Low-temperature physical properties of heavy-fermion CeRh$_2$Sn$_2$

A M Strydom
Physics Department, University of Johannesburg, PO Box 524, Auckland Park 2006, South Africa
E-mail: amstrydom@uj.ac.za

Abstract. Intermetallic f-electron compounds comprising rare earth elements Ce or Yb are known to exhibit a variety of complex electronic and magnetic features that originate essentially from hybridization between localized f-electrons and conduction electrons. The isostructural tetragonal stannide series Ce$_M$$_2$Sn$_2$ in which $M$ is a d-electron element capture the essential features of the strongly correlated electron class of systems in which the on-site Kondo exchange competes with long-range magnetic interactions. Cerium-based antiferromagnetic order is prevalent among compounds in the series at temperatures typically below about 4 K, but with a particularly large enhancement of the electronic specific heat in the neighborhood of $T_N$. The title compound of this study is CeRh$_2$Sn$_2$, and we present here results of low-temperature studies of the specific heat and magnetic susceptibility in order to map its low-temperature behaviour.

1. Introduction
Rare-earth elements and compounds offer an extraordinary wide variety of magnetic phenomena of interest to fundamental investigations as well as to studies seeking solutions to technological problems. In the two rare-earth elements Ce and Yb, the question of stability of the atomic magnetic moment itself in presence of many-body interactions with its environment in the solid state has been a rich source of new Physics. In compounds of Ce for instance, the tendency towards long-range magnetic ordering driven by conduction-electron mediated coupling between well-defined local moments frequently has to compete with an on-site, shorter-range interaction that tends to screen the magnetic moment and thus weakens its proclivity towards magnetic ordering.

In this work we extend investigations into a member of an interesting group of cerium-based compounds, namely CeRh$_2$Sn$_2$. The existence and crystallographic detail of this group of stannides were first reported by Selsane et al. [1]. Aside from Rh, the series includes compounds with the d-electron elements Ni, Ir, Cu, Pd, and Pt, which affords the opportunity of studying trends and systematics in cooperative behaviours. The common crystal structure adopted by the series is the well ordered CeBe$_2$Ge$_2$ type of tetragonal space group $P4/nmm$. This series of compounds were reported [1] to exhibit stable local-moment behaviour at higher temperatures, but measurements to below 4.2 K eventually exposed low-lying antiferromagnetic order in compounds with Ni, Cu, Pd, Pt, and subsequently in the Ir and Rh derivatives as well [2]. A remarkable attribute is their enormously enhanced specific heat $C_P(T)$ at low temperatures, and in particular the electronic contribution $C_P(T)/T$ [2]. The concept of a very heavy effective
electron mass was argued [2] as a plausible origin for the large specific heats. Far away from the magnetic ordering at high temperatures the Kondo interaction starts screening the Ce local $4f$–electron spin as a natural consequence of spin-spin interaction. As the thermal energy is washed out of the spin-conduction electron ensemble the spin-spin interaction builds up a high density of states close to the Fermi energy. This is brought about by a limited yet finite contact between the typically very sharp f-electron resonance and the much broader and degenerate conduction band. Thus, in this series the low-temperature magnetic ordering is thought to develop from heavy quasiparticles [2]. Magnetic ordering of an antiferromagnetic type has been reported across the series and ranges from 4.1 K in CeIr$_2$Sn$_2$ to 0.42 K in CeRh$_2$Sn$_2$. Our purpose in this work is to investigate ordering phenomena in the heavy-fermion title compound CeRh$_2$Sn$_2$ to temperatures well below its magnetic ordering, as well as to investigate how applied magnetic fields affect the magnetic ordering.

2. Experimental details and Results

A polycrystalline sample of CeRh$_2$Sn$_2$ and its non-magnetic counterpart LaRh$_2$Sn$_2$ were prepared by direct argon-arc melting of the constituent elements. The elemental purites of all the elements used were better than 99.99 wt.%. Prolonged sample heat treatment for 7 days at 800°C was performed to promote composition homogeneity and phase purification. Powder x-ray diffraction measurements were used to inspect phase formation. Measurements of physical properties were conducted on a commercial PPMS station [4] equipped with a $^3$He/$^4$He dilution refrigerator. A squid-type MPMS magnetometer from the same suppliers was used to measure magnetic properties in static fields up to 7 T, while temperatures below 1.9 K and typically down to 460 mK were made possible by a $^3$He insert [5]. Powder X-ray diffraction results confirmed that the annealed specimens of CeRh$_2$Sn$_2$ and LaRh$_2$Sn$_2$ formed as single-phase compounds. As part of phase purity assessment we determined lattice parameters for CeRh$_2$Sn$_2$: $a = 4.491(1)$ Å, $c = 10.413(1)$ Å, $V = 210.09(1)$ Å$^3$, as well as for the previously unreported compound LaRh$_2$Sn$_2$: $4.519(1)$ Å, $c = 10.471(1)$, and $V = 213.841(1)$ Å$^3$. The unit-cell volume of CeRh$_2$Sn$_2$ agrees to within 0.5% with the published value [1].

