



## Effects of La dilution on the CeNiAl<sub>4</sub> Kondo lattice

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### ARTICLE INFO

#### Article history:

Received 28 January 2010  
Received in revised form 9 April 2010  
Accepted 24 April 2010  
Available online 6 May 2010

#### Keywords:

Intermetallics  
Heavy fermions  
Kondo effect  
Magnetic properties  
Electrical resistivity  
Heat capacity

### ABSTRACT

We carried out the magnetic, electrical resistivity and heat capacity measurements for the Ce<sub>1-x</sub>La<sub>x</sub>NiAl<sub>4</sub> compounds. The values of the effective paramagnetic moment  $\mu_{\text{eff}}$  were calculated from the Curie–Weiss constant. For all the solid solutions under study the  $\mu_{\text{eff}}$  values are very close to  $2.54\mu_{\text{B}}$  found for cerium in the pure Ce metal. Resistivity studies on the Ce<sub>1-x</sub>La<sub>x</sub>NiAl<sub>4</sub> alloy system illustrate the transition from the Kondo regime ( $x=0.0$ ) to the single-ion Kondo regime ( $0.05 \leq x \leq 0.8$ ). The substitution of Ce by La reduces the electronic heat capacity coefficient  $\gamma$  values from  $154 \text{ mJ/mol K}^2$  for CeNiAl<sub>4</sub> to  $4.8 \text{ mJ/mol K}^2$  for LaNiAl<sub>4</sub>. At low temperatures  $\gamma$  value depends strongly on the temperature range used for the extrapolation.

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### 1. Introduction

The anomalous nature of *f* electrons in the compounds and alloys of lanthanides is affected not only by the distance between the atoms, but also by the electronic structure of the neighbors. A strong interaction between the *4f* electrons and conduction electrons is competitive for both intra-atomic (correlation effects) and inter-atomic interactions. These impacts lead to the emergence of such phenomena as the Kondo effect, heavy fermions, mixed valence, spin fluctuations and unusual magnetic orderings. The occurrence of these phenomena depends primarily on the location of the level of *4f* electrons relative to the Fermi level, hence it is important to know the band structure of the compound. In contrast to the well-localized *4f* electrons of heavy rare earths, in the Ce compounds a high degree of hybridization with the conduction electrons occurs, which can cause delocalization of the *4f* states. Depending on the strength of the hybridization and the position of the *4f* level in respect to the Fermi level, these compounds may exhibit a different type of properties.

Previously, CeNiAl<sub>4</sub> was reported to crystallize in the orthorhombic YNiAl<sub>4</sub>-type structure (space group *Cmcm*) and to exhibit properties typical of a Kondo lattice or heavy fermion system [1–4]. The magnetic susceptibility that follows the Curie–Weiss law in the range 30–250 K suggests a localized *4f* electron character [3]. The electrical resistivity  $\rho(T)$  shows a Kondo-like logarithmic

increase down to a maximum at  $T=100 \text{ K}$  and then a steep decrease towards low temperature. This behaviour indicates that the resistivity is caused by the Kondo effect [3]. The thermoelectric power (TEP) of CeNiAl<sub>4</sub> is positive over the whole temperature range. The electrical resistivity and thermopower data are characteristic of the Kondo lattice. The thermoelectric measurements allowed us to locate the *4f* peak slightly above the Fermi level. The TEP and magnetic measurements provide Kondo temperature  $T_K=40 \text{ K}$  and  $33 \text{ K}$ , respectively, in good agreement with that deduced from the  $C_p$  data ( $37 \text{ K}$ ). A large electronic specific heat coefficient  $\gamma=154 \text{ mJ/mol K}^2$  is observed at minimum of the  $C_p/T$  vs.  $T^2$  dependence. Extrapolation of the lowest temperatures range of  $C_p/T(T^2)$  yields the  $\gamma$  value of  $0.5 \text{ J/mol K}^2$ . In combination with the thermoelectric power and resistivity data, a heavy fermion state is confirmed in CeNiAl<sub>4</sub>. Recently the effect of Y dilution in CeNiAl<sub>4</sub> was also investigated [5].

The present paper concerns on the properties of the Ce<sub>1-x</sub>La<sub>x</sub>NiAl<sub>4</sub> system studied by the magnetic susceptibility, electrical resistivity and specific heat measurements.

### 2. Experimental

The Ce<sub>1-x</sub>La<sub>x</sub>NiAl<sub>4</sub> compounds were prepared by induction melting of the stoichiometric amounts of the constituent elements in a water-cooled boat, under an argon atmosphere. The ingot was inverted and remelted several times to ensure homogeneity. The unit cell volume  $V$  for Ce<sub>1-x</sub>La<sub>x</sub>NiAl<sub>4</sub> increases with increasing the La concentration.

