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**PHYSICA B**

# Thermal expansion of the Kondo insulator CeRhSb

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## Abstract

The linear coefficients of thermal expansion,  $\alpha(T)$ , of the Kondo insulator CeRhSb and its non-f-electron analog LaRhSb have been measured on polycrystalline samples in the temperature range 1.5–200 K. After subtracting the phonon contribution ( $\theta_D = 260$  K), the f-electron contribution to the thermal expansion,  $\alpha_m(T)$ , shows two distinct features, a broad maximum is centered at 125 K, while a large shoulder appears below 40 K. In a plot of  $\alpha_m/T$  versus  $T$ , a maximum appears at the temperature where the energy gap opens,  $T_g = 10$  K. For  $T < T_g$ ,  $\alpha_m(T)$  follows an  $aT + bT^2$  law, which is compatible with a description of the energy gap vanishing on lines at the Fermi surface.

## 1. Introduction

In recent years, much attention has been focused on a novel class of heavy-fermion systems, the so-called Kondo insulators. The most striking property of these materials is the opening of an energy gap in the quasiparticle excitation spectrum. Of particular interest are the cerium compounds CeNiSn and CeRhSb, where a small energy gap ( $\Delta \sim 4$ – $10$  K) opens at low temperatures ( $T_g \sim 10$  K) in the enhanced density of states [1,2]. Evidence for the insulating ground state is inferred from the transport, magnetic and thermal properties below  $T_g$ , specifically from the strong increase in the electrical resistivity, the sudden increase in the absolute value of the Hall coefficient ( $|R_H|$ ) and the gradual decrease in the specific heat divided by temperature ( $c/T$ ). Experiments on single-crystalline samples have revealed that the energy-gap is strongly anisotropic. The anisotropic character of the gap is also supported by NMR experiments [3],

which yield a nuclear spin-lattice relaxation rate  $1/T_1$  proportional to  $T^3$ , indicative of a V-shaped gap. Under high pressure or in a strong magnetic field the gap is suppressed and the normal metallic behaviour is restored [1].

A simplified qualitative model for the Kondo insulating state has recently been delineated in Ref. [4]. A hybridization gap is formed by a flat f-band that crosses a single broad conduction band. In the case of an even number of valence electrons (where the f-electron is counted as itinerant) an insulating ground state results. A more quantitative model for the Kondo insulating state, which is based on the interplay between the crystal field and spin excitations, has recently been formulated by Kagan and co-workers [5,6].

In order to investigate the effect of the insulating state on the unit cell parameters, we recently reported a dilatometry study of CeNiSn [6]. The measurements performed on a single-crystalline sample showed a large anisotropy in the coefficients of linear thermal expansion (orthorhombic crystal structure), but the coefficient of volume expansion  $\alpha_v$  showed no specific

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structure upon opening of the gap. However, the  $\alpha_v/T$  versus  $T$  curve revealed a broad maximum centered at  $T_g$ , just as is the case for  $c/T$  versus  $T$  [1,2]. In this paper, we report thermal-expansion measurements of the analogous compound CeRhSb in the temperature range 1.5–200 K. As no single-crystalline sample was available, the measurements were performed on a polycrystal. An estimate for the phonon contribution was obtained by measuring the non-f-electron material LaRhSb.

## 2. Experimental

CeRhSb and LaRhSb were prepared in polycrystalline form by arc-melting the elements in an argon atmosphere. An excess amount of Sb was added in order to compensate for weight losses. The ingots were annealed at 1100°C for 7 days in sealed quartz tubes. X-Ray diffraction showed that the samples were single phase with the proper  $\epsilon$ -TiNiSi structure. In order to measure the coefficient of linear thermal expansion,  $\alpha = L^{-1}dL/dT$ , the CeRhSb sample was shaped in the form of a cube (edge 5 mm) by means of spark erosion. Values for  $\alpha(T)$  were obtained along the three directions of the polycrystalline cube. In the case of LaRhSb only two plane-parallel surfaces were obtained and consequently  $\alpha(T)$  was measured along one direction. The thermal expansion was measured with a sensitive three-terminal capacitance method [7].

