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Magnetic ordering in Fe-substituted AuCr alloys near percolation concentration

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Abstract. We report DC susceptibility measurements on $Au_{82}(Cr_{1-x}Fe_x)_{18}$ alloys ($0 \leqslant x \leqslant 0.4$) in the temperature range 5 K to 300 K. For $x = 0$, an inhomogeneous antiferromagnetic state is observed which also exhibits magnetic irreversibility in the field-cooled condition similar to that of spin glasses. The temperature dependence of the susceptibility in the $x = 0.2$ alloy shows a field-dependent transition while in the $x = 0.4$ alloy a double transition is observed, of which the lower part is field dependent. An attempt to fit the field dependence of this temperature to a power law yields an exponent of value 0.52 ± 0.05. The observed behaviour appears to be controlled by the distribution of magnetic clusters in the alloys.

1. Introduction

The magnetic phase diagram of the **Au**Cr system derived from various experimental studies shows spin-glass behaviour for a dilute concentration of Cr (below about 10 at.%) and antiferromagnetism for a higher Cr concentration (above 15 at.%) [1, 2]. The magnetic and neutron scattering measurements made by Nakai *et al* [1] have revealed that the system changes from a spin glass to an antiferromagnet (AF) with increasing Cr concentration via an intermediate phase at a concentration of 11–13 at.% where AF clusters are frozen in the spin-glass (SG) matrix. A detailed study of this system has been carried out in the concentration range below 10 at.% Cr where it shows a spin-glass behaviour [3]. A similar behaviour is observed in **Au**Fe below 10 at.% Fe. However, this system exhibits a double magnetic transition above 15 at.% Fe where one observes a paramagnetic to ferromagnetic transition followed by a re-entrant spin-glass (RSG) transition at a lower temperature. At the dilute limit both systems show magnetic interactions of the RKKY type, while at higher concentrations direct exchange interactions between the impurity spins dominate. This near-neighbour interaction is ferromagnetic (FM) in **Au**Fe while it is AF in **Au**Cr. Theoretical calculations [4] predict that the concentrations corresponding to the percolation limit for (FM) and (AF) FCC lattices are 15 and 45 at.% respectively. However, the **Au**Cr alloys are found to show long-range AF order above 15 at.% [1]. It therefore seems interesting to study the magnetic state of the ternary alloy system **Au**(CrFe) above the percolation limit where competition among the FM and AF exchange interactions exists along with RKKY-type interaction. We earlier reported an AC susceptibility study of Fe-rich $Au_{82}(Fe, Cr)_{18}$ alloys where evolution of cluster-glass/spin-glass behaviour has been observed with substitution of Cr [5]. Electrical resistivity studies have been reported earlier in the same alloy system [6]. In the present paper, we report DC susceptibility measurements on $Au_{82}(Cr_{1-x}Fe_x)_{18}$ for $0 \leqslant x \leqslant 0.4$ to study how the AF order is modified due to substitution of Fe. Earlier

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studies on **Au**Cr above 15 at.% exhibited long-range AF order. The present study shows an irreversibility in the magnetic behaviour in small fields below T_N. This behaviour, usually not observed in antiferromagnets, indicates presence of large AF clusters.

2. Experimental details

The samples of $Au_{82}(Cr_{1-x}Fe_x)_{18}$ were prepared by arc melting the 4N pure constituents followed by subjecting them to a homogenizing annealing at 800 °C for a week. They were cold rolled to form foils from which rectangular strips were cut out for measurement. These strips were annealed for 24 h at 850 °C and then quenched in water. They were stored in liquid nitrogen until the measurements were started. The samples were checked for homogeneity through x-ray diffraction using Cu $K\alpha$ radiation which confirmed them to be single-phase materials. DC magnetization measurements were performed on a SQUID magnetometer (MPMS Quantum Design). The DC susceptibility, χ_{DC}, is obtained from the division of the measured magnetization by the applied magnetic field.

