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Low-temperature thermal and high-pressure studies of CePd and $CeAgSb_2$

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Abstract. The anomalous properties of the Kondo lattice $CeAgSb_2$ are compared with the well behaved Kondo system, CePd. The low-temperature thermal expansion of CePd is in agreement with previous heat capacity data, showing a ferromagnetic transition at $T_c = 6.5$ K and a second transition at 3.5 K probably due to reorientation of the magnetic moments. The Grüneisen factor, calculated from thermal-expansion and heat capacity is $\gamma_G \approx 4.5$, the same as the Grüneisen factor γ_P calculated from the pressure dependence and comparable to a previous measurement of $\gamma_P \approx 6$. Thermal expansion measurements (in fields of up to 8 T) and ac susceptibility measurements (under pressures of up to 7.33 kbar) have been made on $CeAgSb_2$ in the temperature range 1.6–40 K. The zero-pressure ac susceptibility measurements confirm that there is an antiferromagnetic transition at $T_N = 9.5$ K, as previously reported. The magnetic contribution to the linear thermal-expansion coefficient of polycrystalline $CeAgSb_2$ (α_p) in zero magnetic field has a maximum value near 17 K and becomes small by 29 K. There is no peak in α_p at the Néel temperature. The Néel temperature is found to decrease under pressure at the rate of $\partial(\ln T_N)/\partial P = -10.1$ Mbar^{-1}, which indicates that $CeAgSb_2$ is a magnetic Kondo lattice (with $T_K > T_N$) on the right-hand side of the Doniach diagram.

1. Introduction

Over the last few years, studies on cerium- and uranium-based intermetallic compounds and alloys have revealed that these systems exhibit a variety of physical properties, such as unusual magnetic order, heavy fermion or mixed valence behaviour [1, 2]. These unusual properties are mainly due to hybridization between the moment-carrying 4f or 5f electrons and the conduction electrons, but may also be strongly influenced by the effects of crystal electric fields [3].

One such exotic family of systems, known as Kondo lattices, is characterized by competition between the magnetic ordering of local moments by the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction [4] and reduction of the magnetic moment by the Kondo interaction. The magnetic behaviour of a Kondo system is characterized by $|\mathfrak{J}N(E_F)|$ where $\mathfrak{J}$ is the magnetic exchange constant between the conduction electrons and the 4f local moments and $N(E_F)$ is the density of conduction electron states at the Fermi energy.

Doniach [5] examined the one-dimensional (1D) 'Kondo necklace' problem in the mean field approximation (which can be extended to 3D systems) and obtained an antiferromagnetic ground state with the magnetic ordering temperature T_M as a function

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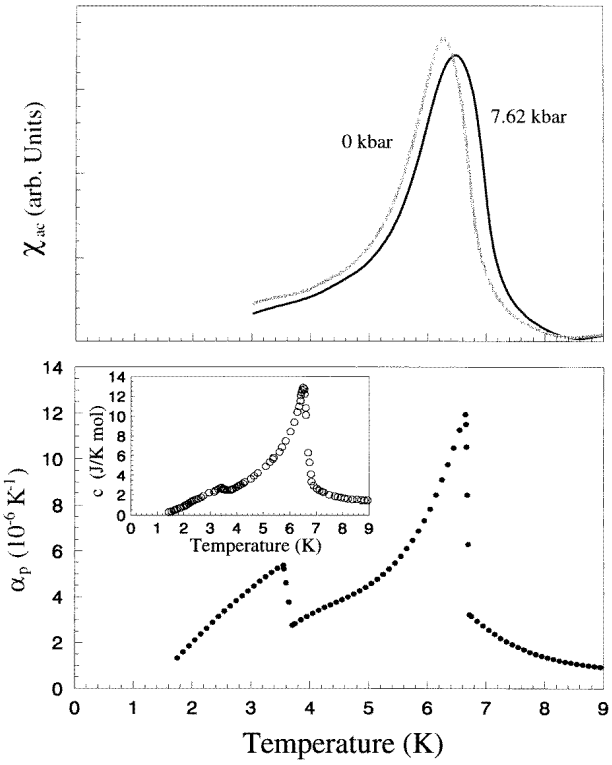

