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Thermal and magnetic properties of $\text{Ce}_{1-x}\text{La}_x\text{RhSn}$ near the critical concentration

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Abstract

The main aim of this work is to investigate the electronic structure, specific heat and magnetic properties of $\text{Ce}_{1-x}\text{La}_x\text{RhSn}$ compounds near the critical concentration $x_0 \cong 0.5$. Near x_0 the magnetic properties for $x > x_0$ correspond to spin fluctuations, while for the Ce-rich samples ($x < x_0$) the micromagnetic-like behaviour indicates the formation of magnetic clusters. Both the magnetic susceptibility and electrical resistivity data obtained for the $\text{Ce}_{0.55}\text{La}_{0.45}\text{RhSn}$, $\text{Ce}_{0.5}\text{La}_{0.5}\text{RhSn}$ and $\text{Ce}_{0.45}\text{La}_{0.55}\text{RhSn}$ compounds obey a non-Fermi liquid temperature dependences due to an atomic disorder and/or the presence of spin fluctuations.

1. Introduction

The low temperature properties of f-electron systems have been a wide field of research for many years. In recent years a new class of strongly correlated f-electron materials whose low-temperature physical properties display non-Fermi liquid (NFL) behaviour has attracted a great deal of attention [1–3]. What characterizes a system as a non-Fermi liquid is not unambiguous. At least a nonconstant specific heat C divided by temperature, $C(T)/T$ (e.g., $C/T \propto -\ln T$ or $C/T \propto T^{-n}$) at low temperature, and deviations from the Fermi liquid (FL) behaviour for magnetic susceptibility χ and electrical resistivity ρ have to be seen (for an overview, see [3, 4]). In particular, the electrical resistivity changes as a function of temperature ($\Delta\rho \propto T^\varepsilon$) with exponents $1 \leq \varepsilon < 2$, and the magnetic susceptibility varies as T^{-n} or $\chi \propto -\ln T$. There are a number of NFL materials in which ρ , C/T , and χ do not all conform to the expected NFL behaviour, i.e. logarithmic or power-law divergences in temperature that necessarily extend over several decades in temperature. For example, in $\text{Y}_{1-x}\text{U}_x\text{Pd}_3$, the first f-electron system in which NFL behaviour was identified [5], and YbRh_2Si_2 [6], $C(T)/T$ exhibits an upward departure from $-\ln T$ behaviour at low temperatures, while $\rho(T)$ remains linear in T throughout this region to very low temperatures.

We have recently shown [7, 8] that CeRhSn exhibits NFL temperature dependencies in its low-temperature physical properties. However, magnetization M and specific heat C , reveal the presence of a very weak magnetic phase transition at 6.2 K. This phase transition observed in the $C(T)$ data is strongly dependent on the annealing of the polycrystalline sample, while magnetic susceptibility data collected in low magnetic fields reveal spin-glass behaviour. For CeRhSn samples χ curves exhibit also at the high-temperature range a knee-shaped phase transition, which is very similar to the case in PdFeMn [9] or LaFe₇Al₆ [10], indicative of a transition to an inhomogeneous ferromagnet ('mictomagnet') [11]. The Curie temperature T_C was found to be ~ 220 K.

Systematic studies of the polycrystalline off-stoichiometry Ce_{*x*}RhSn [12] and chemically substituted Ce_{1-*x*}La_{*x*}RhSn [11] samples suggest that a magnetic ground state could be responsible for the existence of a Griffiths phase [13] in the vicinity of a quantum critical point (QCP). The presence of sufficiently strong chemical phase disorder in CeRhSn can lead to small magnetically ordered regions, where dynamics may dominate thermodynamic properties of the system in a finite region around the QCP due to tunnelling between different configurations. The determined value of the power-law exponent n of about 0.5 obtained through $C(T)$ and $\chi(T)$ measurements agrees, to within the experimental resolution, with the prediction pertaining to the Griffiths model [14].

There is also another NFL model that may be partially applicable to the CeRhSn compound: spin-fluctuation theory for an antiferromagnetic QCP in 3D [15, 16]. The spin-fluctuating model may provide an understanding of the NFL characteristics of $C(T)/T$ and $\rho(T)$ for CeRhSn with some deviations observed at very low temperatures, however, the exponent $n = 0.5$ of $\chi(T)$ does not satisfy the predicted value of 1.5.

