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Journal of Alloys and Compounds 323–324 (2001) 435–439

Journal of ALLOYS AND COMPOUNDS

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Magnetoresistance behaviour of the ternary stannides $CeNi_{0.86}Sn_2$ and $Ce_3Ni_2Sn_7$

B. Chevalier[a,*], L. Durivault[a], J. Rodriguez Fernandez[b], J.C. Gomez Sal[b], J.M. Barandiaran[c], J. Etourneau[a]

[a]*CNRS (UPR 9048), Institut de Chimie de la Matière Condensée de Bordeaux (ICMCB), Avenue du Dr. A. Schweitzer, 33608 Pessac Cedex, France*
[b]*Facultad de Ciencias, Universidad de Cantabria, 39005 Santander, Spain*
[c]*Facultad de Ciencias, Universidad del Pais Vasco, 40080 Bilbao, Spain*

Abstract

In order to obtain more information on the magnetic phase diagram of these stannides, we have performed magnetization and magnetoresistance (MR) measurements. The results are summarized as follows: (i) in the paramagnetic range, the negative curvature of the $MR = f(\mu_0 H)$ curve for $CeNi_{0.86}Sn_2$ reflects the existence of Kondo interactions; for $T < 8$ K, a change of regime appears reminiscent of the onset of ferromagnetic ordering showing that the antiferromagnetic structure of this stannide became easily ferromagnetic under low applied fields; (ii) MR of $Ce_3Ni_2Sn_7$ is positive for $T > 12$ K then changes sign showing that the Kondo effect is rather small for $T < 12$ K; below 7 K MR becomes more negative with decreasing temperature confirming the metamagnetic transition observed at low fields by magnetization measurements.

Keywords: Intermetallics; Kondo effects; Electronic transport; Magnetic measurements

1. Introduction

The ternary stannides $CeNi_{1-x}Sn_2$ and $Ce_3Ni_2Sn_7$ present some structural similarities: for instance in both case a Sn-atom is located inside a trigonal prism [Ce_6] whereas the antiprism [Ce_4Sn_4] is occupied by a Ni-atom [1]. These two intermetallics order antiferromagnetically at low temperature: (i) $CeNiSn_2$ and $CeNi_{0.85}Sn_2$ order antiferromagnetically below $T_N = 3.9$–4.0 K but exhibit a second magnetic phase transition around 3.2–3.0 K [2,3]; (ii) a comparable Néel temperature $T_N = 3.9$ K is determined for $Ce_3Ni_2Sn_7$ [1].

In order to obtain more information on the influence of the Kondo interactions on the magnetic phase diagram of these stannides, we have performed magnetization and magnetoresistance (MR) measurements. We discuss the results considering those described previously on the other ternary stannides $CeNi_5Sn$, $CeNi_2Sn_2$, $CeNi_4Sn_2$ and Ce_2Ni_2Sn [4,5].

2. Experimental procedure

The samples were prepared by melting the constituent elements (purity > 99.9%) in an induction levitation furnace under a purified argon atmosphere. In order to ensure homogeneity, the ingots were turned over and remelted several times. Then they were sealed in evacuated quartz tubes and annealed at 1073 K for 1 month.

Microprobe analysis was used to check both the homogeneity and the composition of the obtained samples. The analysis was based on measurements of the Ce $L\alpha_1$, Ni $K\alpha_1$ and Sn $L\alpha_1$ X-ray emission lines, which were compared with those obtained for CeNiSn used as reference. Our analysis confirms that $Ce_3Ni_2Sn_7$ is obtained as single phase having ideal stoichiometry. In contrast, the $CeNiSn_2$ sample contains two phases: (i) a major phase having $CeNi_{0.86(2)}Sn_2$ as chemical composition; this formula is close to that determined previously by Schobinger-Papamantellos et al. [6] and indicates a slight Ni deficiency; (ii) small amounts of CeNiSn are detected. In contrast, the $CeNi_{0.86}Sn_2$ sample, used in this study, is obtained without impurity phase.