3. Results and discussion

The variation of χ_{DC} in different applied fields for $Au_{82}(Cr_{1-x}Fe_x)_{18}$ with $x = 0$, 0.2 and 0.4 is shown in figures 1, 2 and 3. Except for in the $x = 0$ alloy case, the field-cooled (FC) data have been taken at the same field at which field cooling was done. The temperature variation of χ_{DC} for the $Au_{82}Cr_{18}$ alloy between 5 K and 300 K measured in a field of 1 kOe is shown in figure 1. It exhibits a peak at 206 K. The FC data were also obtained in an applied field of 1 kOe after cooling the sample in a 15 kOe field. These data too show a peak at 206 K but they deviate from the ZFC data at 230 K. The above irreversibility behaviour was also observed when the measurements were repeated in an applied field of 500 Oe with cooling carried out in the same field. Nakai *et al* [1] measured the DC susceptibility of a $Au_{82.5}Cr_{17.5}$ alloy under ZFC and FC conditions. Field cooling was carried out by them in a field of 8 kOe. They did not find any difference between the susceptibilities measured under the two conditions while our data do show a significant difference. The neutron scattering studies on **Au**Cr alloys showed a long-range AF order for a Cr concentration above 15 at.% [2]. In an AF, χ_{DC} shows a peak at a temperature T_N, and field cooling does not cause any significant difference. The difference in ZFC and FC conditions observed in the present data indicates that the system has a weak AF state which gets disrupted with the application of external fields. This could result in canting of the spins leading to a spin-glass-like state showing the observed irreversibility in magnetization. Further, it can result in the creation of ferromagnetic moments which could lead to the observed upturn of the field-cooled susceptibility data at low temperatures. This appearance of FM moments has been observed in magnetic torque measurements on FC **Au**Cr single crystal with Cr concentrations above 15 at.% [7]. A model has been suggested on the basis of computer simulation studies for a two-dimensional triangular lattice, in which the nearest-neighbour AF interaction inevitably induces frustration of spins similar to that in the case of three-dimensional FCC lattices [1]. It has been shown that for magnetic impurity concentrations of 40%, AF clusters embedded in a SG region pervade the entire lattice space. A similar situation may exist in **Au**Cr alloys with $x > 0.15$. The spins within these clusters may get canted in an external field giving rise to a weak FM moment. There could be a RKKY-type interaction between these clusters which may lead to a spin-glass-like freezing below T_N.

In a typical AF, the resistivity shows a minimum preceded by a maximum [8]. In

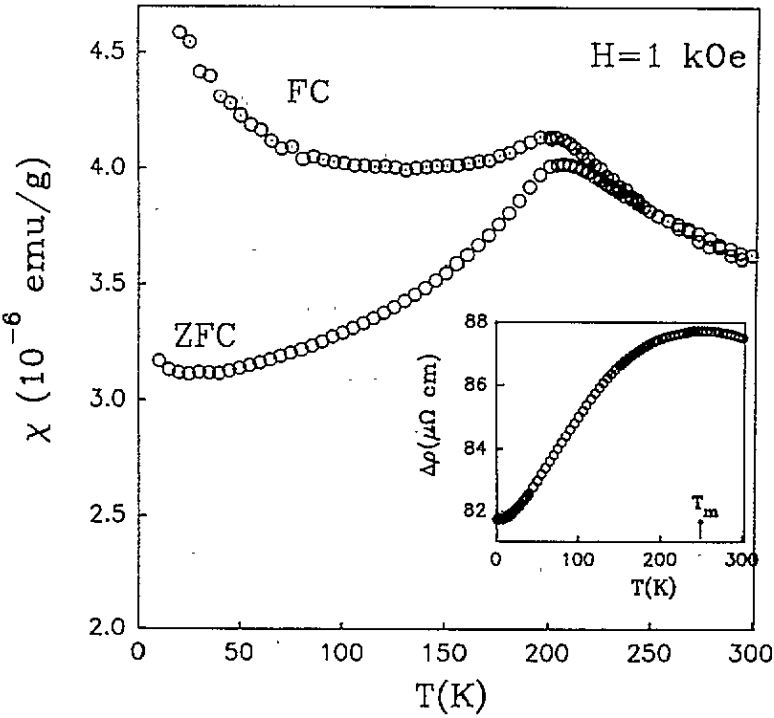

Figure 1. The variation of the DC susceptibility χ (emu g^{-1}) of $Au_{82}Cr_{18}$ alloy as a function of temperature (T) in an applied field of 1 kOe. The inset shows the temperature dependence of $\Delta\rho$ ($\mu\Omega$ cm) for the same alloy.

