

## Nodal to Nodeless Superconducting Energy-Gap Structure Change Concomitant with Fermi-Surface Reconstruction in the Heavy-Fermion Compound CeCoIn<sub>5</sub>

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The London penetration depth  $\lambda(T)$  was measured in single crystals of Ce<sub>1-x</sub>R<sub>x</sub>CoIn<sub>5</sub>,  $R = \text{La, Nd, and Yb}$  down to  $T_{\min} \approx 50$  mK ( $T_c/T_{\min} \sim 50$ ) using a tunnel-diode resonator. In the cleanest samples  $\Delta\lambda(T)$  is best described by the power law  $\Delta\lambda(T) \propto T^n$ , with  $n \sim 1$ , consistent with the existence of line nodes in the superconducting gap. Substitutions of Ce with La, Nd, and Yb lead to similar monotonic suppressions of  $T_c$ ; however, the effects on  $\Delta\lambda(T)$  differ. While La and Nd substitution leads to an increase in the exponent  $n$  and saturation at  $n \sim 2$ , as expected for a dirty nodal superconductor, Yb substitution leads to  $n > 3$ , suggesting a change from nodal to nodeless superconductivity. This superconducting gap structure change happens in the same doping range where changes of the Fermi-surface topology were reported, implying that the nodal structure and Fermi-surface topology are closely linked.

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Magnetically mediated pairing is believed to be responsible for unconventional superconductivity found in materials ranging from the high- $T_c$  cuprates to the iron-based superconductors [1] to heavy fermion compounds [2–4]. For a long time, this pairing was thought to always result in a  $d$ -wave superconducting gap symmetry. While unconventional pairing does require a sign changing gap, nodal lines are not actually required, and most iron-based superconductors have an  $s_{\pm}$  gap structure, where any nodes are merely accidental. Recently, there have been some suggestions of fully gapped [5] or  $s_{\pm}$  [6,7] superconductivity in heavy-fermion materials. In this Letter, we present London penetration depth measurements showing that nodes in the gap of pure CeCoIn<sub>5</sub> are *removed* by substituting Yb for Ce, revealing the clear example of a nodeless heavy-fermion superconductor.

CeCoIn<sub>5</sub> has one of the highest transition temperatures among heavy-fermion superconductors,  $T_c = 2.3$  K [8] and reveals quantum criticality when tuned by either pressure [9] or magnetic field [10–13]. The criticality is thought to be due to magnetic fluctuations, making it an intriguing material in which to study the relationship between magnetism, quantum criticality, and the superconducting energy-gap structure.

Several experimental studies suggested the presence of line nodes in the superconducting gap of pure CeCoIn<sub>5</sub> [14–17]. Magnetic field direction-dependent thermal conductivity and heat capacity [18,19] were interpreted [20] as evidence for a  $d_{x^2-y^2}$  gap. This conclusion is supported by directional point contact spectroscopy [21],  $k$ -space resolved quasiparticle interference scanning tunneling microscopy (STM) [22,23], and the spin resonance found at a three-dimensional  $(\pi, \pi, \pi)$  wave vector [24].

However, some other experiments are difficult to reconcile with the  $d$ -wave scenario. Most importantly, despite very low residual resistivity  $\rho_0 = 0.2 \mu\Omega \text{ cm}$  [25], the London penetration depth of pure CeCoIn<sub>5</sub> has never shown the linear temperature dependence expected in clean  $d$ -wave superconductors. Instead, if it is parametrized by a power law,  $\Delta\lambda(T) = AT^n$ , measurements on crystals from different sources that presumably have different amounts of scattering and by different techniques [16,26–28] yield a variation of the exponent  $n$  between 1.5 and 2, where  $n = 2$  represents the dirty limit in the gapless regime for any pairing symmetry [29]. Similar conclusions about the presence of a large density of uncondensed quasiparticles over an extremely broad temperature and field range were made from doping-dependent thermal conductivity studies [30–32]. The origin of this unusual response in a nominally very clean material remains unclear, and several explanations were put forward, including nonlocal electrodynamics [27] and a temperature-dependent quasiparticle mass enhancement within the superconductor due to a nearby quantum critical point [28,33]. Deviations from a simple  $d$ -wave scenario have stimulated discussions of alternative models in which the Fermi-surface topology plays an important role in the superconducting pairing [6,7], inspired by the ideas put forward for iron-based superconductors [34,35].

