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# Effects of La dilution on the CePt<sub>2</sub>Si<sub>2</sub> Kondo lattice

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# Abstract

Electrical resistivity ( $\rho(T)$ ) and magnetoresistivity (MR) measurements are presented for the (Ce<sub>1-x</sub>La<sub>x</sub>)Pt<sub>2</sub>Si<sub>2</sub> alloy system.  $\rho(T)$  data at low temperatures for alloys in the concentrated Kondo lattice regime  $0 \le x \le 0.2$  conform to a  $\rho(T) = \rho_0 + AT^2$  dependence and Fermi-liquid parameters A and  $T_{coh}$  are presented. The maximum that occurs in  $\rho(T)$  in the concentrated Kondo lattice regime at a temperature of  $T_{max}$  is no longer observed for samples with  $x \ge 0.3$  which show single-ion Kondo behaviour. MR measurements on alloys with x = 0.5, 0.7 and 0.9 yield values of the Kondo temperature  $T_K$  through Schlottmann analysis. The dependences of A,  $T_{max}$  and  $T_K$  on x are discussed in terms of the volume and Kondo hole effects occurring upon alloying. Anomalies in  $\rho(T)$  for LaPt<sub>2</sub>Si<sub>2</sub> and some of the alloy samples are also reported.

(Some figures in this article are in colour only in the electronic version)

# 1. Introduction

A fascinating modification of the Kondo lattice is the depletion of the local moments by replacing magnetic with non-magnetic rare-earth ions. There have been numerous experimental studies on such systems [1–6] and the tuning of it between the dense Kondo and single-impurity regimes by doping the f-ion sites with non-magnetic La, Y or Sc. The non-magnetic impurities in a Kondo lattice are called Kondo holes (see [7]) and an interesting aspect in such Kondo hole systems is the study of the dependence of transport and thermodynamic properties on the hybridization resulting from both dilution and volume effects. In this study effects of dilution by La on the CePt<sub>2</sub>Si<sub>2</sub> Kondo lattice are investigated.

The properties of the parent compound CePt<sub>2</sub>Si<sub>2</sub> are well documented as being a Kondo lattice compound with no magnetic order down to 0.06 K [8–18]. CePt<sub>2</sub>Si<sub>2</sub> crystallizes in a tetragonal structure, but some differences in interpretation exist regarding its appropriate space group, which is indicated to be the P4/nmm CaBe<sub>2</sub>Ge<sub>2</sub> type [8–11] or a new CePt<sub>2</sub>Si<sub>2</sub> type of structure [12]. The electrical resistivity  $\rho(T)$ 

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of polycrystalline CePt<sub>2</sub>Si<sub>2</sub> shows a Kondo-like logarithmic increase up to a maximum at  $T_{\text{max}} = 76$  K upon cooling and then a steep decrease towards low temperatures [9]. Magnetic susceptibility measurements on polycrystalline CePt<sub>2</sub>Si<sub>2</sub> show a normal Curie-Weiss behaviour above 150 K with a large negative paramagnetic Curie temperature ( $\theta_{\rm P} = -86$  K). Upon further cooling a broad maximum in  $\chi(T)$  is observed around 60 K and an almost constant value is attained below 20 K [9, 13, 14]. Measurements on CePt<sub>2</sub>Si<sub>2</sub> single-crystals show the strong effects of anisotropy on the magnetization [14] and electrical resistivity [15, 16]. The ratio of specific heat to temperature C/T drops linearly down to 4 K where the electronic specific heat coefficient  $\gamma$  has a value of 70-86 mJ K<sup>-2</sup> mol<sup>-1</sup> [9, 13, 17] below which C/T departs strongly from linearity, turning up to reach a maximum of 120 mJ  $K^{-2}$  mol<sup>-1</sup> at 2 K [9, 17]. While the specific heat [13, 17] and resistivity [13] exhibit Fermi-liquid behaviour between 4 and 10 K, measurements of specific heat and electrical resistivity below 4 K suggest non-Fermi-liquid behaviour [13, 17].

Previous alloying studies on  $CePt_2(Si_{1-x}Ge_x)_2$  [19],  $Ce(Pt_{1-x}Ni_x)_2Si_2$  [20] and  $CePt_2(Si_{1-x}Sn_x)_2$  [21] ligand systems, as well as a study on a limited number of

 $(\text{Ce}_{1-x}\text{La}_x)\text{Pt}_2\text{Si}_2$  alloys [22] have been interpreted in terms of the effect of the volume changes induced by chemical pressure in these alloys on the Kondo parameters by invoking the compressible Kondo lattice model [23]. The present much more detailed study of  $\rho(T)$  and which is also extended to include magnetoresistance (MR) measurements of the  $(\text{Ce}_{1-x}\text{La}_x)\text{Pt}_2\text{Si}_2$  system enables us not only to interpret the results in terms of volume effects, but also to include the effects due to dilution (Kondo holes) in our description of the results. In addition, anomalous behaviour of  $\rho(T)$  is found in the present study which was not uncovered by the authors of reference [22].

### 2. Experimental details

Polycrystalline samples of  $(Ce_{1-x}La_x)Pt_2Si_2$  ( $0 \le x \le 1$ ) were prepared by arc-melting stoichiometric amounts of the constituent elements on a water-cooled copper hearth in a titanium gettered ultra high purity inert argon gas atmosphere. The purities in wt% of the materials used were Ce: 99.98, La: 99.99, Pt: 99.97 and Si: 99.999. A weight-loss of less than 0.5% was observed during melting for all alloys.

X-ray powder diffraction measurements on the  $(Ce_{1-x}La_x)$  Pt<sub>2</sub>Si<sub>2</sub> alloys ascertained the phase purity of the samples and the absence of unreacted elements. The lattice parameters *a* and *c* of the tetragonal compounds were calculated using standard regression analysis of 22 well-resolved peaks of the powder spectrum of each alloy.

