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Mangnetoresistance of heavy fermion-like compound Ce(Ni$_{1-x}$Cu$_x$)$_2$Al$_3$

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ABSTRACT

$CeNi_2Al_3$ system is a potential candidate for low temperature thermoelectrics. Substitution studies, especially at the Ni site are considered to be of importance due to the drastic tuning of its physical properties. Resistivity in magnetic fields and thermoelectric power measurements of Cu doped $CeNi_2Al_3$ (x=0.0 to 0.4) system is reported in this investigation. This dense Kondo lattice system is investigated with an aim of understanding its basic transport mechanism. Negative magnetoresistance is seen for x=0.3 and 0.4 in the magnetic field up to 14 T. Deviation from the Kondo behavior occurs at temperatures close to 2 K with a down turn in resistivity. The nature of resistivity at low temperatures is investigated in view of the possible evidence for Fermi liquid behavior and also the formation of heavy Fermion in corroboration with specific heat studies. Doping dependence of linear diffusion coefficient and Sommerfeld coefficient of specific heat are analyzed and discussed in connection with the heavy Fermion formation. The results obtained show a promising trend in tuning these materials by way of Kondo route as well as by the substitution especially at the Ni site in the present system. Copyright © 2015 VBRI Press.

Keywords: Magnetoresistance; kondo effect; fermi liquid; heavy fermion.

Sankararao Yadam is a PhD scholar at Low Temperature Laboratory under the supervision of Dr. V. Ganesan, Centre Director, UGC-DAE Consortium for Scientific Research, Indore. He is working on materials with tunable ground states including thermoelectric power properties.

Introduction

Thermo Electric Power (TEP) is considered to be a property of prime interest in materials science especially in the scenario of energy conversion. Recently a variety of novel phenomena are associated as reasons for high figure of merit seen in thermoelectric materials. Examples include Topological Insulator behavior in Bi_2Te_3 based systems, resonant charge relaxation as a source of high TEP in a Kondo Insulator type FeSi, resonant distortion of electronic density of states in Sm doped Zn_4Sb_3 and the possible rattling motion of rare earth (RE) atoms in doped Skutterudites. In addition, Ce based compounds that exhibit unusual ground states and novel phenomena such as heavy Fermion behavior and non-Fermi liquid behavior are considered to be of potential candidate in this arena **[1-4].** Presence of strong hybridization between localized 4f electrons and conduction electrons is responsible for the above phenomena. Competition between Kondo and RKKY interactions are responsible for several magnetic ground states.

The parent compound, $CeNi_2Al_3$, is one of the compounds with strong c-*f* hybridization regime. It crystallizes in the hexagonal $PrNi_2Al_3$-type structure with space group P6/mmm and exhibits metallic behavior **[5]**. Enhancement in the unit cell volume, conduction electrons, thermoelectric power and specific heat were reported upon increasing Cu concentration as well as the system undergoes a transition from Pauli paramagnetic to Curie-Weiss type paramagnetic state respectively at 40% doping in Ce(Ni$_{1-x}$Cu$_x$)$_2$Al$_3$ **[6]**. Upon further increasing in Cu concentration, the system undergoes antiferromagnetic ordered state **[7]**. Like other Ce- and Eu-based heavy Fermion systems, Ce(Pb$_{1-x}$Sn$_x$)$_3$, Ce(Pt$_{1-x}$Ni$_x$), and EuCu$_2$(Ge$_{1-x}$Si$_x$)$_2$, the present system has also been shown to have a proportionality between the initial slope of TEP S(T) curve and the electronic specific heat coefficient as T→0K limit **[8]**. From the thermoelectric measurements, parameters obtained from the two band model fit, it was observed that an evolution from a simple compensated metal (x=0.0) to a paramagnetic one (x=0.4) in this series Ce(Ni$_{1-x}$Cu$_x$)$_2$Al$_3$ **[9]**. The change of transport properties with concentration doping under the effect of magnetic fields, is not yet studied. In this communication, we report on the magnetotransport as well as a note on the enhancement of carrier concentration with Cu doping. The title compound is investigated in view of the possible evidence for Fermi liquid behavior and also the formation of heavy quasi particles in corroboration with specific heat. An attempt has been made to show indirectly that enhancement in the charge carrier concentration using specific heat and thermoelectric measurements as T→ 0 K. A deeper insight is obtained by investigating the system under magnetic fields that corroborates the formation of heavy quasi particles. Such a study establishes the contribution of Kondo type route in enhancing the thermo electric power and the corresponding manifestation in the physical properties.

