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LETTER TO THE EDITOR

The magnetic phases of the itinerant magnetic system $Ce(Fe_{1-x}Co_x)_2$

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Abstract. Magnetic and electrical property results for the pseudobinary compound $Ce(Fe_{1-x}Co_x)_2$ are reported. It is found that as the temperature is decreased the system goes from the para- to the ferro- to the antiferromagnetic phase. The ferro-to-antiferromagnetic phase transition is a first-order transition. An H–T phase diagram for the ferro- to antiferromagnetic transition has been determined. All the experimental results suggest that the magnetic phases in $Ce(Fe_{1-x}Co_x)_2$ may be understood in terms of itinerant electron magnetism. Our experimental results are consistent with the magnetic phase diagram predictions of Moriya and Usami's theory of strongly interacting itinerant electron systems.

Of the series of the cubic laves phase alloys RFe_2 (R = rare earth), the ferromagnetic $CeFe_2$ is of particular interest. Its magnetic moment per formula unit (2.4 μ_B) and its Curie temperature (T_c = 227 K) are significantly lower than for other compounds of the series. This has been attributed to some transfer of the Ce 4f electrons to the conduction band [1]. It has been reported by various authors that the substitution of Fe in $CeFe_2$ by small amounts of Al, Co, Ru, Si, Os and Ir destabilizes the ferromagnetism of $CeFe_2$. In some cases such substitution causes a total loss of ferromagnetism at low temperatures. The lower temperature second magnetic phase transition has been given a number of possible explanations ranging from spin-glass to antiferromagnetism [2–8]. Recent neutron diffraction studies by Kennedy and coworkers [9–11] have concluded that this low temperature phase is antiferromagnetic in $Ce(Fe_{1-x} M_x)_2$ with M = Al, Ru and Co.

In this communication we present a study of the low temperature magnetic phase of the pseudobinary intermetallic compound $Ce(Fe_{1-x}Co_x)_2$. We have carried out experimental measurements of magnetization as a function of temperature and applied magnetic field, AC susceptibility, electrical resistivity and thermal expansion as a function of temperature. An H–T phase diagram of the lower temperature phase transition has been determined and the order of the phase transition established. These results are compared with the Moriya and Usami theory [12, 13] of strongly interacting itinerant electron systems.

The polycrystalline samples of $Ce(Fe_{1-x}Co_x)_2$ were prepared by repeated arc-melting of stoichiometric quantities of the constituent metals in argon atmosphere. The nominal purity of these metals is 99.9% for Ce (Cerac, Inc) and 99.99% for Fe and Co (Johnson Mathey). The arc melted button was annealed in vacuum at 600 °C for 2 days, at 700 °C for 5 days, at 800 °C for 2 days and at 850 °C for 1 day. The DC magnetization measurements as a function of temperature and applied magnetic field was carried out in a SQUID magnetometer (Quantum Design, California). AC susceptibility measurements were made in an AC field of 1.3 G and at a frequency of 80 Hz. The electrical resistivity was measured by a conventional four-probe method. The relative thermal expansitivity ($\Delta L/L$) as a function of temperature was carried out in a capacitance dilatometer [14].