To gain insight into this unusual superfluid response, here we report a systematic study of the London penetration depth in crystals of CeCoIn<sub>5</sub>, with Ce substituted by both magnetic and nonmagnetic rare-earth ions: La, Nd, and Yb. Surprisingly, these three dopants lead to very similar rates of  $T_c$  suppression, despite their very different nature: La acts as a nonmagnetic impurity; excess  $f$  electrons on

Nd ions remain localized and induce long range magnetic order with  $T_N < T_c$  in compositions  $x \geq 0.05$  [36,37]; and Yb substitution provides hole doping, leading to a change in the Fermi-surface topology [38,39]. We found that the low-temperature variation of the London penetration depth with La and Nd substitutions is consistent with the presence of line nodes and evolution from clean to dirty behavior. In stark contrast, Yb substitution leads to a nodal-to-nodeless transformation of the superconducting gap concomitant with the Fermi-surface topology change. This observation is a challenge for the conventional  $d$ -wave picture of magnetically mediated pairing, and difficult to reconcile with the large Coulomb repulsion that should strongly suppress any on-site pairing. A follow-up theoretical paper shows how local, non-Cooper  $d$ -wave pairing can still be consistent with the absence of nodes [40].

Single crystals of  $\text{Ce}_{1-x}R_x\text{CoIn}_5$  ( $R = \text{La, Nd, Yb}$ ) were grown using the In flux method [36,41–44]. In all cases the values of  $x$  were determined using electron-probe microanalysis with wavelength dispersive spectroscopy on the same samples as used in the penetration depth measurements. While compositions for La and Nd substitutions are close to nominal, a large, nearly threefold discrepancy between nominal and actual  $x$  is found for Yb doping [45,46]; note that our actual  $x = 0.015$  and  $x = 0.037$  correspond to nominal  $x = 0.1$  and  $x = 0.2$ . Samples for in-plane London penetration depth measurements were cut and polished into rectangular parallelepipeds with typical dimensions  $\sim 0.6 \times 0.6 \times 0.1 \text{ mm}^3$  ( $a \times b \times c$ ). Details of the tunnel-diode resonator measurements of London penetration depth in a dilution refrigerator and their analysis can be found elsewhere [47–49].

Figure 1 shows the temperature-dependent normalized rf magnetic susceptibility of the samples used in this study over the range from base temperature to  $T_c$ . In all cases, chemical substitution suppresses  $T_c$ , with  $T_c(x)$  in agreement with previous studies [36,41,44,45,46] as shown in Figs. 1(d)–1(f). The transitions remain sharp even in doped samples, suggesting a homogeneous dopant distribution.

In Fig. 2 we show the temperature variation of  $\Delta\lambda(T)$  in three nominally pure samples of  $\text{CeCoIn}_5$ , S1, S2, and S3. For reference we show measurements made in a slightly Yb doped sample,  $x = 0.002$ , with all measurements taken in identical conditions in the same setup and using the same thermometry. This comparison clearly shows that the  $T_c$  of nominally pure samples varies by as much as 0.1 K, possibly due to different amounts of scattering. Not unexpectedly, the low-temperature variation,  $\Delta\lambda(T)$ , changes with  $T_c$ . Fitting data with a power-law function,  $\Delta\lambda(T) = AT^n$ , we find that  $n$  is a strong function of  $T_c$ , as shown in the inset of Fig. 2. In the highest  $T_c$  sample (S1), the exponent  $n = 1.25$  is below 1.5 and is close to 1, as expected for superconductors with line nodes in the clean limit. We use the data for this sample as the reference in the following. For sample S3 and the Yb-doped sample ( $x = 0.002$ ) the exponent is significantly higher, tending