The two samples were characterized by X-ray powder diffraction (Guinier camera, Cu $K\alpha_1$ radiation). This examination shows that $Ce_3Ni_2Sn_7$ and $CeNi_{0.86}Sn_2$ adopt,

*Corresponding author. Tel.: +33-5-5684-6336; fax: 33-5-5684-2761.
E-mail address: chevalie@icmcb.u-bordeaux.fr (B. Chevalier).

0925-8388/01/$ – see front matter
PII: S0925-8388(01)01069-6

respectively, the orthorhombic $La_3Co_2Sn_7$- and $CeNiSi_2$-type structures.

Magnetization, AC- and DC-magnetic susceptibility were carried out down to 1.8 K using a superconducting quantum interference device (SQUID) magnetometer. Magnetoresistivity measurements were performed down to 2 K for applied magnetic fields $0\ T \leq \mu_0 H \leq 9\ T$ using a PPMS quantum design system.

3. Results and discussion

Fig. 1 presents the thermal dependence of the real χ' and imaginary χ'' part of the AC-magnetic susceptibility of $Ce_3Ni_2Sn_7$ measured at different $\mu_0 H_{DC}$-magnetic fields. At low field, for instance $\mu_0 H_{DC} = 0.05\ T$, the curve $\chi' = f(T)$ exhibits a maximum at $T_N = 3.9(1)$ K (Fig. 1a). This magnetic transition is characterized by an absence of peak in the $\chi'' = f(T)$ curve and can be associated to an antiferromagnetic ordering state (Fig. 1b). This maximum observed in the $\chi' = f(T)$ curve shifts to lower temperatures with increasing field; for instance for $\mu_0 H_{DC} = 0.3$ and 0.45 T the curve reaches a maximum respectively at 3.5(1) and 3.0(1) K. For these last values of $\mu_0 H_{DC}$, the $\chi'' = f(T)$ curve shows a positive peak which reflects important energy losses in the magnetically ordered state. This is presumably connected with domain effects appearing for instance in ferromagnetic, ferrimagnetic or canted systems.

Metamagnetic transition of $Ce_3Ni_2Sn_7$ is observed in the magnetization versus field dependencies (Fig. 2). At temperatures close to $T_N = 3.9(1)$ K, for instance at 3.75 K, the magnetization increases linearly up to $\mu_0 H = 0.1$ T and then more rapidly at higher fields. The critical field of this metamagnetic transition is around 0.3–0.35 T for temperature range 2–3 K. In other words, the antiferromagnetic ordering of this ternary stannide is easily disturbed by application of magnetic field.

AC- and DC-magnetization measurements performed by us on $CeNi_{0.86}Sn_2$ stannide suggest the occurrence of antiferromagnetic transition at $T_N = 4.4(2)$ K and the presence of second transition having a ferromagnetic component at $T_1 = 2.8(2)$ K. T_N temperature is somewhat higher than that of 4.0 K (determined using neutron powder diffraction) reported for a $CeNi_{0.86}Sn_2$ sample [3]. The existence of these two magnetic transitions leads to different behaviours in the curves showing the field dependence of the magnetization of $CeNi_{0.86}Sn_2$ compound (Fig. 3): (i) at 4 K, just below T_N, a metamagnetic transition is detected for a critical field of 0.07 T; we note the absence of hysteresis below this field (see inset of Fig. 3); (ii) in contrast at 2 K, below the second magnetic transition appearing at T_1-temperature, the magnetization exhibits always a small hysteresis; at this temperature also a magnetic transition occurs near 0.06 T. This study indicates a different nature for the magnetic ordering detected respectively at T_N and T_1. This can be explained