Figure 1. The ac susceptibility of CePd as a function of temperature at atmospheric pressure and at 7.62 kbar (upper graph). The linear thermal expansion is shown in the bottom graph, with the heat capacity of CePd, after Besnus *et al* [8] in the insert.

of $|\mathfrak{J}N(E_F)|$. The calculations in this study have shown that T_M as a function of $|\mathfrak{J}N(E_F)|$ passes through a maximum, and that this happens when the value of the Kondo energy is very close to the RKKY energy. Experimentally, the strength of the Kondo interaction relative to the RKKY interaction is reflected by the pressure dependence of the Néel temperature [6] $(\partial \ln T_N)/\partial P$ where T_N is the Néel transition temperature.

At small values of $|\mathfrak{J}N(E_F)|$ the RKKY interaction dominates and a normal full moment Néel state occurs below T_N; whereas at large values of $|\mathfrak{J}N(E_F)|$ the Kondo effect dominates entirely, and there is no long-range magnetic order and the ground state is that of a strongly correlated Fermi liquid. Thus, by measuring the pressure dependence of T_N the strength of the RKKY relative to the Kondo interaction can be inferred.

CePd crystallizes in the orthorhombic CrB-type structure, with space group $Cmcm$. Its lattice parameters are $a = 3.88$ Å, $b = 10.89$ Å and $c = 4.69$ Å at room temperature. The saturated moment per Ce atom, estimated from the magnetization data at 4.2 K is 0.84 μ_B [7]. The reduced moment is attributed to the crystal splitting of the $J = 5/2$ state. A small reduction in the magnetic moment is also expected if Kondo interactions exist.

CePd has been described as a magnetic Ce system in which the moment is only weakly hybridized [8]. It undergoes a ferromagnetic transition at $T_c = 6.5$ K and the heat capacity measurements, shown in the insert of figure 1 after Besnus *et al* [8], indicate that there may be a second transition at 3.1 K, possibly due to a spin reorientation process.

$CeAgSb_2$ crystallizes in the tetragonal $UCuAs_2$-type structure with space group $P4/nmm$. A detailed study of the magnetic and transport properties of $CeAgSb_2$ has been completed over a wide temperature range by Houshair *et al* [9]. Their measurements of electrical resistivity and thermopower indicate that the compound is a condensed Kondo lattice system. The magnetic component of the resistivity of $CeAgSb_2$ above 50 K shows the $-\ln(T)$ behaviour expected from a Kondo system. A maximum in the electrical resistivity $\rho(T)$ at 24 K is indicative of the onset of coherence below this temperature, and a sharp drop in the resistivity at 10 K is consistent with the onset of magnetic ordering. The thermopower has a maximum near 80 K. This maximum, together with our preliminary (unpublished) inelastic neutron scattering measurements, indicate that the Kondo temperature T_K for $CeAgSb_2$ is of the order $T_K \sim 60$–80 K.

The magnetization measurements of Houshair *et al* [9] indicate that the material undergoes a transition to a magnetically ordered state below 10 K. This has since been confirmed by Muro *et al* [10], who report on a series of transport measurements made on $CeAgSb_2$ and related compounds. Houshair *et al* [9] suggest that this ordered state may have a complicated antiferromagnetic structure with a small net ferromagnetic moment. They estimate the saturated moment per Ce atom M_{sat} to be only 1.37 μ_B per atom. This is small when compared to the free magnetic moment of Ce^{3+} (2.54 μ_B) [4]. As with CePd, the reduced moment may be due to the crystal splitting of the $J = 5/2$ state.

