LOW-TEMPERATURE SPECIFIC HEAT OF Ce-Ni-Ge COMPOUNDS AND THEIR NONMAGNETIC ANALOGUES *

A.P. Pikul[a], D. Kaczorowski[a], H. Michor[b], P. Rogl[c]
A. Czopnik[a], Yu. Grin[d], E. Bauer[b] and G. Hilscher[b]

[a]Institute of Low Temperature and Structure Research
Polish Academy of Sciences, P.O.Box 1410, 50-950 Wrocław, Poland
[b]Institut für Festkörperphysik, Technische Universität Wien
A-1040 Wien, Austria
[c]Institut für Physikalische Chemie, Universität Wien, A-1090 Wien, Austria
[d]Max-Planck-Institut für Chemische Physik fester Stoffe
01187 Dresden, Germany

(Received July 10, 2002)

The specific heat of $CeNiGe_3$, $Ce_2Ni_3Ge_5$, Ce_3NiGe_2 and $Ce_3Ni_2Ge_7$ and their isostructural analogues with La or Y was studied in the temperature range 2.5–70 K. For all the Ce-based compounds $C_\mathrm{p}(T)$ exhibits pronounced λ-shaped peaks at the magnetic phase transitions. In the paramagnetic range Kondo and Schottky terms notably contribute to the total specific heat. In the ordered region, $C_\mathrm{p}(T)$ is dominated by a spin-wave contribution. The characteristic Kondo and RKKY energy scales in all the compounds are estimated to be of similar magnitude.

PACS numbers: 75.20.Hr, 75.30.Mb

The recent discovery of pressure-induced superconductivity in $CeNi_2Ge_2$ [1] has stimulated investigations on several other phases from the ternary Ce-Ni-Ge system. In this paper we report the results of specific heat measurements carried out on $CeNiGe_3$, $Ce_2Ni_3Ge_5$, Ce_3NiGe_2 and $Ce_3Ni_2Ge_7$, which complete our comprehensive studies on bulk magnetic [2] and electrical transport [3] behaviour of these compounds. All the Ce-based phases and their isostructural analogues with La and Y were prepared and checked in a manner described previously [3]. The specific heat was measured in the temperature range 2.5–70 K by employing an adiabatic step-heating technique.

