Chemical pressure, dilution, and disorder in the heavy fermion compounds $Ce_{3-x}La_xPd_{20}Si_6$

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Introduction

The investigation of quantum criticality has taken a prominent role in condensed matter physics in the past decade. Heavy fermion compounds have emerged as model systems because their intrinsically low energy scales allow us to tune them through a quantum critical point (QCP) with readily accessible control parameters. The best studied heavy fermion compounds all display strong spacial anisotropy, and the relevant spacial dimensionality appears to be close to two. Therefore, investigations on three-dimensional compounds are much needed.

$Ce_3Pd_{20}Si_6$ is one of the rare examples of a cubic system where a QCP can be accessed with a modest magnetic field [1, 2]. A peculiarity of this material is that there are two different Ce sites which might be responsible for the two different low-temperature phase transitions [1]. Ce atoms at the $4a$ ($8c$) site have a Γ_7 (Γ_8) ground state [3] and may therefore give rise to the antiferromagnetic (antiferroquadrupolar) order that is suggested to set in below T_L (T_U) [1, 2, 4]. However, microscopic evidence is still lacking.

This has motivated us to try and disentangle the roles of the two different Ce sites by partial substitutions of Ce with non-magnetic elements. Our investigation of substituted polycrystalline $Ce_{3-x}RE_xPd_{20}Si_6$ (RE = La, Lu; x = 1, 2) revealed that these substitutions are at least partially site selective. In addition, this investigation showed that all La substituted samples with $x \leq 1$ should develop Kondo coherence at low temperatures. In order to study the role of substitutions in the quantum critical behaviour we therefore focus on compounds with $x = \frac{1}{3}$ and $\frac{2}{3}$.

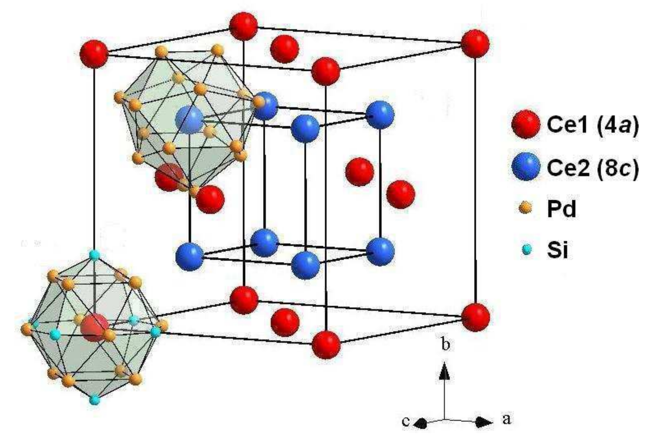

FIGURE 1: Crystal structure of $Ce_3Pd_{20}Si_6$.

