HEAVY ELECTRON BEHAVIOUR OF $CeNi_9Si_4$*

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(Received July 10, 2002)

We have studied the phase formation, thermodynamic and transport properties of $CeNi_9Si_4$. The Rietveld refinement of the X-ray pattern confirms a fully ordered tetragonal crystal structure (space group $I4/mcm$) that is derived from the cubic $NaZn_{13}$ structure type. Resistivity, magnetic susceptibility and specific heat measurements reveal Kondo-lattice behaviour with an enhanced Pauli susceptibility $\chi_0 = 0.005$ emu/mol and a large Sommerfeld value $\gamma = 155\,(5)$ mJ/molK^2. The magnetic contributions to the specific heat and the magnetic susceptibility are well described by the Coqblin–Schrieffer model with a fully degenerate $J = 5/2$ ground state and a characteristic temperature $T_0 \simeq 180$ K.

PACS numbers: 75.30.Mb, 74.25.Bt, 75.30.Cr

1. Introduction

We report on the physical properties of a novel fully ordered compound $CeNi_9Si_4$. This ternary phase is very close to the partially disordered phase $Ce_2Ni_{17}Si_9$ which was reported to be a tetragonal compound with its own structure type (space group $I4/mcm$) that is derived from the cubic $NaZn_{13}$-type [1]. The latter results were confirmed by time-of-flight neutron diffraction by Moze *et al.* [2] who reported a strong site preference of Si to one particular lattice site (fully occupied by Si) and partial occupation of another particular lattice site which is primarily occupied by Ni.

Polycrystalline samples were prepared by induction melting of the elements (Ce and La 99.95%, Ni and Si with 5N) under protective argon

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

atmosphere and subsequent annealing at 990°C for one week. Rietveld refinements were performed on X-ray Guinier image plate data obtained from a flat powder specimen in transmission mode employing the FullProf98 program. Dc susceptibility and specific heat were measured with a SQUID magnetometer and with an adiabatic step heating calorimeter, respectively.

2. Results and discussion

The Rietveld refinement of the X-ray powder diffraction data of single phase $CeNi_9Si_4$ converged satisfactorily for a fully ordered arrangement of Ce, Ni and Si atoms in the sites of the space group I4/mcm with $a = 7.842$ Å and $c = 11.446$ Å. The existence of an ordered 1-9-4 phase is further supported by the much lower residual resistivity $\rho_0 \sim 9\mu\Omega$cm of stoichiometric $CeNi_9Si_4$ and $LaNi_9Si_4$ compared to $\rho_0 > 100\mu\Omega$cm of off-stoichiometric samples (*e.g.* $CeNi_{8.8}Si_{4.2}$) prepared by the same procedure.

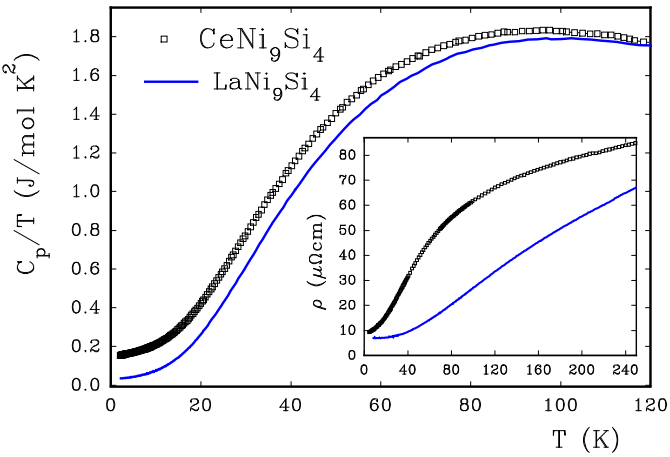

Fig. 1. The specific heat $C_p(T)$ of $CeNi_9Si_4$ and $LaNi_9Si_4$; the inset shows the electrical resistivities $\rho(T)$.

The specific heat $C_p(T)$ of $CeNi_9Si_4$ and its non-magnetic reference $LaNi_9Si_4$ shown in Fig. 1 as C/T *vs.* T reveals a significant enhancement of the T-linear contribution $\gamma = 155\,(5)\,\mathrm{mJ/molK^2}$ of $CeNi_9Si_4$ compared to $\gamma = 33\,(2)\,\mathrm{mJ/molK^2}$ of $LaNi_9Si_4$, reflecting the so-called *heavy electron* behaviour due to the Kondo interaction between the Ce $4f$-states and the conduction electrons. The screening of the Ce-moments revealed by the susceptibility data (see below) together with the temperature dependence of the resistivity, $\rho(T)$, of $CeNi_9Si_4$ compared $LaNi_9Si_4$ (see the inset of Fig. 1) clearly confirms the Kondo lattice behaviour of $CeNi_9Si_4$. Below about 20 K $CeNi_9Si_4$ exhibits a T^2 behaviour, $\rho(T) = \rho_0 + AT^2$ with

$A \sim 0.02\mu\Omega\mathrm{cm/K}^2$, which is characteristic for the regime of coherent Kondo scattering. The Kadowaki–Woods ratio $A/\gamma^2 \sim 10^{-6}\mu\Omega\mathrm{cm(molK/mJ)}^2$ is comparable to that of some Yb Kondo-lattice compounds *e.g.* $YbCu_4Ag$ and *d*-metals [3], but smaller than the typical $10^{-5}\mu\Omega\mathrm{cm(molK/mJ)}^2$ of other Ce and U compounds.