The temperature dependent electrical resistivity  $\rho(T)$  was measured using a standard dc four-probe technique in the temperature range 4–300 K on bar-shaped samples of typical dimension  $8 \times 1 \times 1$  mm<sup>3</sup> cut by spark-erosion. Temperatures were measured using a Au–0.07 at.% Fe versus chromel thermocouple with an absolute accuracy of  $T \pm 0.5$  K. Magnetoresistance measurements have also been performed on selected alloys in fields up to 8 T.

#### 3. Results and discussion

#### 3.1. X-ray diffraction analysis

Powder XRD results indicate a tetragonal structure for all compositions of the  $(Ce_{1-x}La_x)Pt_2Si_2$  system. However, our powder diffraction data cannot distinguish between the CePt<sub>2</sub>Si<sub>2</sub>-type [12] and CaBe<sub>2</sub>Ge<sub>2</sub>-type [8–11] structures since the additional reflections (*hk*0) with h + k = 2n + 1 appearing in the CePt<sub>2</sub>Si<sub>2</sub>-type are very faint and are discernible only from single-crystal data according to [12]. Our powder XRD results are therefore analysed on the basis of the tetragonal CaBe<sub>2</sub>Ge<sub>2</sub>-type structure. Lattice parameters a and c, c/a and the unit-cell volume of the alloy series are shown in figure 1. It is observed that the parameters a and c and the unit-cell volume increase linearly with x following Vegard's rule and the c/a ratio decreases with increasing x. The determined room temperature tetragonal lattice parameters of CePt<sub>2</sub>Si<sub>2</sub>, viz.  $a = 4.253(2) \times 10^{-1}$  nm,  $c = 9.793(4) \times 10^{-1}$  nm and the crystallographic unit-cell volume  $V = 177.1(2) \times 10^{-3} \text{ nm}^{-3}$ are in agreement with the results given in [8, 10, 12] which



**Figure 1.** The tetragonal lattice parameters (a) *a* and *c*, (b) the unit-cell volume *V* and the ratio c/a of alloy compositions  $0 \le x \le 1$  of the  $(\text{Ce}_{1-x}\text{La}_x)\text{Pt}_2\text{Si}_2$  system.

report that V varies in a narrow range of 177.0–177.2  $\times$  $10^{-3}$  nm<sup>3</sup>. Somewhat different values of V for CePt<sub>2</sub>Si<sub>2</sub> are reported by the authors of [9]  $(177.8 \times 10^{-3} \text{ nm}^3)$  and of [22]  $(176.5 \times 10^{-3} \text{ nm}^3)$ . Furthermore, for LaPt<sub>2</sub>Si<sub>2</sub> we obtain  $a = 4.286(1) \times 10^{-1}$  nm,  $c = 9.823(2) \times 10^{-1}$  nm and  $V = 180.4(1) \times 10^{-3} \text{ nm}^3$  and these values compare well with  $a = 4.2871(5) \times 10^{-1}$  nm,  $c = 9.8282(25) \times 10^{-1}$  nm and  $V = 180.63(9) \times 10^{-3}$  nm<sup>3</sup> reported by Rogl [24] and are also in fair agreement with values reported by Gignoux et al [9] ( $a = 4.28 \times 10^{-1}$  nm,  $c = 9.87 \times 10^{-1}$  nm and  $V = 180.8 \times 10^{-3} \text{ nm}^3$ ). It is noted that Bouzianne and du Plessis [22] reported  $V = 182 \times 10^{-3}$  nm<sup>3</sup> which is appreciably larger than what is observed in the present investigation. The authors of [22] also reported that the lattice parameter c is approximately constant across the series whereas we now observe a small continuous increase in c with increase in x. The initial study reported in [22] was undertaken with a x-ray diffractometer of less sensitivity than used in the present study. Hence the volume changes reported in [22] should be viewed as exploratory results in view of the present much more in-depth and higher resolution data.

#### 3.2. Resistivity measurements

The  $\rho(T)$  measurements as a function of temperature on the  $(\operatorname{Ce}_{1-x}\operatorname{La}_x)\operatorname{Pt}_2\operatorname{Si}_2$  alloys are shown in figure 2 which illustrates a transition from the dense Kondo regime  $(0 \le x \le 0.25)$  through the single-ion Kondo regime  $(0.3 \le x \le 0.9)$  to metallic behaviour (x = 1). A well-resolved anomalous kink in  $\rho(T)$  is evident in figure 2(b) for LaPt<sub>2</sub>Si<sub>2</sub>, as well as in some of the other alloys and this phenomenon will be discussed in section 3.2.3. In the dense Kondo regime, single-ion Kondo scattering dominates at higher temperatures and the  $\rho(T)$  curves initially display a logarithmic upturn upon



**Figure 2.** Dependence of the resistivity on temperature for the alloy compositions  $0 \le x \le 1$  of the  $(Ce_{1-x}La_x)Pt_2Si_2$  system. The inset shows the  $T_{max}$  dependence on x.



**Figure 3.** Fermi-liquid  $T^2$  behaviour of the resistivity for the  $(Ce_{1-x}La_x)Pt_2Si_2$  alloys with  $0 \le x \le 0.2$ . The arrows refer to the onset of the  $T^2$ -dependence and hence to  $T_{coh}$  of each alloy.  $T_{coh}$  has been taken as the highest temperature point for which the linear relationship between  $\rho(T) - \rho_0$  and  $T^2$  is still evident.

cooling. Subsequently  $\rho(T)$  goes through a maximum at temperature  $T_{\text{max}}$  as a result of coherence effects, and further cooling gives rise to Fermi-liquid behaviour which is shown in figure 3. As illustrated in the inset in figure 2,  $T_{\text{max}}$  decreases from 64 K for x = 0 to 15.5 K for x = 0.25, extending the single-ion Kondo scattering region to lower temperatures. The solid lines in figure 3 are linear least-squares (LSQ) fits of the relation  $\rho(T) = \rho_0 + AT^2$  ( $\rho_0$  is the residual resistivity) to the data below the coherence temperature  $T_{\text{coh}}$ , the results of which are given in table 1. The determined value  $A = 0.0787 \times 10^{-8} \Omega \text{ m K}^{-2}$  for CePt<sub>2</sub>Si<sub>2</sub> is in fair agreement with previous observations [13, 22].

