

Antiferromagnetic Transition in α -Mn-Fe System

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Abstract

Antiferromagnetic transition in α -Mn-Fe alloys has been studied using resistivity-temperature measurements from 300 to 1.5 K. The results indicate that the transition temperature T_N of the samples studied increases with increasing Fe concentration. This suggests that the Néel temperature is affected by Fe. Also, Fe plays a significant role in the local disorder on the magnetic moment of the ions in the complex manganese structure.

Key Words: resistivity, antiferromagnetic transition, magnetic moment, disorder.

1. Introduction

It is well known that the spontaneous magnetization of 3d-transition metals or alloys is carried by electrons which are more or less itinerant in the crystal and, consequently, the magnetization is largely dependent on features of the individual crystal structure such as the inter-atomic distance, symmetry of the potential, etc. Among the metals, α -Mn stands out as quite unique in this respect. Not only does it have four crystallographic phase changes [1] as it cools from the liquid phase, with an additional paramagnetic-antiferromagnetic phase transition at 95 K in the stable low temperature α -Mn phase, but in that additional phase it has one of the complex crystal structures [2] of the elements with 58 atoms in a body-centered cubic structure of lattice constant 8.91 Å and with 29 atoms in the unit cell. This complicated crystal structure leads to four different atomic configurations in the unit cell: site I with a coordination number of sixteen and one site per unit cell, site II with a coordination number of sixteen and four sites per unit cell, site III with a coordination number of thirteen and twelve sites per unit cell, and site IV with a coordination number of twelve and twelve sites per unit cell.

From neutron diffraction studies by Yamada et al. [3] and nuclear magnetic resonance measurements by Yamagata et al. [4], it is known that the ions at these sites have various

magnetic moments: $1.9\mu_B$ at site I; $1.7\mu_B$ at site II; and $0.6\mu_B$ and $0.25\mu_B$ at sites III and IV, respectively. Consequently, as the packing around a particular ion gets closer, so the magnetic moment reduces practically to zero for a change of inter-ionic spacing from 2.96 to 2.21 Å.

It has been shown [5,6] that pure α -Mn exhibits a sharp cusp in its resistivity-temperature curve around 95 K. This temperature corresponds to the temperature at which antiferromagnetism sets in. Thus, this minimum has been thought to be associated with the ordering temperature T_N . Coles [7] has suggested that such changes in the resistivity-temperature curve can be explained by the presence of a large spin-disorder resistivity above the Néel temperature. The temperature dependence of this resistivity anomaly depends on the amount and type of impurities in the sample.

William and Stanford [8] have made an attempt to understand the nature of the complex magnetic interaction in antiferromagnetic α -Mn by determining the Néel temperature of manganese and some of its alloys. They defined the Néel temperature as that corresponding to the first minimum of the resistivity-temperature curve. To the best of our knowledge, not much work has been done on the electrical resistivity of these alloys after William and Stanford. In this paper, we report on the resistivity-temperature curves of α -Mn alloyed with 0.05 at. % Fe, 1 at. % Fe and 3 at. % Fe and their respective Néel points using the hydrodynamical-critical transition given by Craig and Goldberg [9].

2. Experimental method

The starting materials were 0.05 at. % Fe in Mn, 1 at. % Fe in Mn and 3 at. % Fe in Mn obtained from BDH Chemicals Ltd. Spark-erosion process was used in cutting the samples into the rectangular shapes of dimension 2.0 x 0.4 x 0.2 cm. This process was extremely difficult and in many situations the samples were broken if not cracked. The samples which were obtained in the rectangular form were first cleaned with dilute solution of HCl in methanol to remove surface oxides and other contaminants before the potential and current probes were made to the samples. The potential probes to these specimens were separated by 1.8 cm, while the current probes were made at the ends of the longest dimensions.

The cryostat used for these experiments was of a conventional design and it operates in the temperature range 1.5 to 300 K. Resistance measurements were made by using a constant current supply which was variable in steps from about 1.0 to 20 mA. The precise value of the current flowing through the specimen could be measured since it flowed through a standard resistor in series with the specimen and the potential difference across this could be measured on the same potentiometer as was used for measuring the potential difference across the specimens. The main potentiometer for the measurement of potential difference between the voltage probes was a Tinesley precision vernier potentiometer. A Tinesley photocell amplifier was used in conjunction with the potentiometer and this could provide much more amplification than was necessary.

