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LETTER TO THE EDITOR

Magnetic and electrical properties of YCo_2: effects of Mn substitution

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Abstract. The effects of substitution of Mn on the Co sites in YCo_2 have been studied by AC susceptibility and resistivity measurements. The results support the recent small-angle neutron scattering study showing the absence of long-range ferromagnetic order in $Y(Co_{1-x}Mn_x)_2$ with $x = 0.1$ and 0.15.

The inter-metallic compound YCo_2 is an interesting Pauli paramagnet. Though Co does not carry a moment in pure YCo_2 it is on the verge of being magnetic and a small substitution of Fe for Co rapidly stabilises the Co moment (Burzo 1978). It has been suggested by Cyrot *et al* (1979) that the Stoner criterion is almost satisfied in YCo_2 and slight change of band filling or other parameters can give rise to a large effect on the density of states at the Fermi level. This suggestion has made the study of the effect of alloying in YCo_2 particularly interesting and there has been a great deal of activity in this area in the last few years. Most interesting among recent results is the onset of weak itinerant ferromagnetism produced by about 15% of Al substitution at the Co site (Yoshimura and Nakamura 1985). On the other hand substitution of Mn for Co also gives rise to some interesting but less straightforward results. It is thought that a small amount of Mn, which presumably carries a local moment, can stabilise a moment in the neighbouring Co atoms of YCo_2, rather as iron atoms do when substituted in palladium. Oesterreicher and Parker (1982) have studied the occurrence of magnetic order in the $Y(Co, Mn)_2$ system in some detail. From their study of magnetisation and Arrott plots they have indicated an onset of ferromagnetic order ($T_C = 53$ K) in YCo_2 with 10% substitution of Mn for Co. With further increase in Mn concentration a cluster glass or mictomagnetic behaviour was observed in $Y(Co_{0.6}Mn_{0.4})_2$. The observation of ferromagnetic order in $Y(Co_{0.9}Mn_{0.1})_2$ has been questioned in a recent neutron measurement. In a small-angle neutron scattering measurement, Kilcoyne *et al* (1988) failed to observe critical scattering (due to the onset of ferromagnetic order) at any temperature. Rather their results indicate the presence of ferromagnetic-like correlations over only ten or so unit cells. It is to be noted at this point that Oesterreicher and Parker (1982) performed their magnetisation measurements in a field of 3 to 7.2 kOe. On the other hand small-angle neutron scattering measurements can directly observe spin correlations in the absence of any perturbing external magnetic field. To shed more light on this subject we decided to measure the low-field AC susceptibility and ressistivity in $Y(Co_{1-x}Mn_x)_2$ with $x = 0.1$ and 0.15.

The alloys were prepared by argon arc-melting from metals of at least 99.99% nominal purity and suction chill casting into copper moulds to produce rods of square cross section. The alloys were homogenised *in vacuo* at 800 °C for seven days. The samples were subjected to metallographic analysis to investigate the possible presence of a second phase and a small amount was found in both of the alloys.

AC susceptibility measurements were performed with a driving frequency of 300 Hz and a driving field of 0.7 Oe parallel to the axis of samples. A static external field of up to 320 Oe was provided parallel to the driving AC field, with the help of a pair of Helmholtz coils. The resistivity measurement was performed using a standard DC four-probe method.

