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Electrical resistivity of the Kondo system $Ce_{1-x}La_xPt_2Si_2$

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Abstract. The electrical resistivities of the Kondo system $Ce_{1-x}La_xPt_2Si_2$ ($0 \leqslant x \leqslant 1$) are reported. It is observed that the resistivities of the alloy samples are reduced considerably as a result of annealing the samples. The results furthermore indicate the evolution from dense Kondo behaviour to single-ion incoherent Kondo scattering as x is increased. The resistivity in the dense Kondo regime shows a maximum which drops from $T_{max} = 62$ K for $CePt_2Si_2$ to $T_{max} = 36$ K for $x = 0.2$. Using the relationship $T_{max} \propto T_K \propto \exp(-1/JN(E_F))$ where T_K is the Kondo temperature, J is the exchange integral and $N(E_F)$ is the density of states at the Fermi level E_F, and the experimentally observed values of $T_{max}(x)$ leads to $|JN(E_F)|_0 = 0.0645 \pm 0.0004$.

1. Introduction

The investigation of ternary compounds of the type RT_2X_2, where R is a rare earth element, T is a transition metal and X is Si or Ge, have attracted much interest [1–13]. They exhibit interesting physical properties due to either the localized or de-localized behaviour of the 4f electrons depending on the hybridization strength between 4f electrons and conduction electrons. Their properties include heavy-fermion, Kondo, mixed-valence, magnetic and superconducting behaviour. Of interest in heavy-fermion and related systems are the respective roles played by single-ion effects, like the on-site Kondo mechanism, and by collective phenomena, like the Kondo lattice coherence or the Rudermann–Kittel–Kasuya–Yosida (RKKY) interaction.

The low-temperature properties of $CePt_2Si_2$ are of recurrent interest since it was proclaimed by Gignoux *et al* [7] as a Kondo lattice compound. The compound crystallizes in a tetragonal structure, but some difference in interpretation concerns the appropriate space group which was given as $P4/nmm$ $CaBe_2Ge_2$ type [4, 6, 7] or as a new $CePt_2Si_2$ type structure $P4/mm$ [5]. Resistivity (ρ) measurements on a polycrystalline sample indicate a Kondo-like increase in $\rho(T)$ upon cooling from room temperature, culminating in a peak at $T_{max} = 76$ K and a large drop in $\rho(T)$ towards low temperatures [7]. The peak behaviour in $\rho(T)$ is confirmed in single-crystal studies [11, 12] which also indicate an appreciable anisotropy in $\rho(T)$. Specific heat studies [8] indicate a Kondo temperature $T_K \approx 70$ K. The specific heat $C_P(T)$ [8, 14] and resistivity [8] exhibit Fermi-liquid behaviour between 4 and 10 K, but at lower temperature $C_P(T)$ shows a strong enhancement [8, 14] and $\rho(T)$ tends to a linear temperature dependence [8] suggesting non-Fermi-liquid behaviour [14]. Inelastic neutron scattering indicates a $T_K \approx 170$ K and suggests that its properties be intermediate between those of intermediate-valent and heavy-fermion compounds [9].

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Early work indicated that $CePt_2Si_2$ orders antiferromagnetically at $T_N = 6$ K [6], a temperature where a small peak was also observed in the susceptibility of $CePt_2Si_2$ [4]. The latter authors suggest that the peak may originate from a second phase in their sample. A metamagnetic transition was indicated for a magnetic field of 2.5 T in the basal plane of a $CePt_2Si_2$ single crystal below 4 K [10]. This result was confirmed in a second investigation on an as-grown crystal, but the effect disappeared after the crystal was annealed for 8 days at 950 °C [14] and is hence not considered as an intrinsic property of the compound.

In this paper the results of $\rho(T)$ measurements on the $Ce_{1-x}La_xPt_2Si_2$ series of compounds are reported. The experiments probe primarily the effect of volume dilatation on the Kondo properties of $CePt_2Si_2$. Similar La substitution studies on antiferromagnetic $CePd_2Si_2$ indicate that its magnetic behaviour is rather of localized single-ion character than determined by intersite 4f–4f interaction [15, 16]. The effect of annealing our samples (10 days at 900 °C) on the measured $\rho(T)$ has also been investigated.

