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Superconducting condensation energy in a diluted Kondo lattice

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Abstract. Heavy fermion compounds are host to many interesting but poorly understood strong correlation effects, leading in exceptional cases to unconventional superconductivity. CeCoIn₅ is such an example of an ambient pressure heavy fermion superconductor with a T_c of 2.3K. Such compounds are best described as a Kondo lattice, where Ce, Yb or U ions form a periodic lattice of magnetic moments coupled to the conduction electrons via antiferromagnetic exchange interaction. Dilution, or substitution, of the Kondo ion with a lanthanide, actinide or alkali-earth element that does not participate in the Kondo lattice, results in the so-called "Kondo hole". The electron scattering off the Kondo hole leads to significant changes in both the normal and superconducting properties. We have investigated the suppression of superconductivity in CeCoIn₅ diluted with La, Yb, Y, Ca, Th, Gd, Er, Eu and Lu via specific heat measurements on well characterized single crystals. The suppression of the superconducting condensation energy exhibits a remarkable departure from the Abrikosov-Gorkov impurity pair breaking picture, emphasizing that the Kondo holes are no ordinary pair-breakers.

CeCoIn₅ is an ambient pressure[1], d-wave[2], superconductor with the highest T_c (2.3 K) among the Ce-based heavy fermion superconductors. Together with the parent PuCoGa₅[3] ($T_c = 18$ K), superconductivity in this system forms out of a lattice of incompletely quenched f-electron local moments. This leads to the suggestion that the pairing mechanism directly involves the Kondo interaction[4] in addition to, or rather than antiferromagnetic spin fluctuations. Superconductivity in CeCoIn₅ also displays an unusual interplay with antiferromagnetism. Although no long range magnetic order is reported at H=0 for the pure CeCoIn₅, a resonance peak is observed below T_c in inelastic neutron scattering[5] along the commensurate wavevector $Q = (1/2, 1/2, 1/2)$, corresponding to undamped magnon-like excitations within the superconducting state[6]. A long range antiferromagnetic state is stabilized, with the same commensurate structure[7], when In is substituted by very small amounts of Cd[8] or Hg[9]. When superconductivity is suppressed with magnetic fields, strong deviations from Fermi liquid behavior are observed in the specific heat[10] and resistivity measurements[11], signaling the proximity of a zero temperature magnetic instability, also known as a quantum critical point(QCP).

La substitution for Ce in this compound has been investigated by various groups and the emerging picture is rather counter-intuitive. First, the Kondo coherence temperature is weakly doping-dependent up to about 40%La beyond which it vanishes abruptly[12]. This is hard to reconcile with the idea that the coherence in the Kondo lattice results from intersite coupling

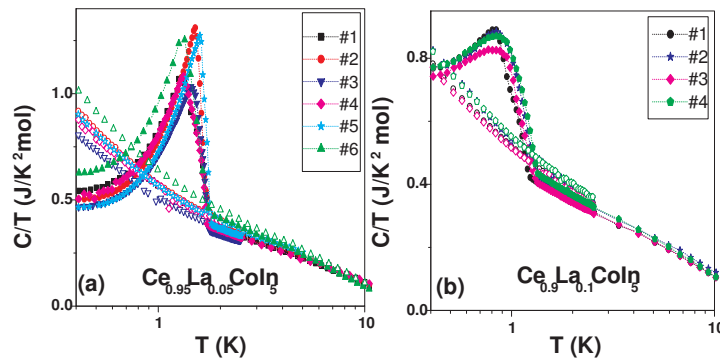


Figure 1. Electronic specific heat vs Temperature at $H = 0$ (closed symbols) and $H = 5T$ (open symbols) in single crystals of $\text{Ce}_{1-x}\text{La}_x\text{CoIn}_5$ for (a) $x = 0.05$ and (b) $x = 0.1$.

among neighboring Ce's. Moreover, the residual thermal conductivity in the superconducting state at $T = 0$ does not have a universal, doping independent, value[13], as expected in the presence of nodal quasiparticles in a d-wave superconductor. This has been interpreted[13] in an extreme multiband scenario in which there remains unpaired electrons on the 3D pockets of the Fermi surface[13], an interpretation that has been contested both on theoretical[14] and experimental[15] grounds. Motivated by these results, we have investigated the effect of Ce-site substitution on superconducting properties of CeCoIn_5 for an extensive range of dopants and concentrations.