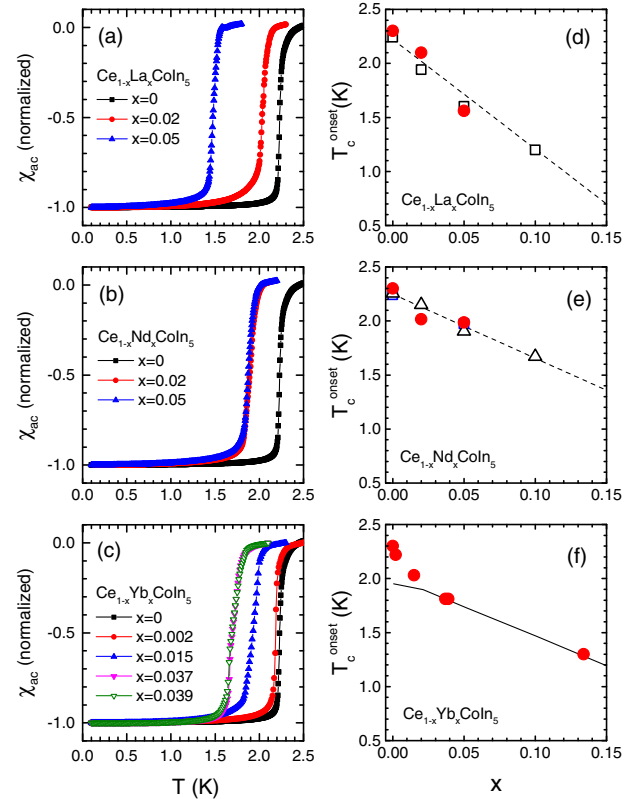


FIG. 1 (color online). Left column panels (a) to (c) show the temperature dependence of normalized rf magnetic susceptibility of  $\text{Ce}_{1-x}R_x\text{CoIn}_5$  for  $R = \text{La}$  (top panel (a),  $x = 0, 0.02$  and  $0.05$  right to left),  $R = \text{Nd}$  (middle panel (b),  $x = 0, 0.02$  and  $0.05$  right to left), and  $x = \text{Yb}$  (bottom panel (c),  $x = 0, 0.002, 0.015, 0.037$ , and  $0.039$ , right to left). Right column panels (d) to (f) show  $T_c(x)$  as determined in our measurements (red solid dots) in comparison with the literature data for  $\text{Ce}_{1-x}\text{La}_x\text{CoIn}_5$  (panel (d), data from Petrovic *et al.* [41]),  $\text{Ce}_{1-x}\text{Nd}_x\text{CoIn}_5$  (panel (e), data from Petrovic *et al.* [36]), and  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  (panel (f), solid line is from Shimozawa *et al.* [46]).

toward  $n = 2$ , consistent with dirty  $d$ -wave behavior [50]. The strong variation of the exponent  $n$  with  $T_c$  may provide an explanation for the unusual exponents found in previous studies.

Figure 3 summarizes penetration depth measurements in  $\text{Ce}_{1-x}R_x\text{CoIn}_5$  ( $R = \text{La, Nd, Yb}$ ). Panel (a) shows data for  $R = \text{La}$  and  $\text{Nd}$ ; Yb substitution data are shown in panel (b). The data are plotted vs a normalized temperature  $(T/T_c)^2$ . For reference, we include data for pure  $\text{CeCoIn}_5$ , S1, which expectedly shows downward curvature consistent with  $n < 2$ . Doping with both La and Nd suppresses  $T_c$  by as much as 0.5 and 0.9 K (see Fig. 1), respectively, and rapidly saturates the exponent at  $n = 2$  for  $x = 0.05$ , as expected for  $d$ -wave superconductors. In contrast, the evolution of  $\Delta\lambda(T)$  with Yb doping is unique. The samples with  $x = 0.037$  and  $0.039$  demonstrate clear saturation at low temperatures, showing high exponents  $n > 2$ , inconsistent with the nodal gap. The increase of the exponent to

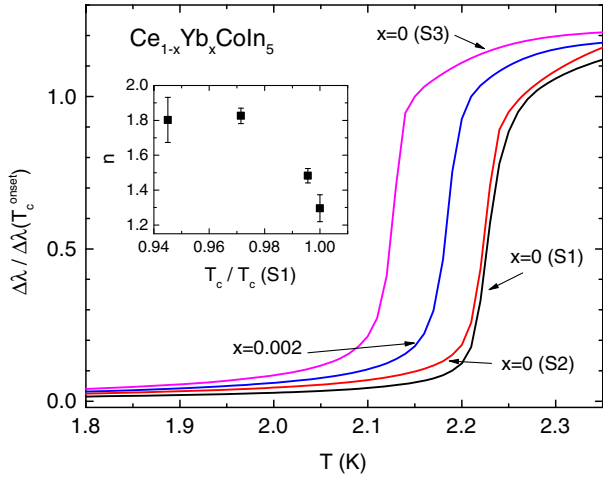


FIG. 2 (color online). London penetration depth  $\Delta\lambda(T)$  in three nominally pure samples of  $\text{CeCoIn}_5$  (S1, S2, and S3) and in a slightly Yb doped sample  $x = 0.002$ . Note that the nominally pure  $\text{CeCoIn}_5$  sample S3 has  $T_c$  lower than the Yb-doped sample. The exponent  $n$  of the power law fit  $\Delta\lambda(T) = AT^n$  (inset), strongly depends on  $T_c$ , tending to  $n = 1$  in the best samples.

values much greater than  $n = 2$  can also be clearly seen in samples with  $x = 0.015$ .