For a direct comparison of the thermodynamic behaviour of $CeNi_9Si_4$ with the thermodynamic equations of the Coqblin–Schrieffer (CS) model [4] solved by Rajan [5] we show in Fig. 2 the magnetic contribution $\Delta C_p(T)$ to specific heat of $CeNi_9Si_4$ (open circles; obtained by subtracting the specific heat of the non-magnetic reference $LaNi_9Si_4$) and the solid line corresponds to the result of the CS model for a 6-fold degenerate ground state ($J = 5/2$) with a characteristic temperature $T_0 = 178\,\mathrm{K}$. The latter is the only free parameter of the model which accounts for the energy scale of the Kondo interaction and has been determined by a simple fit to the experimental data.

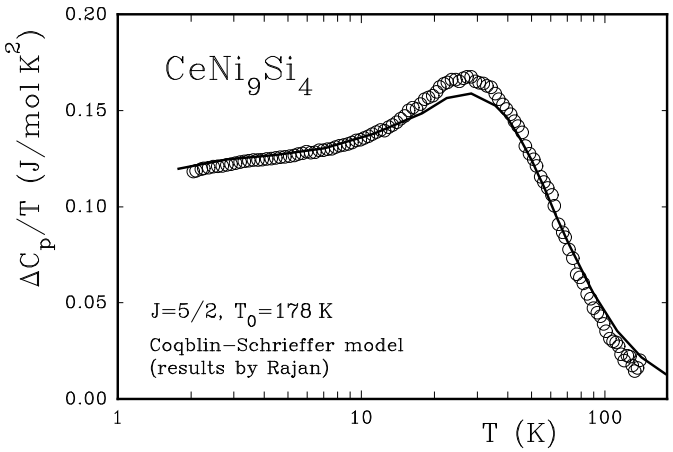

Fig. 2. The magnetic specific heat contribution $\Delta C_p(T)$ of $CeNi_9Si_4$; the solid line corresponds to the Coqblin–Schrieffer model for a 6-fold degenerate ground state with the characteristic temperature $T_0 = 178\,\mathrm{K}$.

The dc magnetic susceptibility $\chi(T)$ of $CeNi_9Si_4$ and $LaNi_9Si_4$ shown in Fig. 3 reveals simple Pauli paramagnetic behaviour with $\chi_0 \simeq 0.6 \times 10^{-3}\,\mathrm{emu/mol}$ for $LaNi_9Si_4$ and a typical *heavy electron* type weakly temperature dependent magnetic susceptibility with $\chi_0 \simeq 5 \times 10^{-3}\,\mathrm{emu/mol}$ for $CeNi_9Si_4$. The solid line in Fig. 3 again shows the result of the CS model for a 6-fold degenerate ground state with the $T_0 = 178\,\mathrm{K}$. The Wilson ratio $R = \chi/\gamma \simeq 1.2$ agrees with the value expected for Ce^{3+}. The rather good agreement between the experimental data, the specific heat and the magnetic susceptibility of $CeNi_9Si_4$, with the simple form of the CS model which

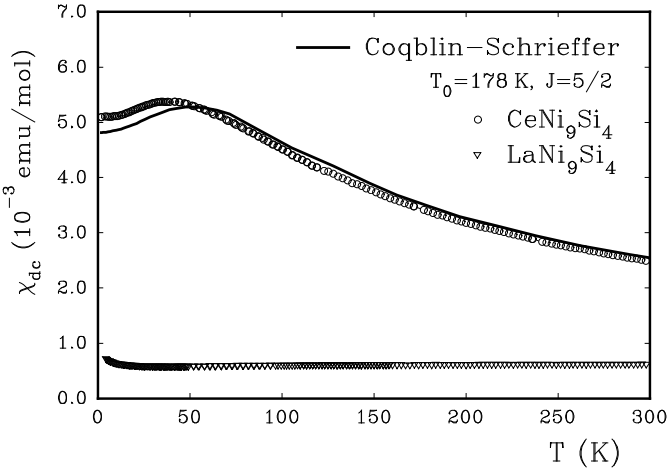

Fig. 3. The dc magnetic susceptibility of $\chi(T)$ of $CeNi_9Si_4$ and $LaNi_9Si_4$ measured at 1 T; the solid line corresponds to the Coqblin–Schrieffer model for a 6-fold degenerate ground state with $T_0 = 178\,\mathrm{K}$.

does not account for any crystalline electric field (CEF) effects indicates that the overall CEF splitting of the Ce^{3+} ground multiplet is smaller than the characteristic energy scale T_0.

The work was supported by the Austrian Science Foundation Fonds under project P-15066-Phy and P-12899-Phy.

REFERENCES

[1] O.I. Bodak, *Sov. Phys. Crystallogr.* **24**, 732 (1979).

[2] O. Moze, C.H. deGroot, F.R. deBoer, K.H.J. Buschow, *J. Alloy. Compd.* **235**, 62 (1996).

[3] N. Tsujii, J. He, K. Yoshimura, K. Kosuge, H. Michor, K. Kreiner, G. Hilscher, *Phys. Rev.* **B55**, 1032 (1997).

[4] B. Coqblin, R.J. Schrieffer, *Phys. Rev.* **185**, 847 (1969).

[5] V.T. Rajan, *Phys. Rev. Lett.* **51**, 308 (1983).