3.2.1. Fermi-liquid parameters A and  $T_{\rm coh}$ . The interplay between Kondo hole and volume effects on the Fermi-liquid parameters A and  $T_{\rm coh}$  is discussed by comparing these



**Figure 4.** Fermi-liquid parameters *A* and  $T_{coh}$  of  $(Ce_{1-x}La_x)Pt_2Si_2$  (this work) and  $Ce(Pt_{1-y}Ni_y)_2Si_2$  [20] systems are plotted against unit-cell volume. The dashed curves are a guide for the eye. Volume dependence of (a) the  $T^2$  coefficient *A* of  $\rho(T)$  (LHS scale); (b) the temperature  $T_{coh}$  below which the  $T^2$  behaviour of  $\rho(T)$  is observed (RHS scale). The contribution of volume and dilution effects on compositions  $0 \le x \le 0.2$  are compared with the results of compositions  $0 \le y \le 0.5$  for which only a volume effect is indicated.



**Figure 5.** Plot of  $A^{1/2}(\propto \gamma)$  and  $T_{\rm coh}$  against unit-cell volume of  $({\rm Ce}_{1-x}{\rm La}_x){\rm Pt}_2{\rm Si}_2$  ( $0 \le x \le 0.2$ ). The curves are a guide for the eye.

**Table 1.** Results of LSQ fits of the relation  $\rho(T) = \rho_0 + AT^2$  to the  $\rho(T)$  data in figure 3.

	-		
x	$T_{\rm coh}~({\rm K})$	$A (10^{-8} \Omega \text{ m K}^{-2})$	$ ho_0 \ (10^{-8} \ \Omega \ \mathrm{m})$
0.00	21.7	0.0787(1)	337.04(2)
0.025	19.5	0.1509(2)	277.09(3)
0.050	19.7	0.1366(3)	344.21(3)
0.075	18.4	0.1182(2)	329.28(2)
0.10	17.4	0.1428(4)	387.57(4)
0.15	13.8	0.0575(2)	388.56(1)
0.20	10.5	0.0452(1)	439.65(1)

parameters for  $(Ce_{1-x}La_x)Pt_2Si_2$  ( $0 \le x \le 0.2$ ) with those obtained for  $Ce(Pt_{1-y}Ni_y)_2Si_2$  ( $0 \le y \le 0.5$ ) [20] in figures 4 and 5. It is seen from figure 4 that *A* increases with a decrease in *y* and hence increase in volume for the  $Ce(Pt_{1-y}Ni_y)_2Si_2$  series. This result is in agreement with

pressure studies on CePt<sub>2</sub>Si<sub>2</sub> which show a rapid decrease in A with pressure [13] and hence an increase in A with increase in volume. Similar results are also obtained for CeCu<sub>2</sub>Si<sub>2</sub> [25], UBe<sub>13</sub> [26], CeInCu<sub>2</sub> [27] and Ce<sub>7</sub>Ni<sub>3</sub> [28] through hydrostatic pressure studies. The results for the  $(Ce_{1-x}La_x)Pt_2Si_2$  system show a large increase in A with increase in volume for the x = 0.025 alloy with respect to the x = 0 parent compound (table 1 and figures 4 and 5). This result is in agreement with the pressure studies mentioned earlier [13]. All the preceding results are considered as due to the dominance of volume effects in the behaviour of the A coefficient. However, A of  $(Ce_{1-x}La_x)Pt_2Si_2$  shows a decrease with further increase in x and this is considered as due to the dominance of the dilution effect over the volume effect as may be motivated by considering the theoretical predicted properties of Kondo hole systems [7, 29, 30]. Substituting Ce by non-magnetic Kondo holes gradually smears out the hybridization (pseudo) gap, which is formed around the renormalized f-level position. Hence the gradual substitution of Ce with La to form Kondo holes results in a gradual diminishing of the coherence thus causing a decrease in A and in  $T_{\rm coh}$  as the f-electron density of states gradually changes from a two-peak coherent state to a single-peak incoherent state [29, 30]. A decrease in A with increase in La concentration and hence volume is also observed in several La based systems such as  $(Ce_{1-x}La_x)Cu_2Si_2$  [31],  $(Ce_{1-x}La_x)Cu_6$  [32] and  $(Ce_{1-x}La_x)Ni_9Si_4$  [33] and may thus also be interpreted as due to the dominance of the Kondo hole effect over the volume effect. Furthermore, the behaviour of  $\gamma (\propto \sqrt{A})$  for the  $(Ce_{1-x}La_x)_3Al$  system which shows a maximum around x = 0.6 [34] is considered to arise due to the competing interplay of these two effects. Theoretical calculations based only on Kondo hole substitution without reference to a volume contribution predict a decrease in A [35]. Turning the attention to  $T_{\rm coh}$ , it is noted that it is expected to decrease with increase in volume (negative pressure) [25-28] as is also seen for the  $Ce(Pt_{1-v}Ni_v)_2Si_2$  system in figure 4. However, an additional contribution to the decrease of  $T_{\rm coh}$  due to dilution is evident for the  $(Ce_{1-x}La_x)Pt_2Si_2$  alloys.