2. Experimental details

Polycrystalline samples of $Ce_{1-x}La_xPt_2Si_2$ were prepared by arc-melting the constituent elements in a purified argon atmosphere on a water-cooled copper hearth. Elements of the following purity in wt% were used: Ce and La, 99.99; Pt, 99.97 and Si, 99.999. The samples were melted three times to improve their homogeneity. X-ray diffraction measurements ascertained the phase purity of the samples and absence of unreacted material. The diffractograms indicated that the parent compounds $CePt_2Si_2$ and $LaPt_2Si_2$, as well as the pseudo-ternary alloys, crystallize in the tetragonal $CaBe_2Ge_2$ type structure. It was found that the lattice constant a increases continuously with La content x, while the lattice constant c remains approximately constant over the series giving an overall increase in unit cell volume from 176.5 Å^3 for $CePt_2Si_2$ to 182 Å^3 for $LaPt_2Si_2$.

The electrical resistivity was measured in the temperature range 4.2–300 K on spark-erosion cut bar-shaped samples using a standard four-probe method. Temperatures were measured with an Au–0.07 at.% Fe versus chromel thermocouple with an absolute accuracy of $T \pm 0.5$ K. Measurements were taken during slow cooling or heating runs in which the temperature changed at a rate of 0.5 K min^{-1}.

3. Results and discussion

Measurements of $\rho(T)$ for an as-cast $CePt_2Si_2$ sample and for the same sample after annealing are indicated in figure 1. At higher temperatures $\rho(T)$ shows the characteristic upturn in magnitude upon cooling as is to be expected for incoherent Kondo scattering. A well defined peak in $\rho(T)$ is observed for both the as-cast and annealed case at temperature T_{max} which is slightly higher for the annealed sample compared to the as-cast case. The overall magnitude of the resistivity is considerably reduced as a result of annealing, but the size of the drop in $\rho(T)$ below T_{max} to low temperatures is larger in the annealed case.

A small anomaly in $\rho(T)$ is evident for the as-cast sample at low temperatures. This may be related to the small peak in susceptibility and suggestion of magnetic ordering observed in some studies of $CePt_2Si_2$ [4, 6]. A possible origin could be the presence of a small amount of a second phase (below the limits of detection by x-rays). Candidates for this are ferromagnetic CePt ($T_c = 6.2$ K) [17] and Ce_7Pt_3 ($T_c = 6.85$ K) [18]. A more fundamental possible origin for such a low-temperature anomaly in Ce compounds is Kondo scattering from crystalline electric field levels as proposed by Maranzana [19] and comprehensively formulated by Cornut

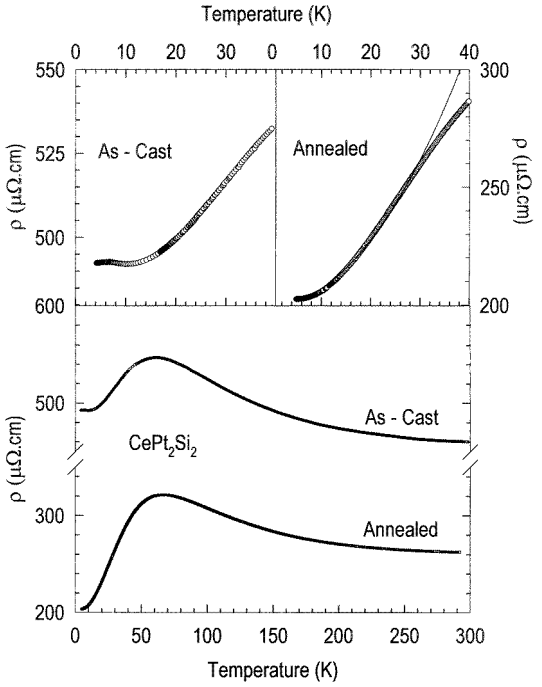

Figure 1. Temperature dependence of the resistivity $\rho(T)$ of $CePt_2Si_2$ for an as-cast sample and also showing the effect of subsequent annealing of the sample. A small anomaly is evident for the as-cast sample and is discussed in the text. After annealing a Fermi liquid $\rho(T) - \rho_0 = AT^2$ dependence is followed.