We present the AC susceptibility (χ) results of $Y(Co_{1-x}Mn_x)_2$ with $x = 0.1$ and 0.15 in figures 1 and 2. For $x = 0.1$, the χ against T plot shows a distinct rise in susceptibility around 60 K followed by a sharp cusp around 14.5 K. One may be tempted to assign a sort of transition temperature (~45 K) from the point of inflection in the χ against T plot but this behaviour is much less well defined than that observed even in a weak itinerant ferromagnet like $Y(Co_{0.85}Al_{0.15})_2$ (figure 2). The most interesting feature is that the initial rise in the AC susceptibility can be suppressed by a small DC biasing field of 80 Oe and can be erased by a field of 320 Oe, leaving behind a spin-glass-like cusp at 14.5 K. The possibility immediately comes to mind that there may be a magnetic second phase. Though we mentioned earlier that our metallographic study indicated the presence of a second phase in our samples, we think the amount is too small to give rise to a distinct observable effect. If we leave this assertion for the time being, the question then arises, if the effect of the second phase is important, of what its composition is. A scan of the literature (Buschow 1980) reveals that the following inter-metallic compounds involving Y and Co or Y and Mn are possible: Y_3Co ($T_C < 2$ K), Y_4Co_3 ($T_C < 4$ K), YCo_3 ($T_C =$ 301 K), Y_2Co_7 ($T_C = 639$ K), YCo_5 ($T_C = 977$ K), Y_2Co_{17} ($T_C = 116$ K), Y_6Mn_{23} ($T_C =$ 486 K), YMn_{12} ($T_N = 120$ K). As we can see, the existence of any of these compounds as a second phase will not give a simple explanation of the results in the $x = 0.1$ compound. (Of course there remains the possibility of an as yet unknown ternary phase involving Y, Co and Mn). Also, why should this second phase give rise to the observed anomaly in the $x = 0.1$ sample only and not in $s = 0.15$? As is shown in figure 2, the χ against T plot shows a cusp around 20 K in the $x = 0.15$ sample. Similar susceptibility behaviour suggesting spin-glass-like freezing has been observed in $Y(Co_{1-x}Fe_x)_2$ for $x < 0.12$ (van Dongen *et al* 1981). These results tend to support the suggestion of the neutron measurements that there is no long-range ferromagnetic order in this concentration regime.

The resistivity results for these two compounds are shown in figures 3 and 4. (The samples were full of microcracks; hence the absolute value of the resistivity should not be taken very seriously. However, the order of magnitude of the high-temperature resistivity is comparable with that of the parent compound YCo_2 (Gratz *et al* 1980).) In each case a resistivity minimum is observed around the temperature where the corresponding susceptibility showed a cusp-like behaviour. There is the indication of a similar type of resistivity minimum in $Y(CO_{1-x}Fe_x)_2$ for $x = 0.08$ and $x = 0.1$ (van Dangen *et al* 1981). Such a correlation between the resistivity minimum and susceptibility cusp has been observed earlier in spin-glass systems, such as PtMn (Sarkissian and Taylor 1974), PtCr (Roshko *et al* 1977), $(U_{1-x}Gd_xAl_2)_2$ (Ping and Coles 1982) and recently in $Ce(Al_{1-x}Fe_x)_2$ (Lees and Coles 1988) systems. The resistivity against temperature plots do not show any knee or sharp change in slope suggestive of a ferromagnetic ordering, rather the high-temperature behaviour of the resistivity shows a strong curvature toward

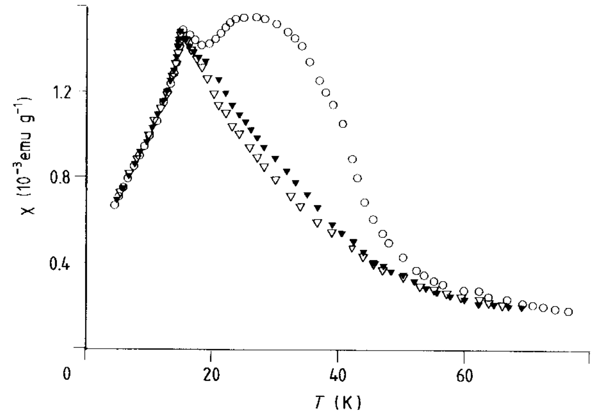

Figure 1. Curves of AC susceptibility plotted against temperature for $Y(Co_{0.9}Mn_{0.1})_2$ in the presence of various static external fields: ○, 0 Oe; ▼, 80 Oe; ▽, 320 Oe.

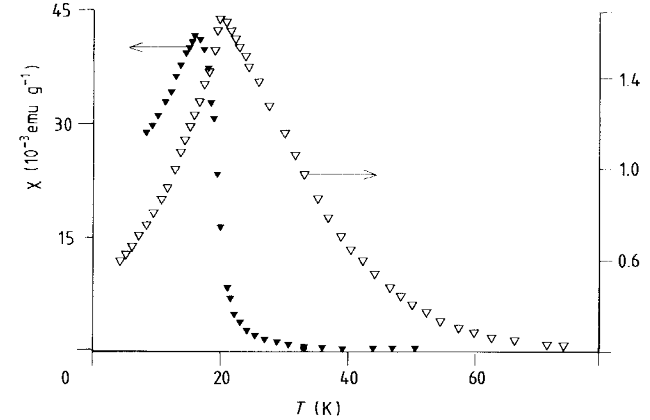

Figure 2. Curves of AC susceptibility plotted against temperature for $Y(Co_{0.85}T_{0.15})_2$: ▽, T = Mn; ▼, T = Al.