Values for the *A* parameter may also be obtained from an earlier study of  $\rho(T)$  for the pseudo-ternary CePt<sub>2</sub>(Si<sub>1-x</sub>Ge<sub>x</sub>)<sub>2</sub> system for which the volume increases with increasing *x* [19]. Since the Ce sublattice is not diluted an increase in *A* with increasing *x* is expected. Measurements of  $\rho(T)$  for CePt<sub>2</sub>(Si<sub>1-x</sub>Ge<sub>x</sub>)<sub>2</sub> were only performed for temperatures above 4 K. In the investigated temperature range distinct Fermi-liquid behaviour is only observed for alloys with x = 0 and 0.1, yielding  $A = 0.06 \times 10^{-8} \Omega$  m K<sup>-2</sup> for x = 0 and  $A = 0.10 \times 10^{-8} \Omega$  m K<sup>-2</sup> for x = 0.1 [19]. These results agree with an expected increase in *A* with volume, but for a more complete verification of this result a study of more alloys down to lower temperatures is needed.

Figure 6 illustrates the evolution from the Kondo lattice regime, through the single-ion Kondo regime to metallic behaviour with increased La concentration. One observes the destruction of coherence as the periodicity of the Ce sublattice is destroyed by the substitution of La, which suppresses the drop of  $\rho(T)$  at low temperatures. The gradual destruction of coherence in the Kondo lattice can be well illustrated by



**Figure 6.** Plot of  $\rho(T)/\rho(297 \text{ K})$  against temperature which illustrates the evolution from the Kondo lattice regime, through the single-ion Kondo regime to metallic behaviour. The inset shows the  $\delta \rho_x(T) = \rho_x(T) - (\rho_{x=0}(T) - \rho_{x=0}(0))$  results of alloy compositions  $0.025 \le x \le 0.1$  normalized to 150 K, which illustrates suppression of coherence with a small amount of La substitution on the CePt<sub>2</sub>Si<sub>2</sub> Kondo lattice.

subtracting  $(\rho_{x=0}(T) - \rho_{x=0}(0))$  of CePt<sub>2</sub>Si<sub>2</sub> from  $\rho_x(T)$  of the dense Kondo  $(0.025 \le x \le 0.1)$  alloys as shown in the inset in figure 6. It is seen that the relative drop in  $\delta\rho(T)/\delta\rho(150 \text{ K})$  decreases and attains a higher  $\delta\rho(4 \text{ K})/\delta\rho(150 \text{ K})$  value with a small increase in La concentration.

3.2.2. Dilution and volume effects on  $T_{\text{max}}$ . For many Ce Kondo systems in the concentrated Kondo region the Kondo energy scale is often characterized by  $T_{\text{max}} \propto T_{\text{K}}$ , where  $T_{\text{K}}$  is the Kondo temperature. The volume dependence of  $T_{\text{K}}$  is addressed by the compressible Kondo lattice (CKL) model [23] which gives the volume dependence of  $|J N(E_{\text{F}})|$ as  $|J N(E_{\text{F}})| = |J N(E_{\text{F}})|_0 \exp[-q(V - V_0)/V_0]$ , where  $|J N(E_{\text{F}})|_0$  indicates the value of the quantity at an initial volume  $V_0$ . J is the on-site Kondo exchange interaction and  $N(E_{\text{F}})$  is the density of states at the Fermi level. Furthermore, q refers to the Grüneisen parameter of  $|J N(E_{\text{F}})|$  (i.e. q = $-\partial \ln |JN(E_{\text{F}})|/\partial \ln V$ ) and it is considered to vary between 6 and 8 [23, 27]. Since  $T_{\text{max}} \propto T_{\text{K}} \propto \exp(-1/|JN(E_{\text{F}})|)$  the volume dependence of  $T_{\text{max}}$  may be described by

$$T_{\max}(V) = T_{\max}(V_0) \exp\left[\frac{q (V_0 - V)}{|JN(E_F)|_0 V_0}\right]$$
(1)

as done in many such studies in the literature [19, 21, 27, 28], the volume changes being effected by appropriate alloying or through application of hydrostatic pressure.

There is on the other hand an additional effect when the Ce sublattice is diluted by La substitution. Theoretical studies based only on the dilution (Kondo hole) effect and not considering the volume effect indicate for this case that an increase in x extends the incoherent Kondo scattering region to lower temperatures [30, 35, 36]. The gradual



**Figure 7.** Unit-cell volume dependence of the position of the resistivity maximum  $T_{\text{max}}$  of the Kondo hole system  $(\text{Ce}_{1-x}\text{La}_x)\text{Pt}_2\text{Si}_2$  and the ligand system  $\text{Ce}(\text{Pt}_{1-y}\text{Ni}_y)_2\text{Si}_2$  [20]. The dashed line is a LSQ fit to equation (1) for the  $\text{Ce}(\text{Pt}_{1-y}\text{Ni}_y)_2\text{Si}_2$  system [20]. The inset shows the *x* dependence of the dilution contribution  $T_{\text{max}}^{Kh}(x) = T_{\text{max}}(V_{\text{ref}}) - T_{\text{max}}(x)$ .

change from a two-peak to a single-peak density of f-electron states associated with a change from coherent to incoherent behaviour takes place around a characteristic concentration  $x_c$  [29]. To provide contact between experiment and theory one may identify  $x_c$  with the concentration of La at which  $T_{\text{max}}$  is just no longer observed upon increased dilution. For the  $(\text{Ce}_{1-x}\text{La}_x)\text{Pt}_2\text{Si}_2$  system  $x_c \approx 0.25-0.3$ . The value of  $x_c$  as introduced here varies for different Kondo systems as deduced from experimental results in the literature and is as diverse as  $x_c \approx 0.03$  for  $(\text{Ce}_{1-x}\text{La}_x)\text{Pd}_3$  [37] and  $x_c \approx 0.6$  for  $(\text{Ce}_{1-x}\text{La}_x)\text{Cu}_2\text{Si}_2$  [31]. Hence, there is an additional contribution  $T_{\text{max}}^{Kh}$  due to dilution with La, which is expected over and above the conventionally considered volume effect.

