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Investigation of the role of Ce atoms in the $CeFe_2$ system

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Abstract

$CeFe_2$ is a ferromagnet (FM) with cubic Laves phase structure and bears some anomalous magnetic properties such as much lower Curie temperature ($T_c = 227$ K) and lower magnetic moment per Fe atom (1.15 μ_B) as compared to the other compounds in the RFe_2 series (R = Y and rare earths). The role of Ce in $CeFe_2$ has been a controversial one. The view ranges from Ce in $CeFe_2$ being in the tetravalent state to its having a non-integral magnetic moment with an antiferromagnetic (AFM) coupling between the iron and cerium sites. It is found that a substitution of Fe sites by Co atoms in $CeFe_2$ destabilizes the FM order at lower temperatures with a FM to AFM transition at $T_N = 78$ K for $Ce(Fe_{0.8}Co_{0.2})_2$. To explore the role of Ce, we substituted Ce with Y in $Ce(Fe_{0.8}Co_{0.2})_2$. It was observed that with the substitution of Y for Ce, the AFM is suppressed and FM is gradually stabilized. In $Ce_{1-x}Y_x(Fe_{0.8}Co_{0.2})_2$, complete restoration of ferromagnetism is achieved down to the lowest temperature measured for 10% of Ce substitution by Y ($x = 0.1$).

1. Introduction

$CeFe_2$ is a ferromagnet with cubic Laves structure and exhibits some anomalous magnetic properties such as a much lower Curie temperature ($T_c = 227$ K) and much lower magnetic moment per Fe atom (1.15 μ_B) than other compounds in the RFe_2 series (R = rare earth) [1]. A number of investigations [2–4] suggest that the anomalous magnetic properties of $CeFe_2$ may be due to some transfer of the Ce 4f electrons to the Fe 3d band. Obviously, the Ce in $CeFe_2$ plays an important role in its magnetic properties.

A small substitution of Fe by other elements such as Co, Al and Ru leads to a total loss of ferromagnetism at a temperature T_N, lower than T_c [5–9]. This second transition was observed for cobalt concentrations $0.03 < x < 0.3$. Neutron diffraction studies [10,11] revealed that this loss is due to the transition from the ferromagnetic state to the antiferromagnetic state. The results of Mössbauer measurements show that the hyperfine field evolves gradually across the second transition temperature [7], suggesting that the magnitude of the Fe moment remains the same across the transition. A high resolution neutron diffraction study shows a rhombohedral distortion of the cubic lattice at the onset of antiferromagnetism [11]. This requires the presence of a magnetic moment due to the 4f electrons of Ce not yet found in the case of the $Ce(Fe_{1-x}Co_x)_2$ system [12]. However, Kennedy and Coles [12] through their powder neutron diffraction data show that the Ce atom in $CeFe_2$ has a magnetic moment which orients antiparallel to the magnetic moment of the Fe atom. The electronic structure calculations made by Khowash [3] show that Ce and Fe moments in $CeFe_2$ are antiparallel with $\mu_{Ce}/\mu_{Fe} = 0.37$, in good agreement with Kennedy and Coles' experimental value of 0.3 [12].

YFe_2 is a ferromagnetic material and has the same crystal structure as $CeFe_2$. The magnetic moment per Fe atom (1.45 μ_B) and the Curie temperature (542 K) of YFe_2 [1] are much higher than those of $CeFe_2$. Cerium has 4f electrons, but yttrium does not. Therefore, substituting Ce with Y could be one way to investigate the role of the Ce atom in $CeFe_2$ as shown by Rastogi *et al.* [8] and Roy and Coles [9]. In this paper, we present the effects of partial substitution of Ce by Y in the $Ce(Fe_{1-x}Co_x)_2$ system mainly on its magnetic properties.

2. Experimental details

The polycrystalline samples of $Ce_{1-x}Y_x(Fe_{0.8}Co_{0.2})_2$ were prepared by repeated arc-melting of stoichiometric quantities of the constituent metals under an argon atmosphere. The nominal purity of these metals is 99.9% for Fe and Y (Cerac Inc.) and 99.99% for Ce and Co (Johnson Matthey). The melted samples were annealed in a vacuum at 600 °C for 2 days, at 700 °C for 5 days, at 800 °C for 2 days, and 850 °C for 1 day. These samples are single phase as determined from X-ray powder diffraction study.

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The dc magnetization measurements were carried out on a SQUID magnetometer (Quantum Design, CA). All measurements were performed with the samples cooled in zero field. The data for magnetization as a function of temperature were collected as the temperature was increased.