In order to further characterize the NFL behaviour close to the QCP, we previously investigated the electronic structure and magnetic properties of the new ternary Ce_{1-*x*}La_{*x*}RhSn system [11]. The T - x phase diagram has spin fluctuations for $x > 0.5$, while with decreasing La concentration x , the spin-glass-type behaviour dominates, which would lead to NFL behaviour in Ce_{1-*x*}La_{*x*}RhSn. We also suggested that the critical composition $x_0 = 0.5$ may correspond to some type of quantum critical point. The measurements reported here were undertaken to obtain heat capacity and magnetic susceptibility data for Ce_{0.55}La_{0.45}RhSn, Ce_{0.5}La_{0.5}RhSn and Ce_{0.45}La_{0.55}RhSn to study the thermodynamic properties of the Ce_{1-*x*}La_{*x*}RhSn system of alloys near the critical concentration x_0 .

2. Experimental details

Polycrystalline samples of Ce_{0.55}La_{0.45}RhSn, Ce_{0.5}La_{0.5}RhSn and Ce_{0.45}La_{0.55}RhSn have been prepared by arc melting the constituent elements (Ce 99.99%, La 99.9%, Rh 99.9%, Sn 99.999% in purity in units of at.%) on a water cooled copper hearth in a high purity argon atmosphere with a Zr getter. Each sample was remelted several times to promote homogeneity and annealed at 800 °C for two weeks, and then quenched in water. The samples were carefully examined by x-ray diffraction analysis and found to consist of a single-phase. Analysis of the x-ray diffraction patterns with the Powder-Cell 2.1 program revealed that the samples crystallize in a hexagonal unit cell of the Fe₂P-type (space group $P\bar{6}2m$).

The dc magnetization was measured using a commercial SQUID magnetometer from 1.8 to 400 K in magnetic fields up to of 5 T.

Specific heat measurements have been performed in a fully adiabatic calorimeter in magnetic fields up to of 3 T between 2.7 and 70 K.

The XPS spectra were obtained with monochromatized Al $K\alpha$ radiation at room temperature using a PHI 5700 ESCA spectrometer. The spectra were measured immediately

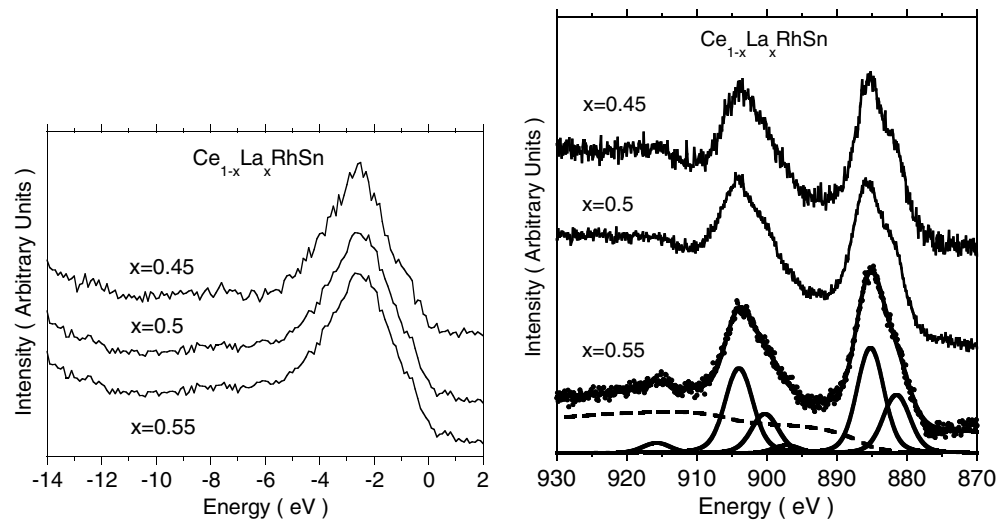


Figure 1. The XPS valence-band spectra for $Ce_{0.55}La_{0.45}RhSn$, $Ce_{0.5}La_{0.5}RhSn$ and $Ce_{0.45}La_{0.55}RhSn$.

Figure 2. Ce 3d XPS spectra for $Ce_{1-x}La_xRhSn$. For $Ce_{0.45}La_{0.55}RhSn$ the separation of the overlapping peaks attributed to $4d^9f^2$, $4d^9f^1$ and $4d^9f^0$ final states and corrected for background (dotted curve) is presented. The Sn 3s peak at 885 eV provides $\sim 15\%$ of the total $4d^9_{5/2}f^1$ peak intensity.

after cleaving the sample in a vacuum of 10^{-10} Torr. A very small amount of oxygen could barely be detected in the noise of the XPS spectra. Calibration of the spectra was performed according to [17]. Binding energies were referenced to the Fermi level ($\epsilon_F = 0$).

3. Results and discussion

3.1. Electronic structure of $Ce_{1-x}La_xRhSn$

In figure 1 are shown the XPS valence band (VB) spectra of $Ce_{0.55}La_{0.45}RhSn$, $Ce_{0.5}La_{0.5}RhSn$ and $Ce_{0.45}La_{0.55}RhSn$. These spectra are similar. The spectra reveal a valence band that has a major peak mainly due to the d-states of Rh located near the Fermi level. The second peak centred at about 7 eV is due to the Sn s states³.