The cryostat was cooled in two steps, namely the precooling stage and the helium transfer stage. The nitrogen dewar of the cryostat was filled with liquid nitrogen and the

system cooled slowly until nitrogen temperatures were obtained. The liquid level of the nitrogen dewar was maintained by an automatic filler. The helium transfer commenced by first flushing the transfer tube with helium gas before being inserted into the cryostat. The helium storage vessel was pressurized by squeezing a football bladder attached to it. Warm helium gas was blown into the vessel. This boiled the liquid helium increasing the pressure and thus forcing liquid helium over into the dewar.

The temperature of the specimen between 300 and 60 K was monitored with a copper resistance thermometer. Below 60 K, carbon resistance thermometer was used since copper resistance thermometers are insensitive at these temperatures.

3. Results and discussion

Figure 1(a,b,c) illustrates the temperature dependence of the electrical resistivity of the samples. On cooling the specimen, the resistivity goes through a minimum and then a maximum. In the 0.05 at. % Fe sample, the minimum in the resistivity is observed at about 101 K and the maximum at about 57 K. For the 1 at. % Fe sample, the resistivity drops at about 110 K and rises to a maximum at about 60 K. In the case of the 3 at. % Fe sample, the resistivity minimum is observed at about 130 K and a maximum at about 55 K. The temperatures corresponding to the resistivity minima have been taken by others to be the transition temperature for these alloys. Thus, it appears that as the Fe concentration increases, the transition temperature shifts towards higher values, as reported by other workers [5,8].

The difference between the temperature at which the maxima occur, $T_{\rho(\max)}$, and that at which the minima occur, $T_{\rho(\min)}$, which can be taken as the width of the transition, ΔT , is found to increase with increasing Fe concentration. The values of ΔT for the specimens are 44 K for the 0.05 at. % Fe sample, 50 K for the 1 at. % Fe sample, and 75 K for the 3 at. % Fe sample. The increase in ΔT with Fe concentration gives evidence of an increase in disorder as the Fe concentration increases. The broadening of the transition could be related to the distribution of the transition temperature among the grains of the samples [10].

The resistivity-temperature curves of the 0.05 and 1 at. % Fe samples drop sharply after $T_{\rho(\max)}$. This rapid decrease in the resistivity is due to magnetic ordering of the spins. For the 3 at. % sample, there is a large excess resistivity of $1.675 \mu\Omega\text{m}$. The origin of this large excess resistivity can be associated with the effect of local disorder on the magnetic moments of the complex α -Mn structure. The excessive scattering could be due to the Fe ions acting as scattering centers or could be connected with the lattice distortion in the high-Fe-concentration alloy. Collins and Low, in their work on the magnetic moment distribution around impurities [11], observed that the magnetic moment disturbance in Fe-Ni alloys could be due to two sources: first from changes in the moments on atoms in the vicinity of an impurity and secondly as a result of displacements of these atoms from their lattice sites.