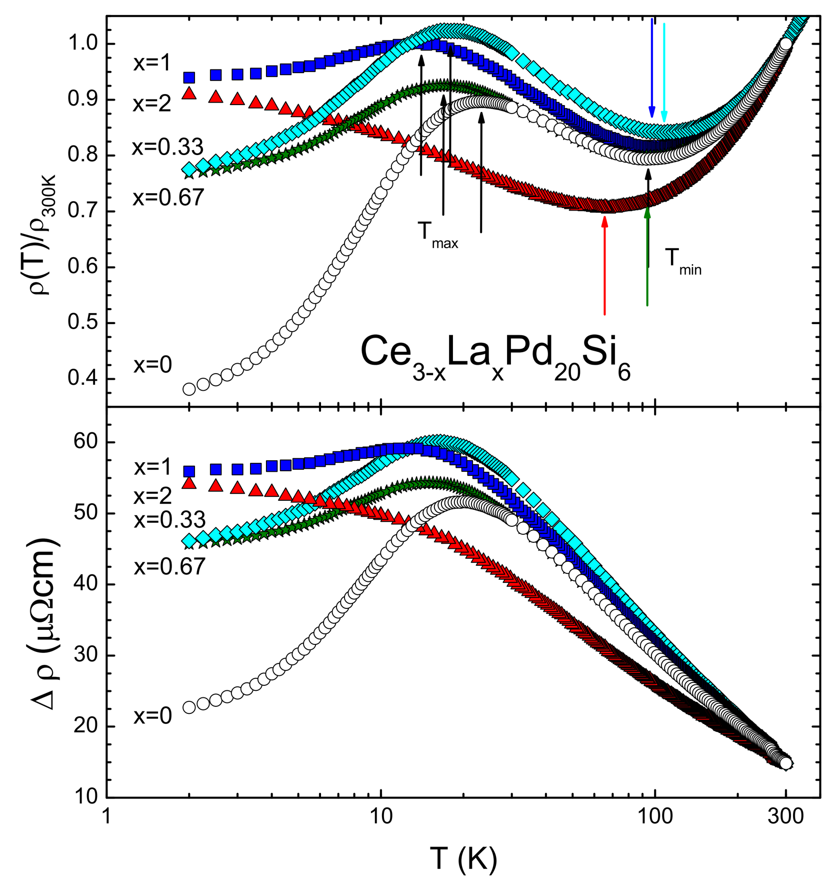

FIGURE 2: Electrical resistivity versus temperature (2 - 300K) of $Ce_{3-x}La_xPd_{20}Si_6$ (x = 0, $\frac{1}{3}$, $\frac{2}{3}$, 1 and 2), normalized to the respective value at 300 K, $\rho(T)/\rho_{300K}$ (top) and "magnetic contribution" $\Delta\rho$ (bottom). The arrows mark the positions of T_{min} and T_{max}.

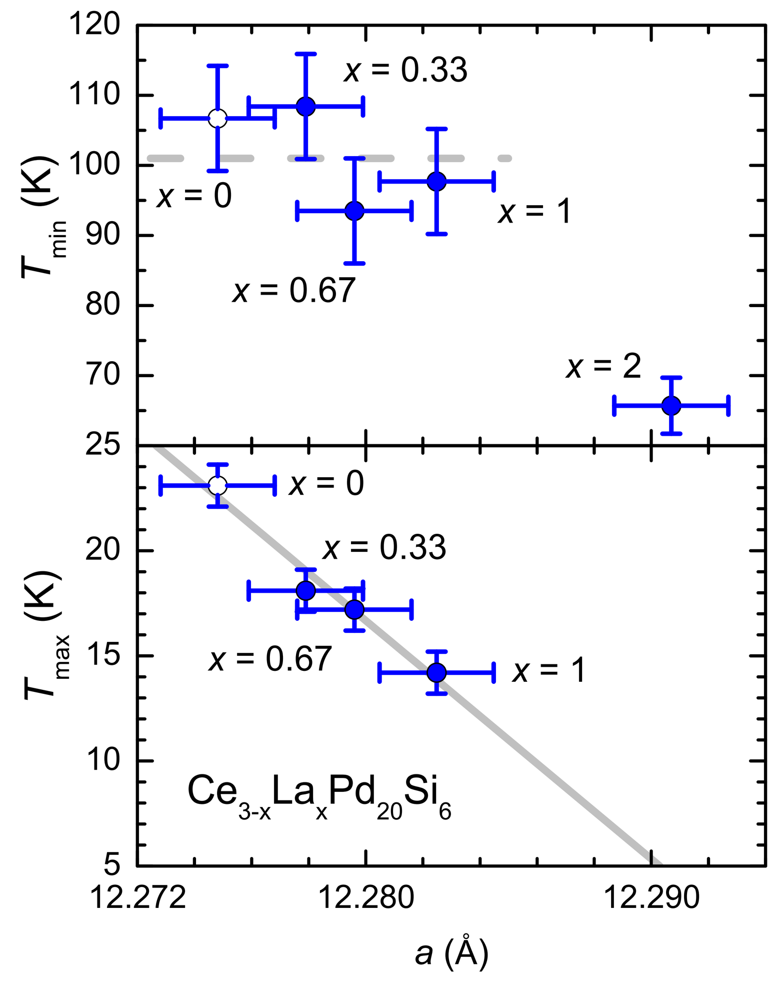

FIGURE 3: T_{min} (top) and T_{max} (bottom) denoted by arrows in the upper panel of Fig. 2 versus the lattice parameter a. The dashed line in the upper panel is a constant fit to the data at $x \leq 1$. The line in the lower panel is a linear fit to the data.

