Coexistence of Kondo effect and Ferromagnetism in Ce$_{1.5}$Nd$_{1.5}$Al

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Abstract. The possibility for a coexistence of Kondo effect and ferromagnetism in Ce$_{1.5}$Nd$_{1.5}$Al has been studied using heat capacity and resistivity. The sample is of polycrystalline in nature. Heat capacity data confirms the heavy fermion behavior of this compound with a Sommerfeld coefficient $\gamma = 190 \text{mJ/mol-K}^2$ and Debye temperature $\sim 180 \text{K}$. An upturn in resistivity and heat capacity observed near $\sim 24 \text{K}$ is attributed to the paramagnetic to ferromagnetic transition as reported. Kondo like behavior is observed below 10K and vanishes for magnetic fields of the order of 2T. Around 60K a prominent jump or cusp like behaviour is seen and observed to be robust against magnetic fields up to 14T. Heat capacity below 20K is found to be consistent with equation $C_p(T) = \gamma T + \beta T^3 + \alpha T^{3/2} e^{-\Delta T}$ that signifies the presence of a gap in magnon excitation energy of the order of 70K.

1. Introduction

Cerium based intermetallic compounds show many properties like Fermi and non-Fermi liquid behavior, heavy fermion behavior, Kondo effect, valence fluctuation, quantum criticality, superconductivity, RKKY interaction and various types of magnetic ground states due to the hybridization between localized 4f electrons and conduction electron band. Quantum critical behavior appeared as a result of competition between RKKY interaction and Kondo Effect [1-5]. Conventionally Kondo effect opposes the magnetic order but there are many cerium and uranium based intermetallic systems where metallic ferromagnetism exists in Kondo Lattice system [5], examples are CeCrGe$_3$ [6], CeTiGe$_3$ [7], UTe [8] and UC$_{0.5}$Sb$_2$ [9]. Here in all above mentioned compounds ferromagnetic ordering temperature ($T_C$) is less than Kondo minima temperature ($T_K$). Parent compound Ce$_3$Al shows Kondo effect, heavy fermion behaviour, antiferromagnetism and first order structural phase transition from hexagonal to monoclinic with space group P 6$_3$/m m c [10 -11]. Doping of Nd at the site of Ce in Ce$_3$Al up to 35%, vanishes the Kondo effect as well as the structural transition [12]. But at 50% Nd doping again shows a Kondo behavior that consistent with the co-existence of ferromagnetic ordering.

2. Experimental Detail

Polycrystalline Ce$_{1.5}$Nd$_{1.5}$Al sample was prepared by arc melting in high purity argon gas using constituent elements of purity $> 99.99\%$. For homogeneity we have melted the ingots three times and then make rectangular rod for resistivity measurement with a special type of dye. Then rod sealed in a quartz tube at high vacuum with pressure (P) $< 10^{-3}$ mbar followed by annealing at 500°C for one
week and at 200°C for two weeks. X-ray diffraction has been carried out using Cu-Kα radiation with a Bruker D8 Advance X-ray Diffractometer for phase analysis. Resistivity measurements were carried out down to 2K in magnetic fields 0-14T with conventional four probe method. Heat capacity in 0T field is also reported in support of the ferromagnetic and heavy fermion behaviour and is carried out by a relaxation calorimetry. QD-PPMS system has been used for both of these measurements.

3. Results and Discussions

Resistivity of the sample up to 14T and temperatures down to 2K are shown in figure 1. The resistivity curve shows Kondo effect at low temperature as evident by the shallow upturn (inset of figure 1). Around 24K there is a drop in resistivity whose behaviour is attributed to the ferromagnetic nature of transition. An interesting behaviour has also been observed where Kondo effect appears below 10K in an otherwise ferromagnetic state. As in the nonmagnetic metal the action of an applied magnetic field on the resistance is associated with the curving of the electronic orbits, and results in an increase of electrical resistance. If there are magnetic impurities in a metal, then the applied magnetic field will polarize the spins of impurity atoms. Thus the direction of spins become fixed and the probability of spin-flip scattering declines i.e. breakdown of Kondo effect. As a result, the resistivity falls off in field. Since this compound is dense Kondo system hence Kondo singlet breakdown is happening here also and resistivity is decreases with magnetic field. A more interesting feature at 60K has been observed in 0T resistivity, which is robust against magnetic field. The dispersion in resistivity has also been visualized upon the application of magnetic fields. Above 60K all the curves coincide in the fields. Even though at this composition of Nd, the structural transition is ruled out, as evident from the nil hysteresis seen, this seems to be a transition of unknown origin.

