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A neutron polarization analysis study of Ce_2Fe_{17} in its paramagnetic phase

Sk MOHAMMAD YUSUF, L MADHAV RAO and K B BHAT*

Solid State Physics Division, *Chemistry Division, Bhabha Atomic Research Centre, Bombay 400 085, India

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Abstract. Spin-flip (paramagnetic) scattering and neutron depolarization studies were performed on Ce_2Fe_{17} in its paramagnetic phase on the Dhruva neutron polarization analysis spectrometer. The absence of normal Q dependence of the scattered spin flip intensity shows that Ce_2Fe_{17} is not a normal paramagnetic and there exist superparamagnetic clusters of sufficiently large dimensions (~ 100 Å). The observed neutron depolarization gives an indication of the dynamics of these Ce_2Fe_{17} superparamagnetic clusters.

Keywords. Ce_2Fe_{17}; superparamagnetic clusters; neutron polarization analysis; spin-flip.

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1. Introduction

The R_2Fe_{17} compounds (R = rare earth) are the most iron rich of all binary rare earth iron intermetallics. They exist across the whole lanthanide series except for La itself, crystallizing in the hexagonal Th_2Ni_{17} structure for R heavier than Dy and in the rhombohedral Th_2Zn_{17} structure for R lighter than Gd; both structures may coexist for R = Gd, Tb and Dy. Compounds of the R_2Fe_{17} class have large magnetization values but low ordering temperatures and magnetocrystalline anisotropies. These make the R_2Fe_{17} compounds less attractive for applications as permanent magnet materials. By alloying these systems with carbon (Gubbens *et al* 1989; Zhong *et al* 1990) and nitrogen (Coey and Sun 1990; Jaswal *et al* 1991), considerable enhancement of the Curie temperature and anisotropy can be achieved. While magnetization and Mössbauer studies exist in these alloyed compounds (Gubbens *et al* 1989; Zhong *et al* 1990; Coey and Sun 1990; Hu *et al* 1991), very few neutron investigations have been reported (Jaswal *et al* 1991). Our interest therefore is in elucidating the magnetic structures of these alloyed compounds using the neutron diffraction technique. In this context, the compound Ce_2Fe_{17} was prepared. This paper reports some interesting features observed by us in Ce_2Fe_{17} in its paramagnetic phase, using the neutron polarization analysis technique.

2. Experimental details

The compound Ce_2Fe_{17} was prepared from the constituent elements of at least 99·9% purity by repeated arc melting in purified argon atmosphere. After arc melting, the ingots were sealed into an evacuated quartz tube and annealed for about 150 hours at

Table 1. Structural parameters derived from fitting the neutron diffractogram obtained for the compound Ce_2Fe_{17} at 300 K. Values in parentheses indicate uncertainties in least significant digits. n.r. denotes not refined.

Space group	$R\bar{3}\ m$
$a = b$(Å)	8·4941 (3)
c(Å)	12·3999 (4)
$\alpha = \beta$	90°, $\gamma = 120°$
Ce(6C)	$x = y = 0$(n.r.), $z = 0{\cdot}3383(3)$
Fe(6C)	$x = y = 0$(n.r.), $z = 0{\cdot}0965(2)$
Fe(18F)	$x = 0{\cdot}2900(3)$, $y = z = 0$(n.r.)
Fe(18H)	$x = 0{\cdot}5007(4)$, $y = \bar{x}$, $z = 0{\cdot}1526(3)$
Fe(9D)	$x = 0{\cdot}5$(n.r.), $y = 0{\cdot}0$(n.r.), $z = 0{\cdot}5$(n.r.)