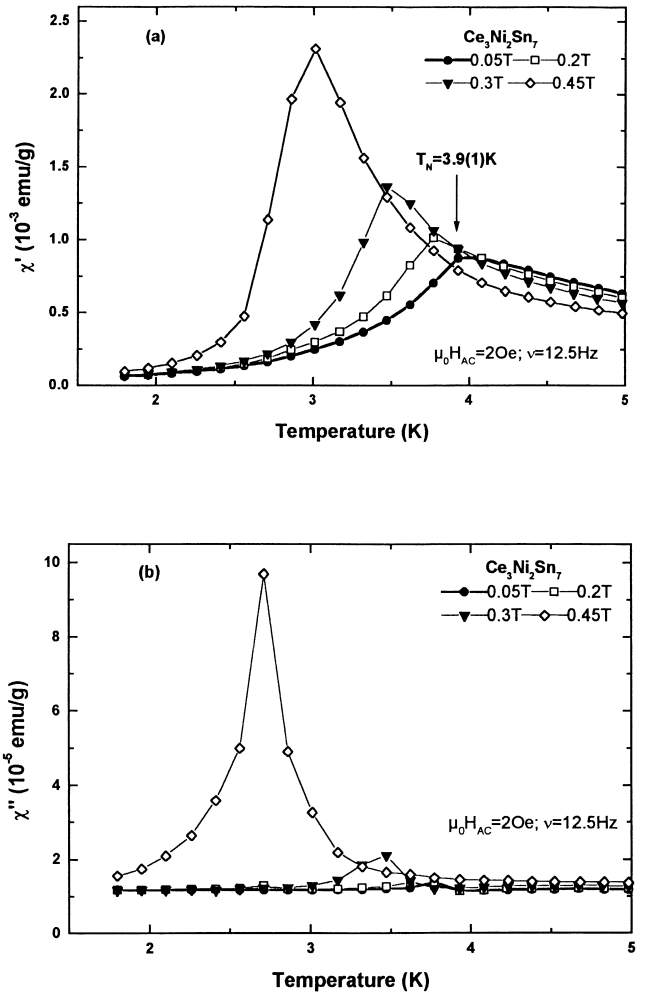


Fig. 1. Temperature dependence of the real (a) and imaginary (b) parts of the AC-magnetic susceptibility of $Ce_3Ni_2Sn_7$ measured at various $\mu_0 H_{DC}$-magnetic fields.

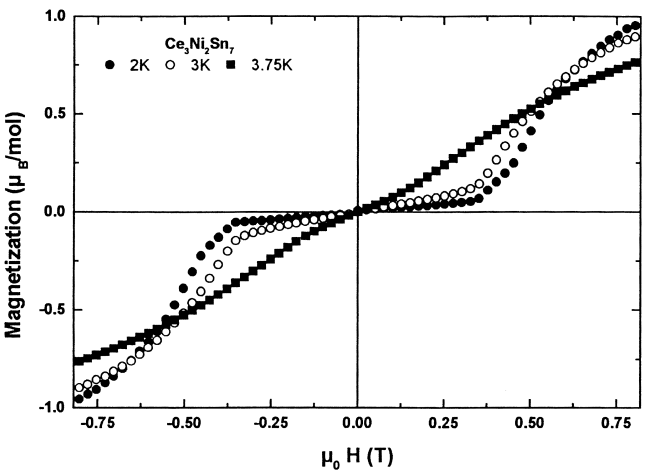


Fig. 2. Field dependence of the magnetization of $Ce_3Ni_2Sn_7$ measured at 3.75, 3 and 2 K.