Alloying studies on the  $Ce(Pt_{1-y}Ni_y)_2Si_2$  system show the dominance of pressure (i.e. volume) effect on  $T_{max}$  as compared with other compounds investigated earlier with hydrostatic pressure studies and displayed in figure 10 of [20]. Hence, the ligand system  $Ce(Pt_{1-y}Ni_y)_2Si_2$  has been used as a reference for the volume effect by applying the CKL model with which to obtain a measure of  $T_{\text{max}}^{Kh}$  for the  $(\text{Ce}_{1-x}\text{La}_x)\text{Pt}_2\text{Si}_2$ system. Data from [20] for  $Ce(Pt_{1-y}Ni_y)_2Si_2$  are reproduced in figure 7 and a LSQ fit of (1) to the experimental data (solid circles) is shown as a dashed line and gives  $q/|JN(E_F)|_0 =$ 21.7(2). This curve has been extrapolated to the negative pressure region and used as a reference  $T_{\text{max}}(V_{\text{ref}})$  of the volume effect on  $T_{\text{max}}$  for CePt<sub>2</sub>Si<sub>2</sub> based systems. Hence, the dilution contribution  $T_{\max}^{Kh}(x) = T_{\max}(V_{\text{ref}}) - T_{\max}(x)$  has been calculated for the  $(Ce_{1-x}La_x)Pt_2Si_2$   $(0 \le x \le 0.25)$  system and plotted in the inset to figure 7. As seen in the inset,  $T_{\text{max}}^{Kh}$ increases as x approaches the characteristic concentration  $x_c$ at which the resistivity peak disappears. For larger values of  $x, x > x_c$ , the Kondo energy scale needs to be obtained from other means to further ascertain the effect of dilution. Values



**Figure 8.** (a) The kink in the  $\rho(T)$  data of  $(Ce_{1-x}La_x)Pt_2Si_2$ ( $0.3 \le x \le 1.0$ ) alloys and (b) an enlarged figure of the kink in LaPt\_2Si\_2, showing hysteresis between cooling and heating runs. (c) Plot of  $T_M$ , the temperature at which a kink with a hysteresis appears (taken approximately at the position with maximum hysteresis of  $\rho(T)$ ) against unit-cell volume and La concentration *x* of the (Ce<sub>1-x</sub>La<sub>x</sub>)Pt\_2Si\_2 ( $0.3 \le x \le 1.0$ ) alloys, which shows approximately a linear increase of  $T_M$  with unit-cell volume.

of  $T_{\rm K}$  are obtained for alloys that are in the single-ion Kondo regime through MR measurements as described in section 3.3.

A low temperature resistivity anomaly in La rich 3.2.3. alloys. Measurements on bars cut from several different ingots confirm a kink in  $\rho(T)$  for our LaPt<sub>2</sub>Si<sub>2</sub> samples that has not been observed in previous studies of this compound [9, 22]. This kink is also seen in other  $(Ce_{1-x}La_x)Pt_2Si_2$  alloys in figures 2, 6 and 8, and is shifted towards lower temperatures with the decrease of La concentration. It is shown in figures 8(a) and (b) that there is a marked hysteresis associated with this kink, with the kink occurring at a lower temperature during the cooling run than during the heating run. Similar kinks with hysteresis in the electrical resistivity have been reported by Besnus et al [38] in ternary RInAu<sub>2</sub> intermetallics with R = La, Ce, Pr and Nd. It has been shown in their work that a phase transition from the high temperature cubic to low temperature tetragonal structure occurs at a transformation temperature  $T_M$ . Such anomalies in the resistivity have also been reported in numerous cubic CsCl-type compounds like  $LaAg_{1-x}In_x$  [39],  $CeAg_{1-x}In_x$  [40] or RCd, where R is a rare-earth element [41]. LaAg<sub>1-x</sub>In<sub>x</sub> and CeAg<sub>1-x</sub>In<sub>x</sub> have the cubic CsCl structure at room temperature and undergo a martensitic transition to a tetragonal phase at low temperatures, which is ascribed to the Jahn–Teller effect [40]. In figure 8(c) it is shown that the transformation temperature  $T_M$  increases linearly with unit-cell volume for our  $(Ce_{1-x}La_x)Pt_2Si_2$  alloys. As an approximation values of unit-cell volume derived from XRD measurements at room temperature were used in figure 8 since we lacked the facilities to study the temperature dependence of the lattice parameters. Extrapolation of our  $T_M$ points shows that such a kink should appear around 62 K in  $\rho(T)$  of CePt<sub>2</sub>Si<sub>2</sub>. However, the anomaly is not observed in CePt<sub>2</sub>Si<sub>2</sub>, and since the hysteresis associated with the kink is

more prominent in La rich samples, it suggests that the La in the alloy is responsible for this anomaly. Temperature-dependent measurements of the structural parameters of the alloy compositions exhibiting the anomaly are called for to ascertain if a structural phase transition is associated with the anomaly in  $\rho(T)$ .

It is furthermore noted that the shape of the  $\rho(T)$  anomaly in LaPt<sub>2</sub>Si<sub>2</sub> is similar to the anomaly in  $\rho(T)$  that is observed at the first-order charge-density wave (CDW) transition that occurs at 83 K in single-crystal samples of Lu<sub>5</sub>Ir<sub>4</sub>Si<sub>10</sub> [42, 43] and which has first been observed in polycrystalline samples of this compound at 79 K [44]. The CDW occurring along the tetragonal *c*-axis of Lu<sub>5</sub>Ir<sub>4</sub>Si<sub>10</sub> below 83 K is characterized by superlattice x-ray reflections with  $\vec{q} \approx (0, 0, \frac{3}{7})$  and it coexist with superconductivity below 3.9 K [42, 43]. It would be of interest to explore the possibility if the  $\rho(T)$  anomaly in LaPt<sub>2</sub>Si<sub>2</sub>, which exhibits a superconducting transition at 2.2 K [9], could be associated with a CDW transition by studying its structural properties, heat capacity and magnetic susceptibility.