850°C. Standard X-ray pattern taken on the specimen showed that the phase was well-formed. A neutron diffraction pattern was taken at room temperature. Using the Rietveld profile refinement technique (Rietveld 1969) the single phase (Ce_2Fe_{17} phase) model analysis indicated the presence of a small amount of unreacted α-Fe. So a quantitative analysis of the proportion of the two phases (phase 1:Ce_2Fe_{17}, phase 2:α-Fe) was simultaneously done using the two phase model of the same technique (Hill and Howard 1987). This analysis showed the specimen to be in phase 1 with 1·4% of unreacted α-Fe. Table 1 gives the crystallographic data. It is seen that the atomic position parameters are very close to those obtained earlier (Isnard *et al* 1990).

AC susceptibility measurements were also performed to ensure that the magnetic phases of the sample are also well-formed (Givord and Lemaire 1974).

Neutron spin-flip and depolarization measurements were carried out on Ce_2Fe_{17} at 300 K in its paramagnetic phase on the neutron polarization analysis spectrometer installed at the Dhruva reactor (Madhav Rao *et al* 1990).

3. Experimental results and discussion

Givord and Lemaire (1974) had studied Ce_2Fe_{17} in its two magnetically ordered phases, one at 140 K and the other at 4·2 K. Our AC susceptibility measurements also showed the specimen to be in two magnetically ordered phases: one below 198 K and the other below 77 K. However, a surprising feature seen in the diffraction pattern taken on the specimen at room temperature using unpolarized neutrons, was the absence of Q dependent paramagnetic scattering over the expected low angle (low Q) region (see figure 1). As outlined by us elsewhere (Madhav Rao *et al* 1990) measurement of neutron spin-flip scattering using the polarization analysis technique can uniquely pick out magnetic correlations. Hence, on the same specimen at room temperature spin-flip scattering which in this case uniquely measures paramagnetic scattering was measured on the Dhruva polarization analysis spectrometer. Figure 2 shows this measurement indicating the absence of any significant scattering wave vector dependence (Q dependence) of paramagnetic scattering. On the same figure is plotted (on an arbitrary scale) the theoretically calculated paramagnetic scattering from Ce_2Fe_{17}. It is seen from this curve that if Ce_2Fe_{17} were a normal paramagnet the scattered spin flip neutron intensity should have fallen by about 60% over the Q range 0

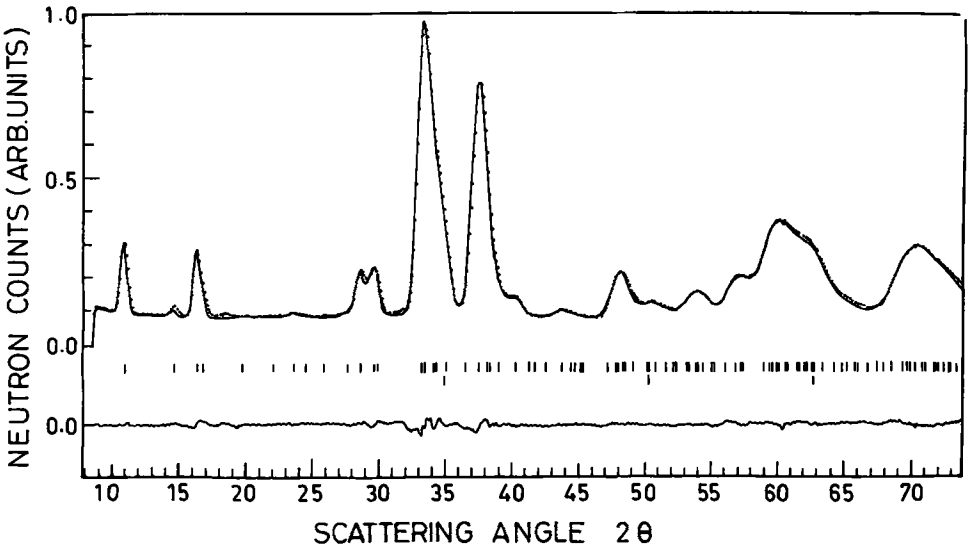

Figure 1. Neutron diffraction diagram of powder Ce_2Fe_{17} obtained at 300 K. Observed (calculated) profiles are given by the dotted (solid) curves. The short vertical lines below the pattern represent the positions of all possible Bragg reflections for Ce_2Fe_{17} (top row) and minor α-Fe (bottom row). The lower curve is the difference between the observed and calculated intensity at each step, plotted on the same scale.