As  $\text{CeCoIn}_5$  is a multigap system [32], we must be careful in our analysis. In single gap  $s$ -wave and  $d$ -wave superconductors, the characteristic behavior of  $\Delta\lambda(T)$  is observed for temperatures  $T < T_c/3$ , where the temperature dependence of the gap  $\Delta(T)$  can be neglected. This assumption is generally not valid for multiband systems, in which the smallest of the gaps determines the low-temperature limit. Since the range over which the smaller gap can be considered as constant is not known *a priori*, it is important to vary the range of the power-law

fitting. We adopted a procedure in which the high temperature end of the fitting interval,  $T_{\text{up}}$ , was varied and the exponent  $n$  was plotted as a function of  $T_{\text{up}}$ , as shown in panel (c) of Fig. 3. Several conclusions can be drawn from inspecting  $n(T_{\text{up}})$  and its evolution with Yb substitution. In samples with  $x = 0.015$ ,  $x = 0.037$ , and  $x = 0.039$  the data are inconsistent with the existence of nodes in the superconducting gap for any  $T_{\text{up}}$ . Moreover, the exponent in the highest doped sample attains values which are practically indistinguishable from the exponential behavior observed in full gap superconductors [49]. Hence, we conclude that the superconducting gap in  $\text{CeCoIn}_5$  undergoes a topological transition from nodal to nodeless with Yb substitution, but not with La or Nd substitutions.

We summarize our study of the evolution of the London penetration depth and  $T_c$  in rare-earth substituted  $\text{CeCoIn}_5$  in Fig. 4. We plot the exponent  $n$  of the power-law analysis as a function of  $T_c$  (left panel) and of  $x$  (right panel). In La and Nd substituted compounds,  $n(x)$  saturates at  $n = 2$ , as expected for superconductors with line nodes. In contrast, Yb substitution brings the exponent above 2, indicating a gap without nodes. Comparison with nonmagnetic La and magnetic Nd clearly shows that this effect is neither due to the spin-flip pairbreaking, nor due to doping-induced magnetism. Instead, we conclude that the hole-doping effect of Yb substitution [45], and the resulting change in the electronic structure is an important factor. Therefore, it is natural to link the change of the superconducting gap to a change in the Fermi-surface topology as suggested by de Haas–van Alphen studies finding the disappearance of the intermediately heavy  $\alpha$  sheet between  $x = 0.015$  and  $0.04$  [38,39,51], exactly where we find the appearance of a nodeless gap. STM studies of  $\text{CeCoIn}_5$  [22] indicate

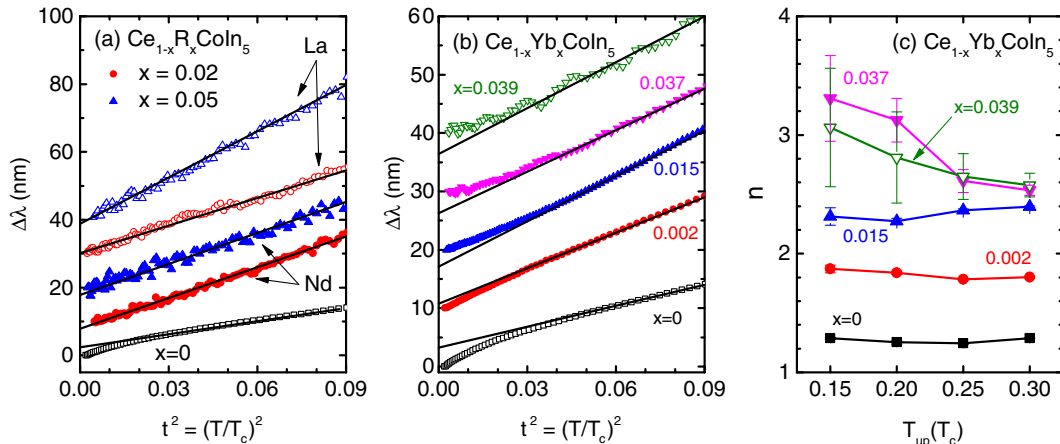


FIG. 3 (color online). London penetration depth of (a) La-, Nd-, and (b) Yb-substituted  $\text{CeCoIn}_5$ , plotted vs a normalized  $(T/T_c)^2$  scale. The data for the pure material ( $x = 0$ , S1) shows a clear downturn, consistent with  $n = 1.25 < 2$ . The data for La- and Nd-doped samples closely follow a  $T^2$  dependence, expected in dirty nodal superconductors for all doping levels. In Yb-substituted samples, there is a clear crossover from sublinear to superlinear, suggesting a rapid increase of the exponent  $n$ , and  $n > 2$  for samples with  $x = 0.015$ ,  $0.037$ , and  $0.039$ . (c) Floating fitting range analysis in pure and Yb-substituted  $\text{CeCoIn}_5$  samples. The data were fit using a power-law function over the temperature range from base temperature to  $T_{\text{up}} < T_c/3$ , and the resultant exponent  $n$  was plotted as a function of  $T_{\text{up}}$ .