Heat capacity measurements were performed by PPMS commercial device (Quantum Design) in the temperature range 1.9–300 K by relaxation method using the two- $\tau$  model. The magnetic susceptibility and the magnetization curves were measured also on the PPMS system. The electrical resistivity was measured on a bar-shaped sample using a standard four-probe technique.

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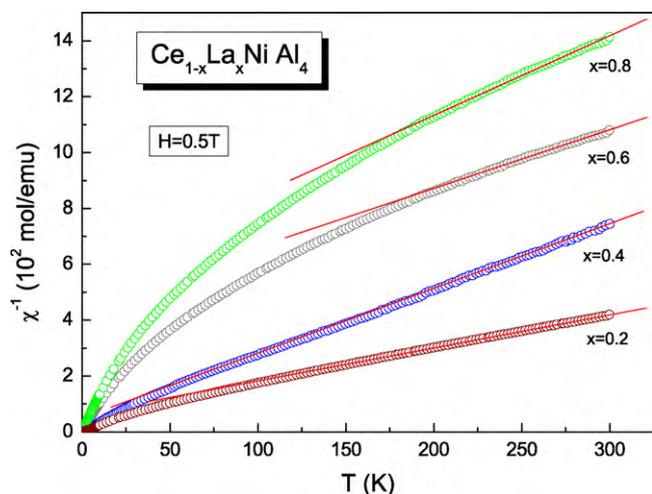


Fig. 1. Inverse magnetic susceptibility  $\chi^{-1}(T)$  of the  $\text{Ce}_{1-x}\text{La}_x\text{NiAl}_4$  compounds.

### 3. Results and discussion

Fig. 1 shows the temperature dependence of the inverse magnetic susceptibility  $\chi^{-1}(T)$  for various compositions of  $\text{Ce}_{1-x}\text{La}_x\text{NiAl}_4$  measured in an applied field of 0.5 T. Typically, the susceptibility of cerium compounds in the paramagnetic region follows the Curie–Weiss behavior  $\chi(T) = C/(T - \theta_p)$ . Here  $C$  is the effective Curie constant of the Ce ions and  $\theta_p$  is the paramagnetic Curie temperature. The resulting values of  $\mu_{\text{eff}}$  and  $-\theta_p$  are given in Table 1. For  $\text{CeNiAl}_4$  above about 30 K,  $\chi^{-1}(T)$  follows the Curie–Weiss law with the effective magnetic moment  $\mu_{\text{eff}} = 2.45 \mu_{\text{B}}/\text{f.u.}$  and the paramagnetic Curie temperature  $\theta_p = -66 \text{ K}$  [4]. For all the solid solutions under study the  $\mu_{\text{eff}}$  values calculated from the Curie–Weiss constant are very close to the value of  $2.54 \mu_{\text{B}}$  found for cerium in the pure Ce metal. Values of the paramagnetic Curie temperatures  $\theta_p$  vary in an irregular manner across the alloy series. This anomalous result is likely due to different degrees of preferred crystalline orientation in the different alloy samples. However, the general tendency is that  $\theta_p$  increases with  $x$ , which results from the transition to the single-ion Kondo regime for  $x > 0.0$ . For many cerium compounds, substitution of Ce by La leads to a decrease of both the Kondo temperature (due to the lattice expansion) and the magnetic transition temperature (due to the dilution), which in turn can lower the overall value of  $\theta_p$ . Large negative values of paramagnetic Curie temperature indicates on the existence of a strong negative interaction between the  $4f$  spins of electrons and the conduction electrons. The slight deviation of  $\mu_{\text{eff}}$  from the free  $\text{Ce}^{3+}$  value is ascribed to the effect of crystal electric field (CEF).

Since  $\text{CeNiAl}_4$  is a Kondo lattice (as inferred from the thermal variation of its resistivity, discussed below) there is a contribution to negative  $\theta_p$  from the Kondo interaction.