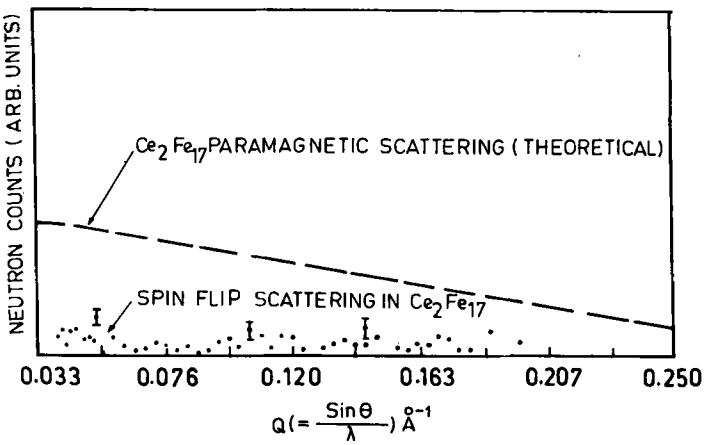

Figure 2. Spin flip (paramagnetic) scattering of Ce_2Fe_{17} obtained at 300 K. The observed data are indicated by dotted points and the theoretically expected calculated profile from paramagnetic Ce_2Fe_{17} is the broken line. The vertical lines over some observed dotted points represent the error bar.

to 0·2 $Å^{-1}$. The observed flat response of the measured spin flip intensity over this Q range leads us to speculate that Ce_2Fe_{17} is not in the normal paramagnetic state, but there exists clusters of Ce_2Fe_{17} which are big enough (~ 50 to 100 Å) so that no significant Q dependent paramagnetic scattering is seen in the normal Q range covered by the spectrometer.

Another independent measurement performed was the depolarization study of the

transmitted neutron beam in the Ce_2Fe_{17} specimen, at room temperature. Generally speaking, a neutron polarization vector senses fluctuating magnetic fields averaged over a time scale t_N, where t_N is a measure of the Larmor precession time. The static vertical magnetic field imposed on the specimen was varied from 1 kOe to 10 kOe. In any normal paramagnet, spins fluctuate so rapidly with characteristic time $\sim 10^{-12}$ sec, that the neutron polarization vector feels a fluctuating field as zero. The neutron polarization vector is preserved along the direction of the static vertical field and hence the z–z polarization of the transmitted beam is left undisturbed (Mistuda and Endoh 1985). Essentially, therefore, if Ce_2Fe_{17} were a normal paramagnet, no depolarization in the transmitted neutron beam should be observed. On the other hand, a severe neutron depolarization of the order 50% was observed in the transmitted neutron beam. This interesting observation gives a clue therefore about the dynamics of these Ce_2Fe_{17} clusters. In a classical picture, the depolarization of a neutron can be visualized as occurring as a result of a torque exerted by a transverse magnetic field on the neutron spin vector. Clearly this transverse field should be finite at least during the period of the Larmor precession of the neutron.

The severe depolarization observed in Ce_2Fe_{17} at room temperature can be explained qualitatively as arising due to the superparamagnetic clusters relaxing sufficiently slowly so as to produce a finite fluctuating transverse magnetic field which gets coupled to the neutron polarization vector.

4. Summary and conclusion

Two quite independent observations using polarized neutrons have been made on Ce_2Fe_{17} in its paramagnetic phase. First, the flat response in the spin-flip scattering (which samples uniquely the paramagnetic scattering) over a Q range up to 0·25 $Å^{-1}$ gives us an idea about the size of the superparamagnetic clusters ($\sim$ 50 Å to 100 Å). Second, the severe depolarization of the transmitted polarized neutron beam throws light on the dynamics of these clusters.

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