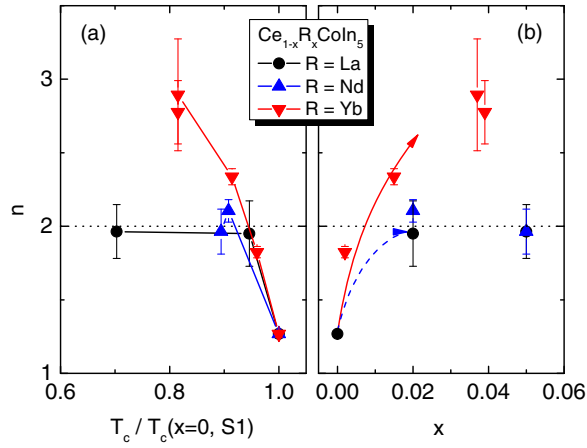


FIG. 4 (color online). The average exponent  $n$  of the power-law fit of  $\Delta\lambda(T)$  as a function of  $T_c$  for La (black circles), Nd (blue up-triangles) and Yb substitution (red down-triangles) (left panel). The values are obtained by the averaging the exponent obtained from the range fitting, see Fig. 3(c). The right panel shows the same data plotted as a function of  $x$ .

that the  $\alpha$  Fermi-surface sheet plays a key role in superconductivity, and a change in the gap structure with its disappearance seems entirely plausible. However, it is difficult to understand why this transformation does not lead to an anomaly in  $T_c(x)$ .

We can think of three possible scenarios to explain the observed transition from nodal to nodeless superconductivity in Yb-substituted  $\text{CeCoIn}_5$ . The simplest possibility is that the original nodes are accidental and disappear as the Fermi surface changes with hole doping. This is similar to accidental nodes evolving with doping in some iron pnictides [52]. However, scattering lifts accidental nodes [53] and this is inconsistent with our results in La- and Nd-substituted samples in which substitution does not change the electron count and the nodes are preserved.

A second possibility is a topological transition from  $d$ -wave to  $s$ -wave pairing at  $x_c$ , where the disappearance of the  $\alpha$  sheet induces a change in the superconducting gap structure. A related scenario involves a transition to  $d_{x^2-y^2} + id_{xy}$  pairing, or another time-reversal symmetry breaking mixture of two gap symmetries that fully gaps out the Fermi surface [54,55], where the  $d_{x^2-y^2}$  pairing is still dominant, but  $d_{x^2-y^2} + id_{xy}$  pairing turns on at a lower temperature  $T_{c2} < T_c$ , leading to a gapped behavior in the low-temperature penetration depth. This second order phase transition should be visible, for example, in specific heat measurements. Both these scenarios should result in an anomaly in  $T_c(x)$ , which is not observed, at least in our experiments.

A third, more exotic, but attractive possibility, discussed in a follow up theory Letter, is that the underlying Fermi surface is unimportant [40], and that the main pairing mechanism is local *composite* pairing, not Cooper pairing. Here, superconductivity arises from cooperative Kondo screening, where two electrons screen the same local

moment to form a composite pair [56]. This process is local and does not require an underlying Fermi surface, allowing Yb substitution to tune the heavy Fermi liquid toward a Kondo insulator without affecting the pairing strength or  $T_c$ . The resulting superconductivity is still  $d$  wave, but it is nodeless due to the removal of the Fermi surface at  $x_c$ , leading to an exponential penetration depth at low temperatures. In reality,  $\text{CeCoIn}_5$  has many bands and will not become a Kondo insulator, as only the dominant  $\alpha$  band is removed, while its superfluid stiffness remains. However, the remaining bands have unobservably small gaps [22] and the signal from any remaining nodal quasiparticles will be within the experimental resolution.

In conclusion, by performing systematic measurements of the London penetration depth in  $\text{Ce}_{1-x}\text{R}_x\text{CoIn}_5$ ,  $R = \text{La}$ ,  $\text{Nd}$ , and  $\text{Yb}$ , we find an anomalous evolution of the superconducting gap structure in Yb-substituted compounds from nodal to nodeless, possibly linked with the Fermi-surface topology change.

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