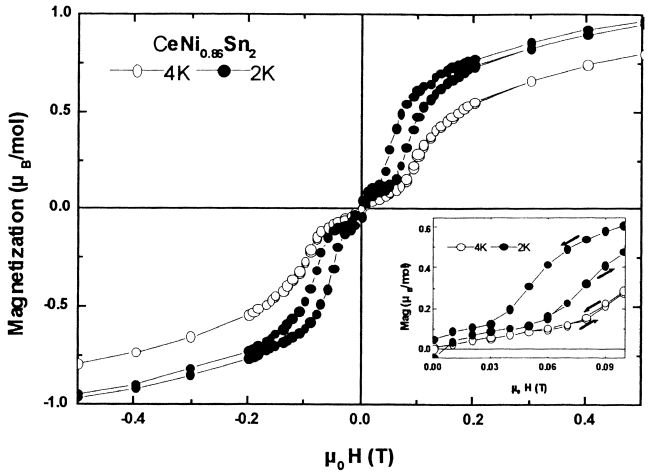
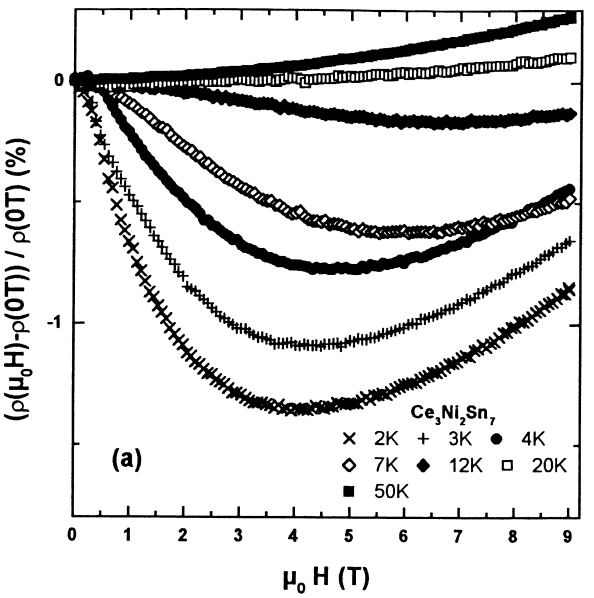


Fig. 3. Field dependence of the magnetization of $CeNi_{0.86}Sn_2$ measured at 4 and 2 K.

considering the neutron investigation performed on $CeNi_{0.86}Sn_2$ sample [3]: below T_1 two magnetic phases coexist, one is ferromagnetic and the second is a modulated antiferromagnet whereas above T_1 only an incommensurate antiferromagnet phase occurs.

Fig. 4 shows the low temperature part of the electrical resistivity of $Ce_3Ni_2Sn_7$ at various magnetic fields. The small decrease observed near 4 K is correlated with the occurrence of the antiferromagnetic ordering. This anomaly disappears completely with the application of the magnetic field of 3 or 6 T. This result indicates that the gap opening of the Fermi surface (formed due to antiferromagnetic ordering) disappears at higher fields. The decrease of the electrical resistivity with increasing magnetic field detected above T_N, in the temperature range T_N–7 K (Fig. 4), has two possible origins: the suppression of Kondo effect and/or the spin fluctuation due to the magnetic field.

The normalized magnetoresistance $(\rho(\mu_0 H) - \rho(0T))/\rho(0T)$ for $Ce_3Ni_2Sn_7$ as a function of applied field at various temperatures is given in Fig. 5a. Above 20 K, the magnetoresistance takes positive values. This behaviour indicates that the Kondo effect is inexistent in the temperature range $T > 20$ K and the magnetoresistance is domi-



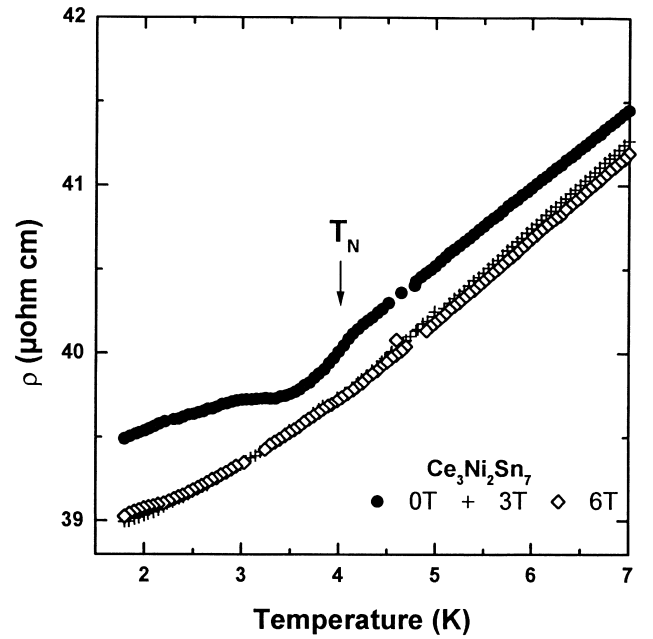


Fig. 4. Temperature dependence of the electrical resistivity of $Ce_3Ni_2Sn_7$ measured at various magnetic fields.

