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Wilson ratio in Yb-substituted CeCoIn₅

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Abstract – We have investigated the effect of Yb substitution on the Pauli-limited, heavy-fermion superconductor, CeCoIn₅. Yb acts as a non-magnetic divalent substituent for Ce throughout the entire doping range, equivalent to hole doping on the rare-earth site. We found that the upper critical field in (Ce,Yb)CoIn₅ is Pauli limited, yet the reduced (H, T) phase diagram is insensitive to disorder, as expected in the purely orbitally limited case. We use the Pauli-limiting field, the superconducting condensation energy and the electronic specific-heat coefficient to determine the Wilson ratio (R_W), the ratio of the specific-heat coefficient to the Pauli susceptibility in CeCoIn₅. The method is applicable to any Pauli-limited superconductor in the clean limit.

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Introduction. – Heavy-fermion (HF) systems have been an ideal playground for investigating unconventional superconductivity (SC) since the discovery of SC in this class of materials [1,2]. Much of the attention has been focused on the symmetry of the superconducting order parameter and the interplay/competition between SC and magnetism [3]. Such investigations laid ground for magnetism as the origin of Cooper pairing, since SC in HF seems to occur invariably in close proximity to a $T=0$ magnetic instability [4]. Their large specific-heat (C) anomaly at the SC transition and the absence of SC in the non-magnetic La analogs imply that Cooper pairs form out of heavy quasiparticles (QP). Thus, the heavy mass and SC appear to originate from the same mechanism. Yet, the relationship between SC and the Kondo lattice physics, at the heart of the HF problem, has only recently come to spotlight. In particular, a new model of superconductivity for the 115 family of heavy fermions shows that the composite heavy quasiparticles form only when the system becomes superconductor [5].

CeCoIn₅ is an ambient pressure SC with $T_c = 2.3$ K [6] and has the unique feature of an antiferromagnetic (AFM) quantum critical point located near the upper critical field H_{c2} , indicating that AFM is superseded by SC [7,8].

Moreover, the change of the SC transition from second to first order for transition temperatures $T_c \leq T^* \sim 0.7 T_{c0}$ [9] combined with the discovery of a second SC phase in the large B/T region of the phase diagram lead to the suggestion that CeCoIn₅ is the first realization [10,11] of a Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state [12,13]. Subsequent NMR measurements have shown that only some of the NMR lines broadened within the second SC phase, consistent with local moment ordering [14]. A more recent neutron diffraction experiment has found that this second SC phase carries AFM order that collapses at the same upper critical field at which SC is destroyed [15]. CeCoIn₅ is thus the first example of magnetic order being stabilized by superconductivity, suggesting a ground state differing from the one proposed by FF and LO.

The unconventional $d_{x^2-y^2}$ gap symmetry in CeCoIn₅ has been established based on the angular dependence of C [16] and thermal conductivity [17,18], as well as point contact spectroscopy [19]. Recently, a resonance peak has been discovered below T_c in inelastic neutron scattering [20], suggesting a magnetically mediated pairing in analogy with the high- T_c cuprates. CeCoIn₅ has also been a model system for investigating the emergence of coherence in a Kondo lattice. A phenomenological two-fluid model has been successfully used to describe the crossover from single-ion Kondo behavior to coherent heavy-fermion ground state in Ce_{1-x}La_xCoIn₅ [21].

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Here we report a specific-heat investigation of CeCoIn_5 as a function of Yb substitution to Ce. Yb acts as a non-magnetic divalent substituent for Ce throughout the entire doping range, equivalent to hole doping on the rare-earth site. The divergence of the specific heat in the normal state (at $H = 5\text{ T}$) is moderately suppressed with Yb doping, as a result of the Ce Kondo-lattice dilution. The upper critical field in $(\text{Ce},\text{Yb})\text{CoIn}_5$ appears to be Pauli limited, as in the pure compound, yet it exhibits a scaling expected in the purely orbital limit. To the best of our knowledge, this is the first report that the upper critical field is insensitive to the amount of doping in a Pauli-limited superconductor. Furthermore, we show how the Wilson ratio (R_W), the ratio of the specific-heat coefficient to the Pauli susceptibility, can be determined from the superconducting condensation energy in a Pauli-limited superconductor. The method yields a Wilson ratio consistent with the expected value in pure CeCoIn_5 .