3. Results and discussion

The results of magnetization measurements as a function of temperature for $Ce_{1-x}Y_x(Fe_{0.8}CO_{0.2})_2$ with $0<x<0.1$ are shown in Fig. 1. It is known that $CeFe_2$ orders ferromagnetically below $T_c=227$ K. The magnetization data for $Ce(Fe_{0.8}Co_{0.2})_2$ shows a ferromagnetic transition at $T_c=165$ K and an antiferromagnetic transition below $T_N=75$ K [5]. A partial replacement of Fe by Co in $CeFe_2$ destabilizes the ferromagnetic state at low temperatures. The ferro-to-antiferromagnetic transition is a first order phase transition [5] with a rhombohedral distortion of the cubic unit cell [11]. The ferromagnetic state in $CeFe_2$, in the low temperature regime, is on the verge of magnetic instability. This has been recently suggested by the neutron diffraction studies of Kennedy and Coles [12] and Yang *et al.* [13]. They observed an antiferromagnetic component developing below $T\approx 80$ K in $CeFe_2$. Therefore, a partial substitution of Fe by Co in $CeFe_2$ precipitates the instability of the FM state and hence a ferro-to-antiferromagnetic transition appears at low temperatures. Such re-entrant magnetic transitions have been studied theoretically by Moriya *et al.* [14] and Isoda [15]. Our starting pseudobinary alloy was $Ce(Fe_{0.8}Co_{0.2})_2$ and we substituted Ce by Y in $Ce_{1-x}Y_x(Fe_{0.8}Co_{0.2})_2$. We observed (Fig. 1) that as the concentration of Y increased, the ferromagnetic transition temperature gradually increased and the ferro-to-antiferromagnetic transition temperature decreased. For the Y concentration of 10% ($x=0.1$) in $Ce_{1-x}Y_x(Fe_{0.8}Co_{0.2})_2$, the value of T_c increased to 185 K with no evidence of ferro-to-antiferromagnetic transition down to 5 K. A magnetic phase diagram for $Ce_{1-x}Yx(Fe_{0.8}Co_{0.2})_2$ as a function of Y concentration is presented in Fig. 2. It is quite evident from Figs. 1 and 2 that the partial substitution of Ce by Y in $Ce_{1-x}Y_x(Fe_{0.8}Co_{0.2})_2$ gradually reduces the instability of the ferromagnetic state and for 10% (*i.e.* $x=10\%$) Y in place of Ce, a complete restoration of the ferromagnetic state down to 5 K is achieved. Therefore, one may conclude that the instability of the ferromagnetic state is directly related to Ce in this system.

The results of isothermal (at $T=10$ K) magnetization measurements as a function of applied magnetic field for various Y concentrations is presented in Fig. 3. The magnetization increases very sharply at a critical field (for $x=0$, 0.02 and 0.06) with large field hysteresis.

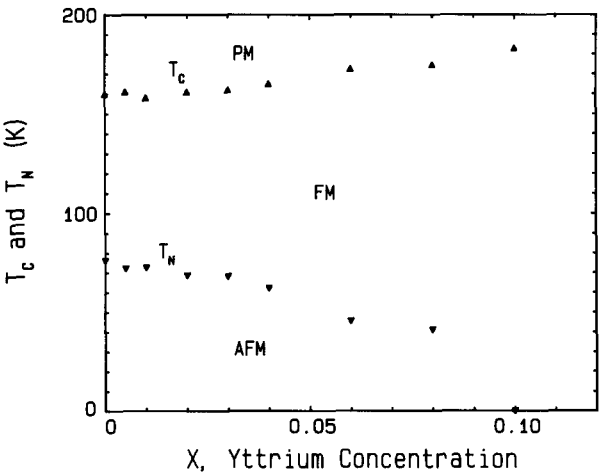

Fig. 2. The values of T_N and T_c as a function of Y concentration (x) for the $Ce_{1-x}Y_x(Fe_{0.8}Co_{0.2})_2$ system. PM, paramagnetic; FM, ferromagnetic; AFM, antiferromagnetic.

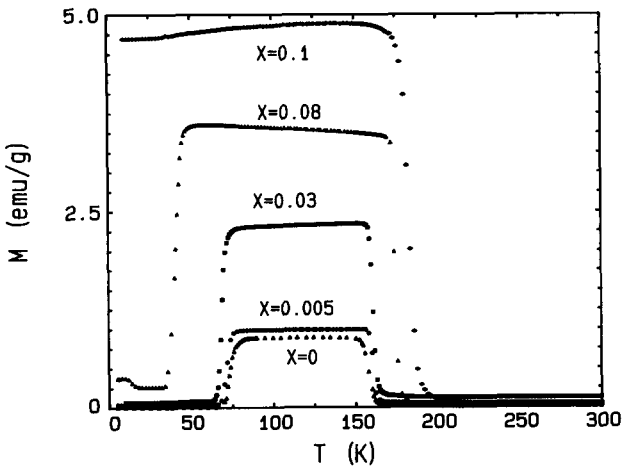

Fig. 1. Magnetization (M) as a function of temperature (T) at 100 G for the $Ce_{1-x}Y_x(Fe_{0.8}Co_{0.2})_2$ system. The x values are labeled on the curves.