#### 3.3. Magnetoresistivity measurements

The magnetoresistivity defined as MR =  $[\rho(T, \mu_0 H) - \rho(T, 0)]/\rho(T, 0)$  was measured for isotherms between 1.5 and 60 K for alloys of composition x = 0.5, 0.7 and 0.9 which all are in the single-ion Kondo regime as is evident from figure 6. The MR isotherms were measured in transverse fields  $\mu_0 H$  up to 8 T. A typical example of the results is shown in figure 9 for the (Ce<sub>0.5</sub>La<sub>0.5</sub>)Pt<sub>2</sub>Si<sub>2</sub> sample. Results taken during increasing fields always fall on the same curve with data taken during decreasing fields. The behaviour in figure 9 is typical of the suppression of the incoherent Kondo scattering in a magnetic field. Except for the isotherm at the lowest temperature of 1.5 K, it is observed that the data are well described by the results of the Bethe ansatz calculations of the Coqblin–Schrieffer model as predicted by Andrei [45] and Schlottmann [46]

$$\frac{\rho(B)}{\rho(0)} = \left[\frac{1}{2j+1}\sin^2\left(\frac{\pi n_{\rm f}}{2j+1}\right)\sum_{\ell=0}^{2j}\sin^{-2}(\pi n_{\ell})\right]^{-1}.$$
 (2)

As discussed in more detail in [20], at low enough temperatures  $(T \ll \Delta_{\rm CF}, \text{ where } \Delta_{\rm CF} \text{ is the overall crystal field splitting})$ in systems having  $T_{\rm K} < \Delta_{\rm CF}$ , only the ground state crystal field level is populated and hence at such low temperatures the system behaves as if in the spin- $\frac{1}{2}$  state. The overall crystal field splitting in the CePt<sub>2</sub>Si<sub>2</sub> compound is reported to be  $\Delta_{\rm CF} \approx 100$  K [47]. Results of specific heat and susceptibility experiments indicate Kondo temperature  $T_{\rm K}$  values ranging from 45 K [9] to 70 K [13] for  $CePt_2Si_2$ . Hence for the  $(Ce_{1-x}La_x)Pt_2Si_2$  (0.5  $\leq x \leq 0.9$ ) alloys (since  $T_K$  is expected to decrease due to volume and dilution effects as observed in the decrease of  $T_{\text{max}}$  with increasing x)  $T_{\text{K}} < \Delta_{\text{CF}}$  and as an approximation only the ground state doublet may effectively operate  $(j = \frac{1}{2})$  at low temperatures  $(T < T_{\rm K})$ . The solid lines in figure 9 are LSQ fits for the  $j = \frac{1}{2}$  case of (2) to the experimental data. The exact solutions for  $j = \frac{1}{2}$  in the



**Figure 9.** The magnetic field dependence of the electrical resistivity at a number of sample temperatures for the alloy  $(Ce_{0.5}La_{0.5})Pt_2Si_2$ . The data have been measured in increasing and decreasing fields without evidence of hysteresis. The solid curves in the main figure are LSQ fits of the Bethe ansatz theory of magnetoresistivity given by (2) to the experimental data. The data for the 1.5 K isotherm deviate from (2) (depicted as a dotted line). The inset shows the temperature variation of the characteristic field  $B^*(T)$ , and the solid line is a LSQ fit of (3) to the data.

**Table 2.** Values of the Kondo temperature  $T_{\rm K}$  and the magnetic moment of the Kondo ion  $\mu_{\rm K}$  as obtained from LSQ fits of (2) and (3) to the MR data of the alloy compositions  $0.5 \le x \le 0.9$  of the (Ce<sub>1-x</sub>La<sub>x</sub>)Pt<sub>2</sub>Si<sub>2</sub> system.

x	0.5	0.7	0.9
$\frac{T_{\mathrm{K}}\left(\mathrm{K}\right)}{\mu_{\mathrm{K}}\left(\mu_{\mathrm{B}}\right)}$	$\begin{array}{c} 11.4 \pm 0.4 \\ 0.111 \pm 0.002 \end{array}$	$5.6 \pm 1.0$ $0.078 \pm 0.005$	$2.6 \pm 0.6$ $0.039 \pm 0.003$

above model indicate that the MR is completely determined by a single parameter, the characteristic field  $B^*$  [46], which is expected to have the following temperature dependence [48]

$$B^{*}(T) = B(0) + \frac{k_{\rm B}T}{g\mu_{\rm K}} = \frac{k_{\rm B}(T_{\rm K}+T)}{g\mu_{\rm K}}.$$
 (3)

Values of  $B^*(T)$  obtained from the LSQ fits of (2) to the data in the main figure, are plotted in the inset in figure 9. The linear relation predicted by (3) is borne out at low temperature, but there is some deviation from linearity of the  $B^*$  value of the 60 K isotherm which may be associated with an increased population of the first excited crystal field level with increasing temperature. A LSQ fit of (3) to the  $B^*(T)$  points, yields  $B^*(0)$ , hence  $T_K$ , and also the corresponding magnetic moment  $\mu_K$  of the Kondo ion. Values of  $T_K$  and  $\mu_K$  thus obtained for the investigated alloys are given in table 2. It is shown in figure 10 that all isotherms between 5 and 20 K conform to the universal scaling of  $B^*(T)$ . A related scaling behaviour encompassing both the x = 0 and 0.5 members of the Ce(Pt<sub>1-x</sub>Ni<sub>x</sub>)Sn alloy system has been previously reported [49].



**Figure 10.** The universal scaling of  $B^*(T)$  observed for alloy compositions  $0.5 \le x \le 0.9$  of the  $(\text{Ce}_{1-x}\text{La}_x)\text{Pt}_2\text{Si}_2$  system. Data for isotherms between 5 and 20 K for the concentrations x = 0.5, 0.7 and 0.9 all fall on one curve.