Single crystals of $\text{Ce}_{1-x}\text{R}_x\text{CoIn}_5$ were grown from excess In flux with $R = \text{La}^{3+}, \text{Yb}^{2+}, \text{Y}^{3+}, \text{Th}^{4+}, \text{Lu}^{3+}, \text{Ca}^{2+}, \text{Eu}^{2+}, \text{Er}^{3+}, \text{Gd}^{3+}$ as described elsewhere[1]. Samples were etched in dilute HCl to remove excess In and carefully screened for free-In inclusions. Energy dispersive X-ray spectroscopy (EDS) shows that Ce has been effectively substituted by R , although an accurate determination of the actual concentration was not possible due to the non-uniform distribution of dopants. Figure 1 shows the electronic specific heat, measured in In-free single crystals of $\text{Ce}_{1-x}\text{La}_x\text{CoIn}_5$ with nominal La concentrations of $x = 0.05$ (1a) and $x = 0.1$ (1b). The electronic specific heat is obtained by subtracting the lattice contribution, determined from LaCoIn_5 . At zero field, the specific heat transition has the same onset temperature of $T_c = 1.8\text{K}$ ($x = 0.05$) and 1.3K ($x = 0.1$) for all samples from a given batch, whereas the size of the specific heat anomaly, as well as the $T = 0$ intercept of $\frac{C}{T}$, γ_0 , is sample-to-sample dependent. As also shown in Fig. 1, a field of $H = 5T$ applied along [001] suppresses the SC transition and reveals a divergent $\frac{C}{T}$ in all samples, similar to the non-Fermi Liquid behavior reported in pure CeCoIn_5 [10]. Similar observations hold when Ce is substituted by $R = \text{Yb}^{2+}, \text{Th}^{4+}, \text{Y}^{3+}, \text{Lu}^{3+}, \text{Eu}^{2+}, \text{Er}^{3+}, \text{Gd}^{3+}$ and Ca^{2+} , as seen in Figure 2: the specific heat jump broadens and is suppressed as the T_c decreases, while γ_0 systematically increases.

Figure 3 summarizes our main results. The specific heat jump ΔC is shown in Fig. 3a as a function of the critical temperature T_c , both quantities normalized with the values of 3.544 J/K mol and 2.25 K found in the pure compound. Regardless of the nature of R (magnetic or not, isovalent or not), the data for all $\text{Ce}_{1-x}\text{R}_x\text{CoIn}_5$ compounds fall on a single line which is clearly distinct from the prediction of Abrikosov-Gorkov (AG) theory[17], also shown in Fig. 3a as a solid line. In contrast to Ce-site substitution, In-site substitution by Sn[16] do follow the AG theory fairly well. The difference between Ce vs In site substitution is a compelling evidence that the deviation we observe is due to the Kondo hole scattering. Fig. 3b shows the doping dependence of T_c . The doping x is obtained via EDS analysis and the non-uniform spatial distribution of doping within samples prevented an accurate determination of x . We attribute

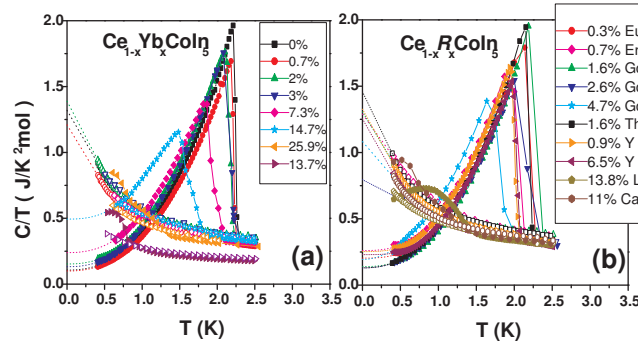


Figure 2. Electronic specific heat vs Temperature at $H = 0$ (closed symbols) and $H = 5T$ (open symbols) in single crystals of (a) $Ce_{1-x}Yb_xCoIn_5$ and (b) $Ce_{1-x}R_xCoIn_5$ ($R = Th, Lu, Eu, Er, Gd, Ca, Y$) for the indicated concentrations. Dashed lines correspond to power law (linear) fits to the $H = 0$ ($H = 5T$) data.

