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2009 J. Phys.: Condens. Matter 21 176004

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A new Kondo antiferromagnet $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$

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Received 29 December 2008, in final form 11 March 2009

Published 30 March 2009

Online at stacks.iop.org/JPhysCM/21/176004

Abstract

We find that $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ crystallizes in the hexagonal AlB_2 -type structure with lattice parameters $a = 0.4850(5)$ nm and $c = 0.3908(5)$ nm. Magnetic susceptibility, electrical resistivity and low-temperature specific heat data reveal that the bulk phase transition at 3.7 ± 0.2 K in $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ is to an antiferromagnetic state. The magnetic contribution to the resistivity ρ_{mag} of $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ increases as $\ln(T)$ when temperature is lowered from room temperature and reaches a plateau at 9 K, followed by a rapid decrease around 4 K. These results associated with a reduction of the Ce magnetic moment and of the magnetic entropy at T_N suggest that $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ could be a Kondo antiferromagnet. The Kondo temperature is estimated to be of order 6 K.

(Some figures in this article are in colour only in the electronic version)

The low-temperature electronic, magnetic, and thermal properties of dense Kondo systems have attracted much interest during the past decades [1, 2]. In the course of our search for new materials compounds, we have found that $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ displays properties of both antiferromagnetic ordering and dense Kondo systems. In this paper, we report the crystal structure, the temperature dependence of the magnetic susceptibility, electrical resistivity and heat capacity of $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$. As determined from ac susceptibility, electrical resistivity and heat capacity measurements, the compound $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ exhibits antiferromagnetic order below 3.7 ± 0.2 K. In addition to experiments on $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$, electrical resistivity and heat capacity measurements on the isostructural $\text{La}(\text{Ni}_{0.25}\text{In}_{1.75})$ have been made in order to estimate the phonon contribution to the resistivity and heat capacity of $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$.

Polycrystalline samples investigated for this work were synthesized by arc melting together stoichiometric amounts of the constituent elements on a water-cooled Cu hearth in 1 atm of ultrahigh-purity argon gas in which a Zr button used as an oxygen getter had been previously arc melted. The 3N purity rare-earth elements, 4N purity nickel (Ni) and 5N purity indium (In) were purchased from Alfa Aesar, a Johnson Matthey company. Weight losses

during arc melting were less than 0.2%. To ensure that no impurity phase was present in the sample, each sample was wrapped in tantalum foil, sealed under argon in a quartz tube and annealed for four weeks at $590 \pm 20^\circ\text{C}$. This heat treatment was followed by a water quench to room temperature. Analysis of powder x-ray diffraction patterns indicates that the samples crystallize in a hexagonal AlB_2 -type structure [3] with space group $P6/mmm$. No traces of secondary phase were observed, as shown in figure 1. One formula unit of $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ (or $\text{LaNi}_{0.25}\text{In}_{1.75}$) is present in the unit cell. The crystallographic parameters were determined by using the PowderCell program [4]. The lattice parameters $a = 0.4850(5)$ nm, $c = 0.3908(5)$ nm for $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ and $a = 0.4850(2)$ nm, $c = 0.3963(8)$ nm for $\text{La}(\text{Ni}_{0.25}\text{In}_{1.75})$ were then obtained. In this refinement to determine lattice parameters, only the positions of the x-ray lines were required. A comparison of the calculated line intensities with those established experimentally showed reasonable agreement, especially considering the nature of powder x-ray data for a quantitative determination of line intensities. In this likeness, we used standard atomic positional parameters for the atoms in the unit cell. Specifically, the rare-earth atom was placed in the 1a position while the atoms Ni and In were randomly located in the 2d position of the space group $P6/mmm$ (D_{6h}^1) with the following fractional coordinates:

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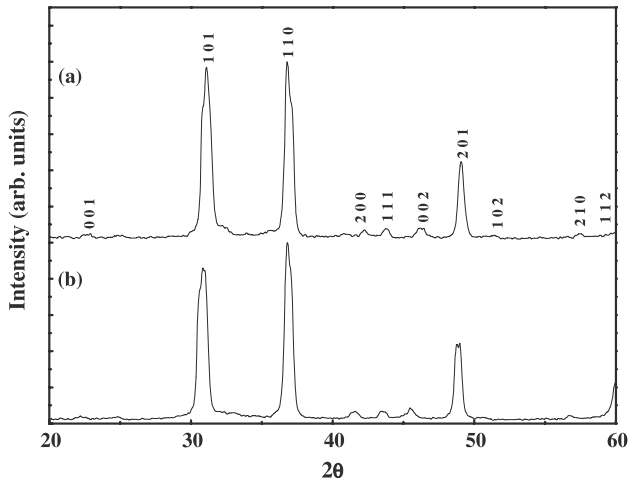


Figure 1. Room-temperature powder x-ray diffraction patterns of (a) $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ and (b) $\text{La}(\text{Ni}_{0.25}\text{In}_{1.75})$.

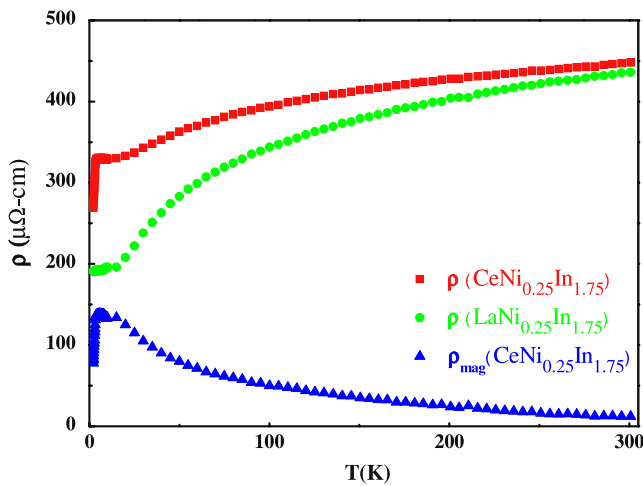


Figure 2. Resistivity versus temperature of $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$, $\text{La}(\text{Ni}_{0.25}\text{In}_{1.75})$, and the difference between these two compounds from 1.8 to 300 K.

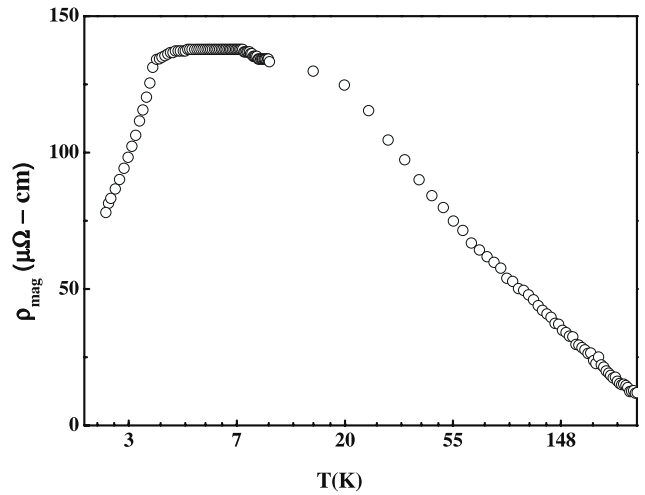


Figure 3. Magnetic resistivity ρ_{mag} versus $\ln(T)$ of $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ between 1.8 and 300 K.