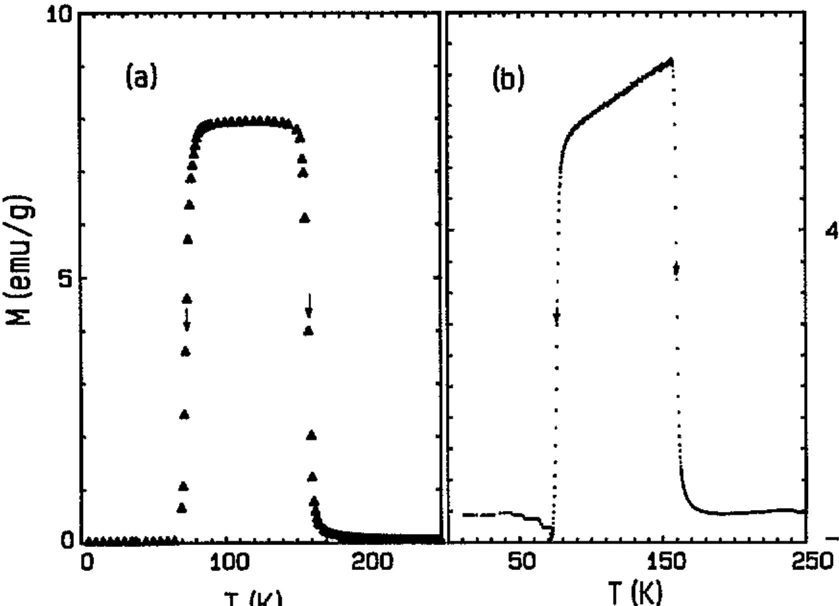

Figure 1. (*a*) Magnetization (M) as a function of temperature (T) in a constant magnetic field of 100 G, and (*b*) AC susceptibility (χ_{AC}) as a function of temperature (T) for $Ce(Fe_{0.8}Co_{0.2})_2$.

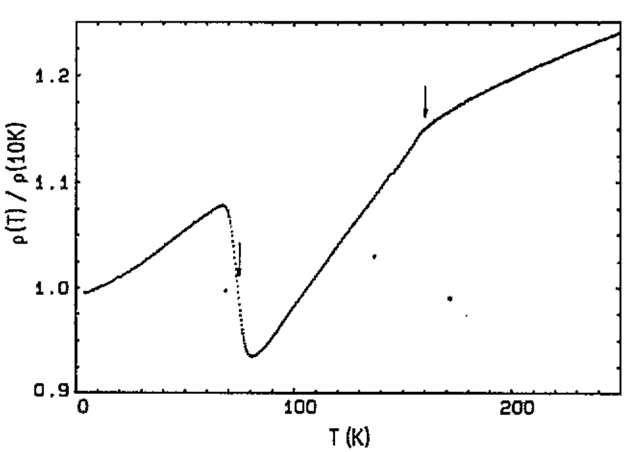

Figure 2. Normalized electrical resistivity ρ as a function of temperature (T) for the $Ce(Fe_{0.8}Co_{0.2})_2$ sample.

The magnetization (M) and AC susceptibility (χ_{AC}) as a function of temperature for $Ce(Fe_{0.8}Co_{0.2})_2$ are presented in figure 1. As the temperature decreases the system goes from the paramagnetic to the ferromagnetic state at $T_c \simeq 160$ K. On further decreasing the temperature there is a sudden and sharp drop in magnetization below $T_N = 75$ K. It is evident that a partial substitution of Fe by Co causes the same sample to undergo paramagnetic to ferromagnetic to antiferromagnetic phase transitions as the temperature of the sample is reduced. The temperature dependence of the electrical resistivity (figure 2) also clearly shows these magnetic transitions. These results are consistent with the earlier studies by Rastogi *et al* [15]. A slope change in the resistivity at 162 K is identified as being due to a paramagnetic to ferromagnetic transition. As the temperature is further decreased the resistivity of $Ce(Fe_{0.8}Co_{0.2})_2$ sharply rises below 75 K, goes through a maximum, and then decreases with decreasing temperature. Such behaviour in the electrical resistivity is reminiscent of the superzone boundary effect in antiferromagnetic transitions as observed for various rare earth metals. The behaviour of ρ versus T at the lower temperature phase transition, along with the magnetization data suggest that the magnetic transition below $T_N = 75$ K is a transition from a ferromagnetic to an antiferromagnetic state. Our *magnetization* AC susceptibility and resistivity measurements and their interpretations are in good agreement with earlier work [2–5, 15].