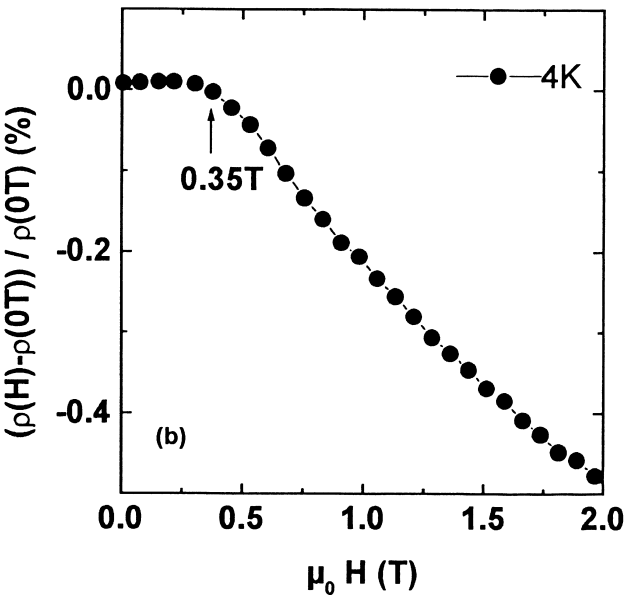


Fig. 5. Field dependence of the normalized magnetoresistance for $Ce_3Ni_2Sn_7$ measured at various temperatures (a) and in the magnetic ordered state 4 K $< T_N = 4.4$ K (b).

nated by the positive contribution due to the Lorentz force on the conduction electrons. In contrast, at 12 K the magnetoresistance changes sign at $\mu_0 H \cong 2$ T and always becomes negative in the temperature range $T < 12$ K. It is however clear that the magnitude of the magnetoresistance (MR) increases with decreasing temperature down to 2 K. Also, at 7 K, MR exhibits a minimum near 6 T and above this critical field MR increases, in positive direction, with field up to 9 T, the field at which MR exhibits a minimum decreases with decreasing temperature. This behaviour is typical of a ferromagnet [7]; in such a case, MR due to spin-disorder scattering would be very small and its upturn would result from the dominating influence of the magnetic field on the Fermi surface.

The characteristic feature, in the magnetic ordered state, for the field dependence of the magnetoresistance of $Ce_3Ni_2Sn_7$ is given in Fig. 5b. At 4 K, MR is practically independent of $\mu_0 H$ up to a critical field $\cong 0.35$ T, above which MR decreases rapidly. This behaviour is a signature of a metamagnetic transition revealed also by magnetization measurements (Fig. 2).

At low temperature and without applied field, the electrical resistivity of $CeNi_{0.86}Sn_2$ increases slightly between 17 and 5 K, and then decreases strongly (Fig. 6). This behaviour is typical of a magnetically ordered Kondo system. We note also that the electrical resistivity decreases always with increasing magnetic field in the temperature range 30–2 K. This fact is confirmed by Fig. 7 showing the field dependence of the normalized magnetoresistance (MR) of this stannide. At 25 K, in the paramagnetic range, the negative curvature of the $MR = f(\mu_0 H)$ curve reflects the existence of Kondo interactions as observed previously