## 3. Results

The experimental results are shown in Fig. 1. The three  $\alpha(T)$  curves for CeRhSb show the same behaviour qualitatively, i.e. a broad maximum is centered near 140 K and a pronounced shoulder is found below  $T \sim 40$  K. The absolute values differ somewhat (ranging from 12 to  $16 \times 10^{-6} \text{ K}^{-1}$  near 200 K), which indicates that crystallites with preferred orientations are present in the polycrystal. The anisotropy in  $\alpha(T)$  changes sign near 25 K. The coefficient of volume expansion  $\alpha_v(T)$  was obtained by averaging over the three directions. The coefficient of thermal expansion of LaRhSb was considerably smaller and could be nicely fitted with a Debye function yielding a Debye temperature  $\theta_D = 260$  K (See Fig. 1). The conduction electron contribution to  $\alpha(T)$  of LaRhSb can be neglected. Specific-heat measurements [8], performed on a part of the sample used for the thermal expansion

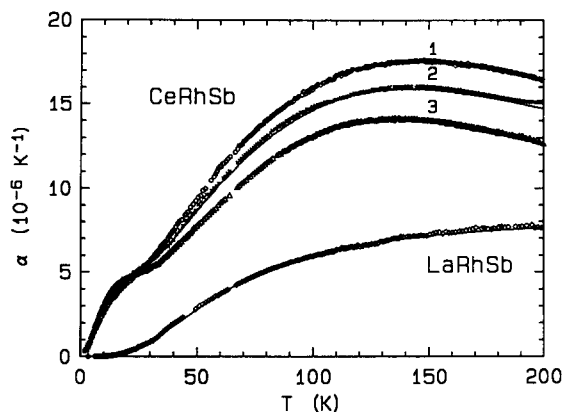


Fig. 1. The coefficients of linear thermal expansion of CeRhSb and LaRhSb. Curves labelled 1, 2 and 3 show  $\alpha(T)$  for three different directions of the polycrystalline cube, while  $\alpha_v/3$  is given by the solid line. The solid line through the data for LaRhSb is a Debye fit with  $\theta_D = 260$  K.

measurements, showed a somewhat smaller  $\theta_D$  of 210 K. Superconductivity was observed at  $T_c = 2.6$  K, in agreement with the literature [9,10].

## 4. Discussion

The f-electron contribution to the thermal expansion of CeRhSb was obtained by subtracting the data for LaRhSb and is shown in Fig. 2. The broad maximum, now centered at  $T_{\max} = 125$  K, concurs with both the maximum in the electrical resistivity and

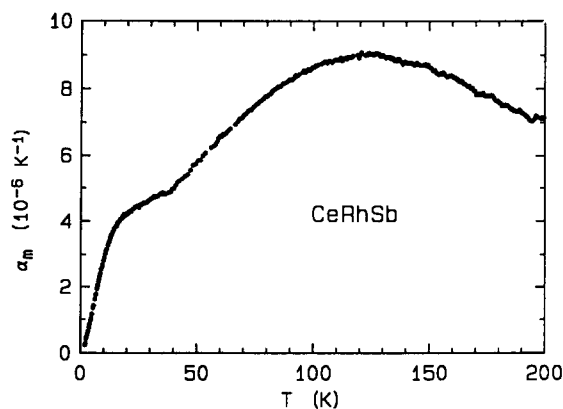


Fig. 2. The f-electron contribution ( $\alpha_m$ ) to the thermal expansion of CeRhSb.

the magnetic susceptibility. The crystalline electric field splits the  $J = 5/2$  multiplet into three doublets. However, when comparing the data in the temperature range ( $T > 50$  K) with a Schottky expression for a doublet-doublet splitting ( $\Delta = T_{\max}/0.417$ ) of about 300 K, a large disagreement is found below  $T_{\max}$ . Instead, it is more likely that the maximum originates from the admixture of the Kondo and crystalline electric field effects. A comparison of the specific-heat data ( $T < 60$  K) to the  $S = 1/2$  single-impurity Kondo model yielded a Kondo temperature  $T_K \approx 96$  K [11]. Below 38 K,  $\alpha_m(T)$  shows a second distinct contribution. The sharp kink in  $\alpha_m$  at 38 K, coincides with a change in the slope of the electrical resistivity [1,8]. However, no anomaly has been found in the specific-heat data published so far and the nature of the contribution below 40 K remains to be elucidated.