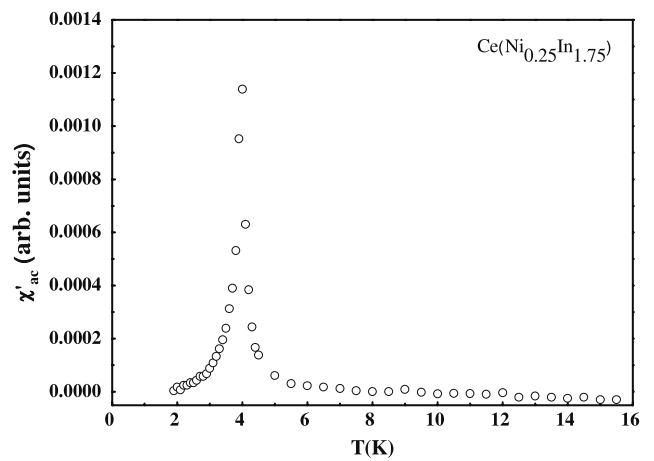


Figure 4. The temperature dependence of the real component of ac susceptibility for $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ in a driving field of 1.0 G at 1000 Hz.

Ce or La (0, 0, 0), 1/8 Ni and 7/8 In (1/3, 2/3, 1/2). The agreement of intensities indicates that no significant vacancy is present in our samples. Very interestingly, neither CeNi_2 (Cu_2Mg type) [5] nor CeIn_2 (CeCu_2 type) [6] is the parent compound of $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$.

Dc electrical resistivity measurements were made between 1.8 and 300 K using a standard four-probe technique in a system fully automated for temperature stability and data acquisition [7]. Figure 2 displays the temperature dependence of the resistivity of $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$, $\text{La}(\text{Ni}_{0.25}\text{In}_{1.75})$, and the magnetic resistivity ρ_{mag} of $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$. The magnetic resistivity was estimated by subtracting the resistivity of isostructural $\text{La}(\text{Ni}_{0.25}\text{In}_{1.75})$ from that of $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$. The data for ρ_{mag} are replotted as a function of $\ln(T)$ in figure 3. It is seen that the curve in figure 3 shows a logarithmic increase with decreasing temperature in the high-temperature region and reaches a plateau around 9 K. The $\ln(T)$ dependence in ρ_{mag} is due to the scattering processes of the conduction electrons with independent Kondo centers and is one of the

characteristic features of dense Kondo systems. Upon further cooling, ρ_{mag} decreases rapidly below ~ 4 K. This decrease of ρ_{mag} may be ascribed to both the onset of coherence between Kondo states at Ce sites and the magnetic ordering, because the rapid decrease of ρ_{mag} commences near the Néel temperature T_N as determined from the ac susceptibility and the specific heat data. We must point out here that the overall $\rho_{\text{mag}}(T)$ of $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ is reminiscent of that of $\text{CeCu}_{1.54}\text{Si}_{1.46}$ [8]. However, the compound $\text{CeCu}_{1.54}\text{Si}_{1.46}$ has larger residual magnetic resistivity ($\sim 150 \mu\Omega \text{ cm}$ below 3 K), because of the disorder in the Cu and Si sublattice, which weakens the coherence between Kondo states, and behaves as an incoherent Kondo system even at low temperatures.

Figure 4 presents the real part of ac susceptibility χ'_{ac} for the compound $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ measured in a driving field of 1.0 Oe and at a frequency 1000 Hz between 1.8 and 16 K with a commercial SQUID magnetometer. A very sharp transition from nonmagnetic to magnetic ordering is seen at 3.9 K (peak value). The data curve of the imaginary part