Figure 2 shows the Ce 3d XPS spectra obtained for $Ce_{0.55}La_{0.45}RhSn$, $Ce_{0.5}La_{0.5}RhSn$ and $Ce_{0.45}La_{0.55}RhSn$. Three final-state contributions f^0 , f^1 and f^2 are observed, which exhibit a spin-orbit splitting $\Delta_{SO} = 18.7$ eV. The appearance of an f^0 component in the 3d XPS spectra is clear evidence of the mixed valence behaviour of Ce. Based on the Gunnarsson-Schönhammer (GS) theoretical model [18, 19], the intensity ratio $I(f^0)/(I(f^0) + I(f^1) + I(f^2))$, which should be directly related to the f-occupation probability in the final states, indicates an f-occupation number n_f^{XPS} , which is about 0.9 (table 1). The separation of the overlapping peaks in the Ce 3d XPS spectra was made on the basis of the Doniach-Šunjić theory [20].

The f^2 components located at the low-binding energy side of the f^1 components are attributed within the GS model to the hybridization between the f states and the conduction

³ In [11] we reported the results of the study of the electronic structure of $Ce_{1-x}La_xRhSn$ by the self-consistent tight binding linearized muffin-tin orbital (LMTO) method for different local environments of Ce atoms. The LMTO calculations yield a magnetic moment per formula unit only for the disordered alloys, and have shown that there is considerable hybridization between the Ce 4f and Rh 4d bands. The Ce and La DOS show no localized f states.

Table 1. 3d XPS peak binding energies for $\text{Ce}_{0.55}\text{La}_{0.45}\text{RhSn}$, $\text{Ce}_{0.5}\text{La}_{0.5}\text{RhSn}$ and $\text{Ce}_{0.45}\text{La}_{0.55}\text{RhSn}$ (all values in eV with respect to E_F), and the $r = f^2/(f^1 + f^2)$, and $n_f^{\text{XPS}} = 1 - f^0/(f^0 + f^1 + f^2)$ peak intensities ratio.

Compound	3d _{5/2}			3d _{3/2}			r	n_f^{XPS}
	f^2	f^1	f^0	f^2	f^1	f^0		
$\text{Ce}_{0.55}\text{La}_{0.45}\text{RhSn}$	881.6	885.5	897.6	900.9	904.4	916.2	0.21	0.9
$\text{Ce}_{0.5}\text{La}_{0.5}\text{RhSn}$	882.0	885.9	897.5	900.2	904.2	916.0	0.20	0.9
$\text{Ce}_{0.45}\text{La}_{0.55}\text{RhSn}$	881.6	885.3	898.0	900.9	904.2	915.8	0.30	0.9

band. The hybridization energy $\Delta = \pi V_{\text{sf}}^2 \rho_{\text{max}}$ describes the hybridization part of the Anderson impurity Hamiltonian [21], where ρ_{max} is the maximum in the DOS and V_{sf} is the hybridization matrix element. Since the intensity ratio $r = I(f^2)/(I(f^1) + I(f^2))$ has been calculated as a function of Δ in [19], it is possible to estimate the hybridization energy Δ , when the peaks of the Ce 3d XPS spectra that overlap in figure 2 are separated [20]. The intensity ratios r give a crude estimate of the hybridization energy Δ of ~ 120 meV. The GS parameters n_f and Δ , obtained from the XPS measurements, do not include the surface to bulk emission ratio I_s/I_b [22–24]. Since the intensity ratio I_s/I_b depends on the electron escape depth λ , it varies strongly with the electron kinetic energy. I_s/I_b is estimated to be ~ 0.15 at Ce 3d XPS kinetic energies [24]. Therefore, the Ce 3d XPS spectra allows an estimate of the occupation number n_f , and energy Δ within an accuracy limited by the I_s/I_b ratio, i.e. 15%.

3.2. Magnetic properties

The magnetic susceptibility plotted as χ versus T between 1.8 and 400 K in the magnetic field 0.5 T is shown in figure 3. The magnetic susceptibility deviates markedly from a Curie–Weiss law and depends on the concentration of La. For both samples $\text{Ce}_{0.55}\text{La}_{0.45}\text{RhSn}$ and $\text{Ce}_{0.5}\text{La}_{0.5}\text{RhSn}$ the susceptibility is hysteretic under zero-field (ZFC) and field (FC) cooling and also exhibits at the high-temperature range a knee-shaped phase transition, which is very similar to the case in PdFeMn [9], or LaFe₇Al₆ [10], indicative of a transition to an inhomogeneous ferromagnet (‘mictomagnet’). The high-temperature inhomogeneous ferromagnet may be understood as a consequence of magnetic clusters which are formed in the disordered paramagnetic (Ce, La)–Rh–Sn alloy. In [11] we discussed the possible origin of magnetism in the clusters. The magnetism can arise from Rh clusters [25–27], or Ce 4f electrons strongly interacting with the Rh and Sn d electrons (discussed later). The Curie temperature T_C is taken to be the temperature at which $d\chi/dT$ has its maximum value, T_C is defined to be ~ 230 K.