Experimental

Materials

Polycrystalline $Ce(Ni_{1-x}Cu_x)_2Al_3$ with x=0.0,0.1,0.3,0.4 have been prepared by taking constituent elements, Ce (99.9%), Ni (99.995%) supplied by Leico from USA and Mateck from Germany respectively. Cu (99.9%) and Al (99%) are supplied by Cerac from USA. The stoichiometric amounts were arc melted in high purity Argon atmosphere. For phase homogeneity flipping was done three times and finally rods of few centimeters were prepared.

Method

The Phase purity was checked on unannealed powder specimens with the Bruker D8 Advanced X-ray diffractometer using Cu-Kα radiation. Resistivity measurements were carried out using four probe technique with the 14T/2K PPMS, (QD-USA) temperature down to 2K and magnetic field up to 14T. Thermoelectric power measurements were done by the differential method (Chromel/Au+0.07% Fe thermocouple as detector) using a homemade setup **[10]**.

Results and discussion

Fig. 1 shows resistivity vs temperature of $Ce(Ni_{1-x}Cu_x)_2Al_3$ with x=0.0-0.4 in the magnetic field up to 14 T. Sample x=0.0 shows metallic behavior **[5]**, whereas x=0.1 shows broad hump at high temperature. This feature is gradually enhanced and shifted towards low temperature with doping concentration. The broad hump around 150 K and low temperature rise is shown for x=0.4 is due to crystalline electric field effect on Ce^{+3} **[11]**. Apart from x=0.4, x=0.3 also shows a clear rise (for clarity refer to figure 2) at low temperatures. Positive magnetoresistance (PMR) is observed for the samples x=0.0 and 0.1 as can be observed in insets of Figure 1. Since these samples show Pauli paramagnetic and metallic behavior **[6]** one can expect the above said PMR. From the literature, for x=0.3 the magnetic susceptibility (χ) values are improved compared to x=0.0 and 1/χ apparently approached towards a linear dependence **[6]**. Interestingly above ~15K x=0.3, shows PMR, indicating dominant character of Pauli paramagnetism. However, crossover of magnetoresistance from negative to positive happens at ~15 K. Further, for 40% Cu doping, the system exhibits negative magnetoresistance throughout the temperature range measured because of Curie-Weiss type paramagnetism **[6]** and is as expected.

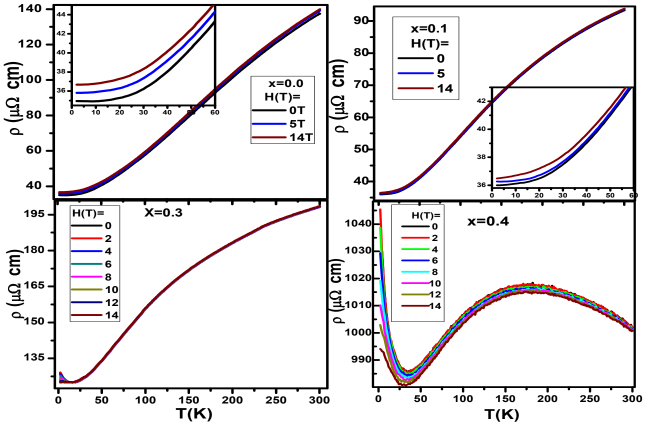

Fig. 1. Resistivity ρ(T) of $Ce(Ni_{1-x}Cu_x)_2Al_3$ for x=0.0-0.4 in the magnetic field up to 14 T. Insets: Low temperature data of x=0.0 and 0.1.

Fig. 2 and **3** shows magnetic field effect on the ρ(T). At low temperature –lnT (Kondo effect) rise of resistivity with decreasing the temperature is observed and is fitting to the equation 1.

$$\rho = \rho_0 - b\ln(T) \tag{1}$$

where ρ_0 and b are constants (shown in the **Fig. 4**).

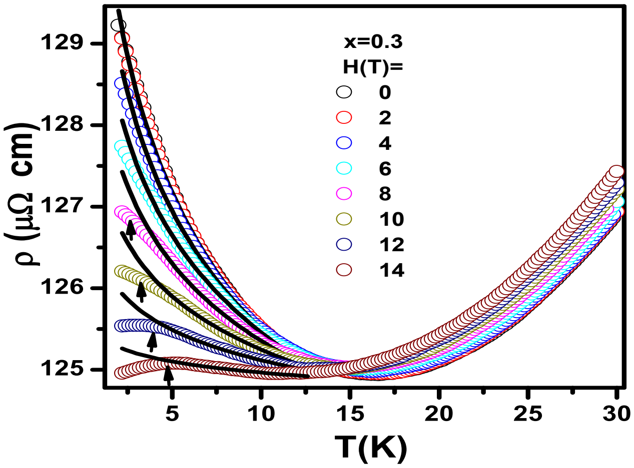

Fig. 2. Resistivity as function of temperature for x=0.3 which shows a clear rise at low temperatures and in magnetic fields up to 14 T which is fitted to the kondo model (Equation 1).