and Coqblin [20]. This could lead to more than one Kondo related maximum in resistivity as observed for $CeCu_2Si_2$ [21]. This latter explanation has also been advanced to account for the presence of a second peak in $\rho(T)$ in the dense Kondo system $CePtSi_2$ at low temperature [22]. It is indicated in figure 1 that the annealed sample follows a Fermi liquid $\rho(T) - \rho_0 = AT^2$ temperature dependence (solid line) for temperatures below 30 K with $A = 0.07\ \mu\Omega$ cm K^{-2} in agreement with previous observations [8].

Measurements of $\rho(T)$ showing the evolution from dense Kondo behaviour to single-ion incoherent Kondo scattering as x is increased for the $Ce_{1-x}La_xPt_2Si_2$ system are shown in figure 2 for the as-cast samples and in figure 3 for the same samples after annealing. As for the $CePt_2Si_2$ sample, all other samples in the series show a significant reduction of resistivity due to annealing. This beneficial lowering in the magnitude of $\rho(T)$ may firstly be associated with enhancing the metallurgical quality of the samples through the reduction of defects. On the other hand it has been indicated for RPt_2Si_2 compounds, and more specifically for $ErPt_2Si_2$, that Pt and Si atoms are statistically distributed among the 4(d) and 4(e) positions in the $ThCr_2Si_2$-type structure [23]. Annealing possibly affects this distribution and hence results in a change in the atomic periodicity in the crystal resulting in a change in the amount of electron scattering. These aspects pertaining to annealing are not further pursued in the present study, apart from observing that the peak in $\rho(T)$ is observed more clearly in the annealed samples for $x = 0.1$ and 0.2 than for the corresponding as-cast samples. This facilitated the extraction of values of T_{max} for these alloys, the significance of which will be discussed later.

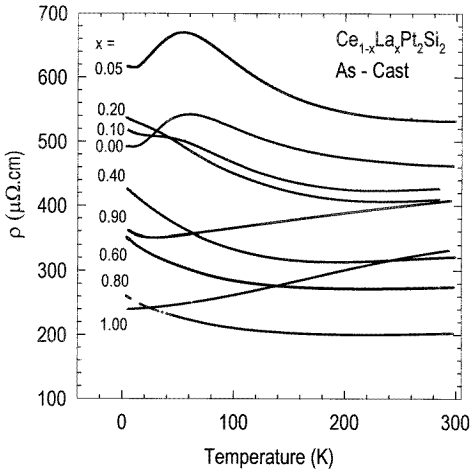

Figure 2. Resistivity against temperature for the as-cast series of $Ce_{1-x}La_xPt_2Si_2$ compounds.

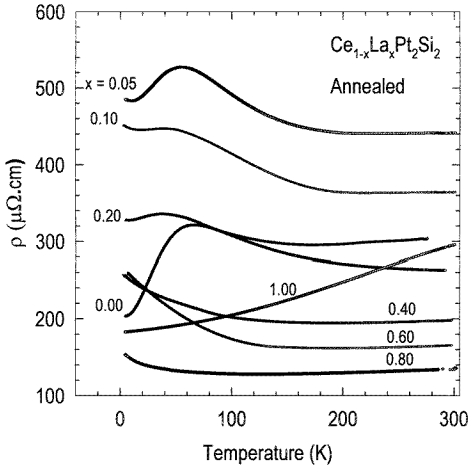

Figure 3. Resistivity against temperature for the annealed (10 days at 900 °C) series of $Ce_{1-x}La_x$ Pt_2Si_2 compounds.