In contrast, the magnetic susceptibility of $\text{Ce}_{0.45}\text{La}_{0.55}\text{RhSn}$ is not hysteretic and without any maximum, and suggests the spin-fluctuation contribution.

The origin of the magnetic clusters in $\text{Ce}_{1-x}\text{La}_x\text{RhSn}$ compounds where $x \leq 0.5$ is of great interest because of the high value of Curie temperature T_C . Rhodium clusters of between 9 and 11 atoms have been found to be superparamagnetic at 93 K with magnetic moment of $0.8 \mu_B$ per atom [25, 26]. Structural properties of the Fe₂P-type unit cell do not exclude small size rhodium clusters formed in the disordered (Ce, La)–Rh–Sn alloy. However, the Curie temperature of this ferromagnetic cluster should be smaller by a factor of ~ 2.5 than the measured value. If the high- T_C magnetism is attributed to Rh clusters, then the similar cluster-effect should also be observed for the La-rich $\text{Ce}_{1-x}\text{La}_x\text{RhSn}$ samples which, however, is not the case.

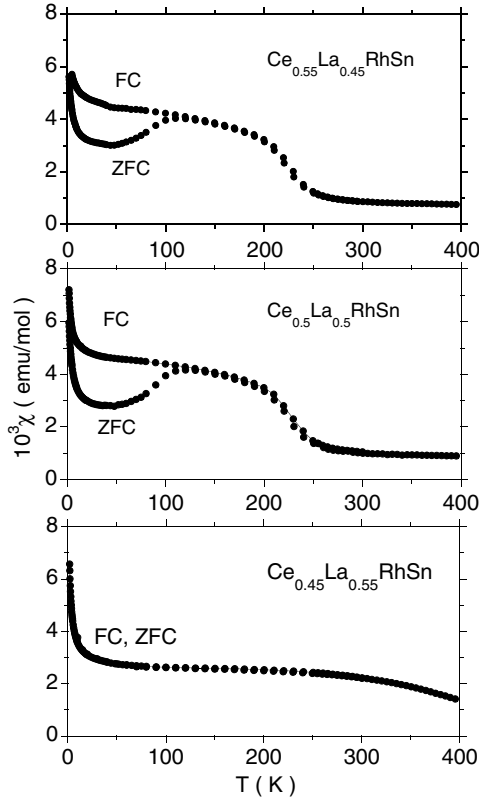


Figure 3. FC and ZFC susceptibility $\chi \equiv M/H$ versus T in a magnetic field of 0.5 T for $\text{Ce}_{0.55}\text{La}_{0.45}\text{RhSn}$, $\text{Ce}_{0.5}\text{La}_{0.5}\text{RhSn}$ and $\text{Ce}_{0.45}\text{La}_{0.55}\text{RhSn}$.

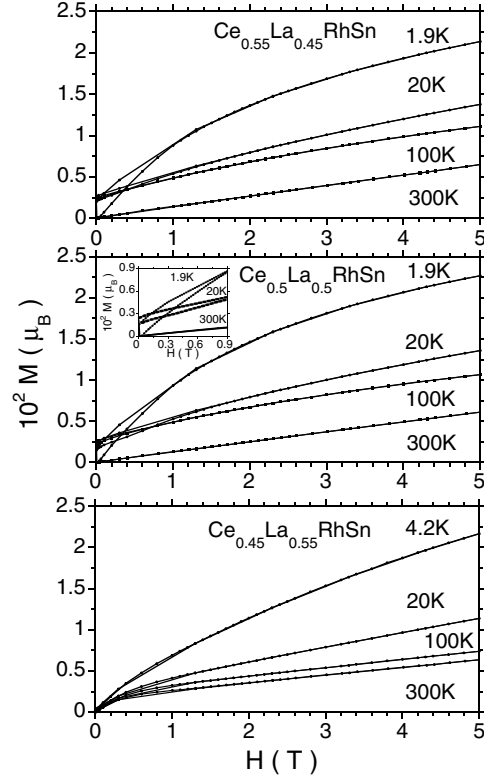


Figure 4. Magnetization M versus H at different temperatures for $\text{Ce}_{0.55}\text{La}_{0.45}\text{RhSn}$, $\text{Ce}_{0.5}\text{La}_{0.5}\text{RhSn}$ and $\text{Ce}_{0.45}\text{La}_{0.55}\text{RhSn}$. Shown in the inset is an expanded plot of M versus H .