Magnetoresistance (MR) vs temperature curve up to 14T is shown in figure 2. Maximum MR is 18% at 2K and 14T. There is a deep in MR at ~24K which vanishes after 6T indicated by a red arrow in figure 2. Above 60K all the MR curves are coincide means magnetic response of the sample is only below 60K. Therefore upturn at the 60K may be a magnetic in nature.

Heat capacity curve of the sample is shown in figure 3. Inset of figure 3 shows the close interval points measurement around 25K. Inset curve shows an upturn near 25K resulting in a magnetic transition from PM to FM as reported [12]. This compound has hexagonal structure up to lowest measured

\[ \rho = a - b \ln T \]

**Figure 1.** \( \rho \) vs. T in magnetic field up to 14T and temperature range 2-120K. Inset shows the 0T resistivity vs. \( \ln T \) below 50K and blue line is fitted with equation \( \rho = a - b \ln T \)

**Figure 2.** MR (%) vs. T up to 14T. Red arrow is pointing the dip in MR around 24K attributed to the FM ordering.
temperature. For a ferromagnetic metal with a noncubic crystal structure; the heat capacity well below $T_C$ is generally expressed as

$$C_p(T) = a^*T + b^*T^3 + c^*T^{1.5}\exp(-\Delta/T)$$

Where first and second terms represent the electronic and phonon contributions in heat capacity respectively. The third term represents the conventional form of a magnon contribution, where $\Delta$ is an anisotropy gap in magnon energy. We have fitted the heat capacity data up to 20K (inset of figure 3) with the above given equation and we have found $\gamma = 190.8\text{mJ/mol-K}^2$, $\beta = 1.24\text{ml/mol K}^4$, $\alpha = -1.4 \times 10^3\text{ml/mol} - K^2$ and $\Delta \sim 70K$. From the fitting with equation (1) we conclude that system is moderate heavy fermion with Debye temperature $\sim 180K$.

A small and broad upturn in heat capacity curve observed near $\sim 24K$ may represent a short range magnetic correlation. Ce has a strong Kondo nature while Nd has a ferromagnetic tendency. Hence for temperatures $T_K < T_C (= 25K)$ there may be a hybridization between conduction electron band of Ce and the uncompensated excess Nd magnetic moments, giving rise to a possible FM correlations.

4. Conclusions
A good quality polycrystalline sample of $\text{Ce}_{1.5}\text{Nd}_{1.5}\text{Al}$ was prepared using arc melting in Argon atmosphere. XRD pattern shows the sample has single phase. Resistivity and specific heat measurements have been done by the QD 14T/2K PPMS. Heat capacity curve is showing heavy fermion behavior with $\gamma = 190\text{mJ/mol-K}^2$ with Debye temperature ($\Theta_D = 180K$). The Magnetic moment of Nd and of Ce in this compound is $(3.62*1.5 = 5.43\mu_B)$ and $(2.54*1.5 = 3.81\mu_B)$ respectively. Therefore magnetic moment in the compound is not fully compensated and hence a small amount of Nd magnetic moment is available for magnetic ordering. MR vs T plot is showing the negative behavior below 40K. This is a clear signature of FM ordering. Kondo effect is vanishing in the magnetic field after 2T. The deep in MR around 25K is a direct evidence of ferromagnetic nature of the material.

![Figure 3. Heat capacity (Cp) vs. Temperature in 0T. Inset shows the Cp curve below 30K and red line is fitting curve with equation (1).](image-url)
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