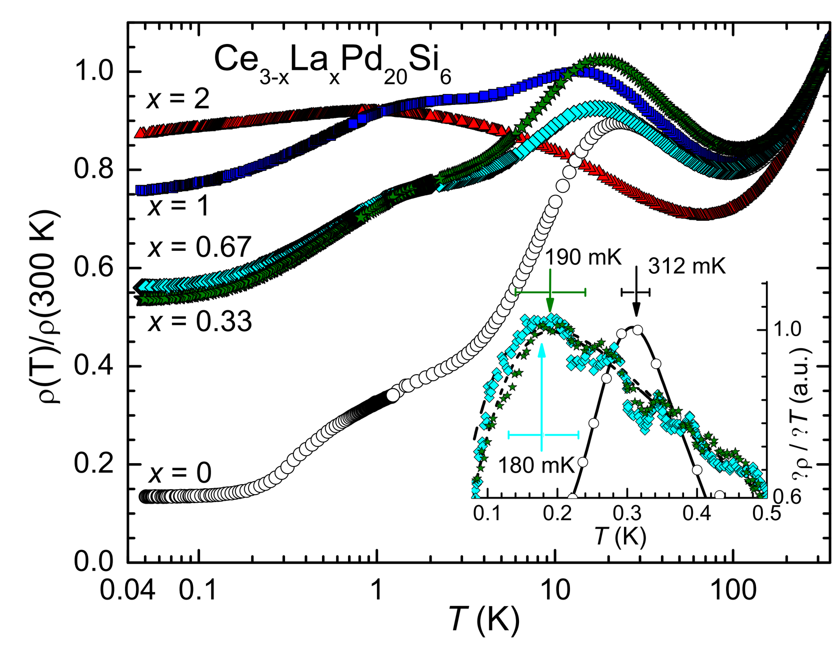

FIGURE 4: Electrical resistivity versus temperature (50mK - 300K) of $Ce_{3-x}La_xPd_{20}Si_6$ (x = 0, $\frac{1}{3}$, $\frac{2}{3}$, 1 and 2), normalized to the respective value at 300 K, $\rho(T)/\rho(300\text{ K})$. The inset shows the temperature derivative of ρ, normalized to the respective values of the local maxima, $\partial\rho/\partial T_{norm}$.

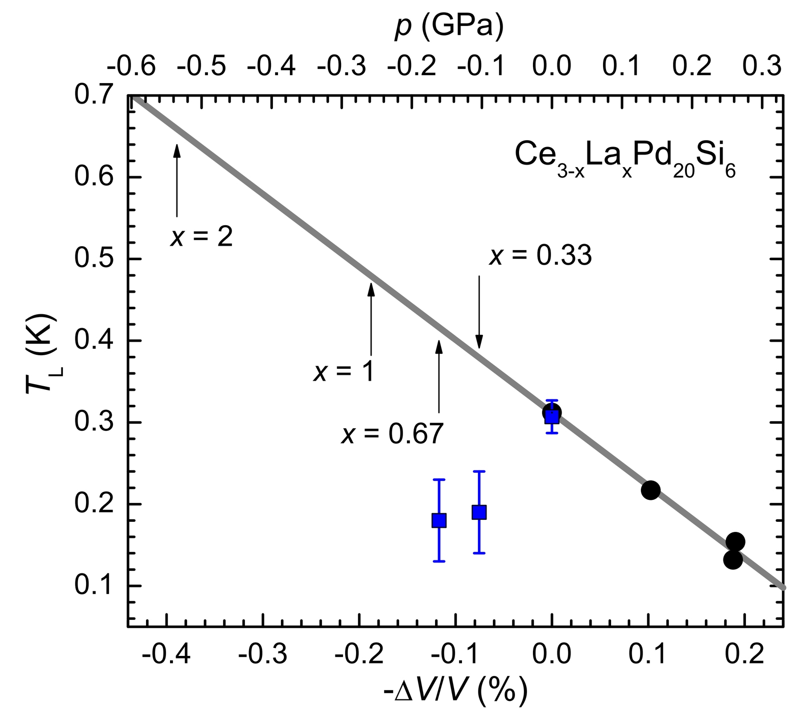

FIGURE 5: Lower transition temperature T_L (black dots) determined in [5] from specific heat data for various off-stoichiometric $Ce_3Pd_{20}Si_6$ single crystals as a function of the relative volume change ΔV/V (bottom) or pressure p (top). The straight line is a linear fit from [5], and its extrapolation towards larger volumes. The blue squares are the temperatures of the maxima in $\partial\rho/\partial T$.