Fig. 4 shows the fitting parameters ρ_0 and b from the Equation 1 which is fitted to the x=0.3 (**Fig. 2**) and 0.4 (**Fig. 3**). The decreasing trend of ρ_0 and b indicates weakening of spin flip scattering between conduction electrons and Ce-4f electrons while increasing the applied magnetic field strength. Commonly, for x=0.3 and 0.4, the low temperature resistivity takes a down turn after an initial rise in presence of higher magnetic fields at ~6 T (x=0.3) and ~8 T (x=0.4), respectively. The temperature at which down-turn ($T_{\text{down-turn}}$) occurs, is gradually shifted towards high temperatures upon increasing the magnetic field strength. Such a behavior can be understood in the following manner. In general, this down turn is a common feature of heavy Fermion compounds (based on Kondo route) **[12-16]** which show Fermi liquid behavior. The external magnetic field gradually shifts the $T_{\text{down-turn}}$ to high temperatures. It suggests that compounds with x=0.3 and 0.4 have heavy Fermion like character. In order to see this sharp fall of resistance at zero field one has to extend measurements below 2 K.

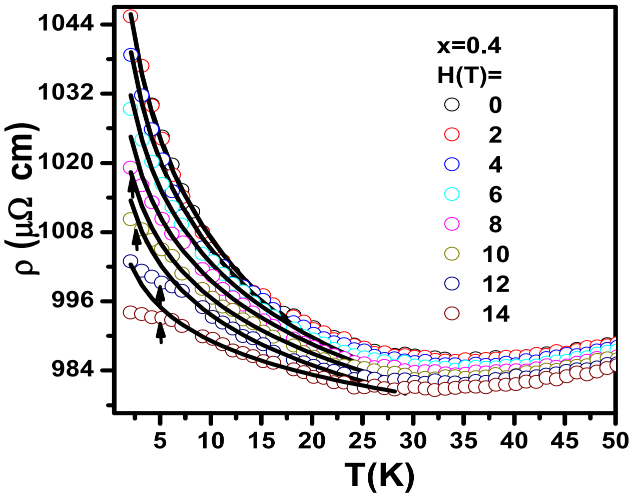

Fig. 3. Resistivity vs temperature of x=0.4 at low temperatures in the magnetic field up to 14 T are fitted to the kondo model with Equation 1.

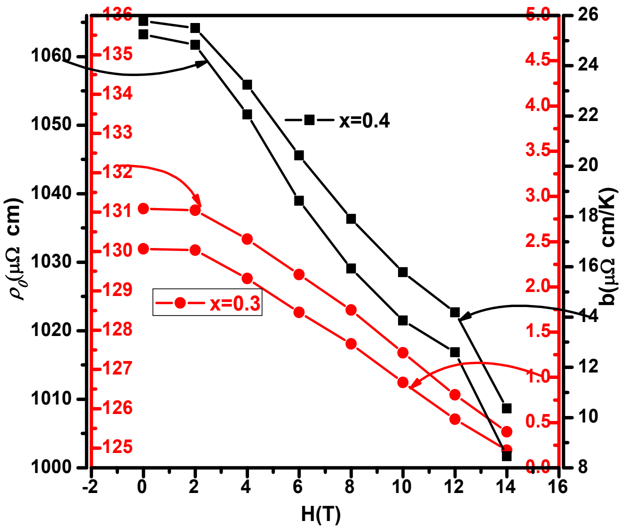

Fig. 4. ρ_0 and b vs H of $Ce(Ni_{1-x}Cu_x)_2Al_3$ with x=0.3(red) and 0.4(black) using equation 1.

Linear temperature coefficient of thermoelectric power which is obtained from the linear fit to the data (as shown in **Fig. 5**) is plotted in **Fig. 6**. The γ_0, the enhanced electronic specific heat coefficient, is estimated by extrapolating the low temperature rise of C_p/T vs T^2 down to 0K. The plots have been reported in our earlier communication **[9]**. Obtained γ_0 values are in agreement with literature **[6, 8]**. Such enhancement of γ_0 up to 269 mJ/mole-K^2 suggests $Ce(Ni_{1-x}Cu_x)_2Al_3$ compound to be heavy Fermion-like character.