The magnetic field dependence of magnetization of  $\text{Ce}_{0.2}\text{La}_{0.8}\text{NiAl}_4$  and  $\text{Ce}_{0.8}\text{La}_{0.2}\text{NiAl}_4$  for different temperatures

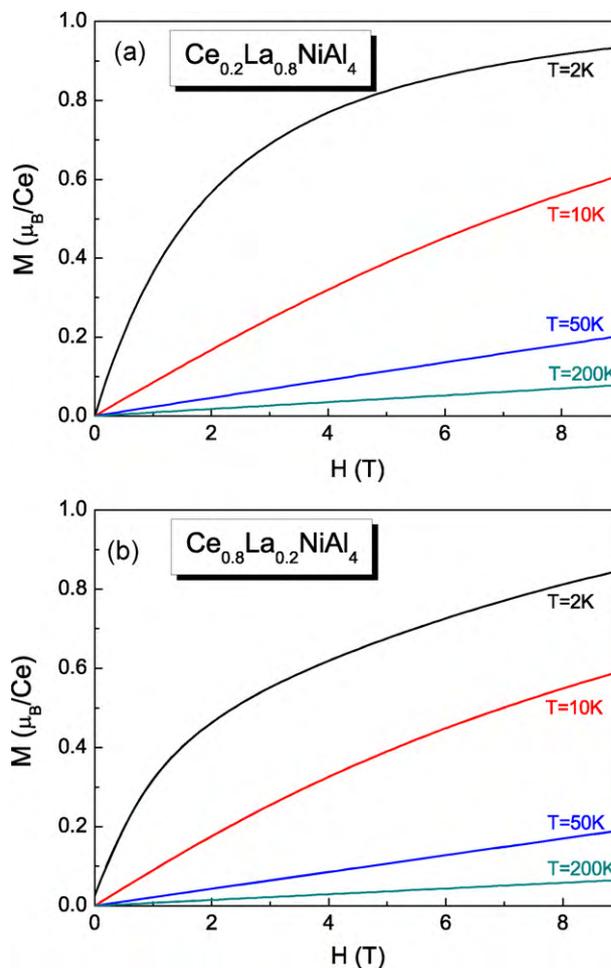


Fig. 2. The magnetic field dependence of magnetization for  $\text{Ce}_{0.2}\text{La}_{0.8}\text{NiAl}_4$  and  $\text{Ce}_{0.8}\text{La}_{0.2}\text{NiAl}_4$  at different temperatures.

is presented in Fig. 2a and b. The temperature evolution of these curves confirms the absence of magnetic ordering down to 2 K.

The isothermal magnetization per Ce ion of selected  $\text{Ce}_{1-x}\text{La}_x\text{NiAl}_4$  at 2 K, measured up to 9 T is shown in Fig. 3. At  $T = 2 \text{ K}$  and at the maximum applied field of 9 T, the magnetization  $M$  decreases marginally from  $0.93 \mu_{\text{B}}/\text{f.u.}$  for  $\text{Ce}_{0.2}\text{La}_{0.8}\text{NiAl}_4$  to  $0.84 \mu_{\text{B}}/\text{f.u.}$  for  $\text{Ce}_{0.8}\text{La}_{0.2}\text{NiAl}_4$ . Saturation moments at  $H = 9 \text{ T}$  are much smaller than the theoretical values calculated for the free  $\text{Ce}^{3+}$  ion. The effect can be attributed to the important role of crystal electric fields.

The  $\rho(T)$  measurements as a function of temperature for the  $\text{Ce}_{1-x}\text{La}_x\text{NiAl}_4$  compounds are shown in Fig. 4, which illustrates a transition from the Kondo dense regime ( $x = 0.0$ ), through the single-ion Kondo regime ( $x \geq 0.05$ ), to the metallic behavior ( $x = 1.0$ ). The  $\rho(T)$  for the parent compound  $\text{CeNiAl}_4$  shows a Kondo-like logarithmic increase down to a maximum at  $T_{\text{max}} = 100 \text{ K}$  and then a steep decrease towards low temperatures. In the dense

**Table 1**  
The Weiss constants  $\theta_p$  and the effective magnetic moment  $\mu_{\text{eff}}$  obtained by fits of the Curie–Weiss relation to the data in Fig. 1. Values of  $A$  and  $c_K$  are extracted from fits with Eq. (1) shown by solid line in Fig. 5. These results correspond to measurements for  $x$  changing by 0.2 for  $\text{Ce}_{1-x}\text{La}_x\text{NiAl}_4$ .

$x$	$\theta_p$ (K)	$\mu_{\text{eff}}$ ( $\mu_{\text{B}}/\text{f.u.}$ )	Fitted range (K)	$A$ ( $\mu_{\Omega} \text{ cm}$ )	$c_K$ ( $\mu_{\Omega} \text{ cm}$ )	$\gamma$ (mJ/mol K <sup>2</sup> )
0.0	−66	2.45	30–250	400.6	61	154
0.2	−48	2.61	70–300	37.4	1.8	52
0.4	−35	2.58	50–300	40.4	1.5	22
0.6	−171	2.46	150–300	42.9	2.0	12
0.8	−184	2.42	170–300	33.2	1.4	8

Values of  $\theta_p$  and  $\mu_{\text{eff}}$  for  $\text{CeNiAl}_4$  are taken from Ref. [4].

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