In this paper, we report on the low-temperature thermal expansion and the ac susceptibility of CePd and $CeAgSb_2$. The ac susceptibility measurements, performed under a range of applied hydrostatic pressures, confirm that $CeAgSb_2$ is indeed a Kondo lattice material on the right-hand side of the Doniach diagram [5, 6], shown in figure 2, however the low-temperature thermal expansion measurements made on polycrystalline samples of $CeAgSb_2$ show no indication of magnetic ordering at the Néel temperature of 10 K. This

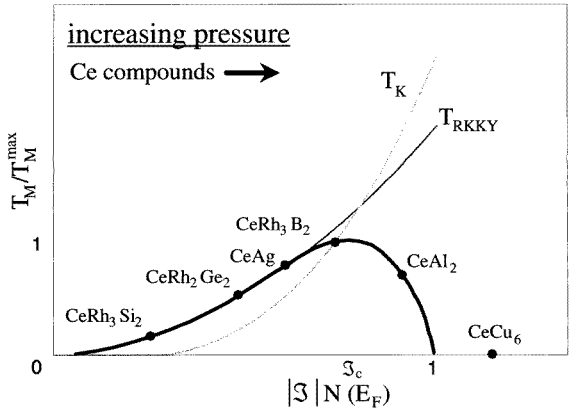

Figure 2. Schematic representation of the effect of competing Kondo and RKKY interactions, plotted as temperature against the exchange parameter $\mathfrak{J}$ times the conduction band density of state N. At small $\mathfrak{J}N$, RKKY interaction dominates and a normal, full-moment Néel state is expected below T_N; whereas at large $\mathfrak{J}N$ the Kondo effect partially compensates local moments before they order antiferromagnetically. At still larger $\mathfrak{J}N$, the Kondo effect dominates entirely, and there is no long-range magnetic order and the ground state is that of a strongly correlated Fermi liquid. With the application of pressure Ce compounds will move along the curve from left to right and Yb compounds will move along the curve from right to left (adapted after Cornelius *et al* [6]).

effect has also been observed in polycrystalline CePdSb [11–13], but in the case of CePdSb, a broad maximum is observed in the thermal expansion *below* the ordering temperature of 17 K, whereas in the case of $CeAgSb_2$ the broad maximum occurs *above* T_N. On the other hand, the low-temperature thermal expansion of CePd is in agreement with previous heat capacity data and shows a ferromagnetic transition at $T_c = 6.5$ K and a second transition at 3.5 K probably due to reorientation of the magnetic moments. The Grüneisen factor, calculated from thermal-expansion and heat capacity, is comparable to the Grüneisen factor calculated from the pressure dependence.

2. Experimental

The thermal expansion measurements were made in a parallel plate capacitance cell that had been calibrated using the known thermal expansivity of copper and sapphire. A magnetic field of up to 8 T could be applied parallel to the measured direction. The lowest temperature attainable in the variable temperature insert was 1.6 K. The ac susceptibility as a function of temperature was measured in an uncalibrated mutual-inductance apparatus.

The pressure dependence of the Néel temperatures for both CePd and $CeAgSb_2$ was determined by observing changes in the ac susceptibility, measured as a function of temperature. The ac susceptibility coils were mounted on the end of the fixed piston of a Be–Cu clamp cell. This could be pressurized at room temperature to up to 10 kbar prior to cooling. In this system the highest pressure available at 2.6 K is approximately 8 kbar. The pressure was monitored with a calibrated semiconducting pressure transducer and the temperature of the sample was monitored with a calibrated carbon-glass thermometer mounted at the top of the cell.

The polycrystalline samples were made by repeated arc melting of stoichiometric amounts of the high-purity constituent elements in a purified argon atmosphere. Measurements were performed on discs cut by spark erosion from the ingots. In order to analyse the magnetic contribution of $CeAgSb_2$, the thermal expansion of the non-magnetic analogue $LaAgSb_2$ was subtracted from that of the Ce compound to obtain the magnetic contribution to the thermal expansion, as shown in figure 3. No suitable non-magnetic analogue was available for CePd.

3. Experimental results

The ac susceptibility $\chi_{ac}(T)$, and the thermal expansion $\alpha_p(T)$ of CePd were measured from 1.6–9 K, as shown in figure 1. The heat capacity $c(T)$ after Besnus *et al* [8] is reproduced in the insert for reference. The ac susceptibility at ambient pressure showed a maximum at $T_c = 6.21$ K. A small shoulder was also observed at around 3.3 K. On application of 7.62 kbar the maximum appeared at 6.49 K and the shoulder was found to have moved to around 3.6 K. These measurements give a value of $(\partial \ln T_c)/\partial P = 4.5$ Mbar^{-1} close to the previously reported value [14] of 6 Mbar^{-1}.

