

# Universal heat conduction in $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$ : Evidence for robust nodal $d$ -wave superconducting gap

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In the heavy-fermion superconductor  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$ , Yb doping was reported to cause a possible change from nodal  $d$ -wave superconductivity to a fully gapped  $d$ -wave molecular superfluid of composite pairs near  $x \approx 0.07$  (nominal value  $x_{\text{nom}} = 0.2$ ). Here we present systematic thermal conductivity measurements on  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  ( $x = 0.013, 0.084$ , and  $0.163$ ) single crystals. The observed finite residual linear term  $\kappa_0/T$  is insensitive to Yb doping, verifying the universal heat conduction of the nodal  $d$ -wave superconducting gap in  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$ . Similar universal heat conduction is also observed in the  $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$  system. These results reveal a robust nodal  $d$ -wave gap in  $\text{CeCoIn}_5$  upon Yb or Cd doping.

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$\text{CeCoIn}_5$  is an archetypal heavy-fermion superconductor with superconducting transition temperature  $T_c \simeq 2.3$  K [1]. With many similarities to the high- $T_c$  cuprates, including quasi-two-dimensionality (quasi-2D), proximity to antiferromagnetism, and a non-Fermi-liquid normal state, the unconventional superconductivity in  $\text{CeCoIn}_5$  has long been thought to be due to spin fluctuation pairing [2–5]. The superconducting gap structure  $\Delta(k)$  of  $\text{CeCoIn}_5$  has been widely studied with various experimental probes ever since it was discovered. These include specific heat [6], thermal conductivity [7–10], and surface-sensitive techniques such as point contact spectroscopy and scanning tunneling microscopy (STM) [11–13]. A  $d$ -wave superconducting gap with symmetry-protected nodes has been well established in  $\text{CeCoIn}_5$ .

The investigation of impurity effects can give a better understanding of the exotic normal and superconducting state of  $\text{CeCoIn}_5$  [14,15]. Recently, the anomalous phenomena observed in the  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  system have attracted much attention [16–26]. At first, the violation of Vegard's law was found in  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  single crystals, together with the robustness of  $T_c$  and the Kondo-coherence temperature  $T_{\text{coh}}$  upon Yb doping [16]. However, later measurements on the  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  thin films demonstrated a verification of Vegard's law and strong suppression of  $T_c$  and  $T_{\text{coh}}$  with Yb doping [17]. To solve this discrepancy, Jang *et al.* carefully determined the actual Yb concentration  $x_{\text{act}}$  in  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  single crystals and found that  $x_{\text{act}}$  is only about 1/3 of the nominal value  $x_{\text{nom}}$  up to  $x_{\text{nom}} \approx 0.5$  [18]. With  $x_{\text{act}}$ , the rate

of  $T_c$  suppression with Yb concentration for the single crystals is nearly the same as that observed in the thin films [18].

Nevertheless, the remarkable anomalies observed at  $x_{\text{nom}} = 0.2$  are still very puzzling, including Fermi surface topology change [19], Yb valence transition [20], significant quasiparticle effective mass reduction, and suppression of the quantum critical point [19,21]. Moreover, a recent London penetration depth study by Kim *et al.* suggested the nodal  $d$ -wave superconductivity becomes fully gapped beyond the critical Yb doping  $x_{\text{nom}} = 0.2$  [22]. To explain this, an exotic scenario was proposed in which the nodal Fermi surface undergoes a Lifshitz transition upon Yb doping, forming a fully-gapped  $d$ -wave molecular superfluid of composite pairs [23].

To examine such an exotic scenario, more experiments are highly desired to investigate the superconducting gap structure of the  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  system. Low-temperature thermal conductivity measurement is an established bulk technique to probe the gap structure of a superconductor [27]. According to the magnitude of residual linear term  $\kappa_0/T$  in zero field, one may judge whether gap nodes exist or not. The field dependence of  $\kappa_0/T$  can give further information about nodal gaps, gap anisotropy, and multiple gaps.

In this paper, we report a systematic heat transport study of  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  ( $x_{\text{nom}} = 0.05, 0.2$ , and  $0.4$ ) single crystals. Finite  $\kappa_0/T$  is observed in all three samples, which does not support a fully gapped superconducting state at  $x_{\text{nom}} \geq 0.2$ . Furthermore,  $\kappa_0/T$  manifests a nearly constant value upon doping, i.e., the universal heat conduction, which is an important property of a nodal  $d$ -wave superconducting gap. Similar universal heat conduction is also observed in the  $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$  ( $y_{\text{nom}} = 0.05, 0.075$ , and  $0.1$ ) system. These results demonstrate that the nodal  $d$ -wave gap in  $\text{CeCoIn}_5$  is robust against Yb or Cd doping.