Another explanation would be a large Kondo coupling parameter $J_{sf} \propto -V_{sf}^2/E_{4f}$ in a simple Kondo-lattice picture. As a Ce ion approaches the mixed-valence state, the position of the localized 4f level $E_{4f} \rightarrow 0$ and J_{sf} can grow appreciably in magnitude. However, the magnetic ordering temperature $T_{\text{RKKY}} \propto J_{sf}^2$ would be expected to be anomalously large in the system, where the excitation energy between the two configurations $E_{\text{ex}} = |E_{4f}^1 - E_{4f}^0|$ is larger than Δ , where E_{4f}^1 and E_{4f}^0 are energies of the configurations $4f^1$ and $4f^0$, respectively. Otherwise, the criterion for the formation of a local moment in the Ce-intermediate valence systems which undergo interconfigurational fluctuations (ICF) between the configurations $4f^1$ and $4f^0$ is roughly given by the relation $\Delta/E_{\text{ex}} \lesssim 1$ [28] (this would mean that the mixed valence state requires a reasonably large value for the V_{sf} term, relative to E_{4f}). CeRhSn and $\text{Ce}_{1-x}\text{La}_x\text{RhSn}$, which have $\Delta \sim 120$ meV and $E_{\text{ex}} \sim T_{\text{K}} \cong 30$ meV, do not follow this criterium. In reality, however, $\text{Ce}_{1-x}\text{La}_x\text{RhSn}$ are disordered. The LMTO calculations yield DOS at ϵ_{F} for the disordered system two or three times larger than that obtained from the numerical calculations for the ordered compound (e.g., for Ce–Rh–Sn) [11]. Thus for a disordered alloy the s–f hybridization strength V_{sf} could be reduced two-fold. The crude analysis predicts $V_{sf} < 50$ meV which is comparable to the value of E_{ex} . In addition, the interatomic hybridization, mainly between Ce 4f and Rh 4d states, could also enhance magnetic interactions between Ce ions.

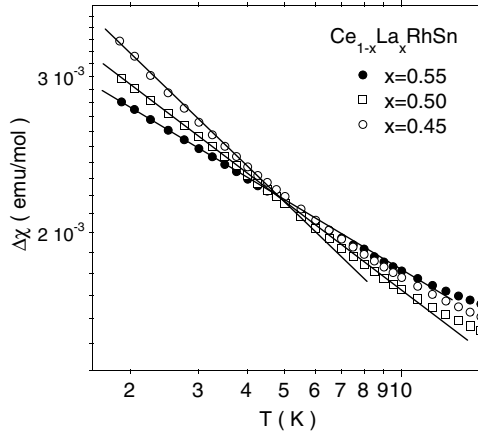


Figure 5. Temperature variation of the ZFC magnetic susceptibility $\Delta\chi = \chi(\text{Ce}_{1-x}\text{La}_x\text{RhSn}) - \chi(\text{LaRhSn})$ on a double-logarithmic plot for $\text{Ce}_{1-x}\text{La}_x\text{RhSn}$ compounds.

Probably both mechanisms, the on-site exchange J_{sf} and Rh itinerant magnetism, could be responsible for this anomalous magnetic properties at ~ 220 K.

The susceptibility behaviour for $\text{Ce}_{0.5}\text{La}_{0.5}\text{RhSn}$ presented in figure 3 differs substantially from what has been published in [11] for the same composition, especially with respect to the present observation of a well-developed knee in $\chi(T)$ near 220 K. This difference in $\chi(T)$ could be attributed to the strong field dependence of the magnetic susceptibility χ . A knee in $\chi(T)$ near 220 K was observed in the magnetic field of 5 mT, while it was only slightly visible in the field of 1 T [11]. In this presentation the $\chi(T)$ data are obtained in a magnetic field of $B = 0.5$ T compared to the $B = 1$ T of the $\chi(T)$ data in [11].