It was found that the thermal expansion showed features at around 3.5 K and 6.6 K in agreement with heat capacity results [8]. A non-magnetic analogue was not available for this system, so the curves in figure 1 include both a magnetic and phonon contribution. However, in this low-temperature regime, the phonon contribution is expected to be negligible compared with the large magnetic contribution at the peak values of α_p and $c(T)$. Using the maximum values, an estimate of the Grüneisen factor γ_G was made for CePd for the whole ferromagnetic regime. This was done by scaling α_p to $c(T)$ (with a

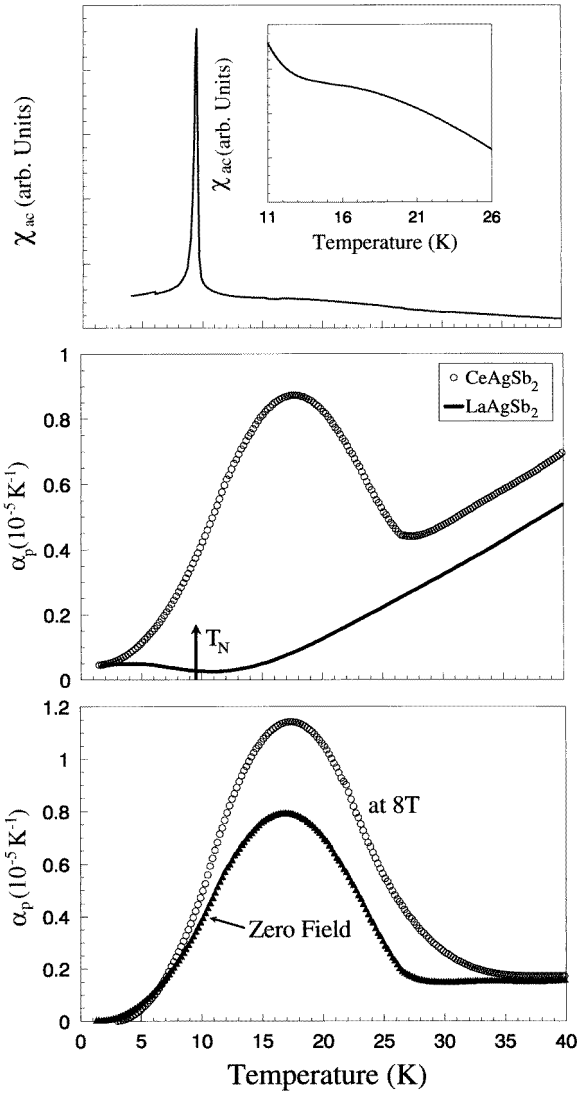

Figure 3. The upper graph shows ac susceptibility as a function of temperature of $CeAgSb_2$. The insert shows ac susceptibility in the region in which coherence appears to set in, showing a distinct shoulder at around 16.7 K. The centre graph shows the linear thermal expansion α_p of $CeAgSb_2$ and $LaAgSb_2$ as a function of temperature. The magnetic contribution to the linear thermal expansion of $CeAgSb_2$ as a function of temperature in zero field and in 8 T is shown in the bottom graph.

scaling factor of $1 \times 10^{-6} \equiv 1.086$ J mol^{-1} K^{-1}). The molar volume V_{mol} of CePd is 1.49×10^{-5} m^3 mol^{-1} [7], the value of the bulk modulus B_T was taken to be of the order of 10^{11} Pa, so that $\gamma_G = 3\alpha_p V_{mol} B_T/c \approx 4.5$. This is larger than the value ($\sim$2) typically found in simple metals. This value is the same as that found from the change in Néel temperature with applied pressure, $\gamma_P \approx 4.5$, and to the value $\gamma_P \approx 6$, derived from the pressure coefficient reported by Huber *et al* [14]. For the low-temperature feature, a similar analysis gave $\gamma_G \approx 8.6$ and $\gamma_P \approx 11$.