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

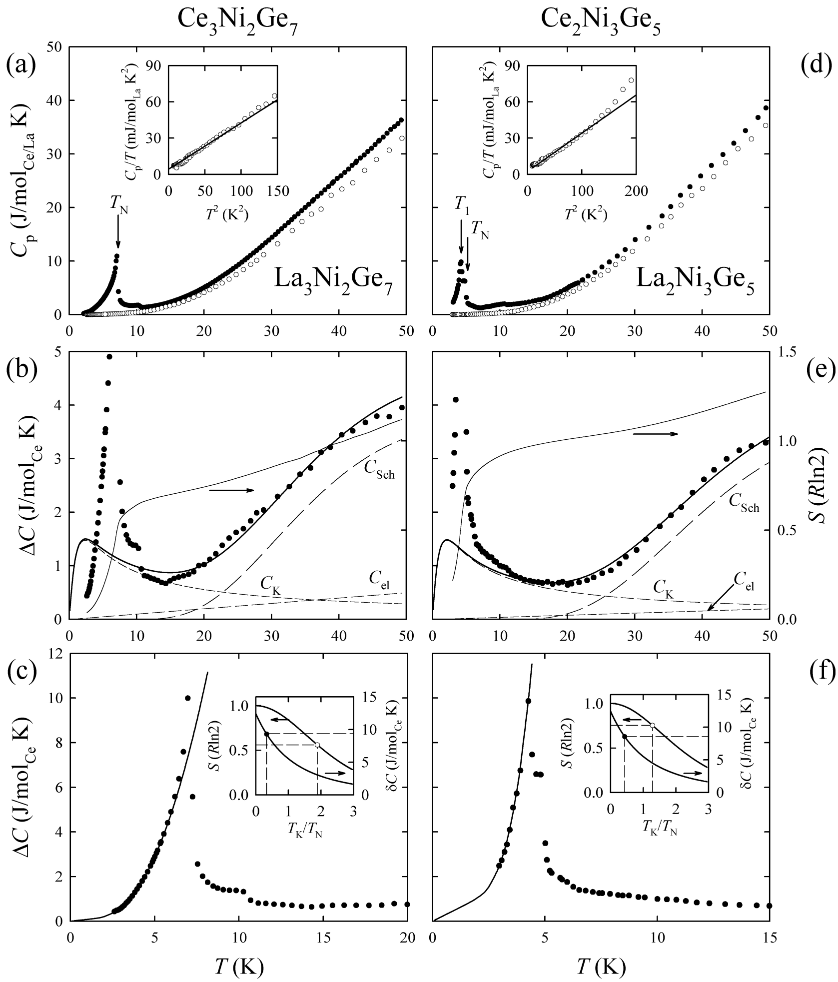

Fig. 1. (a), (d) Specific heat of $(Ce/La)_3Ni_2Ge_7$ and $(Ce/La)_2Ni_3Ge_5$. The arrows mark magnetic phase transitions. Insets: $C_p(T)$ *vs* T^2 for La compounds. (b), (e) Non-lattice specific heat and entropy *vs* T (left and right axes, respectively). The dashed lines represent the electronic, Kondo and Schottky contributions. The thick solid lines are fits to Eq. (1). (c), (f) Low-temperature ΔC. The solid lines are fits to Eq. (2). Insets: estimation of the Kondo temperatures according to Ref. [7] and [8].

The magnetic phase transitions in $CeNiGe_3$, $Ce_2Ni_3Ge_5$, Ce_3NiGe_2 and $Ce_3Ni_2Ge_7$ manifest themselves in $C_p(T)$ as pronounced λ-shaped peaks at $T_{N,C}$ = 5.5, 5.1, 6.2 and 7.5 K, respectively. As an example, Fig. 1 displays the results obtained for $Ce_3Ni_2Ge_7$ and $Ce_2Ni_3Ge_5$ (the data for

$CeNiGe_3$ and Ce_3NiGe_2 will be given elsewhere). In the case of $Ce_2Ni_3Ge_5$ a subsequent order–order transition is seen at $T_1 = 4.4$ K in agreement with Ref. [4]. Tiny anomalies in $C_p(T)$ at about 10 K may result from a small amount of magnetic impurity, presumably Ce_2NiGe_6 [5].

The $C_p(T)$ curves for the La(Y)-based phases are typical for nonmagnetic metals (see Fig. 1). The experimental data below 10 K follow the dependence $C_p = \gamma T + \beta T^3$ with the Sommerfeld coefficients γ of the order of a few mJmol^{-1}K^{-2} and the Debye temperatures of about 300 K. Assuming that the phonon contribution to the specific heat, C_{ph}, of the nonmagnetic analogues is a good approximation of C_{ph} in the Ce compounds, the non-lattice contribution $\Delta C = C_p - C_{ph}$ was extracted, as shown in Fig. 1. Then the ΔC curves were analysed in the paramagnetic region as a sum

$$\Delta C = C_{el} + C_K + C_{Sch}, \tag{1}$$

were the subsequent terms denote the electronic, Kondo and Schottky specific heat, respectively. For $C_{el}(T)$ a simple proportionality $C_{el} = \gamma_p T$ was assumed. The Kondo contribution $C_K(T)$ with the characteristic temperature T_K was applied, as derived theoretically by Desgranges and Schotte [6]. For $C_{Sch}(T)$ a doublet–doublet crystal field scheme with an energy gap Δ_{CEF} was adopted for $Ce_3Ni_2Ge_7$ and $Ce_2Ni_3Ge_5$, because the magnitude of the magnetic entropy at 50 K is for both compounds only slightly larger than $R\ln 2$ (see Fig. 1). In the case of $CeNiGe_3$ a doublet–doublet scheme appeared to be not appropriate and the third doublet originated from the $^2F_{5/2}$ Ce^{3+} ground multiplet was taken into account. In turn, $\Delta C(T)$ for Ce_3NiGe_2 does not reveal any clear Schottky contribution and thus the C_{Sch} term was neglected. Fitting the experimental specific heat curves to Eq. (1) yielded the parameters γ_p, Δ_{CEF} and T_K given in Table I. It is worthwhile noting that for all the compounds the values of Δ_{CEF} and T_K are close to those estimated from the electrical resistivity data [3, 4].