Returning to the CKL model it can readily be shown [27] that

$$\ln\left(\frac{\theta(V_0)}{\theta(V)}\right) = \frac{q(V - V_0)}{V_0 |JN(E_{\rm F})|_0} \tag{4}$$

where  $\theta$  is a quantity that characterizes the Kondo energy scale and which can be taken as either  $T_{\rm K}$  or  $T_{\rm max}$ . In figure 11 results for the  $(Ce_{1-x}La_x)Pt_2Si_2$  system are given of the dependence on  $(V - V_0)/V_0$  of  $\ln[T_{\max}(V_0)/T_{\max}(V)]$  for  $0 \le x \le 0.25$ and of  $\ln[T_{\rm K}(V_0)/T_{\rm K}(V)]$  for 0.5  $\leq x \leq 0.9$ . For  $T_{\rm K}(V_0)$ one may use either  $T_{\rm K}$  = 45 K [9] or  $T_{\rm K}$  = 70 K [13] as given for the  $CePt_2Si_2$  parent compound. However, for consistency of presentation we used the value  $T_{\text{max}}(V_0) = 64 \text{ K}$ observed for the parent compound, also as the value for  $T_{\rm K}(V_0)$ in figure 11. It is observed that the  $T_{\rm K}$  values as derived from MR measurements follow (4) well, whereas the  $T_{\text{max}}$  values near  $x = x_c$  deviate somewhat from (4). The linear LSQ fit through the  $T_{\rm K}$  data points (solid line) passes close to the origin which justifies our choice of  $T_{\rm K}(V_0) = T_{\rm max}(V_0)$  used for the data presentation. The LSQ fit gives  $q/|JN(E_{\rm F})|_0 = 194(2)$ which is an order of magnitude more than the value of 21.7(2)observed for this quantity in a study of  $T_{\text{max}}$  values for the  $Ce(Pt_{1-v}Ni_v)_2Si_2$  system. It is also noted that if the values of  $T_{\rm K}$  from the literature for the parent compound are used, namely 45 K [9] or 70 K [13] the values for  $q/|JN(E_F)|_0$ derived using these for  $T_{\rm K}(V_0)$  are only changed to 171(2) and 199(1) respectively. Thus, irrespective of the particular choice of  $T_{\rm K}(V_0)$  one observes that  $q/|JN(E_{\rm F})|_0$  for La diluted system is much larger than for the ligand modified alloys. One may also consider  $T_{\rm K}$  results for the CePt<sub>2</sub>(Si<sub>1-x</sub>Ge<sub>x</sub>)<sub>2</sub> system [19] derived from MR and this yields  $q/|JN(E_{\rm F})|_0 =$ 50 which is approximately four times smaller than the value obtained for this quantity upon La dilution. The extraordinary large value of  $q/|JN(E_{\rm F})|_0$  obtained for the (Ce<sub>1-x</sub>La<sub>x</sub>)Pt<sub>2</sub>Si<sub>2</sub> system is considered to originate from the influence of the



**Figure 11.** The volume dependence of  $\ln[T_{\max}(V_0)/T_{\max}(V)]$  for the  $(Ce_{1-x}La_x)Pt_2Si_2$  alloys with  $0 \le x \le 0.25$  (squares) and of  $\ln[T_K(V_0)/T_K(V)]$  for  $T_K$  values as derived from MR measurements with  $0.5 \le x \le 0.9$  (circles). The solid line fit to the data is discussed in the text. The dashed line is an extrapolated curve derived from results for the Ce(Pt\_{1-y}Ni\_y)\_2Si\_2 system (see figure 7) and is shown for comparison.

dilution (Kondo hole) effect in addition to the conventionally considered volume effect on the Kondo energy scale.

# 4. Conclusion

Resistivity studies on the  $(Ce_{1-x}La_x)Pt_2Si_2$  alloy system illustrate the transition from the dense Kondo regime ( $0 \le x \le 0.25$ ) to the single-ion Kondo regime. The behaviour of the Fermi-liquid parameter A(x) in the  $\rho(T) = \rho_0 + AT^2$ dependence and the position  $T_{max}(x)$  of the maximum in  $\rho(T)$ in the dense Kondo regime of the alloy system are qualitatively described by considering both volume and Kondo hole effects. Magnetoresistivity studies in the single-ion Kondo regime are interpreted in terms of the Schlottmann formalism. An anomaly in  $\rho(T)$  of LaPt<sub>2</sub>Si<sub>2</sub> and some of the other alloys is reported and further investigation by other techniques is required to resolve its origin.