The ac susceptibility ac of $CeAgSb_2$ was measured from 3–40 K, as shown in figure 3. The magnetic susceptibility shows a sharp peak at the phase change at 9.5 K. This second-order phase change is indicative of antiferromagnetic ordering. There is no change in the ac susceptibility between 30–40 K but below 30 K the magnitude of $\partial\chi/\partial T$ increases to a shoulder at around 16.7 K. The insert in figure 3 helps to illustrate this.

The coefficient of thermal expansion of polycrystalline $CeAgSb_2$ in the temperature range below 40 K is shown in figure 3. Although the magnetic fluctuations appear to contribute to $\alpha_p(T)$ from below ∼29 K, there is no trace of the transition at 9.5 K. However, a maximum in the magnetic contribution to the linear thermal-expansion is seen at 16.9 K rising to a maximum value of 7.9×10^{-6} K^{-1}.

The application of an 8 T magnetic field parallel to the measurement direction increases the maximum value of α_p to 1.15×10^{-5} K^{-1} and the value of the temperature at which it occurs is increased to 17.3 K. It is found that the area underneath the 8 T curve is greater than the zero-field curve by 45%. An increase in area may indicate that on the application of a magnetic field the Grüneisen factor increases due to an associated increase in Ce intersite correlations, though the measurement of the field-dependent heat capacity is required for a full analysis.

The strength of the Kondo interaction relative to the RKKY interaction is reflected by the pressure dependence of the magnetic transition temperature $\partial(\ln T_N)/\partial P$ of the Néel temperature T_N: If the Kondo effect is sufficiently weak, the Néel temperature $T_N \sim \mathfrak{J}^2\mu_{eff}^2$ is expected to increase with pressure, owing to the increase in $|\mathfrak{J}|$, the absolute value of the exchange constant. However, if the Kondo effect is sufficiently strong, then the increase in magnitude of the exchange constant is smaller than the reduction of the effective Ce moment μ_{eff}, and this will result in $\partial(\ln T_N)/\partial P < 0$. Thus, a pressure measurement on $CeAgSb_2$, assuming that it is a concentrated Kondo lattice, shows where it is on the Doniach diagram (figure 2).

The pressure dependence of the Néel temperature, $\partial(\ln T_N)/\partial P$ was determined by measuring the ac susceptibility for a range of temperatures at pressures of up to 7.33 kbar. At ambient pressure a large peak is observed at around 9 K and a small kink is observed at around 6 K (figure 3). It is of interest to consider the shift in temperature of these two features under pressure. The peak at 9.5 K decreases to 8.8 K under a pressure of 7.33 kbar, this gives a value for $\partial(\ln T_N)/\partial P$ of -10.1 $Mbar^{-1}$, taking a typical bulk modulus B_T of 1 Mbar then the Grüneisen factor γ_P has a value of -10.1. This is comparable to $CeAl_2$, a well studied magnetic Kondo lattice material, in which the Néel temperature also decreases, with $\partial(\ln T_N)/\partial P$ of the order of -10 $Mbar^{-1}$. The low-temperature feature moves from 6.09 K at zero pressure to 6.96 K under a pressure of 7.33 kbar, so that $\partial(\ln T_c)/\partial P$ has a value of $+20.2$ $Mbar^{-1}$.

The pressure measurements, therefore, indicate that the main transition at 9.5 K reduces because of strong Kondo interaction. The Kondo interaction causes an increase of the exchange constant between the 4f and conduction electrons and overcompensates the reduction of the effective Ce moment μ_{eff}^2, resulting in a decrease in the Néel temperature. To resolve the $CeAgSb_2$ behaviour, neutron diffraction or specific-heat at different pressures investigations need to be carried out.

4. Discussion

CePd shows no unexpected behaviour around its Curie temperature (6.5 K). Both the thermal expansion data and the ac susceptibility measurements reflect the heat capacity results of Besnus *et al* [8], showing the presence of the ferromagnetic transition at 6.5 K,

and of a second transition, probably due to a magnetic spin reorientation at around 3.5 K. We have calculated the Grüneisen factor associated with the ferromagnetic ordering from our measurements by two methods. From the thermal expansion data and heat capacity measurements we obtain a Grüneisen factor $\gamma_G \approx 4.5$, and from the change in Néel temperature with applied pressure we also obtain $\gamma_P \approx 4.5$. The large positive value of $\partial(\ln T_c)/\partial P \sim +4.5$ Mbar^{-1} tends to indicate that CePd is a Kondo material, since except for rare occasions $\partial(\ln T_c)/\partial P$ is negative for a metallic ferromagnet with low T_c.