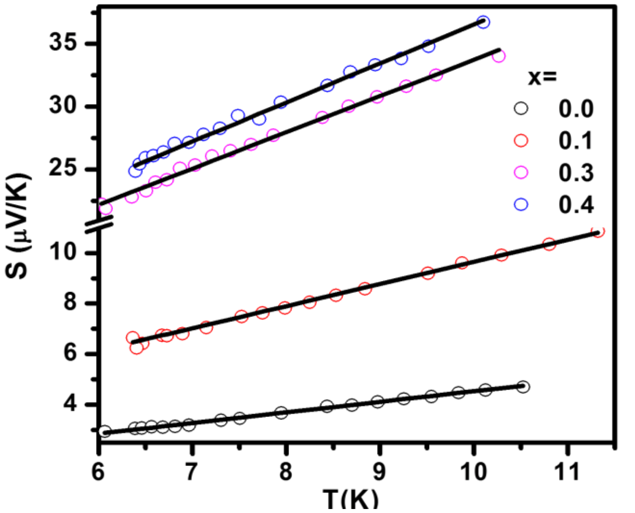

Fig. 5. Seebeck coefficient vs temperature of Ce(Ni1-xCux)2Al3 with x=0.0-0.4 below 11 K, which is fitted with linear equation.

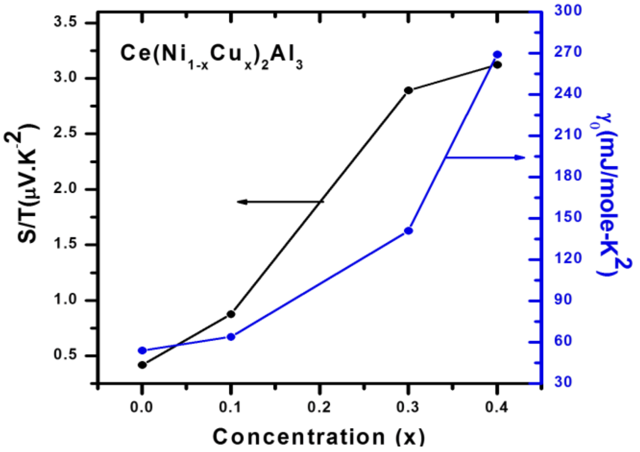

Fig. 6. Slope of S(T) and electronic specific heat coefficient (γ_0) vs doping concentration (x) of $Ce(Ni_{1-x}Cu_x)_2Al_3$ with x=0.0-0.4.

We have extracted carrier concentration 'n' using specific heat and thermoelectric power. In this regard, the Sommerfeld coefficient is estimated by extrapolating the high temperature specific heat. The relation between Sommerfeld coefficient (γ) of heat capacity and linear temperature coefficient of thermoelectric power as T→0 K, which gives the carrier concentration 'n' from the Equation 3 **[17]**. The γ values are obtained from our previous reports **[9]** on this series are shown in table.

$$\frac{S}{T} = \frac{C/T}{ne} \qquad (2)$$

$$n = \frac{\gamma}{(S/T)e} \qquad (3)$$

At low temperatures, as the temperature decreases TEP decreases linearly which is shown in figure 6. The linear fit to the temperature dependent thermoelectric power below about ~11 K is shown in figure 6. Obtained fit parameters along with γ are shown in **Table 1**.

Table 1. Sommerfeld coefficient γ and linear temperature coefficient of the thermoelectric power S/T of $Ce(Ni_{1-x}Cu_x)_2Al_3$ with x=0.0-0.4 are obtained from the our earlier report [9] and linear fit from Fig. 5 respectively.

Concentration	γ	S/T	*n*
x	(J/mole-K^2)	(μV/K^2)	per gr
0.0	0.00271	0.418	1.2×10^{20}
0.1	0.00953	0.870	2.0×10^{20}
0.3	0.0505	2.893	3.2×10^{20}
0.4	0.0835	3.124	4.9×10^{20}

From the **Table 1** it is clear that the carrier concentration ‘n’ increases with increasing Cu doping at Ni site and it falls in between common metals and semiconductors. Especially this region of concentration is suitable for obtaining the figure of merit. The suggested ‘n’ value for achieving high figure of merit (ZT) is between 10^{19} and 10^{21} carriers per cm^3 **[18]**.

Conclusion

The resistivity of polycrystalline $Ce(Ni_{1-x}Cu_x)_2Al_3$, for x=0.0 to 0.4 is studied in the magnetic fields up to 14 T. Positive magnetoresistance is seen for compensated metallic samples (x=0.0 and 0.1) throughout the whole temperature range, whereas x=0.3 has shown negative magnetoresistance due to suppression of –ln*T* behavior and positive magnetoresistance because of its Pauli paramagnetic nature below and above 15 K, respectively. For x=0.4, it’s Ce^{+3} ion with local 4f moment is exposed. Hence, negative magetoresistance is observed. Deviation from the –ln*T* rise at low temperatures is clearly seen for x=0.3 and 0.4. The resistivity down-turn at low temperatures in magnetic fields can be understood as formation of Fermi liquid. Carrier concentrations are obtained from the doping dependence of linear diffusion coefficient of thermopower and the Sommerfeld coefficient of specific heat. They are indicative towards the formation of heavy Fermion character of the carriers. The study points toward a future prospective in rare earth based intermetallic materials for low temperature thermo electric applications.

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