Crystal growth and characterization. – CeCoIn_5 crystallizes in the tetragonal HoCoGa_5 structure. Single crystals of $\text{Ce}_{1-x}\text{Ln}_x\text{CoIn}_5$ ($\text{Ln} = \text{Yb}, \text{Lu}$) were grown from excess In flux [6]. The lattice parameters were determined from Rietveld refinement of powder X-ray diffraction patterns, using Si standard. The effective concentrations were determined with energy dispersive X-ray spectroscopy (EDS) on the measured single crystals, as well as proton-induced X-ray emission microprobe (PIXE) on a mosaic of crystals from the same batches. The magnetic susceptibility was measured using a vibrating sample SQUID magnetometer in a field of 1T or higher applied perpendicular to [001]. The heat capacity was measured using the standard adiabatic heat pulse technique in a ^3He -PPMS. The resistivity (ρ) was measured using the standard four wire technique with an AC resistance bridge. Low resistance contacts were obtained by spot-welding Au wires.

Figure 1 shows the doping evolution of characteristic parameters in $\text{Ce}_{1-x}\text{Ln}_x\text{CoIn}_5$ ($\text{Ln} = \text{Yb}, \text{Lu}$). The effective Yb concentrations, as determined with either EDS or PIXE, are close to the nominal values for small x_{nom} but show large distribution around $x_{\text{nom}} = 0.5$, as indicated by the error bars in fig. 1(a). Phase separation between pure YbCoIn_5 and $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$ is likely the reason why we could not reach effectively $x_{\text{Yb}} \geq 0.3$. In fact, for $x_{\text{nom}} \geq 0.7$, the batches yield essentially pure YbCoIn_5 , with very few Yb-substituted CeCoIn_5 crystals. The difference between EDS and PIXE values reflects this difference between a single crystal and the average concentration of the mosaic of crystals. For simplicity, nominal concentrations will be used in the rest of the paper. The lattice volume decreases systematically with Yb and Lu doping due to the lanthanide contraction (see fig. 1(c)). The Curie-Weiss moment μ_{eff} (per formula unit), obtained from linear fits to inverse magnetic susceptibility is suppressed below the Ce^{3+} moment ($2.54\mu_B$) with Yb as with Lu doping (see fig. 1(d)). This indicates