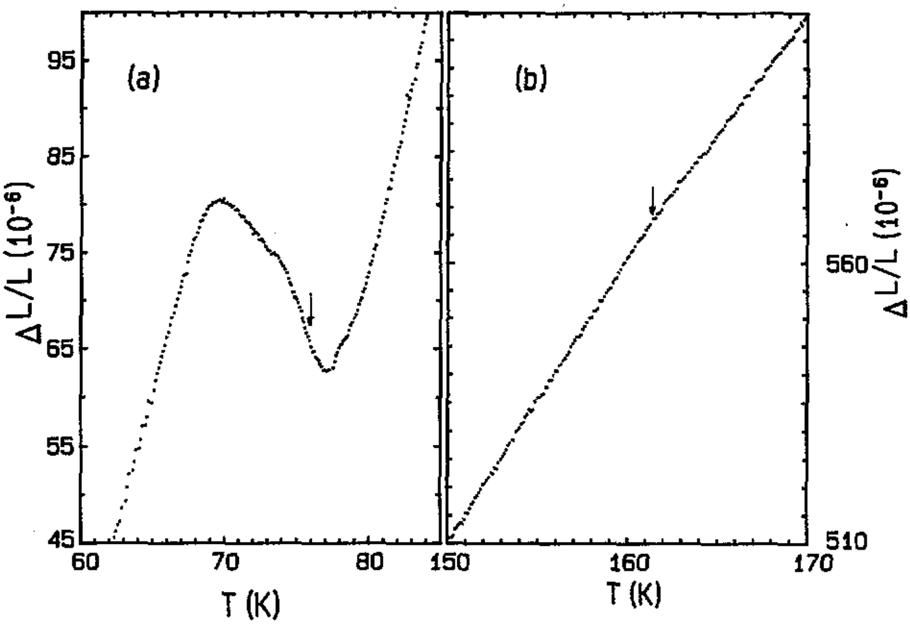

Figure 3. (*a*) Thermal expansivity ($\Delta L/L$) as a function of temperature (T) in the neighbourhood of T_N and (*b*) thermal expansivity ($\Delta L/L$) as a function of temperature (T) in the neighbourhood of T_c for $Ce(Fe_{0.8}Co_{0.2})_2$.

To explore the order of the phase transitions at $T_c \simeq 162$ K and $T_N \simeq 75$ K, we measured thermal expansitivity ($\Delta L/L$) as a function of temperature for $Ce(Fe_{0.8}Co_{0.2})_2$. Figure 3 shows $\Delta L/L$ versus T in the vicinity of $T_c \simeq 162$ K and $T_N = 75$ K. We observe a slope change in $\Delta L/L$ versus T at $T_c = 162$ K and a step change in $\Delta L/L$ versus T around $T_N = 75$ K. The $\Delta L/L$ versus T data clearly show that the phase transition at $T_c \simeq 162$ K is of second order and that the ferromagnetic to antiferromagnetic transition at $T_N \simeq 75$ K is of first order. One expects a sharp step in $\Delta L/L$ at a first-order phase transition temperature. The first-order phase transition around $T_N = 75$ K in the $\Delta L/L$ versus T curve has a width of about 10 K (figure 3(*a*)). This may be due to lack of compositional uniformity of the sample. This lack of compositional uniformity may cause a spatial distribution of T_N value in the sample and hence manifest itself as a wide step in $\Delta L/L$ versus T around T_N rather than as a vertical step.

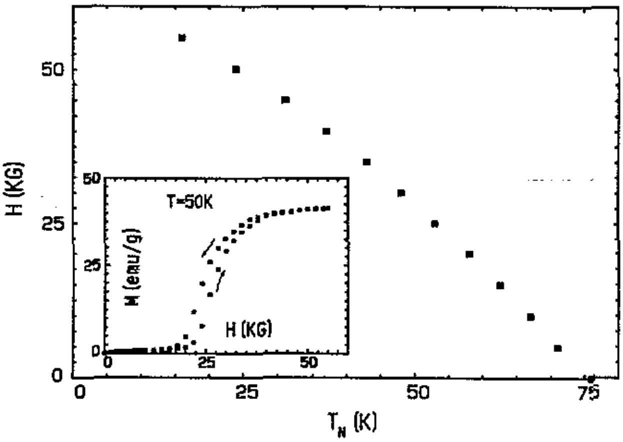

Figure 4. The magnetic field (H) versus T_N for $Ce(Fe_{0.8}Co_{0.2})_2$. The inset shows the magnetic field induced transition from the anti- to ferromagnetic state at 50 K for the $Ce(Fe_{0.8}Co_{0.2})_2$ sample.