Shown in figure 4 are magnetic curves measured up to 5 T at various temperatures between 1.9 and 300 K. The magnetization M versus H curves exhibit very similar features in the Ce-rich $\text{Ce}_{1-x}\text{La}_x\text{RhSn}$ compounds with $x = 0.45$ and 0.5 . The $M(H)$ data clearly exhibit hysteresis at $T = 1.9$ K and a linear scaling of M with H at 300 K, while the M versus H curves for $\text{Ce}_{0.45}\text{La}_{0.55}\text{RhSn}$ are different and indicative of spin-fluctuations. The square of the spontaneous magnetization M_0 obtained in the zero-field limit for this compound varies as $T^{4/3}$, indicating that $\text{Ce}_{0.45}\text{La}_{0.55}\text{RhSn}$ is close to a ferromagnetic instability [29]. Shown in figure 5 are ZFC $\Delta\chi = \chi(\text{Ce}_{1-x}\text{La}_x\text{RhSn}) - \chi(\text{LaRhSn})$ versus T data on a double logarithmic plot which reveal that $\Delta\chi$ varies as T^{-n} between 1.8 and 11 K for $\text{Ce}_{0.55}\text{La}_{0.45}\text{RhSn}$ and $\text{Ce}_{0.5}\text{La}_{0.5}\text{RhSn}$ samples, and between 1.8 and ~ 3 K for $\text{Ce}_{0.45}\text{La}_{0.55}\text{RhSn}$, while the FC $\Delta\chi$ varies as T^{-n} between 1.8 and ~ 4 K for each sample. The value of n obtained for the low- T ZFC (and FC) susceptibility is, respectively, 0.34 (0.25) for $\text{Ce}_{0.55}\text{La}_{0.45}\text{RhSn}$, 0.33 (0.25) for $\text{Ce}_{0.5}\text{La}_{0.5}\text{RhSn}$, and 0.40 (0.43) for $\text{Ce}_{0.45}\text{La}_{0.55}\text{RhSn}$. The least-square determined n -exponent value of between 0.40 and 0.45 for $\text{Ce}_{0.5}\text{La}_{0.5}\text{RhSn}$ depending on field cooling or zero-field cooling data, differs somewhat from the value of $n = 0.34$ obtained in [11]. We attribute this difference to the different measuring field of χ .

3.3. Specific heat

In order to better understand the low-temperature state in the complex of $\text{Ce}_{1-x}\text{La}_x\text{RhSn}$ alloys we have performed specific-heat measurements in a magnetic field. The lowest temperature of

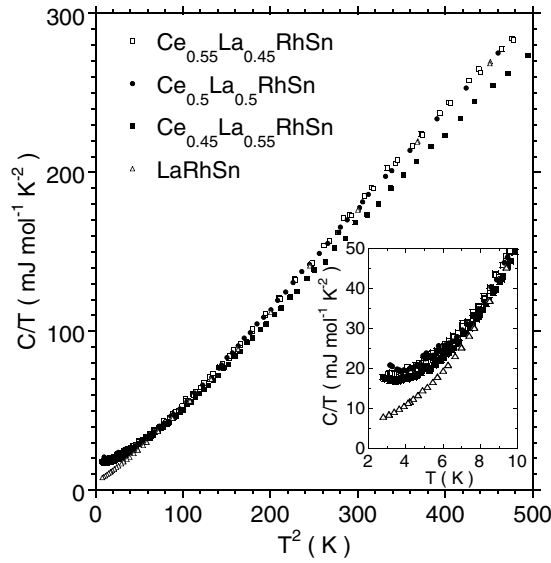


Figure 6. Specific heat C divided by temperature T , C/T versus T^2 for the $Ce_{1-x}La_xRhSn$ compounds. In the inset is C/T versus T between 2.7 and 10 K.

Table 2. The coefficients deduced from a least-squares fit to the zero-field specific heat of $Ce_{0.55}La_{0.45}RhSn$, $Ce_{0.5}La_{0.5}RhSn$ and $Ce_{0.45}La_{0.55}RhSn$ in the temperature range 2.7–17 K, employing the relation $C/T = \gamma^* + \beta_{ph}T^2 + \delta T^2 \ln(T/T^*)$.

$Ce_{1-x}La_xRhSn$	γ^* (mJ K ⁻² mol ⁻¹)	β_{ph} (mJ K ⁻⁴ mol ⁻¹)	δ (mJ K ⁻⁴ mol ⁻¹)	T^* (K)
$x = 0.55$	22.4	0.73	0.39	30.1
$x = 0.5$	21	0.74	0.41	28
$x = 0.45$	19.1	0.73	0.39	31
$x = 1$	6.2	0.68	0.18	32.9

this investigation was 2.7 K and special emphasis was on the explication of the specific-heat behaviour in the system of $Ce_{1-x}La_xRhSn$ alloys near the critical concentration $x_0 \cong 0.5$.

There is remarkable similarity between the zero-field specific-heat data for the $Ce_{1-x}La_xRhSn$ compounds near the critical concentration x_0 ; the zero-field specific-heat is characteristic of the spin-fluctuation compounds. In figure 6 we plot the specific-heat data as C/T versus T^2 , which show that a description with a linear and T^3 term is far from appropriate. The specific heat data obey the relationship $C/T = \gamma^* + \beta_{ph}T^2 + \delta T^2 \ln(T/T^*)$, representing a spin-fluctuation contribution $T^3 \ln(T/T^*)$ to the specific heat [30] in the temperature range 2.7–17 K. Here γ^* is the enhanced linear coefficient, β_{ph} is the usual phonon coefficient ($T \ll \theta_D$), and T^* is a spin-fluctuation cut-off temperature. The best-fit parameters are listed in table 2.