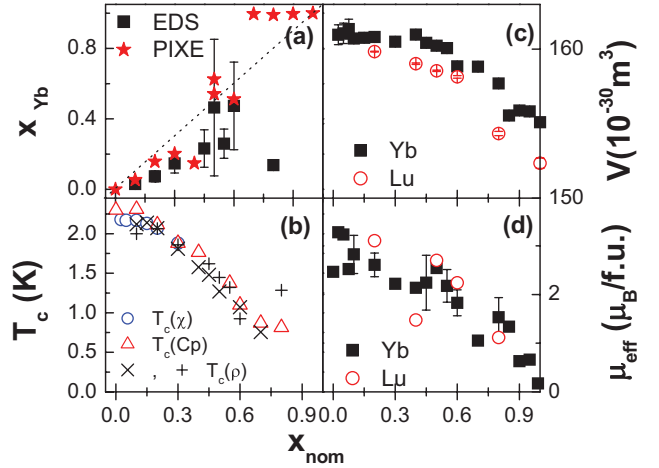


Fig. 1: (Color online) (a) Effective (x_{Yb}) vs. nominal (x_{nom}) concentrations of Yb in $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$, as determined from EDS and PIXE. (b) Critical temperature T_c vs. x_{nom} in $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$, as determined from magnetic susceptibility (χ), specific heat (C) and resistivity (ρ). (c) Lattice volume vs. x_{nom} in $\text{Ce}_{1-x}\text{Ln}_x\text{CoIn}_5$ ($\text{Ln} = \text{Yb}, \text{Lu}$), as determined from powder X-ray diffraction. (d) Effective Curie-Weiss moment μ_{eff} (in units of Bohr magneton) vs. x_{nom} in $\text{Ce}_{1-x}\text{Ln}_x\text{CoIn}_5$ ($\text{Ln} = \text{Yb}, \text{Lu}$), as determined from χ .

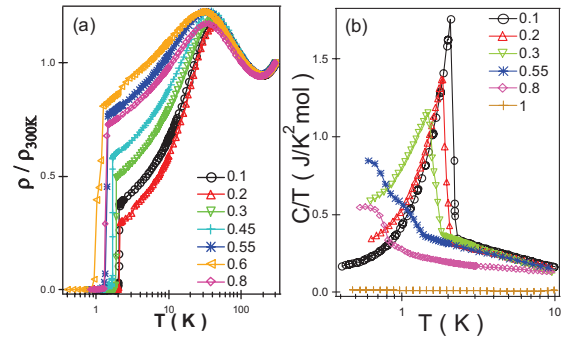


Fig. 2: (Color online) (a) ρ vs. T in single crystals of $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$. (b) Electronic specific heat $\frac{C}{T}$ vs. T in the same crystals at $H = 0$. The indicated concentrations are nominal.

that Yb substitutes as a non-magnetic Yb^{2+} ion, resulting in a dilution of the Ce lattice. As such, the Yb doping is *not* an isoelectronic substitution, unlike La or Lu, but is equivalent to hole doping. The absence of Curie-Weiss behavior in pure YbCoIn_5 and its small Sommerfeld coefficient ($\approx 11\text{ mJ/K}^2\text{ mol}$) shows that it is not a heavy fermion.

Results and discussion. – Figures 2(a) and (b) show the zero-field superconducting transition in ρ and C for various Yb (EDS) concentrations. The transitions in ρ are sharp, except for $x = 0.6$ which shows a double jump, indicative of an inhomogeneous sample, consistent with the large error bars found in the EDS (see fig. 1(a)). Two different crystals have been measured in ρ from most batches and they exhibit very similar T_c 's, determined

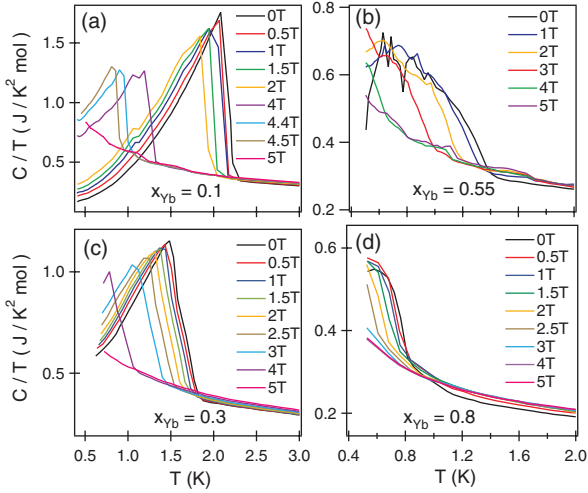


Fig. 3: (Color online) Electronic specific heat $\frac{C}{T}$ vs. temperature for nominal Yb concentrations of (a) $x_{Yb} = 0.1$, (b) $x_{Yb} = 0.3$, (c) $x_{Yb} = 0.55$ and (d) $x_{Yb} = 0.8$, and with magnetic fields ranging from 0 to 5 T, applied parallel to [001].