The magnetic susceptibility $\chi(T)$ of CeRh$_2$Sn$_2$ follows Curie-Weiss behaviour over a remarkably wide temperature range and to as low as $\approx 10$ K (see solid line in main panel, fig. 1). The fit yields an effective moment of $\mu_{\text{eff}} = 2.26(1)$ μ$_B$/Ce and a Weiss temperature $\theta_\text{P} = -5.56(5)$ K. The net exchange between stable local moments of Ce$^{3+}$ is thus of antiferromagnetic nature. There is little or no perturbation evident on the paramagnetic state of this compound until a weak anomaly appears at $T_N \approx 0.5$ K. Specific heat results shown further below will provide further substantiation for asigning this anomaly to magnetic ordering in CeRh$_2$Sn$_2$ as is expected from existing data on this compound [2]. Magnetization results shown in fig. 2 illustrate a clear change in $M$ vs. $B$ curvature between isotherms obtained at $T = 1.8$ K $> T_N$ and at $T = 0.464$ K which is very close to $T_N$ (see discussion further below). Casting the $T = 0.464$ K data of $M(B)$ in the form $\partial M(B)/\partial B$ (not shown) reveals a near-constant slope up to $\approx 1$ T, beyond which the slope of the magnetization rapidly diminishes. A near-saturation magnetization amounting to $M_{\text{sat}} \approx 1.3 \mu_B$ is found in a field of $B = 7$ T. We note that the full $J = 5/2$ multiplet of Ce$^{3+}$ would lead to $M_{\text{sat}} = 2.14 \mu_B$/Ce. The shortfall is likely due to magnetocrystalline entropy, and our present data on a polycrystalline permits extraction of only about 2/3 of the full ground-state magnetic moment. A near-linear $M$ vs. $B$ progression is evident up to a field of $\approx 1.5$ T, with pronounced negative curvature developing towards higher fields.

Shown in fig. 3(a) is an upturn in the specific heat $C_P(T)$ of CeRh$_2$Sn$_2$ below 2.5 K from an already much enhanced 1 J/(mol.K). Taking the maximum slope of the upturn as $T_N = 0.5$ K, $C_P(T)$ finally peaks at 3.2 J/(mol.K). An applied magnetic field of 2 T was found to suppress and broaden the phase transition anomaly, which is seen especially from the specific heat plot.
Figure 1. (main panel) Magnetic susceptibility $\chi(T)$ of CeRh$_2$Sn$_2$ together with fit of the Curie-Weiss law (red line). (inset) Semi-log plot of low-temperature $\chi(T)$ showing magnetic ordering as a weak anomaly at $T_N \approx 0.5$ K.

Figure 2. Magnetization of CeRh$_2$Sn$_2$ at a number of successively lower temperatures down to 0.464 K, which is just below $T_N$ (see fig. 1). Notice the change in curvature in the lowest temperature $T < T_N$ curve for fields greater than $\sim 1.5$ T.

Figure 3. (main panel) Specific heat of CeRh$_2$Sn$_2$ in various fields.

Figure 4. (main panel) Entropy of CeRh$_2$Sn$_2$ in zero field and 2 T. (inset) Estimation of the Sommerfeld coefficient of CeRh$_2$Sn$_2$ well above $T_N$, yielding $\gamma = 271(5)$ mJ/mol.K$^2$.

cast in the form of $C_P(T)/T$ shown in fig. 3(b). In 6 T the anomaly is no longer visible. In fig. 4 an estimation of the 4f–electron spin entropy $S(T) = \int_0^T C_P(T)/T \, dT$ is illustrated with $S$ in units of $R \ln 2$. Note that at the temperatures of interest for calculating $S(T)$ from fig. 3, the lattice specific heat (approximated by $C_P(T)$ of LaRh$_2$Sn$_2$, see fig. 5) is at least two
orders of magnitude smaller than that of CeRh$_2$Sn$_2$. At sufficiently low temperatures only the lowest doublet electronic levels of Ce$^{3+}$ out of the crystal-electric field split $J = 5/2$ multiplet are expected to be populated, and hence responsible for the magnetic ordering in CeRh$_2$Sn$_2$. However, fig. 4 suggests that this amount of entropy is evidently only released at a much higher temperature of $\simeq 5$ K. This is an indication of precursory short-range correlations which occupy the spin system somewhat above $T_N$. The inset of fig. 4 also shows calculation of the electronic specific heat of CeRh$_2$Sn$_2$. The temperature range $7 \, \text{K} < T < 17 \, \text{K}$ is amenable to calculation of the Sommerfeld coefficient in terms of $C_P(T) \propto \gamma T^2$ and yields $\gamma = 271(5) \, \text{mJ/(mol.K)}$. We note that the temperature region of this proportionality does not extend down to the temperature of $\simeq 5$ K where the ground-state entropy is fully released. Finally in fig. 5 we turn to the low-temperature $C_P(T)$ of LaRh$_2$Sn$_2$. A well-defined anomaly peaking at $T_{SC} = 0.67$ K has been established as the onset of superconductivity in this material. Further measurements on this compound are in progress and will be presented elsewhere [6].

3. Conclusions

Specific heat and magnetic susceptibility data confirm antiferromagnetic type ordering in CeRh$_2$Sn$_2$ below $T_N = 0.5$ K. From entropy considerations a picture emerges of displacement of a considerable portion of the ground-state entropy to well above $T_N$. This spin-correlation entropy is largely responsible for the enhanced electronic specific heat above $T_N$. Our findings establish CeRh$_2$Sn$_2$ as a moderately heavy fermion system. A magnetic field suppresses the anomaly associated with the Néel transition, but a strong influence exerted by the $T > T_N$ short-range correlations causes further thermal broadening of the phase transition until the ordering finally becomes inconspicuous in 6 T.

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