LaRhSn follows $C(T)/T$ versus T^2 between 2 and 6 K, while it does not follow a T^2 dependence over an extended range in temperature. These data have also been fitted against a spin-fluctuating model, however, the least-square determined parameters in table 2, γ , β_{ph} and δ are distinctly different from that obtained for the $Ce_{1-x}La_xRhSn$ samples. The anomalous behaviour found in $C(T)$ of LaRhSn could be related to the anomalous temperature dependence that was reported in [11] either for $\chi(T)$ or $\rho(T)$ of LaRhSn as a result of an s–d scattering

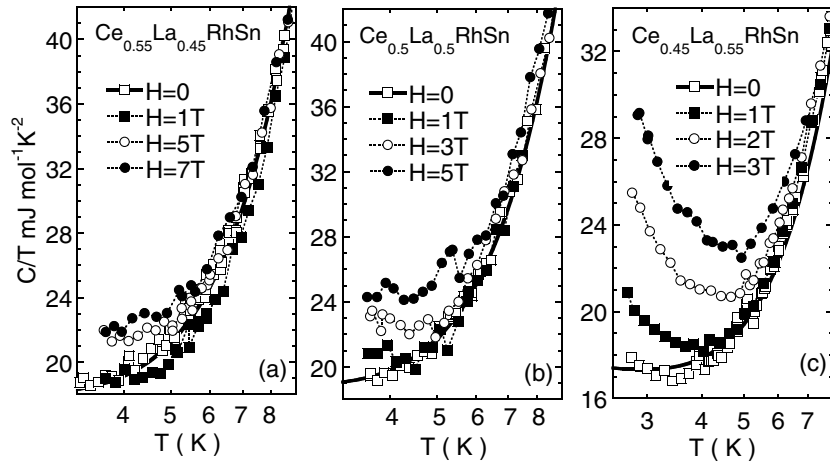


Figure 7. C/T versus $\ln T$ under an external magnetic field for: $\text{Ce}_{0.55}\text{La}_{0.45}\text{RhSn}$ (a), $\text{Ce}_{0.5}\text{La}_{0.5}\text{RhSn}$ (b) and $\text{Ce}_{0.45}\text{La}_{0.55}\text{RhSn}$ (c). The relation $C/T = \gamma^* + \beta_{\text{ph}}T^2 + \delta T^2 \ln(T/T^*)$ has been fitted to the $C(T)$ data at $B = 0$ (curve). γ , β_{ph} and δ obtained from the best fit to $C(T)/T$ are listed in table 2.

mechanism considered by Mott for d-electron type metals [31]. We also suggest that the Rh-clusters are formed in LaRhSn , and, furthermore, that these clusters could give rise to spin fluctuations.

However, a different kind of anomaly is exhibited by specific heat under an external magnetic field, corresponding to the lower ($x < 0.5$) or higher ($x \geq 0.5$) concentration range. It is evident (figure 7) that the anomalous contribution to the heat capacity C/T is dependent on magnetic field for the $\text{Ce}_{0.45}\text{La}_{0.55}\text{RhSn}$ sample. Shown in figure 7 is the specific heat C plotted as C/T versus $\ln T$, which displays a logarithmic dependence in the temperature range $T < 3.5$ K at magnetic fields 2 and 3 T. In contrast, for the Ce-rich samples ($x = 0.45$ and 0.5) the field-effect on the specific heat is small, and hardly exceeds the experimental uncertainty. The field-dependent upturn observed in the specific heat coefficient C/T for $\text{Ce}_{0.45}\text{La}_{0.55}\text{RhSn}$ is not typical for the heavy-fermion materials. The electronic specific heat can also be enhanced by the effect of exchange-enhanced spin fluctuations [32, 33] and it is generally significantly suppressed by a strong magnetic field. However, some itinerant magnetic materials are known for which the influence of magnetic field is quite varied (e.g., for Sc_3In C/T is enhanced for $H < 5$ T, while for $H > 5$ T the spin fluctuations are quenched) [34]. Specific heat results (in figure 7) and magnetic properties as a function of field and concentration x have shown that the concentration $x_0 \cong 0.5$ separates the spin-fluctuation effect which dominates the properties of $\text{Ce}_{0.45}\text{La}_{0.55}\text{RhSn}$ sample and the magnetic behaviours dominated by the magnetic clusters in the Ce-rich samples of $\text{Ce}_{1-x}\text{La}_x\text{RhSn}$ alloys.