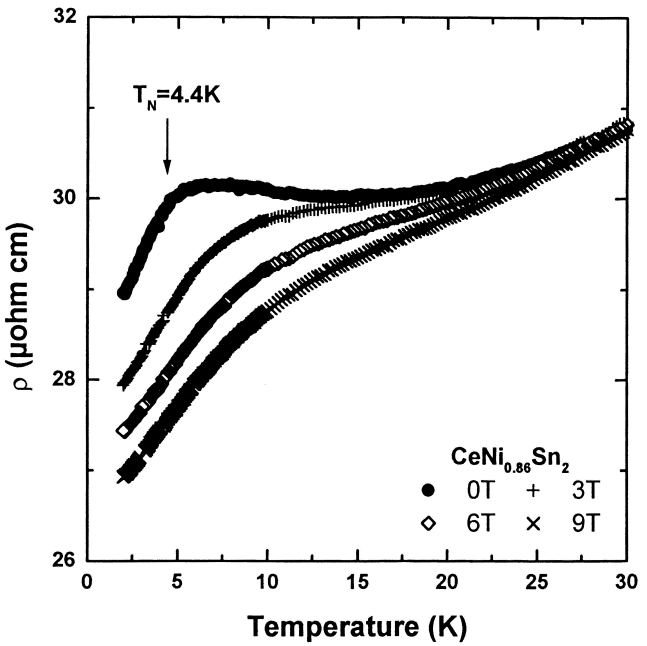


Fig. 6. Temperature dependence of the electrical resistivity of $CeNi_{0.86}Sn_2$ measured at various magnetic fields.

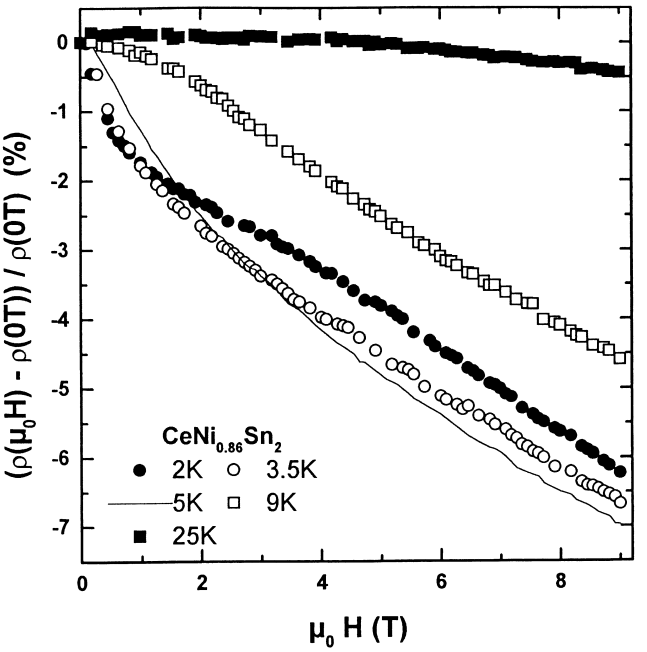


Fig. 7. Field dependence of the normalized magnetoresistance for $CeNi_{0.86}Sn_2$ measured at various temperatures.

for Ce_2Ni_2Sn compound [5]. As the temperature is lowered to 5 K, the magnetoresistance takes a large negative value of -7% at 9 T which is consistent with the magnetic field quenching of incoherent Kondo scattering. In the magnetic ordered state, for instance for 3.5 and 2 K, the magnetoresistance is negative showing that the magnetic structure of $CeNi_{0.86}Sn_2$ stannide became easily ferromagnetic under low applied field.

In conclusion, our study demonstrates that the antiferromagnetic ground state of $Ce_3Ni_2Sn_7$ and $CeNi_{0.86}Sn_2$ is easily destroyed; metamagnetic transition appears at very low fields. Moreover, the magnetoresistance measurements suggest that the Kondo interaction is rather small in $Ce_3Ni_2Sn_7$ but influences more strongly the physical properties of $CeNi_{0.86}Sn_2$ (just above $T_N = 4.4$ K, the magnetoresistance of this ternary stannide is largely negative).

Acknowledgements

The work was carried out with the financial support of the programme ‘Fonds Commun de Coopération Aquitaine/Euskadi/Navarre’.

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