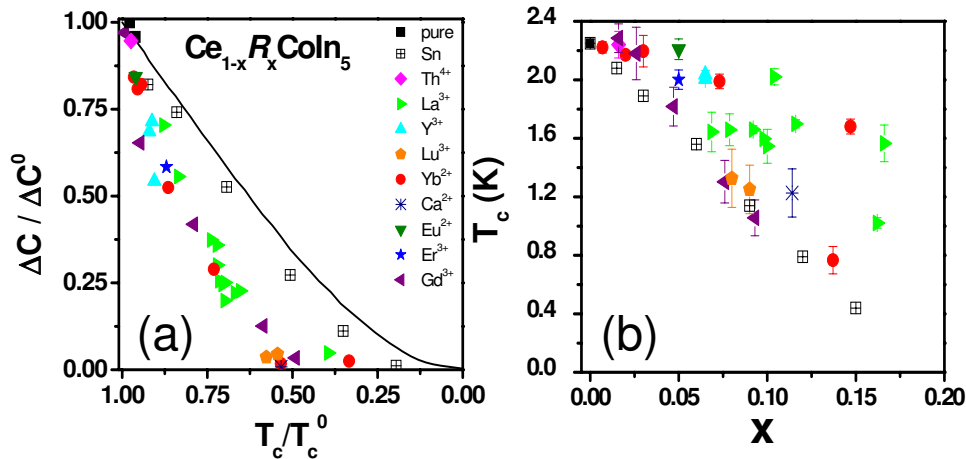


Figure 3. Normalized specific heat jump vs normalized T_c at $H = 0$ in single crystals of $Ce_{1-x}R_xCoIn_5$ with $R = La^{3+}, Yb^{2+}, Y^{3+}, Th^{4+}, Lu^{3+}, Ca^{2+}, Eu^{2+}, Er^{3+}, Gd^{3+}$, as indicated. The values of $\Delta C = 3.544 J/K \text{ mol}$ and $T_c = 2.25 K$ for pure $CeCoIn_5$ were used for normalization. For comparison, results of Sn substitution for In are also shown[16]. Solid line corresponds to the prediction based on Abrikosov-Gorkov theory[17]. (b) Doping dependence of T_c in the same crystals.

the spread in the data to the inaccurate determination of x via EDS. A previous investigation of rare-earth substitution, via resistivity, claimed a universal suppression of T_c with increasing residual resistivity, irrespective of the magnetic nature of the substituent[18]. Nevertheless, there is some spread in T_c vs ρ_0 as well. Taken together, these results suggest that the pair-breaking effect due to the Ce-site substituent is not sensitive to the nature of this substituent.

The term Kondo Hole has been coined[19] to describe the creation of a vacancy in the Kondo lattice by chemical substitution of Ce with an element that does not have a local moment which could couple to the conduction band via Kondo effect. While a marked upturn in resistivity observed in $CePd_3$ doped with as little as 3% La has been attributed to Kondo Hole scattering[20], the transport and thermodynamic signatures of Kondo Holes have remained elusive and hard to disentangle from the characteristic properties of the heavy fermion metal in

the normal state. Heavy fermion superconductivity, on the other hand, offers an unequivocal playground to investigate the physics of the Kondo Hole, as the non s-wave pairing is known to be extremely sensitive to any source of scattering. Previous thermodynamic investigations of the suppression of superconductivity upon Ce- or U-site dilution[19, 21, 22, 23, 24] have revealed deviations from the Abrikosov-Gorkov theory[17] in the doping dependence of both T_c and the specific heat jump. However, the universality we observe in $Ce_{1-x}R_xCoIn_5$ with respect to the magnetic or electronic nature of R has been missed in earlier dilution studies in other heavy fermion superconductors.

In conclusion, we have investigated the suppression of the superconductivity via specific heat measurements in well-characterized single crystals of $Ce_{1-x}R_xCoIn_5$ with $R = La^{3+}, Yb^{2+}, Y^{3+}, Th^{4+}, Lu^{3+}, Ca^{2+}, Eu^{2+}, Er^{3+}, Gd^{3+}$. The suppression of the specific heat anomaly at T_c is universal, independent of the valence or the magnetic nature of the substituent. Moreover, we found a striking deviation from the Abrikosov-Gorkov theory for the Ce-site substitution, in contrast to In-substitution, giving compelling evidence for the Kondo hole scattering. More theoretical work is needed in order to account for the suppression of superconductivity in presence of Kondo holes in heavy fermion superconductors.

Acknowledgments

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