The observed $\rho(T)$ curves in figures 2 or 3 originate from electron scattering from defects (ρ_0), from phonons (ρ_{ph}) and a magnetic term due to incoherent or coherent Kondo scattering (ρ_{mag})

$$\rho(T) = \rho_0 + \rho_{ph} + \rho_{mag}. \tag{1}$$

Ideally one would like to obtain ρ_{mag} for the various alloys by subtracting ρ_0 (considered to be primarily temperature independent) and $\rho_{ph}(T)$ from the measured $\rho(T)$. However, it has already been indicated that annealing of these samples leads to a significant reduction in their resistivities and consequently in their ρ_0 values. For the end-members of the series assignments of $\rho_0 = 202\ \mu\Omega$ cm for $x = 0$ and $\rho_0 = 183\ \mu\Omega$ cm for $x = 1$ are made in the case of the annealed samples. Unfortunately such estimates of ρ_0 are not feasible for samples with $0.1 \leqslant x \leqslant 0.8$. Therefore we rather consider the combination $\rho_0 + \rho_{mag}$ obtained by subtracting $\rho_{ph}(T)$ from the measured $\rho(T)$ data. We approximate $\rho_{ph}(T)$ for alloys across

the series by considering the temperature dependence of the resistivity of the non-magnetic $LaPt_2Si_2$ homologue. Thus $\rho_0 + \rho_{mag}(T)$ for the different alloys are obtained as

$$\rho_0 + \rho_{mag}(T) \approx \rho(T) - (\rho_{LaPt_2Si_2}(T) - \rho_{LaPt_2Si_2}(4\mathrm{K})). \tag{2}$$

In figure 4 we plot $\rho_0 + \rho_{mag}(T)$ on a $\log_{10}T$ scale for some annealed samples. All alloys exhibit a linear behaviour at higher temperature in agreement with a $(-\ln T)$ non-coherent single-ion Kondo scattering. The resistivity of the $CePt_2Si_2$ sample shows a peak and the effect of coherence at lower temperature. For low Ce concentrations one obtains a monotonic increase in resistivity with decrease in temperature and with a tendency towards saturation as expected for single-ion Kondo systems.

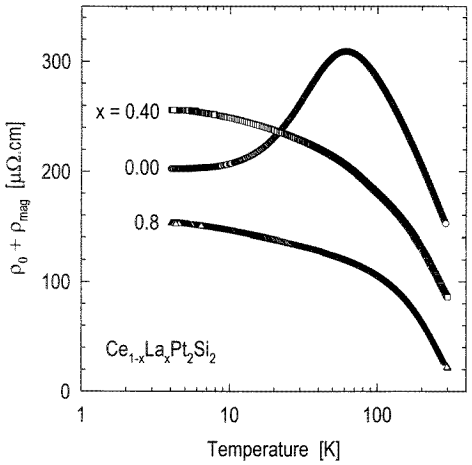

Figure 4. Comparison of the quantity $\rho_0 + \rho_{mag} = \rho(T) - (\rho_{LaPt_2Si_2}(T) - \rho_{LaPt_2Si_2}(4K))$ for representative samples of annealed $Ce_{1-x}La_xPt_2Si_2$ alloys.

Finally, the change in T_{max} with La substitution is addressed. It is evident comparing figures 2 and 3 that annealing enhances the occurrence of the Kondo peak and more readily enable the identification of T_{max} for compositions $x = 0.1$ and 0.2. Figure 4 shows the decrease in T_{max} with increase of La content and hence in unit cell volume of the system. This result is consistent with that of the pressure dependence of resistivity of $CePt_2Si_2$ which indicated an increase of T_{max} with pressure and hence with reduction in volume of the unit cell [8]. Our results will now be used to estimate $|JN(E_F)|$ for the system where $N(E_F)$ is the density of states at the Fermi level E_F and the exchange integral J is given by Coqblin and Schrieffer [24] as

$$J = \langle V_{k,f}^2 \rangle U/\varepsilon_f(\varepsilon_f + U). \tag{3}$$

$V_{k,f}$ is the matrix element that characterizes the hybridization between 4f and conduction electrons at E_F, U is the Coulomb integral and ε_f is the position of the 4f level relative to the Fermi level. Lavagna *et al* [25] indicated the close relation between T_{max} and T_K for Kondo-lattice systems.