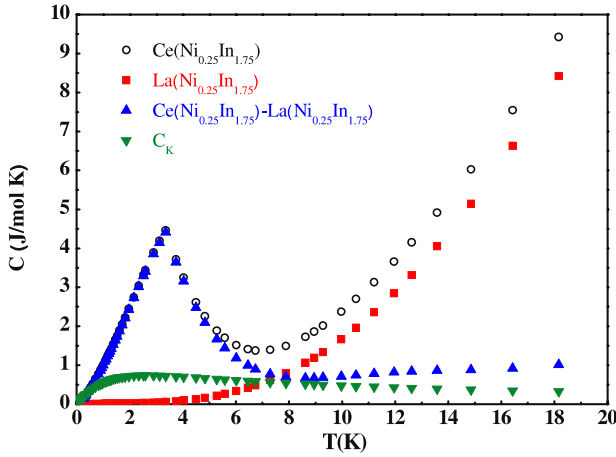


Figure 5. Heat capacity of $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$, $\text{La}(\text{Ni}_{0.25}\text{In}_{1.75})$, the difference between these two compounds C_{mag} and the Kondo model fit C_K between 0.35 and 20 K. The data curve for the sample $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ shows a rather broad λ type of antiferromagnetic ordering peak centering at 3.6 K and a minimum around 7 K. It is implied that the inhomogeneity of the sample is due to the disordered Ni and In atoms arranged in the 2d position of the space group $P6/mmm$.

χ''_{ac} is not shown here because it has similar shape and its peak occurs at slightly different temperature (~ 3.8 K) from that of the real part χ'_{ac} . Since no frequency dependence of the position of the peak value in the $\chi'_{\text{ac}}(T)$ or $\chi''_{\text{ac}}(T)$ curve is observed, the possibility of spin-glass behavior in this compound $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ is excluded. In fact, the high-temperature dc magnetic susceptibility data ($T > 100$ K) can be fitted to a linear Curie–Weiss law with an effective moment of $2.6 \pm 0.1 \mu_B$ per cerium atom and a paramagnetic Curie temperature -22 K. This value of μ_{eff} is sufficiently close to the free Ce^{3+} ion value to preclude intermediate valence. Earlier experience may tell us that for a compound containing 4f light rare earth and 3d (e.g. Mn, Fe, Co, Ni) intermetallics the spins S couple anti-parallel between S_{3d} and S_{4f} . The magnetic moment, however, depends upon the coupling between S_{3d} and J_{4f} . $J = L - S$ for the light rare earths from Hund's rule and as a result J_{4f} is coupled parallel to S_{3d} . We must point out here that there seems to be not much influence of Ni on total magnetic moment in the compound $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$, probably due to the low Ni concentration (~ 8.3 at.%). In fact, the low concentration of Ni in the compound $\text{La}(\text{Ni}_{0.25}\text{In}_{1.75})$ also shows neither magnetic order nor superconductivity above 350 mK according to specific heat measurements. It is noted here that for the higher Ni concentration compound $\text{Ce}(\text{Ni}_{0.5}\text{In}_{1.5})$ (or Ce_2NiIn_3) with the same AlB_2 -type structure [9], spin-glass-like features arise below the freezing temperature $T_{\text{sf}} \sim 2.8$ K together with the Kondo and crystal field effects [10].

The low-temperature specific heat measurements were made using a thermal relaxation microcalorimeter. Detailed measurement procedures can be found in [11, 12]. As illustrated in figure 5, the heat capacity versus T data curve for the sample $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ measured in zero field between 0.35 and 20 K shows a λ type of antiferromagnetic ordering

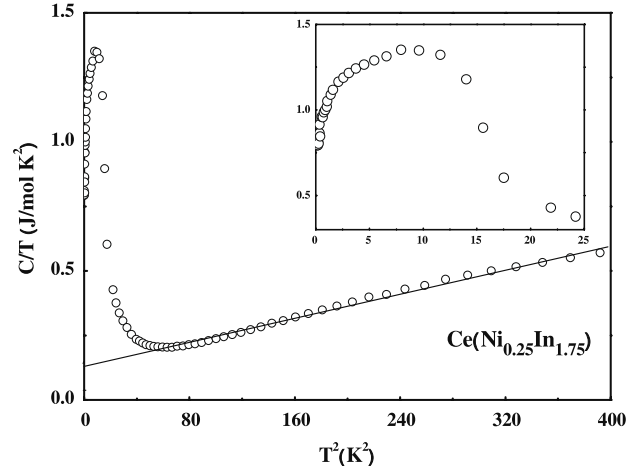


Figure 6. Specific heat divided by temperature C/T versus T^2 of $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ between 0.35 and 20 K; inset, C/T versus T^2 between 0.35 and 5 K.