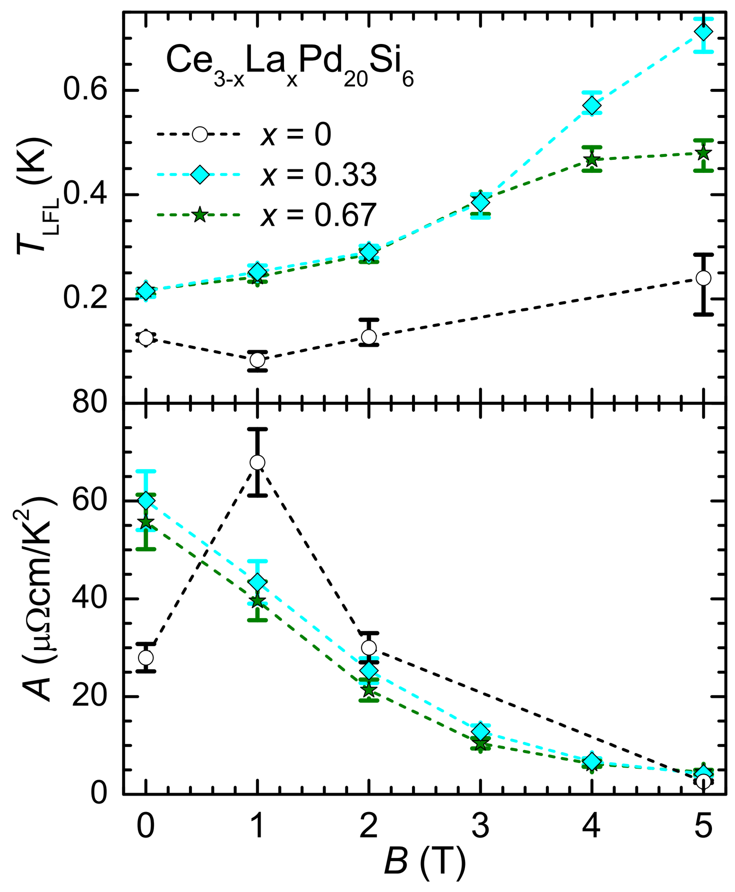

FIGURE 6: Temperature T_{LFL} up to which Landau Fermi liquid (LFL) behaviour prevails, and A coefficient as determined from the fits ($\rho(T) = \rho_0 + AT^2$) , versus magnetic field B. Results for x = 0 are taken from [2].

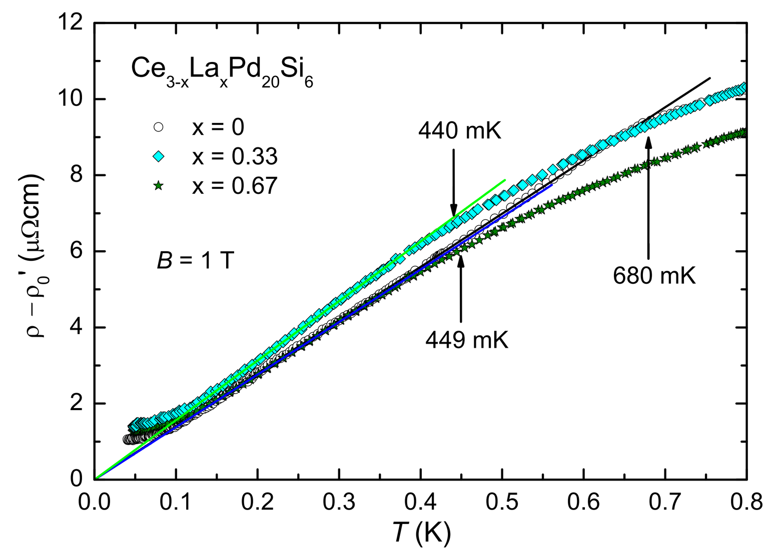

FIGURE 7: Difference of electrical resistivity ρ and the residual resistivity $\rho_0{}'$ and fits (lines) to the NFL form $\rho(T) = \rho_0{}' + A'T$ versus temperature T. Results for x = 0 are taken from [2].

Conclusion

Electrical resistivity measurements on $Ce_{3-x}La_xPd_{20}Si_6$ in a wide temperature range (50mK - 300K) in magnetic fields up to 5T are presented. The evolution of the incoherent Kondo scattering at high temperatures suggests that La substitutes preferentially at the crystallographic $4a$ site and that this site plays only a minor role in the incoherent Kondo scattering due to a lower Kondo temperature. Off-stoichiometry in single crystalline $Ce_3Pd_{20}Si_6$ disturbs the Kondo lattice at the $8c$ site much more strongly than La substitution, as long as x does not exceed 1. The temperature and field dependent electrical resistivity data at the lowest temperatures suggest that quantum critical points in the $x = \frac{1}{3}$ and $\frac{2}{3}$ samples occur at magnetic fields below 1T. Specific heat measurements are needed to confirm the ordering temperatures of about 0.19 K and 0.18 K for the $x = \frac{1}{3}$ and $\frac{2}{3}$ samples, respectively.

Acknowledgements

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