Unlike CePd, the Kondo system $CeAgSb_2$ displays an extraordinary disparity between the low-temperature thermal expansion and ac susceptibility. In particular, the absence of a thermal expansion peak at $T_N \sim 9.5$ K corresponding to the phase transition, and the existence of a maximum at the higher temperature of ~17 K are the most striking features of the low-temperature behaviour of $CeAgSb_2$. Possible explanations for these phenomena are discussed later.

It is likely that the origin of the maximum in the magnetic component of the linear thermal expansion at 16.9 K is related to the Kondo effect and the stabilization of coherence between the Ce sites. The onset of coherence is marked by the maximum in the electrical resistivity $\rho(T)$ at 24 K [9]. As the temperature decreases below this temperature, there is an increase in both χ_{ac} and α_p.

According to the Kondo model of periodic magnetic impurities, proposed by Kaga *et al* [15], coherent magnetic states start to form between Ce sites as the temperature decreases below a critical temperature. At very low temperatures $T \ll T_K$, the coherence between the independent scattering sites has fully developed, and a band of quasi-particles with large effective masses is formed, causing a pseudo-gap near the Fermi energy. This and other models [16, 17] suggest that a maximum in the thermal expansion will occur once the coherence between the Ce sites has stabilized with an energy gap ΔE. For temperatures below 10 K we have found that a function of the form

$$\alpha_p \propto T^3 \exp(-\Delta E/k_B T)$$

produces a good fit to the data. This is the expected form of behaviour for an antiferromagnetic structure with an energy gap. The best fit to our data is obtained with an energy gap of $\Delta E = 5.3$ K.

In order to put the results for $CeAgSb_2$ into perspective we will briefly consider three other ordered compounds, $CeAl_2$, $GdCu_5$ and CePdSb which also exhibit unusual behaviour in their transport properties as a function of temperature [18–20]. It should be noted, however, that though Houshair *et al* suggest that $CeAgSb_2$ orders into a canted antiferromagnet with a small ferromagnetic component [9], the magnetic structure of $CeAgSb_2$ is not known, therefore, such a comparison may be of limited value. It is, however, worth considering the more general problem of understanding the thermal properties of the whole class of cerium compounds. The problem arises from the complexity of the interaction between Kondo, magnetic and crystal electric field (CEF) interactions. Each of these can lead to unusual thermal behaviour. It is possible for a sharp peak to occur in α_p at low temperature, for example, even in a material that does not exhibit magnetic order, for example $CeCu_2Si_2$, Lacerda *et al* [21]. They attempted to fit the peak they observed at 9 K using a single ion Kondo theory of Schottke and Schottke [22] but it was later pointed out, Kumar *et al* [3], that the peak temperature must be a strong function of the CEF, which splits the $(2J + 1)$ degenerate ground state of the Ce ion into three doublets. The peak temperature calculated for zero CEF by Kumar *et al* moved from 6 K (i.e. from below the experimental value) to around 30 K using the experimental value of 228 K for the energy (D_1) of the first excited CEF energy level. This effect is quite separate from the

expected broad weak peak in α_p due to the CEF, which occurs in the temperature range near $D_1/2$. When the complication of magnetic ordering is added it is therefore perhaps not surprising that the shape and position of any peaks in α_p do not correspond to normal critical behaviour.

A system which may be analogous in many ways to $CeAgSb_2$ is $CeAl_2$. This system was investigated by Schefzyk *et al* [18]. The $\alpha_p(T)$ measurements show two distinct peaks which would appear to be two transitions: a sharp peak appearing at 3.8 K, and a broader feature appearing at 3.7 K. It is found that only the 3.8 K feature can be detected by ac susceptibility and is thought to correspond to the formation of commensurate magnetic structures near lattice defects in which the Kondo effect is weak. Only the 3.7 K feature can be detected by heat capacity measurements and is thought to originate from the well ordered regions of the material, in which incommensurate, long-range magnetic structures form with a strong associated Kondo effect.