$Au_{82}Cr_{18}$, a broad maximum is seen at 247 K which is similar to that observed in spin glasses. This also indicates that the alloy is not a simple AF but rather a disordered AF consisting of AF clusters coupled through RKKY interaction.

For the $x = 0.2$ alloy, the ZFC data in a 1 kOe field show a broad peak at 78.9 K and the FC data deviate from the ZFC data around the same temperature. Nakai *et al* observe a peak in susceptibility at around 90 K in a similar alloy [4]. In our data, we find that this peak shifts to lower temperatures, T_i, with increasing external fields; it is found to be at 66 K in a field of 4 kOe and at 50 K in a field of 15 kOe. In a $Au_{75}(Cr_{1-x}Fe_x)_{25}$ alloy with $x = 0.2$, neutron diffraction studies show a sharp AF reflection while the susceptibility is different for the ZFC and FC conditions below the ordering temperature [4]. This indicates the presence of large AF clusters along with free spins which are frozen in a spin-glass-like manner. The observed shift of T_i to lower temperatures in our $x = 0.2$ alloy due to the magnetic field could result from the growing of AF clusters where Cr and Fe spins are antiferromagnetically coupled, thereby reducing the net cluster moment. This reduction in the net cluster moment weakens the RKKY-type inter-cluster interaction leading to a decrease in T_i.

Figure 3 shows the χ_{DC} variation in a field of 500 Oe for the $x = 0.4$ alloy in ZFC and FC conditions, where two distinct magnetic transitions are observed. The higher-temperature transition T_g occurs at 65 K. Nakai *et al* reported the same T_g though they have not specified the field in which the measurements were carried out [4]. Figure 4 shows χ_{DC} in higher applied fields where one observes that while the transtion at 65 K is unaffected by applied fields, the other transition shifts to lower temperatures, T_i with increasing fields. This variation of T_i with field is shown in the inset of figure 4. Here, the irreversibility occurs

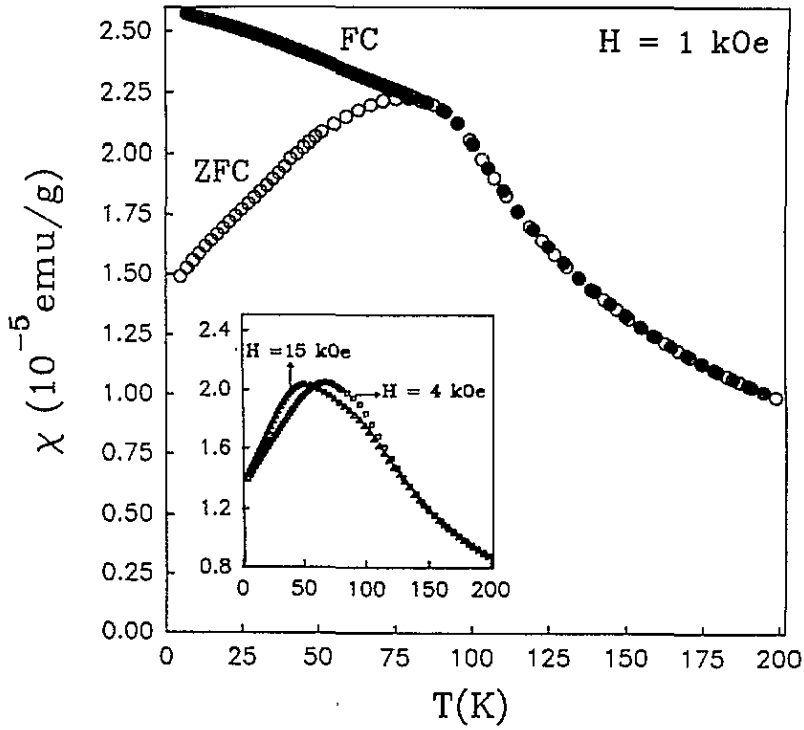

Figure 2. The temperature (T) dependence of χ (emu g^{-1}) for $Au_{82}(Cr_{0.8}Fe_{0.2})_{18}$ alloy in a field of 1 kOe under ZFC and FC conditions. The inset shows the temperature variation of χ in fields of 4 and 15 kOe.