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#### References

- [1] Schilling J S 1986 Phys. Rev. B 33 1667 and references therein
- [2] Sumiyama A, Oda Y, Nagano H, Ōnuki Y, Shibutani K and Komatsubara T 1986 J. Phys. Soc. Japan 55 1294
- [3] Medina A N, Hayashi M A, Cardoso L P, Gama S and Gandra F G 1998 Phys. Rev. B 57 5900
- [4] Rojas D P, Cardoso L P, Coelho A A and Gandra F G 2001 *Phys. Rev.* B 63 165114
- [5] Petrovic C, Bud'ko S L, Kogan V G and Canfield P C 2002 Phys. Rev. B 66 054534
- [6] Dickey R P, Freeman E J, Zapf V S, Ho P C and Maple M B 2003 Phys. Rev. B 68 144402
- [7] Sollie R and Schlottmann P 1991 J. Appl. Phys. 69 5478
- Sollie R and Schlottmann P 2000 *J. Appl. Phys.* **70** 5803 [8] Dommann A, Hulliger F, Ott H R and Gramlich V 1985
- J. Less-Common Met. 110 331
  [9] Gignoux D, Schmitt D, Zerguine M, Ayache C and Bonjour E 1986 Phys. Lett. A 117 145
- [10] Palstra T T M, Menovsky A A, Nieuwenhuys G J and Mydosh J A 1986 J. Magn. Magn. Mater. 54–57 435
- [11] Menovsky A A, Snel C E, Gortenmulder T J, Tan H J and Palstra T T M 1986 J. Cryst. Growth 74 231
- [12] Hiebl K, Horvath C and Rogl P 1986 J. Less-Common Met. 117 375
- [13] Ayache C, Beille J, Bonjour E, Calemczuk R, Creuzet G, Gignoux D, Najib A, Schmitt D, Voiron J and Zerguine M 1987 J. Magn. Magn. Mater. 63/64 329
- [14] Gignoux D, Schmitt D and Zerguine M 1988 Phys. Rev. B 37 9882
- [15] Bhattacharjee A K, Coqblin B, Raki M, Forro L, Ayache C and Schmitt D 1989 J. Physique 50 2781
- [16] Steeman R A, Dirkmaat A J, Menvovsky A A, Frikkee E, Nieuwenhuys G J and Mydosh J A 1990 Physica B 163 382
- [17] Damas de Réotier P, Yaouanc A, Calemczuk R, Huxley A D, Marcenat C, Bonville P, Lejay P, Gubbens P C M and Mulders A M 1997 Phys. Rev. B 55 2737
- [18] Gignoux D, Schmitt D, Zerguine M and Murani A P 1988
   J. Magn. Magn. Mater. 76/77 401
- Tchoula Tchokonté M B, du Plessis P de V, Strydom A M and Kaczorowski D 2001 J. Magn. Magn. Mater. 226–230 173
   Tchoula Tchokonté M B 2002 PhD Thesis University of the Witwatersrand, Johannesburg, South Africa
- [20] Ragel F C and du Plessis P de V 2004 J. Phys.: Condens. Matter 16 2647
- [21] Tchoula Tchokonté M B, du Plessis P de V and Strydom A M 2005 Solid State Commun. 136 450
- [22] Bouziane K and du Plessis P de V 1999 J. Phys.: Condens. Matter 11 3161
- [23] Lavagna M, Lacroix C and Cyrot M 1982 Phys. Lett. A 90 210 Lavagna M, Lacroix C and Cyrot M 1983 J. Phys. F: Met. Phys. 13 1007

- [24] Rogl P 1984 Handbook on the Physics and Chemistry of Rare Earths vol 7, ed K A Gschneidner Jr and L Eyring (Amsterdam: Elsevier) chapter 51
- [25] Bellarbi B, Benoit A, Jaccard D, Mignot J M and Braun H F 1984 Phys. Rev. B 30 1182
- [26] Thompson J D, McElfresh M W, Willis J O, Fisk Z, Smith J L and Maple M B 1987 Phys. Rev. B 35 48
- [27] Kagayama T, Oomi G, Takahashi H, Mori N, Onuki Y and Komatsubara T 1991 Phys. Rev. B 44 7690
- [28] Umeo K, Kadomatsu H and Takabatake T 1996 *Phys. Rev.* B 54 1194
- [29] Li Z-Z and Qiu Y 1991 Phys. Rev. B 43 12906
- [30] Wermbter S, Sabel K and Czycholl G 1996 *Phys. Rev.* B 53 2528
- [31] Aliev F G, Brandt N B, Moshchalkov V V and Chudinov S M 1984 J. Low Temp. Phys. 57 61
- [32] Sumiyama A, Oda Y, Nagano H, Ōnuki Y, Shibutani K and Komatsubara T 1986 J. Phys. Soc. Japan 55 1294
- [33] Sengupta K and Sampathkumaran E V 2006 J. Phys.: Condens. Matter 18 L115
- [34] Medina A N, Hayashi M A, Cardoso L P, Gama S and Gandra F G 1998 Phys. Rev. B 57 5900
- [35] Yoshimori A and Kasai H 1986 Solid State Commun. 58 259
- [36] Mutou T 2001 Phys. Rev. B 64 245102
- [37] Lawrence J M, Graf T, Hundley M F, Mandrus D, Thompson J D, Lacerda A, Torikachvili M S, Sarrao J L and Fisk Z 1996 Phys. Rev. B 53 12559
- [38] Besnus M J, Kappler J P, Meyer A, Sereni J, Siaud E, Lahiouel R and Pierre J 1985 *Physica* B 130 240
- [39] Balster H, Ihrig H, Kockel A and Methfessel S 1975 Z. Phys. B 21 241
- [40] Gratz E and Zuckermann M J 1982 Handbook on the Physics and Chemistry of Rare Earths vol 5, ed K A Gschneidner Jr and L Eyring (Amsterdam: Elsevier) p 117 and references therein
- [41] Aléonard R and Morin P 1984 J. Magn. Magn. Mater. 42 151
- [42] Becker B, Patil N G, Ramakrishnan S, Menovsky A A, Nieuwenhuys G J, Mydosh J A, Kohgi M and Iwasa K 1999 *Phys. Rev.* B **59** 7266
- [43] Ramakrishnan S and Mydosh J A 2007 J. Magn. Magn. Mater. 310 207
- [44] Shelton R N, Hausermann-Berg L S, Klavius P, Yang H D, Anderson M S and Swenson C A 1986 Phys. Rev. B 34 4590
- [45] Andrei N 1982 Phys. Lett. A 87 299
- [46] Schlottmann P 1983 Z. Phys. B **51** 223
- [47] Evans S M M, Bhattacharjee A K and Coqblin B 1991 *Physica* B 171 293
- [48] Batlogg B, Bishop D J, Bucher E, Golding B Jr, Ramirez A P, Fisk Z, Smith J L and Ott H R 1987 J. Magn. Magn. Mater. 63/64 441
- [49] Adroja D T, Rainford B D, Neville A J and Jansen A G M 1996 Physica B 223/224 275