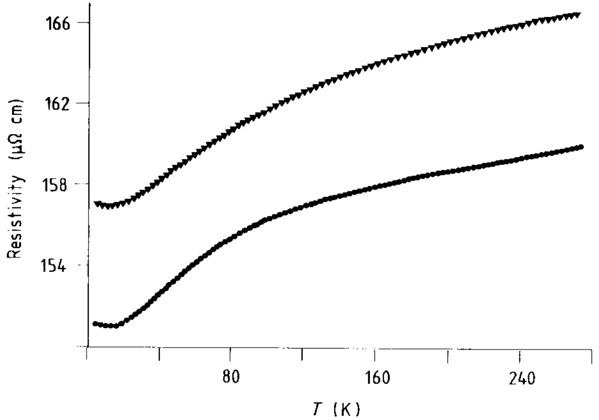

Figure 3. Resistivity plotted against temperature for $Y(Co_{1-x}Mn_x)_2$: ▼, $x = 0.1$; ●, $x = 0.15$.

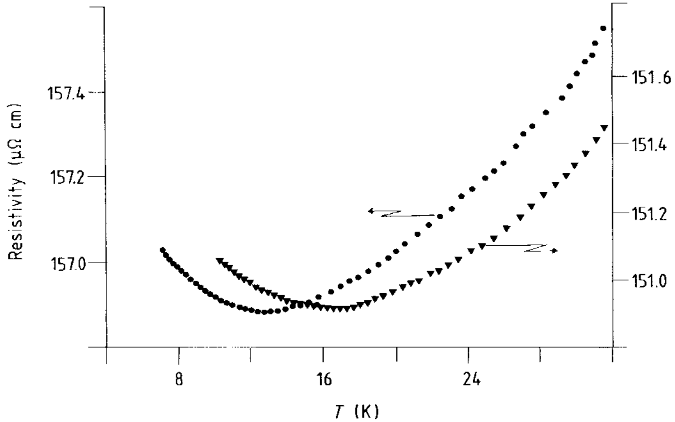

Figure 4. Low-temperature resistivity minimum in $Y(Co_{1-x}Mn_x)_2$: ●, $x = 0.1$; ▼, $x = 0.15$.

the temperature axis. The latter feature is a characteristic of the parent compound YCo_2 (Gratz *et al* 1980). Though such behaviour is often associated with the weakly or nearly ferromagnetic character of the samples (Hilscher and Gratz 1978), an alternative explanation (Ogawa 1976) in terms of spin fluctuations also exists.

A simplistic explanation of the various observed features may be the following. In the concentration range of present interest, Mn atoms probably carry a local moment and locally polarise the Co matrix which, in turn, forms a giant moment. These giant moments have ferromagnetic interactions between them, whereas Mn atoms approach each other as the concentration grows and interact antiferrromagnetically. In the presence of a high magnetic field (as used by Oesterreicher and Parker 1982) these giant moments are aligned and give rise to a field-induced quasi-ferromagnetism. Such a field-induced effect has indeed been observed in the spin-glass to ferromagnetic crossover region of $Fe_{80-x}Ni_xCr_{20}$ (Majumdar and Blanckenhagen 1984). In the absence of any strong external field the competition between ferromagnetic and antiferromagnetic interaction gives rise to spin-glass-like behaviour. Of course the explanation of the effect of a small static biasing field on the AC susceptibility for $x = 0.1$ needs to be supported by something more than the present results. In conclusion we would like to say that Mn (unlike Al) cannot give rise to long-range ferromagnetic order in YCo_2 (at least in the concentration range studied here), rather it gives rise to an inhomogeneous spin-glass-like magnetic order. A careful study of low-field DC magnetisation (effect of field cooling and zero-field cooling) and magnetoresistance would be useful to clarify the matter further.

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