from the onset of non-zero resistance, and shown in fig. 1(b). A fairly sharp, λ -like anomaly is observed in C , measured on the same single crystals for $x = 0.1, 0.2$ and 0.3 , also shown in fig. 2(b). The SC anomaly is smaller and broader for $x = 0.55$ and 0.8 . The onset of the jump in $\frac{C}{T}$ defines T_c for all samples. Two samples have been measured for $x = 0.55$ and both exhibit a broad jump (but with similar T_c 's) possibly due to the doping inhomogeneity. Overall, the resistive T_c is in good agreement with the thermodynamic T_c determined from $\frac{C}{T}$ and χ , as shown in fig. 1(b), except for $x = 0.8$ with $T_c^p > T_c^C$. The ratio $\frac{\Delta C}{\gamma_0 T_c}$ has been determined with γ_0 taken as the $\frac{C}{T}$ value linearly extrapolated to $T = 0$ in the normal state at $H = 5$ T. In the standard BCS theory, this ratio is expected to be 1.43 in the weak coupling regime. For Yb concentrations $x = 0.1, 0.2$ and 0.3 , the SC appears to be bulk, in contrast to $x = 0.55$ and 0.8 with significantly smaller $\frac{\Delta C}{\gamma_0 T_c}$ values.

The temperature (and field) dependence of the electronic specific heat $\frac{C}{T}$ in Yb-doped CeCoIn₅ is shown in fig. 3, for $x_{Yb} = 0.1, 0.3, 0.55$ and 0.8 (nominal). The electronic contribution is obtained by subtracting the phonon contribution, determined from C of YbCoIn₅. $\frac{C}{T}$ has a divergent T -dependence down to $T_{c2}(H)$ for all Yb concentrations, with little change in slope with increasing magnetic field. Moreover, the divergence of $\frac{C}{T}$ extends down to lowest $T \simeq 0.5$ K at $H = 5$ T in these samples, a characteristic shared with the pure CeCoIn₅. This divergence has been attributed to a field tuned QCP near H_{c2}^0 in CeCoIn₅ [7,8]. As seen in fig. 2(b) and fig. 3, the divergence of $\frac{C}{T}$ becomes weaker with increasing Yb concentration and the corresponding γ_0 , obtained from linear extrapolation of $\frac{C}{T}$ vs. T at $H = 5$ T, decreases systematically with x_{Yb} , consistent with a dilution of the Kondo lattice (see table 1).

Table 1: Doping dependence of characteristic parameters in Ce_{1-x}Yb_xCoIn₅. x_{Yb} : nominal Yb concentration, T_c : critical temperature at $H = 0$ from heat capacity, γ_0 : the $T = 0$ linear extrapolation of $\frac{C}{T}$ at $H = 5$ T in J/K² mol, U_c : the superconducting condensation energy in J/mol, H_{c2}^0 (T) and H_P (T): the orbital upper critical field and Pauli-limiting field.

x_{Yb}	T_c (K)	γ_0	U_c	H_{c2}^0 (T)	H_P (T)
0	2.3	1.2	1.43	13.6	5.3
0.1	2.19	1.15	1.05	13.2	5.2
0.2	1.98	0.96	0.69	12.1	4.7
0.3	1.68	0.96	0.47	12.3	4.8
0.55	1.45	0.84	–	10.2	4.0
0.8	0.77	0.53	–	8.9	3.5