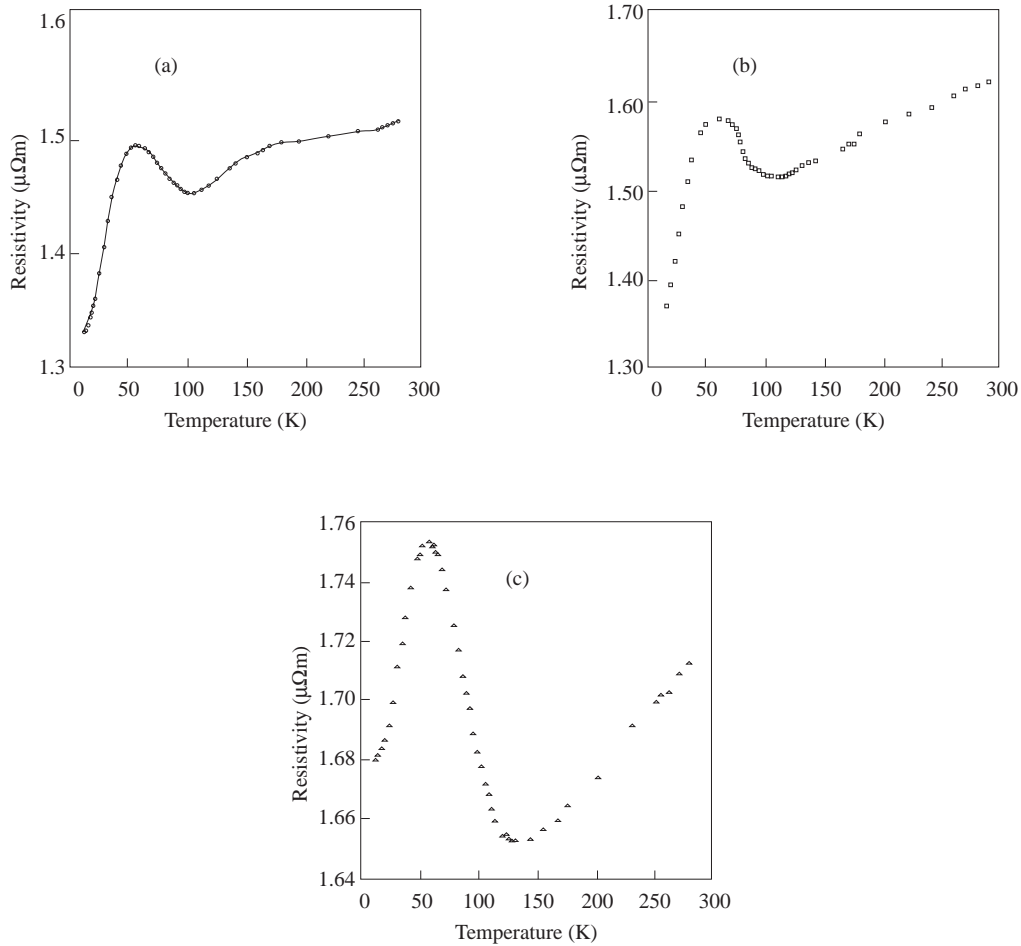


Figure 1. Temperature dependence of the electrical resistivity of the α -Mn-Fe alloys: (a) 0.05 at. % Fe, (b) 1 at. % Fe, and (c) 3 at. % Fe

The significant increase in the value of the resistivity at a particular temperature with Fe is also worth mentioning. This is further evidence of the introduction of disorder in the system. It is known that both magnetic and non-magnetic disorder affect the magnetic and electrical properties of materials in several theoretical models [12,13], which predict an increase in the residual and normal-state resistivity due to impurity scattering introduced by disorder.

Zunsteg and Parks [14] have defined the Néel point as the temperature at which the temperature dependent of the magnetic coherence length $\xi(T)$ approximately equals the phonon limited mean free path of the conduction electrons. Craig and Goldberg [9]

have suggested that the anomaly in $\delta\rho/\delta T$ versus T that gives a singularity marks the transition from the hydrodynamic regime to the critical regime predicted by magnetic scaling theory, and the anomaly lies in the strong temperature dependence of the phonon limited mean free path in the vicinity of the ordering temperature. The hydrodynamical-critical transition is defined by the position of the singularity in the $\delta\rho/\delta T$ versus T curve. Consequently, the maximum or minimum in the plot of $\delta\rho/\delta T$ versus T determines the Néel temperature in antiferromagnetic materials.

The resistivity-temperature relationship presented in this paper is useful for the determination of the Néel temperature of these alloys, as demonstrated in the $\delta\rho/\delta T$ versus T plots in Fig. 2 (a,b,c). This analysis has been employed in determining the Néel temperature of some α -Mn thin films [15]. For a film deposited at a pressure of 10^{-6} Torr