High-quality  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  ( $x_{\text{nom}} = 0.05, 0.2$ , and  $0.4$ ) and  $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$  ( $y_{\text{nom}} = 0.05, 0.075$ , and  $0.1$ ) single crystals were grown by a standard indium self-flux method [28,29]. Samples were etched in dilute hydrochloric acid to remove the In flux on the surfaces. The actual

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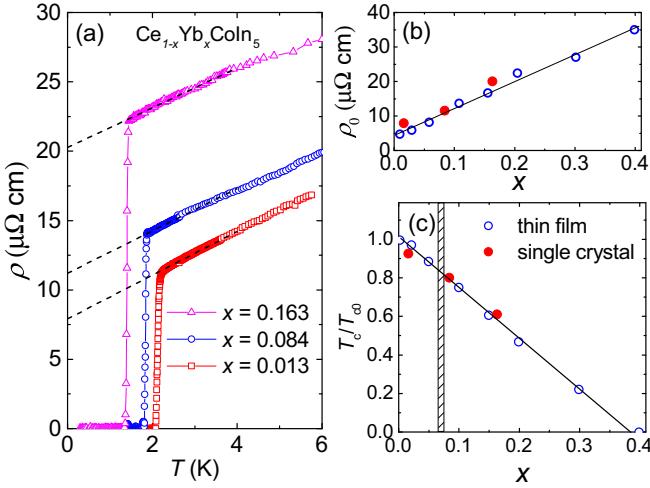


FIG. 1. (a) Low-temperature resistivity of  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  single crystals. Here  $x$  is the actual Yb concentration. The dashed lines are linear extrapolations to get the residual resistivity  $\rho_0$ . (b) and (c) Doping dependence of  $\rho_0$  and normalized  $T_c/T_{c0}$  ( $T_{c0} = 2.3$  K for pure  $\text{CeCoIn}_5$ ) for our single crystals and the thin films (from Ref. [17]). The shadowed narrow bar marks  $x \approx 0.07$ , corresponding to  $x_{\text{nom}} \approx 0.2$ , where several anomalies were reported, including the Yb valence transition.

Yb concentration  $x_{\text{act}} = 0.013$ , 0.084, and 0.163 and the actual Cd concentration  $y_{\text{act}} = 0.004$ , 0.008, and 0.011 were determined by wavelength-dispersive spectroscopy (WDS), utilizing an electron probe microanalyzer (Shimadzu EPMA-1720H). Hereafter,  $x$  and  $y$  represent  $x_{\text{act}}$  and  $y_{\text{act}}$ , respectively. The samples were cut and polished into a rectangular shape. Contacts were made with indium solder for  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  and spot welding for  $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$ , respectively, which were used for both in-plane resistivity and thermal conductivity measurements. The resistivity measurements were made in a  $^3\text{He}$  cryostat. The thermal conductivity was measured in a dilution refrigerator, using a standard four-wire steady-state method with two  $\text{RuO}_2$  chip thermometers, calibrated *in situ* against a reference  $\text{RuO}_2$  thermometer. Magnetic fields were applied along the  $c$  axis and perpendicular to the heat current. To ensure a homogeneous field distribution in the samples, all fields were applied at a temperature above  $T_c$ .

Figure 1(a) shows the low-temperature in-plane resistivity  $\rho(T)$  of  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$ . For  $x = 0.013$ , 0.084, and 0.163, the normal-state resistivity  $\rho(T)$  below 4 K is roughly linear, and the residual resistivities  $\rho_0 = 7.9$ , 11.2, and 20.2  $\mu\Omega$  cm are obtained by linear extrapolation. The  $T_c$ , defined as the midpoint of each resistive transition, is 2.13, 1.84, and 1.40 K, respectively. Figures 1(b) and 1(c) plot the doping dependence of  $\rho_0$  and normalized  $T_c/T_{c0}$  ( $T_{c0} = 2.3$  K for pure  $\text{CeCoIn}_5$ ), respectively. For comparison, the data for  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  thin films from Ref. [17] are also plotted. The curves for the single crystal and thin film samples nearly overlap with each other. This suggests that our determination of the actual Yb concentrations for our single crystals is accurate and further confirms the conclusion of Ref. [18]. According to Ref. [17], the suppression of  $T_c$  can be well reproduced by the Abrikosov-Gorkov (AG) pair breaking curve, suggesting