$$T_{max} \propto T_K \propto \exp(-1/|JN(E_F)|). \tag{4}$$

For $Ce_{1-x}La_xPt_2Si_2$ the substitution of Ce with La increases the volume of the unit cell, which may lead to a decrease in $|JN(E_F)|$ and a decrease in T_K. The following volume dependence is assumed in the compressible Kondo model [25]

$$|JN(E_F)| = |JN(E_F)|_0 \exp(-n(V - V_0)/V_0) \tag{5}$$

where $|JN(E_F)|_0$ indicates the value of the quantity at initial volume V_0 and $|JN(E_F)|$ corresponds to volume V. The numerical constant n takes a value between 6 and 8 [25–27]. The exponential in equation (5) is expanded for small values of $\Delta V = V - V_0$ and the result for $|JN(E_F)|$ substituted in equation (4), yielding

$$\ln[T_{max}(V)/T_{max}(V_0)] = (n\Delta V/V_0|JN(E_F)|_0. \tag{6}$$

A least-squares fit analysis of the experimental $T_{max}(x)$ data against equation (6) is plotted as a solid line in figure 5. For ΔV we use values for the unit cell volumes of $CePt_2Si_2$ (176.5 Å^3) and $LaPt_2Si_2$ (182 Å^3) as measured with x-ray diffraction on our samples and assumed the validity of Vegard's rule to scale linearly in order to obtain $\Delta V(x)$. Assuming $n = 6$, the fit gives $|JN(E_F)|_0 = 0.0645 \pm 0.0004$ for $CePt_2Si_2$ in the limit of $x = 0$. A similar calculation may be performed by using the experimentally measured pressure dependence of $T_{max}(\mathrm{d}T_{max}/\mathrm{d}P = 2.2$ K kbar$^{-1})$ for $CePt_2Si_2$ [8]. However, we are not aware of measurements of the magnitude of the compressibility of $CePt_2Si_2$ and hence are unable to make quantitative comparison between the results of their experiment and ours. Our value of $|JN(E_F)|_0 = 0.0645$ may be compared with that of previous studies on heavy-fermion compounds $CeCu_6$ ($|JN(E_F)|_0 = 0.09$ [26]) and $CeInCu_2$ ($|JN(E_F)|_0 = 0.081$ [27] or 0.047 [28]).

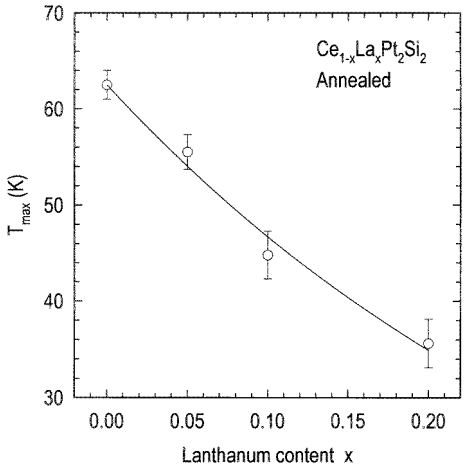

Figure 5. T_{max} as a function of La content for $Ce_{1-x}La_xPt_2Si_2$. The solid line is a least-squares fit of the data against a theoretical expression based on the compressible Kondo model (equation (6), see text).

4. Conclusions

Electrical resistivity measurements on the $Ce_{1-x}La_xPt_2Si_2$ ($0 \leqslant x \leqslant 1$) Kondo system indicates a large difference in the observed $\rho(T)$ between as-cast and annealed samples. A small anomaly in $\rho(T)$ is observed at low temperatures for the as-cast $CePt_2Si_2$ sample, but after annealing of the sample the resistivity follows a Fermi liquid $\rho(T) - \rho_0 = AT^2$ temperature dependence. The study furthermore demonstrates for the $Ce_{1-x}La_xPt_2Si_2$ alloy series the evolution from dense Kondo behaviour to single-ion incoherent Kondo scattering: at higher temperatures the resistivity, after subtraction of the contribution due to scattering from phonons (ρ_{ph}), follows a $(-\ln T)$ dependence. The Ce-concentrated samples ($0 \leqslant x \leqslant 0.2$) develops a

peak at T_{max} when cooled to lower temperatures. A value of $|JN(E_F)|_0 = 0.0645 \pm 0.0004$ (J is the exchange integral and $N(E_F)$ is the density of states at the Fermi level) has been obtained for $CePt_2Si_2$ using the compressible Kondo model and the observed dependence of T_{max} on La substitution. Future work may entail the effects of substitution on the Pt or Si sites.

Acknowledgments

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