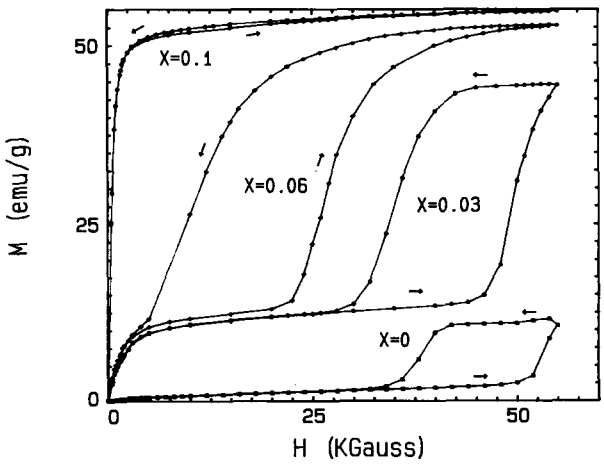

Fig. 3. The magnetization (M) as a function of applied magnetic field (H) for the $Ce_{1-x}Y_x(Fe_{0.8}Co_{0.2})_2$ system at 10 K for various x values.

This sharp rise in magnetization at a critical field is associated with a field induced antiferromagnetic to ferromagnetic first order phase transition. The magnetic hysteresis loops are located symmetrically about the origin (not presented here). We notice that critical field decreases with increasing concentration of Y in $Ce_{1-x}Y_x(Fe_{0.8}Co_{0.2})_2$ and it vanishes for 10% Y concentration. The hysteresis loops for the samples with $x=0$ and $x=0.03$ in Fig. 3 are not complete due to our limitation of a maximum magnetic field of 55 kG. Therefore, we are unable to compare the areas of the hysteresis loops between different x value samples in Fig. 3. However, from the curves for $x=0.1$ and 0.06, it is reasonable to expect that with increasing concentration of Y, the hysteresis loop area decreases and disappears for $x=0.1$.

To determine the temperature dependence of the critical field and the hysteresis loop area, we measured the magnetization as a function of applied magnetic fields at various constant temperatures for the $Ce_{0.97}Y_{0.03}(Fe_{0.8}Co_{0.2})_2$ sample. These data are presented in Fig. 4. It is evident from Fig. 4 that both the critical field and the area of hysteresis loop decrease with increasing temperature.

The field-induced transition and the magnetic hysteresis as seen in our results suggest the presence of magnetic anisotropy [16]. A rhombohedral distortion of the cubic unit cell observed at the ferro-to-antiferromagnetic transition temperature could be accounted for by the magnetoelastic coupling model of Cullen and Clark [17]. However, this requires a local moment on the Ce sites. The neutron scattering studies on $Ce(Fe_{1-x}Co_x)_2$ by Kennedy and Coles [12] suggest no moment ($<0.1\ \mu_B$) at the Ce sites for this system.

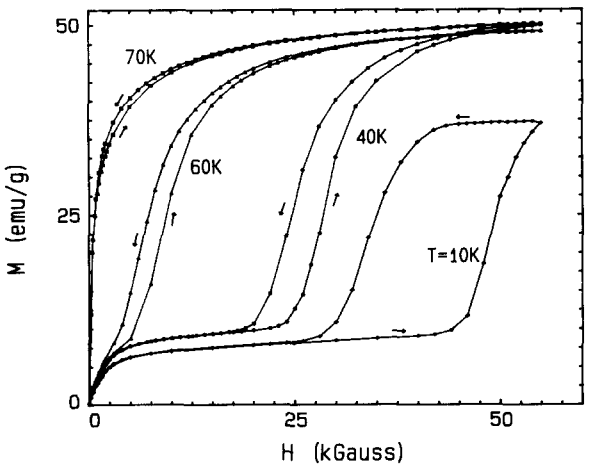

Fig. 4. The magnetization (M) as a function of applied magnetic field (H) at various temperatures for the sample $Ce_{0.97}Y_{0.03}(Fe_{0.8}Co_{0.2})_2$.

Summarizing, our observations are as follows: a partial replacement of Ce by Y in $Ce(Fe_{1-x}Co_x)_2$ gradually restabilizes the ferromagnetic state and for 10% Y, it completely restores the ferromagnetic state; and the temperature dependence of the critical field (for the field induced transition) and the temperature and field dependence of the area of the hysteresis loops, suggest a strong temperature dependence of magnetic anisotropy. In addition, it has been observed by Kennedy *et al.* [11] that an applied field can reverse not only the antiferromagnetic state but also structural distortion. Therefore, it is likely that a competition between the exchange interaction energy and magnetic anisotropy energy, which is temperature dependent, is responsible for the ferro-to-antiferromagnetic transition in $Ce(Fe_{1-x}Co_x)_2$. This suggests that the magnetic anisotropy increases with decreasing temperature and destabilizes the ferromagnetic state. The partial replacement of Ce by Y in $Ce(Fe_{1-x}Co_x)_2$ enhances the stability of the ferromagnetic state, and hence it would appear that a decrease in the concentration of Ce (replaced by Y) in $Ce(Fe_{1-x}Co_x)_2$ would lead to a decrease in the magnetic anisotropy in the system. The temperature dependence of magnetic anisotropy in this system needs further investigation.

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