TABLE I

The specific heat characteristics for the Ce–based compounds.

	param. region			ord. region		
Compound	γ_p $\left(\frac{\text{mJ}}{\text{mol}_{\text{Ce}}\text{K}^2}\right)$	Δ_{CEF} (K)	T_K (K)	γ^* $\left(\frac{\text{mJ}}{\text{mol}_{\text{Ce}}\text{K}^2}\right)$	Δ (K)	A $\left(\frac{\text{mJ}}{\text{mol}_{\text{Ce}}\text{K}^5}\right)$
$CeNiGe_3$	45	116, 163	4.5	193	4	9.1
$Ce_2Ni_3Ge_5$	4	173	4.7	504	17	6.4
Ce_3NiGe_2	25	—	12	—	—	—
$Ce_3Ni_2Ge_7$	10	148	5.2	78	10	1.1

For comparison the Kondo temperatures have also been calculated from the magnetic entropy S at $T_{N,C}$ [7] and the specific heat jump δC at $T_{N,C}$ [8]

(see the graphical analysis of T_K in $Ce_3Ni_2Ge_7$ and $Ce_2Ni_3Ge_5$, presented in the insets to Figs. 1(c) and 1(f), respectively). The values of T_K derived from S (δC) are 6.2 (3.7), 6.5 (2.2), 10 (8.4) and 14 (2.5) K for $CeNiGe_3$, $Ce_2Ni_3Ge_5$, Ce_3NiGe_2 and $Ce_3Ni_2Ge_7$, respectively. Despite some discrepancies between the values obtained by different methods it is clearly seen that in all the compounds studied the energy scales for Kondo and RKKY interactions are of similar magnitude.

In the ordered region $C_p(T)$ of an antiferromagnetic Kondo lattice is given by the formula [9]

$$\Delta C(T) = \gamma^* T + A\Delta^{7/2}T^{1/2}e^{-\Delta/T}[1+(39/20)(T/\Delta)+(51/32)(T/\Delta)^2], \quad (2)$$

where $\gamma^* T$ is an electronic term, Δ is an energy gap in the spin-wave spectrum and A is a constant. The least-squares fitting parameters for $CeNiGe_3$, $Ce_2Ni_3Ge_5$ and $Ce_3Ni_2Ge_7$ (ferromagnetic Ce_3NiGe_2 will be analysed separately) are given in Table I (see also the solid lines in Figs. 1(c) and 1(f)). The enhanced values of γ^* hint at strong electronic correlations in all the compounds studied, and thus, together with the behaviour of $C_p(T)$ in the paramagnetic state, support the dense Kondo picture postulated for these ternaries in the previous studies [2–4].

The work was supported by the Austrian–Polish Scientific–Technical Exchange Program (project No. 14/2001), the Max-Planck Society and the Polish State Committee for Scientific Research (KBN) grant No. 2P03B 028 23.

REFERENCES

[1] F.M. Grosche, P. Agarwal, S.R. Julian, N.J. Wilson, R.K.W. Haselwimmer, S.J.S. Lister, N.D. Mathur, F.V. Carter, S.S. Saxena, G.G. Lonzarich *J. Phys. Condens. Matter* **12**, L533 (2000).

[2] A.P. Pikul, D. Kaczorowski, P. Rogl, *Physica B* **312-313**, 422 (2002).

[3] A.P. Pikul, D. Kaczorowski, P. Rogl, Yu. Grin, Proc. European Conference "Physics of Magnetism '02", 1-5 July, 2002, Poznań, Poland.

[4] Z. Hossain, S. Hamashima, K. Umeo, T. Takabatake, C. Geibel, F. Steglich, *Phys. Rev.* **B62**, 8950 (2000).

[5] B. Chevalier, J. Etourneau, *J. Magn. Magn. Mater.* **196–197**, 880 (1999).

[6] H.U. Desgranges, K.D. Schotte, *Phys. Lett.* **91A**, 240 (1982).

[7] H. Yashima, H. Mori, N. Sato, T. Satoh, *J. Magn. Magn. Mater.* **31–34**, 411 (1983).

[8] J.A. Blanco, M. de Podesta, J.I. Espeso, J.C. Sal, C. Lester, K.A. McEwen, N. Patrikios, J.R. Fernández, *Phys. Rev.* **B49**, 15126 (1994).

[9] M.A. Continentino, S.N. de Medeiros, M.T.D. Orlando, M.B. Fontes, E.M. Baggio-Saitovitch, *Phys. Rev.* **B64**, 012404 (2001).