peak centering at 3.6 K and a peak value approaching $4.5 \text{ J mol}^{-1} \text{ formula K}$. The agreement of transition temperatures measured by ac susceptibility and heat capacity techniques are clear evidence of bulk antiferromagnetism in $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$. We note here that the jump in the heat capacity is much smaller than the value $15 \text{ J mol}^{-1} \text{ Ce K}$ as expected on the basis of the mean-field theory. Two factors may affect this large reduction in heat capacity jump at the magnetic transition. One is the inhomogeneity of the sample, which results in a rather broad λ transition, due to the disordered Ni and In atoms arranged in the 2d position of the space group $P6/mmm$, and the other one is attributed to the Kondo effect. A determination of the electronic and lattice heat capacity contributions is complicated by the existence of the antiferromagnetic ordering peak. The specific heat data in the form of C/T versus T^2 for $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ are plotted in figure 6. It is seen that the heat capacity $C(T)$ of $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$, in the paramagnetic state at temperatures between 8 and 20 K, can be fitted to the expression $C_n = \gamma T + \beta T^3$ by a least squares analysis, which yields a γ value of about $130 \text{ mJ mol}^{-1} \text{ K}^{-2}$ and a β value of about $1.2 \text{ mJ mol}^{-1} \text{ K}^{-4}$ or a Debye temperature $\Theta_D = 170$ K. As was previously pointed out [13], such high γ values, which exceed those of normal metals by nearly two orders of magnitude, are indications of Kondo interactions. Similar γ values have been observed in the Kondo lattice systems CeAl_2 [13], CeCuAl_3 [14], CeCu_2 [15], CeCu_5 [16], CePtSi_2 [17] etc. Since the Ruderman–Kittel–Kasuya–Yoshida (RKKY) interactions are responsible for long-range magnetic ordering and the Kondo-type interactions favor a nonmagnetic ground state, the sharp drop in the $\rho_{\text{mag}}(T)$ curve that marks most of the magnetic order at low temperatures tends to obscure the Kondo maximum when $T_K \sim T_m$. Thus, as shown in figure 3, a rough estimate of Kondo temperature T_K by using the temperature of the maximum [18] in the $\rho_{\text{mag}}(T)$ curve should occur between 4 and 9 K for the compound $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$. Another estimation comes from the low-temperature heat capacity data [19, 20]. As depicted in figure 5, we mark the low-temperature magnetic

contribution as $C_{\text{mag}} = C_{\text{tot}} - C_{\text{ph}} - C_{\text{cryst}} = C_{\text{K}} + C_{\text{AF}}$. The C_{cryst} term can be neglected because no Schottky type anomaly in the specific heat data is seen below 20 K for the compound $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$. This Kondo model fit leads to $T_{\text{K}} = 6$ K, which is consistent with the estimate based on magnetic resistivity data. Using numerical integration, we estimate the magnetic entropy as a function of temperature from $S_{\text{M}} = \int_0^T (C_{\text{mag}}/T) dT$. The magnetic entropy $S_{\text{M}}(T_{\text{N}})$ associated with a modulated magnetic structure for $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$ is only 62% of $R \ln(2)$, corresponding to a reduction of 38% of the cerium moment. The temperature at which $S_{\text{M}} = R \ln 2$ is 14 K, in good agreement with estimates of the Kondo temperature for this material.

In conclusion, the intermetallic compound $\text{Ce}(\text{Ni}_{0.25}\text{In}_{1.75})$, which crystallizes in the AlB_2 -type structure, exhibits antiferromagnetic order below $T = 3.7 \pm 0.2$ K. This compound is found to be a Kondo lattice compound with $T_{\text{K}} \sim 6$ K from the unusual resistivity and specific heat data.

Acknowledgment

Research at Chung Cheng University was supported by the National Science Council of the Republic of China under contract no NSC 96-2112-M-194-008-MY3.

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