration where ferromagnetism is thought to set in). In contrast, the ordering temperatures deduced from the resistivity results (the temperatures of the maxima in dp/dT) agree with those obtained from bulk magnetisation measurements over the whole concentration range [14]. The results are illustrated in figure 1 where we reproduce a diagram from a paper by Cannella and Mydosh [1].

![Figure 1](image-url)

**Fig. 1.** Magnetic ordering temperatures $T_o$ vs. concentration $C$ for Au-Fe alloys as compiled by Cannella and Mydosh [1]. The triangles are from the magnetisation data of Crangle and Scott [7]; closed circles from the Mössbauer data [12 and 13]; crosses represent the maxima in the susceptibility data of Lutes and Schmit [19]; and the open circles are the temperatures of the maxima in dp/dT from Mydosh et al. [14].

In the following we present results of magnetisation measurements in low fields on a Au 15 at. % alloy which enable us to resolve this discrepancy and to show that both ordering temperatures have interesting physical significance. The present results also demonstrate unambiguously for the first time through bulk magnetisation measurements, the micromagnetic or spin glass type of magnetic behaviour of a Au-Fe alloy in the concentration range above 12 at. %

The alloy was prepared by argon arc melting appropriate amounts of high purity Au (5N) and Fe (4N8) metals on a water cooled copper hearth. The melting was repeated a total of four times after turning over the alloy each time. The alloy was subsequently cold worked, homogenized for 72 hours at 900 °C, quenched in water and kept under liquid nitrogen until used. Careful metallographic examination of a portion of the sample showed no traces of any second phase precipitation. The equilibrium solid solubility of Fe in Au is more than 50 at. % at 900 °C [15], so that at a concentration as low as 15 at. % Fe the above treatment should certainly yield a good random solid solution.

In figure 2 we present the magnetisation results for the Au-15 at. % Fe alloy measured in two different
One very important respect in which the above magnetisation curves differ from those of a common spin glass is the rapid rise around 110 K to very large magnetisations as shown by the low field measurements curves a and b. Such behaviour, by itself, has been interpreted as indicating the onset of ferromagnetism [1]. Isothermal magnetisation measurements at 78 K, however, show no detectable remanence after cycling the alloy in a field of 8 kOe. For a polycrystalline specimen this clearly implies the absence of long range ferromagnetic order. The magnetic isotherms between 50 and 110 K reveal the existence of giant super-paramagnetic moments consisting of several thousand Fe atoms [17]. We suggest that these are formed by the (predominantly) ferromagnetic freezing of Fe spins (whether single or in statistical near neighbour clusters) on a localized microscopic scale which is marked by the sharp increase in the magnetisation around 110 K. The giant clusters so formed are, however, not large enough on a macroscopic scale to behave as ferromagnetic entities [18] and will be free to rotate until they freeze relative to each other (with random orientations) over macroscopic dimensions, at some lower temperature. If the thermal relaxation rate of the clusters is fast compared with the long time life of the Mössbauer transition ($\sim 10^{-8}$ s) the latter measurements would not detect the short range order within them. Hence it is not until the spin glass temperature of 50 K is reached and the clusters freeze that the Mössbauer spectrum of Fe appears.

As pointed out above, the magnetisation of the alloy around 110 K and below is strongly field dependent and tends to saturation in fields above 20 Oe, hence an analysis of the magnetisation measured in moderate fields in terms of the Arrott plots would yield a ferromagnetic ordering temperature near to where the cluster formation occurs and is the explanation for the results of Crangle and Scott [7].

It is also not difficult to understand why the maximum in the first derivative of the resistivity of the alloy, suggesting the onset of magnetic freezing of spins, should coincide with the temperature of the formation of the clusters, for as far as the conduction electrons are concerned the latter event is the most significant one, if the size of the clusters greatly exceeds the mean free path of the electrons. This is certainly the case for the present alloy where the clusters span a region of about $10^5$ atoms [17], and hence a cube of linear dimension around 50 lattice spacings, whereas the mean free path of the conduction electrons is only of the order of a few lattice spacings (since $\rho \sim 150 \, \mu\Omega \cdot \text{cm}$).

We are now in a position to understand a little more clearly the magnetic behaviour of the Au-Fe system. It would appear that near neighbour interactions between Fe atoms in Au are ferromagnetic (as in metallic Fe) but those between the next near neighbour and beyond are of the oscillatory. R. K. K. Y. type.
In such a situation long range ferromagnetism is established when the concentration of Fe atoms in the alloy is that given by the percolation limit, when the probability of having an infinite chain of near neighbour Fe atoms becomes finite. For the f. c. c. lattice this occurs at a concentration of 19.5 at. % [20]. It would appear from the data in figure 1 that all three types of measurements — Mössbauer effect, magnetisation, and electrical resistivity — would yield the same ordering temperature for an alloy with concentration around 20 at. % Fe, which would then also show long range ferromagnetism. Below this concentration, however, as in the present case of the Au-15 at. % Fe alloy, any magnetic order can have only the spin glass character.

We conclude by saying that the above results are the first bulk magnetisation measurements which demonstrate unambiguously the spin glass type of magnetic behaviour of Au-Fe alloys in the controversial concentration region above 12 at. % Fe. The results also enable us to recognise clearly, for the first time, that the two vastly different ordering temperatures obtained by previous magnetisation and resistivity results, on the one hand, and the Mössbauer effect measurements on the other, are both physically significant.

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References

[17] The isothermal magnetisation of the alloy at 78 K suggests an extreme form of superparamagnetic behaviour with an average effective magnetic moment per Fe atom of about 900 $\mu_B$ hence an average cluster size of about $10^7$ Fe atoms. Full results will be published shortly.