The specific heat data for three $\text{Ce}_{1-x}\text{La}_x\text{RhSn}$ samples are displayed as $\Delta C/T$ versus $\ln T$ in figure 8, where $\Delta C/T = C/T(\text{Ce}_{1-x}\text{La}_x\text{RhSn}) - C/T(\text{LaRhSn})$. By subtracting the heat capacity of pure LaRhSn we observe a logarithmic increase of $\Delta C/T$ for temperatures $2.7 < T < 10$ K for $\text{Ce}_{1-x}\text{La}_x\text{RhSn}$ around the critical concentration x_0 . For three compounds (figure 8) we find a logarithmic increase of $\Delta C/T$, which usually is taken as experimental evidence for a NFL behaviour. Note however, that the entropy related to the NFL behaviour in specific heat amounts roughly to $0.1R \ln 2$ at $T = 10$ K after extrapolation of $\Delta C/T$ down to $T = 0$, while either for the QCP case [35] or for the Griffiths phase [36] ΔS amounts roughly to $0.5R \ln 2$ at $T = T_K$. Within the phenomenological systematics proposed by Sereni *et al*

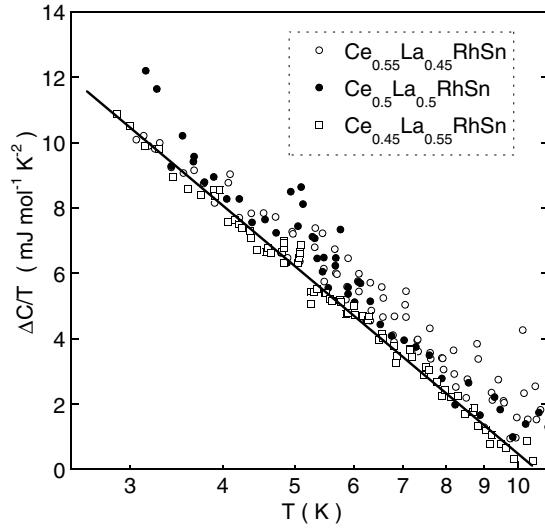


Figure 8. $\Delta C/T$ versus $\ln T$ for: $\text{Ce}_{0.55}\text{La}_{0.45}\text{RhSn}$, $\text{Ce}_{0.5}\text{La}_{0.5}\text{RhSn}$ and $\text{Ce}_{0.45}\text{La}_{0.55}\text{RhSn}$. A line fits the logarithmic temperature dependence of $\Delta C/T$ for the $\text{Ce}_{0.45}\text{La}_{0.55}\text{RhSn}$ sample.

in [35] we try to explain the small value of ΔS for samples in the $\text{Ce}_{1-x}\text{La}_x\text{RhSn}$ ($x = 0.55, 0.5, 0.45$) system for which Kondo temperature T_K is ~ 60 K [11]. The scaling procedure [35], $C/t \sim -\log t$, where $t = T/T_0$ and $T_0 \sim T_K$, suggests a very small ΔS at $T \ll T_K$, e.g., at $t = T/T_K = 10 \text{ K}/60 \text{ K}$ $\Delta S \cong 0.25R \ln 2$. For the $\text{Ce}_{1-x}\text{La}_x\text{RhSn}$ samples investigated here the entropy measured at 10 K is, however, twice as small as the ΔS estimated using this scaling procedure. This suggests incorrect background subtraction attributed to the anomalous magnetic behaviour of LaRhSn that is discussed earlier in the paper.

3.4. Concluding remarks

For CeRhSn the ZFC susceptibility χ exhibits at the high-temperature range a knee-shaped phase transition, indicative of a transition to an inhomogeneous ferromagnet ('mictomagnet') [11]. The magnetic susceptibility of CeRhSn is also hysteretic under ZFC and FC and suggests a spin-glass like magnetic order below $T_f \cong 10$ K. It was found [11] that the substitution of La for Ce suppresses both the mictomagnetic and spin-glass ordering, and emphasizes spin-fluctuations.

The $T-x$ phase diagram obtained for $\text{Ce}_{1-x}\text{La}_x\text{RhSn}$ has a critical concentration $0.5 < x_0 < 0.55$; in the concentration regime $x < x_0$ the magnetic properties are dominated by the existence of spin fluctuations, while for a Ce-rich side spin clusters can form analogous to Griffiths phases [13, 14]. As shown in figure 5, the χ data obey a standard NFL temperature dependence ($\Delta\chi \propto T^{-n}$, where $n \cong 0.4$) and the electrical resistivity $\rho \propto T$ (in [11]). The temperature dependence of the specific-heat coefficient $\gamma \equiv \Delta C/T$ ($\Delta C/T \sim -\ln T$) around the critical concentration x_0 demonstrate clear NFL behaviour. The deviation from the Fermi liquid behaviour can be attributed to disorder and (or) the presence of spin fluctuations in the $\text{Ce}_{1-x}\text{La}_x\text{RhSn}$ compounds.

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