The antiferromagnetic to ferromagnetic phase transition in zero applied magnetic field takes place at $T_N = 75$ K (determined from magnetic data) in $Ce(Fe_{0.8}Co_{0.2})_2$ samples. A magnetic field dependence of T_N has been determined using magnetization measurements as a function of temperature at various constant applied magnetic fields. The T_N values at various applied fields are determined from a series of M versus T measurements like that of figure 1(*a*). The experimentally determined H versus T_N is presented in figure 4. The H versus T_N phase diagram (figure 4) shows a strong H dependence of T_N. At the maximum available magnetic field of 55 kG T_N decreases to 16 K. From figure 4 the extrapolation of the H versus T_N curve shows $T_N = 0$ K at $H \simeq 65$ kG. In other words, for fields above 65 kG, the system $Ce(Fe_{0.8}Co_{0.2})_2$ never enters the antiferromagnetic phase. We have found that the H versus T_N curve (figure 4) does not show a simple field dependence. The entire range of H versus T_N data could be reasonably well fitted to the following equation

$$T_N = a + bH + cH^2$$

where the coefficients a, b and c are found to be $a = 74.04$ K, $b = -0.641$ K kG^{-1} and $c = 7.30 \times 10^{-3}$ K° kG^{-2}. It is noticed that the measured critical field makes a finite angle with the H-axis at $T_N(H = 0)$. The inset of figure 4 shows that at constant temperature when the applied magnetic field is increased (in the antiferromagnetic state) the magnetization has a sharp step increase at a critical field where the system goes from an antiferromagnetic to a ferromagnetic state (see inset of figure 4). We also observe a field hysteresis in the magnetization as shown by arrows in the inset of figure 4.

The temperature dependence of the magnetization of $Ce(Fe_{0.8}Co_{0.2})_2$ samples shows that as the temperature is lowered a transition from paramagnetic to ferromagnetic order occurs at $T_c \simeq 160$ K; a transition from ferromagnetic to an almost complete loss of magnetization then occurs at $T_N \simeq 75$ K. This lower temperature phase is antiferromagnetic and the transition at $T_N = 75$ K is first order. This result is consistent with recent neutron diffraction studies [9–11]. The temperature dependence of electrical resistivity clearly shows the para- to ferromagnetic transition at $T_c \simeq 160$ K as a change in slope of the resistivity. As the system goes from the ferromagnetic to the antiferromagnetic phase below $T_c \simeq 75$ K the resistivity rises sharply, goes through a maximum, and then decreases with decreasing temperature. This sharp rise in the resistivity at the onset of antiferromagnetic ordering is due to the appearance of magnetic superzones as seen in certain rare earths, e.g. Dy, Ho and Er.

Moriya and Usami [12, 13] have theoretically studied a strongly interacting itinerant electron system without magnetic anisotropy and express its free energy as a function of the uniform and staggered components of magnetization M_0 and M_Q. They predicted various magnetic phase possibilities and the coexistence of ferro- and antiferromagnetism in itinerant-electron systems. One possible magnetic phase diagram predicted by Moriya and Usami is the one yielding para- to ferro- to antiferromagnetic phases as the temperature is decreased. For such a system they show that at $T < T_N$, the field dependent magnetization jumps at a critical magnetic field (as shown in figure 4 (inset)). The critical magnetic field versus T_N is predicted to make a finite angle with the H-axis at $T = T_N$ ($H = 0$) and the transition from the ferro- to antiferromagnetic state is predicted to be of first order.

Our experimental results on the temperature dependence of magnetization, AC susceptibility, resistivity and thermal expansion show a para- to ferro- to antiferromagnetic transition in the $Ce(Fe_{0.8}Co_{0.2})_2$ system. The ferro- to antiferromagnetic transition is of first order. The magnetization as a function of field (for $T < T_N$) shows a critical magnetic field at which a large step increase in magnetization is observed when the sample goes from the antiferro- to ferromagnetic state. The H–T curve showing the field dependence of $T_N(H)$ makes a finite angle with the H-axis. It is quite evident that our experimental results on $Ce(Fe_{0.8}Co_{0.2})_2$ realize the magnetic phases predicted by Moriya and Usami [12, 13] using a strongly interacting itinerant electron system.

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