below the lower transition. Also, the maximum susceptibility drops appreciably as the field increases. The neutron diffraction study on $Au_{75}(Cr_{1-x}Fe_x)_{25}$ single crystals for $x = 0.1$ to 0.3 shows that the magnetic moment contributing to the long-range order or to the formation of giant clusters is remarkably small for $x = 0.3$ [4]. The results show that the majority of the impurity spins remain in the spin-glass state and only a small fraction of spins form clusters which are embedded in the SG matrix. The situation in our $x = 0.4$ alloy in zero field will be similar since both these alloys are above the percolation limit. One therefore anticipates that a single peak in susceptibility would be observed at zero field, similar to that of conventional spin glasses. However, in such concentrated systems, the external field would tend to correlate the spins to form FM Fe–Fe clusters along with the AF Cr–Cr and Cr–Fe clusters. The presence of FM clusters is endorsed by the observation of a significant contribution to magnetoresistance in this alloy [9]. The field-independent transition would correspond to the freezing of the uncorrelated spins. The field dependence of the other transition seems to result from freezing of the clusters formed due to magnetic field. With increasing fields, there is a redistribution of cluster sizes and the net moments; the former results in a field-dependent freezing temperature while the latter causes a drop in the maximum susceptibility.

In a few spin glasses such as AgMn [10], ScGd [11] and amorphous Fe–Ni–P [12] alloys, field-dependent magnetic irreversibility transitions similar to those in $Au_{82}(Cr_{0.6}Fe_{0.4})_{18}$ have been observed. The occurrence of these transitions has been predicted in the framework of mean-field theory for Ising and Heisenberg spin glasses. In Ising spin glasses, an $H^{2/3}$-dependence of δT_i ($= T_f - T_i(H)$) is predicted by de Almeida and Thouless (called the AT

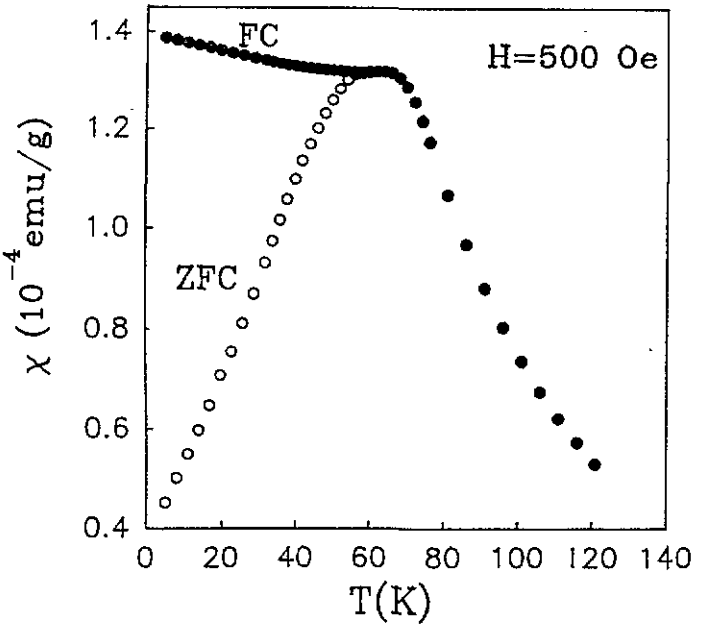

Figure 3. The temperature (T) variation of χ (emu g^{-1}) of $Au_{82}(Cr_{0.6}Fe_{0.4})_{18}$ alloy in a field of 500 Oe in ZFC and FC states.

transition) [13]. In Heisenberg spin glasses (the Gabay–Toulouse or GT model), two field-dependent transitions, showing H^2- and $H^{2/3}$-dependences of δT_i, have been predicted [14]. From various experimental data, precise determination of characteristic temperatures for AT and GT transitions is found to be difficult. These temperatures are generally defined as the temperatures of deviation of FC and ZFC susceptibilities. However, in some cases, a series of characteristic temperatures have been defined corresponding to various magnetic states of the system. The magnetization study of AgMn revealed two characteristic lines showing $\delta T_i \propto H^n$ where $n = 0.70 \pm 0.05$ and 0.51 ± 0.05, corresponding to different cross-over temperatures [10]. In our study of the $x = 0.4$ alloy, we find a power law $\delta T_i \propto H^n$ with $n = 0.52 \pm 0.05$. It is therefore not clear whether the observed field-dependent transition can be identified with the AT transition. Wenger and Mydosh have also derived a similar power law for field-dependent transition based on a superparamagnetic relaxation time approach [15].