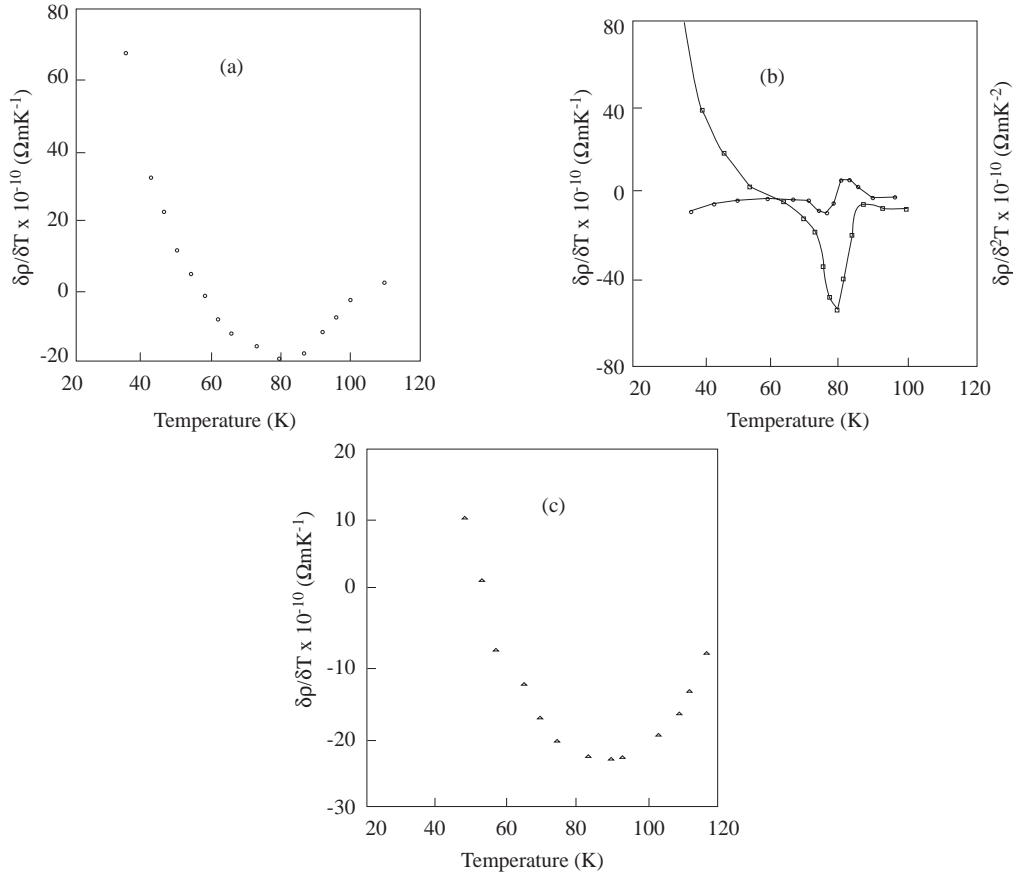


Figure 2. Temperature dependence of $\delta\rho/\delta T$ of the α -Mn-Fe alloys: (a) 0.05 at. % Fe, (b) 1 at. % Fe, and (c) 3 at. % Fe. $\delta^2\rho/\delta T^2$ versus T curve is demonstrated in (b) to give more evidence of a phase transition.

on a glass substrate held at 350 °C, $T_{\rho(\min)}$ was found to be 116 K while T_N determined from the minimum of the $\delta\rho/\delta T$ versus T curve was 90 K. Also for a bulk sample with $T_{\rho(\min)}$ of 95 K [16], a similar analysis gives T_N of 81 K. The values of T_N obtained from the minima of the $\delta\rho/\delta T$ versus T curves for the samples studied in this work occur at about 80 K for the 0.05 at. % and 1 at. % Fe samples and 87 K for the 3 at. % Fe sample. Thus, the Fe concentration shifts the Néel temperature towards higher values. However, it is seen that the lower impurity levels do not affect the transition temperature of the system. It has been observed by Collins and Low [11] that in order to investigate the effects of controlled additions of about 1.5 at. % of impurities it is necessary to start from materials with a much lower total impurity content. From the above analysis it can be said that $T_{\rho(\min)}$ is the temperature of the onset of transition but is not the Néel temperature itself.

The $\delta\rho/\delta T$ curves show broad transition which is consistent with the difference between the temperature at which the maxima and the minima of the resistivity-temperature curves occur. This is true for many polycrystalline samples in which there is a distribution of transition temperature among the grains of the samples [10]. Such specimens could be considered being made up of grains which are coupled by weak links with distribution of critical temperatures [17]. The $\delta^2\rho/\delta T^2$ curves, as demonstrated in Fig 2b, give clear evidence of a phase transition.

4. Conclusion

The present results indicate that the Néel temperature of the α -Mn-Fe alloys increases with increasing Fe concentration. The high excess resistivity of the 3 at. % Fe sample may be associated with the effect of local disorder on the magnetic moment of the ions in the host or be connected with the lattice distortion in the high-Fe-concentration alloy. The increasing values of the resistivity at a particular temperature gives further evidence of the introduction of disorder in the system with increasing Fe concentration.

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