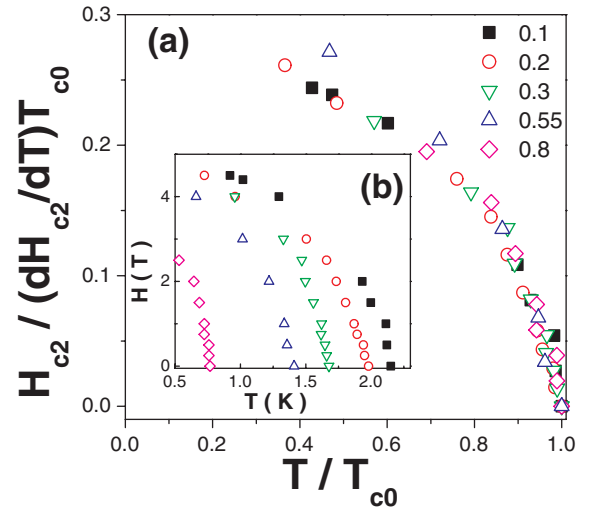


Fig. 4: (Color online) H - T phase diagram in Ce_{1-x}Yb_xCoIn₅. (a) Reduced critical field $\frac{H_{c2}}{dH_{c2}/dT}T_{c0}$ vs. reduced critical temperature $\frac{T}{T_{c0}}$ for various Yb concentrations (nominal). (b) Upper critical field H_{c2} vs. temperature.

Figure 3 also shows the magnetic field suppression of the superconducting transition, for magnetic field applied parallel to the [001] axis, with a broader and smaller jump as the field is increased, due to the presence of vortices. Unlike in the pure case, the SC transition in the Yb-doped compounds does *not* sharpen into a first-order anomaly in the $T = 0$ limit, as seen in fig 3, likely due to disorder introduced by doping [22]. The corresponding H - T phase diagrams (deduced from C) are shown in figs. 4(a) and (b). The upper critical field ($H_{c2}(T)$) is determined from the kink in the entropy S , corresponding to the midpoint of the specific-heat jump seen in fig. 3. S is obtained by integrating $\frac{C}{T}$, following a quadratic [23] extrapolation of C vs. T to $T = 0$. For $x_{Yb} = 0.55$, the average critical field of two samples is shown. In pure CeCoIn₅ the first-order SC transition [9] as well as the $H_{c2}(T)$ fits [24] give clear evidence for a Pauli-limited H_{c2} . Despite the absence of a first-order transition in Yb-doped compounds, the

Pauli limit [25] still applies: the extrapolation based on the standard expression [26], $H_{c2}^0 \simeq -0.7 \frac{dH_{c2}}{dT} T_c$, yields an orbital critical field far in excess of the observed transition field, see table 1.

Figure 4(a) shows that the H_{c2} data for all Yb concentrations collapse onto a single curve when scaled by the initial slope at T_c . Such a scaling is expected in the purely orbital limit, as the reduced critical field *vs.* the reduced temperature $\frac{T}{T_c}$ is only weakly dependent on the disorder level in this limit [26]. In the Pauli limit there is no reason to expect a similar scaling, however the orbital mechanism is still predominant in the zero-field limit. The implication of this scaling is that the Maki parameter [22], the ratio of the orbital critical field to the Pauli-limiting field: $\alpha = \frac{\sqrt{2}H_{c2}^0}{H_P}$, is independent of x_{Yb} . This is not a trivial result, knowing that α decreases under pressure [27]. α was estimated to be 3.6 for $H||[001]$ in pure CeCoIn₅ [9]. We have used this value to determine H_P in the Yb-doped samples from H_{c2}^0 , see table 1).

In a *d*-wave BCS superconductor, the superconducting condensation energy, U_c , is related to the specific-heat jump ΔC at the SC transition via the standard relation [28]: $U_c = \frac{(\Delta C)T_c}{4}$. Since $\frac{C}{T}$ in the normal state is divergent in CeCoIn₅ and since it is likely a strong coupling superconductor [6], the use of this formula is questionable. Alternatively, we have determined U_c directly from integration of $\int_0^{T_c} (S_n - S_s) dT$ up to T_c , following extrapolation of the $\frac{C}{T}$ data to $T=0$. We have excluded the $x=0.55$ and 0.8 samples from the analysis since the lowest temperature (0.4 K) in the data does not allow a proper extrapolation to $T=0$. The results are listed in table 1. The obtained value of U_c for pure CeCoIn₅ (1.43 J mol⁻¹) compares favorably with the value (1.34 J mol⁻¹) determined from magnetization [29]. For the doped samples, we found that the condensation energy decreases with increasing Yb concentration, a trend similar to the one reported for La (not shown) or Sn doping [30].