A similar situation was found to arise in $GdCu_5$. From neutron diffraction Barandianran *et al* [19] showed that $GdCu_5$ ordered to a triangular antiferromagnetic. Such a magnetic structure shows many characteristics of incommensurate magnetic systems. It was found that, though the magnetic ordering of the Gd moments resulted in a significant increase in susceptibility, no anomaly appeared in the specific heat at the Néel temperature. A similar long-range or frustrated magnetic structure in $CeAgSb_2$, thought to order into a canted antiferromagnet with a small ferromagnetic component [9], could result in large susceptibility with no significant change in heat capacity or thermal expansion.

Another system in which there is no indication of the magnetic ordering from the heat capacity is CePdSb. Trovarelli *et al* [20] also suggest that the absence of a specific heat jump at T_c for CePdSb may be due to the incommensurate nature of the magnetic state, though there is no clear evidence for an incommensurate component to the magnetic structure in this compound. They also consider the possibility that the phenomenon is due to the anisotropic (effectively 2D) structure of CePdSb, with their model giving a thermal expansion or heat capacity peak at a lower temperature than the magnetic transition temperature. This is clearly not the case for $CeAgSb_2$. It is interesting to note that though our thermal expansion measurements on polycrystalline samples of CePdSb showed no evidence of any anomaly corresponding to T_c, careful measurements on a good single crystal did reveal the beginnings of normal critical behaviour in the thermal expansion at T_c [10–12]. This indicates that single crystal measurements of heat capacity and thermal expansion on $CeAgSb_2$ would be of interest in the future.

5. Conclusions

The properties of polycrystalline samples of CePd and $CeAgSb_2$ have been measured as a function of temperature using ac susceptibility and thermal expansion techniques. The ac susceptibility measurements were performed under a range of pressures up to $\sim$7.5 kbar and the thermal expansion was measured under different fields of up to 8 T. Both systems appear to be Kondo lattices. The behaviour of $CeAgSb_2$ however, unlike CePd, shows inconsistencies when measured by different transport techniques.

The low-temperature thermal expansion of CePd is in agreement with previous heat capacity data, showing a ferromagnetic transition at $T_c = 6.5$ K and a second transition at 3.5 K which probably arises from reorientation of the magnetic moments. The Grüneisen factor, calculated from thermal expansion and heat capacity is $\gamma_G \approx 4.5$, comparable to the Grüneisen factor calculated from the pressure dependence, $\gamma_P \approx 4.5$ is also comparable with a previous measurement of $\gamma_P \approx 6$.

For $CeAgSb_2$ the zero-pressure ac susceptibility measurements confirm that there is an antiferromagnetic transition, with a sharp peak appearing at $T_N = 9.5$ K. The Néel temperature is found to decrease under pressure at the rate of $\partial(\ln T_N)/\partial P = -10.1\ \mathrm{Mbar}^{-1}$, which indicates that $CeAgSb_2$ is a magnetic Kondo lattice on the right-hand side of the Doniach diagram. The ac susceptibility indicates that some magnetic correlations start to set in for temperatures below ∼26 K and a small shoulder in the curve is observed at around 17 K, and may be related to the Kondo effect, leading to changes in the coherence between Ce sites above T_N.

The magnetic contribution to the linear thermal-expansion coefficient of polycrystalline $CeAgSb_2$ (α_p) in zero magnetic field also reflects this behaviour, with a maximum value near 17 K. However, there is no peak in α_p at the Néel temperature. The behaviour of the system may be comparable to polycrystalline CePdSb where the peak in α_p is also absent at the magnetic transition temperature.

Measurements on single crystals are required to provide further information on the nature of the thermal expansion. It would also be of interest to perform heat capacity measurements in a magnetic field on this system to clarify whether the observed increase in area under the α_p curve with magnetic field is associated with an increase in the Grüneisen factor, due to an associated increase in Ce intersite correlations.

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