The low-temperature part of the data is plotted as  $\alpha_m/T$  versus  $T$  in Fig. 3. For comparison we also show the data for CeNiSn [6]. The corresponding  $c_m/T$  versus  $T$  data are shown in Fig. 4. Taking the temperatures of the maxima in  $\alpha_m/T$  as indicative for the opening of the energy gap, we obtain values for  $T_g$  of 10 K and 8 K for CeRhSb and CeNiSn, respectively, while values of 9 and 6 K result from  $c_m/T$ , respectively. The difference in the values of  $T_g$  as determined by the thermal expansion and the specific heat is within the experimental accuracy. For  $T < T_g$ , both  $\alpha_m$  and  $c_m$  follow an  $aT + bT^2$  law, albeit in a limited temperature interval. This power law behaviour is compatible with a description of the energy gap vanishing

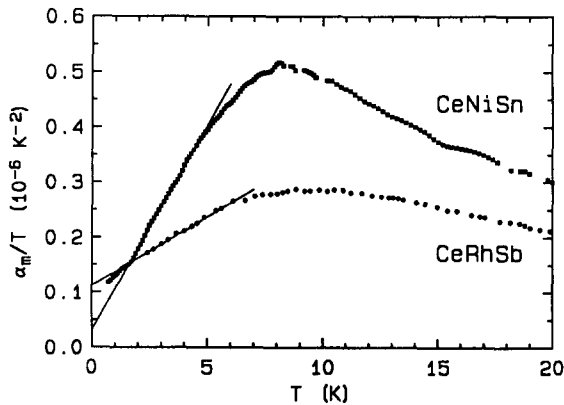


Fig. 3.  $\alpha_m/T$  versus  $T$  for CeRhSb (●) and CeNiSn (■). The solid lines represent fits with a linear and quadratic term (see text).

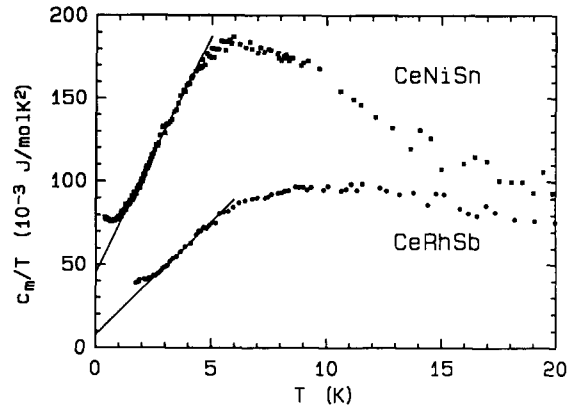


Fig. 4.  $c_m/T$  versus  $T$  for CeRhSb (●) and CeNiSn (■). The solid lines represent fits with a linear and quadratic term (see text).

on lines at the Fermi surface (V-shaped gap). The departure from this behaviour observed below  $T = 1.6$  K for CeNiSn has been attributed to the onset of antiferromagnetic correlations [6]. The energy gap opens in the enhanced density of states as follows from the fairly large values for  $c_m/T$  for  $T > T_g$ . Extrapolating the  $c_m/T$  and  $\alpha_m/T$  data as measured for  $T > T_g$  to  $T = 0$  K, we obtain an enhanced electronic Grüneisen parameter of  $\sim 19$  for CeRhSb and CeNiSn (using a value for the isothermal compressibility of  $1.8 \text{ Mbar}^{-1}$  as measured for CeNiSn [12]). Similarly enhanced values have been reported for other heavy-fermion compounds [13].

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