The combination of U_c and H_P then allows the determination of the Pauli paramagnetic susceptibility (χ_0). In fact, H_P is related to U_c via [25]: $U_c = \frac{1}{2}\chi_0 H_P^2$. This expression is originally derived for an *s*-wave superconductor [25] and overlooks the possibility of a finite Pauli susceptibility at $T=0$ in the superconducting state [31]. For a *d*-wave superconductor, it only remains valid in the clean limit. In the presence of nodal quasiparticles in a *d*-wave superconductor, there should be a contribution to the free energy in the superconducting state and thus the Pauli critical field should be derived from: $F_n - \chi_n H_P^2 = F_s - \chi_s H_P^2$, where the subscripts “*n*” and “*s*” refer to the normal and the superconducting states. This is equivalent to $\Delta F = F_n - F_s = (1 - \frac{\chi_s}{\chi_n})\chi_n H_P^2$ and thus $U_c = \frac{1}{2}(1 - \frac{\chi_s}{\chi_n})\chi_n H_P^2$. It is straightforward to assume that the fraction of excited nodal quasiparticles, $\frac{\chi_s}{\chi_n}$, should be proportional to the Yb concentration, but since we do not have an exact determination of this ratio, we

will not pursue the analysis in the Yb doped compounds. Instead, we focus on pure CeCoIn₅, which is in the clean limit, implying that $\frac{\chi_s}{\chi_n} \ll 1$. For CeCoIn₅, $\chi_0 = 10^{-4}/4\pi$ determined from U_c via this formula is close to the value of the *c*-axis susceptibility [6], $\chi = 1.54 \times 10^{-4}/4\pi$ at 1.8 K. The resulting Wilson ratio is $R_W = \frac{\chi_0}{\gamma_0} = 0.76 R_W^0$, where R_W^0 is the free-electron value. In this method, the error on R_W essentially comes from the uncertainty on U_c (via χ_0).

The Wilson ratio of a free-electron gas is defined as the ratio of the Pauli paramagnetic susceptibility to the electronic specific-heat (Sommerfeld) coefficient and is a universal number: $R_W^0 = \frac{\chi}{\gamma} = \frac{3\mu_B^2}{\pi^2 k_B^2}$. Despite the large mass renormalization, heavy-fermion systems exhibit Wilson ratios very close to the free-electron gas value. This is due to the fact that the mass renormalization corresponds to an enhanced density of states at the Fermi level, and the latter determines both the paramagnetic susceptibility and the specific-heat coefficient [32]. The difficulty in estimating the Wilson ratio in heavy-fermion systems is associated with the experimental determination of the low-temperature Pauli susceptibility. In fact, the magnetic susceptibility is overwhelmingly dominated by the Curie-Weiss contribution of Ce moments. Here we show that this difficulty can be overcome in the case of Pauli-limited heavy-fermion superconductors, making use of the superconducting condensation energy. The R_W we obtained from this method in pure CeCoIn₅ is close to but somewhat lower than the expected value of $R_W = \frac{2J+1}{2J} R_W^0 = 1.2 R_W^0$ for a system of $J=5/2$ local moments (corresponding to Ce), confirming the validity of the method.

Conclusion. – In conclusion, we have investigated the effect of Yb substitution on the superconducting and heavy-fermion properties of CeCoIn₅. Our findings can be summarized as follows: i) the suppression of the condensation energy and Sommerfeld coefficient γ_0 with Yb doping is due to the dilution of the dense Ce Kondo lattice, ii) the upper critical field exhibits a scaling that is unusual for Pauli-limited superconductors, implying a doping-independent Maki parameter. Moreover, we introduce a new method for the determination of the Wilson ratio from the superconducting condensation energy, which is valid for any clean superconductor in the Pauli limit. The value we estimate is consistent with the expected value for pure CeCoIn₅.

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