Both the $x = 0.2$ and the $x = 0.4$ alloys exhibit a maximum in the magnetic contribution to resistivity at 160 K and 200 K respectively [6]. The occurrence of a maximum in the magnetic resistivity at temperatures far above the magnetic transition temperatures is a characteristic of canonical spin glasses [3]. This further supports the assertation of a spin-glass behaviour of these alloys.

All the alloys show a Curie–Weiss behaviour above the magnetic transition temperature. Table 1 shows the values of the effective magnetic moment, p_{eff}, and the Curie–Weiss temperature, Θ, for these alloys. In $Au_{82}Cr_{18}$, p_{eff} and Θ are found to take the values 4.4 μ_B and -400 K respectively—which are close to the values reported earlier [16].

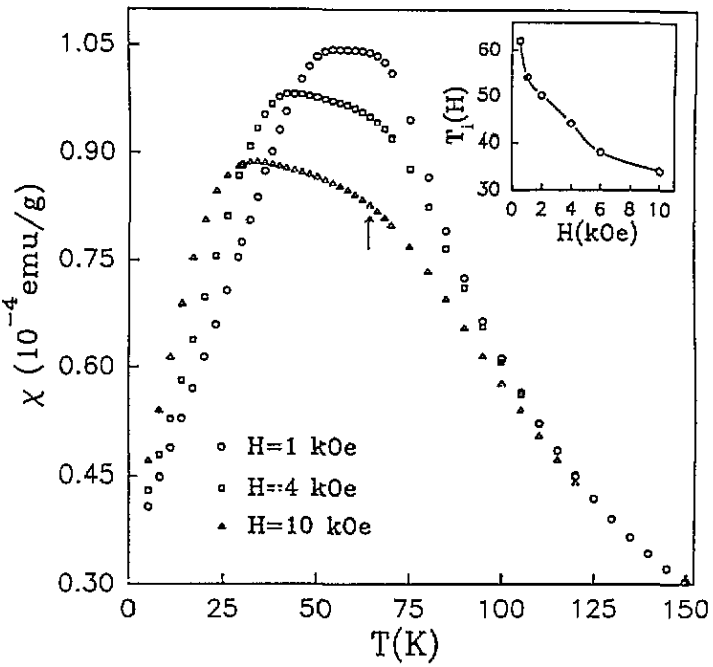

Figure 4. The variation of χ (emu g^{-1}) for the $x = 0.4$ alloy with temperature (T) in different applied fields. The arrow indicates the field-independent transition temperature at 65.8 K. The inset shows the variation of the transition temperature, T_i, with applied field (H). The solid line acts as a guide to the eye.

Table 1. Values of the maximum susceptibility χ (emu g^{-1}) for a field of 1 kOe, average magnetic moment per impurity atom p_{eff} (in units of μ_B) and Curie–Weiss temperature Θ (K) for $Au_{82}(Cr_{1-x}Fe_x)_{18}$ alloys.

x	χ_{max} (emu g^{-1})	p_{eff} (μ_B)	Θ (K)
0	4.02 (10^{-6})	4.4	−400
0.2	2.23 (10^{-5})	3.8	54
0.4	1.04 (10^{-4})	5.5	7.7

4. Conclusions

The magnetic susceptibility and resistivity data on $Au_{82}Cr_{18}$ suggest an inhomogeneous AF state of the alloy where large AF clusters exist, freezing in a spin-glass-like manner. These AF clusters are broken up due to partial substitution for Cr with Fe, modifying the AF behaviour into a spin-glass-like state. It is not clear whether the occurrence of a field-dependent magnetic transition could be associated with the AT-like transition as has been reported in some other spin-glass systems. In the present study, the cluster picture seems to be more appropriate in providing a qualitative explanation of the observed magnetic behaviour in these alloys.

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