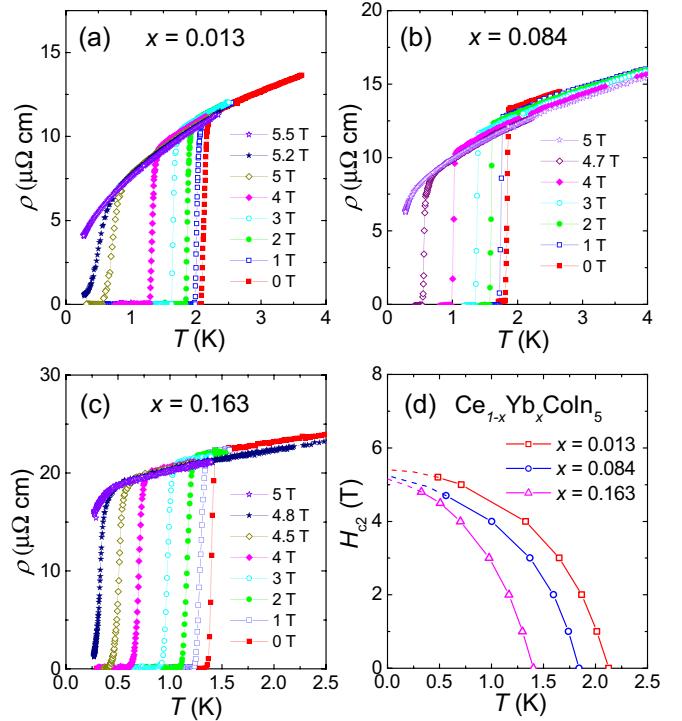


FIG. 2. (a)–(c) Temperature dependence of resistivity for  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  single crystals at various magnetic fields  $H \parallel c$  up to 5.5 T. (d) Temperature dependence of the upper critical field  $H_{c2}(T)$  for all three samples.

that Yb ions act as impurity centers with unitary scattering, regardless of the Yb valence.

Figures 2(a)–2(c) present the resistivity  $\rho(T)$  of  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  single crystals under various magnetic fields. Negative magnetoresistance and sub- $T$ -linear  $\rho(T)$  are observed in the normal state, as in pure  $\text{CeCoIn}_5$  [30]. To determine the zero-temperature upper critical field  $H_{c2}(0)$ , we plot the temperature dependence of  $H_{c2}(T)$  in Fig. 2(d). With rough extrapolation,  $H_{c2}(0) \approx 5.4$ , 5.2, and 5.1 T are obtained for  $x = 0.013$ , 0.084, and 0.163, respectively.  $H_{c2}(0)$  only exhibits a slight decrease with the increase of Yb concentration, in contrast with the strong suppression of  $T_c$ .

The low-temperature in-plane thermal conductivity of  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  single crystals is shown in Figs. 3(a)–3(c). In zero field, all the curves are roughly linear below 0.4 K, with a moderate slope. Previously for pure  $\text{CeCoIn}_5$  at this temperature range, the slope is about 30 times larger than our doped samples and constantly changing [8–10], which impedes an accurate extrapolation of  $\kappa/T$  to zero temperature.  $\kappa_0/T < 2$  mW K $^{-2}$  cm $^{-1}$  and  $\kappa_0/T < 3$  mW K $^{-2}$  cm $^{-1}$  were estimated for pure  $\text{CeCoIn}_5$  in Refs. [8] and [10], respectively. Here, due to the moderate slope of our doped samples, we linearly extrapolate  $\kappa/T$  to zero temperature to obtain  $\kappa_0/T = 1.22$ , 1.19, and 1.08 mW K $^{-2}$  cm $^{-1}$  for  $x = 0.013$ , 0.084, and 0.163, respectively. These values are listed in Table I.

In magnetic fields, linear extrapolations still apply, except for the  $x = 0.013$  and 0.084 samples when  $H > 4.5$  T, as seen in Figs. 3(a) and 3(b). The  $H = 5$  and 5.5 T curves for  $x = 0.013$  and 0.084 samples tend to point to their

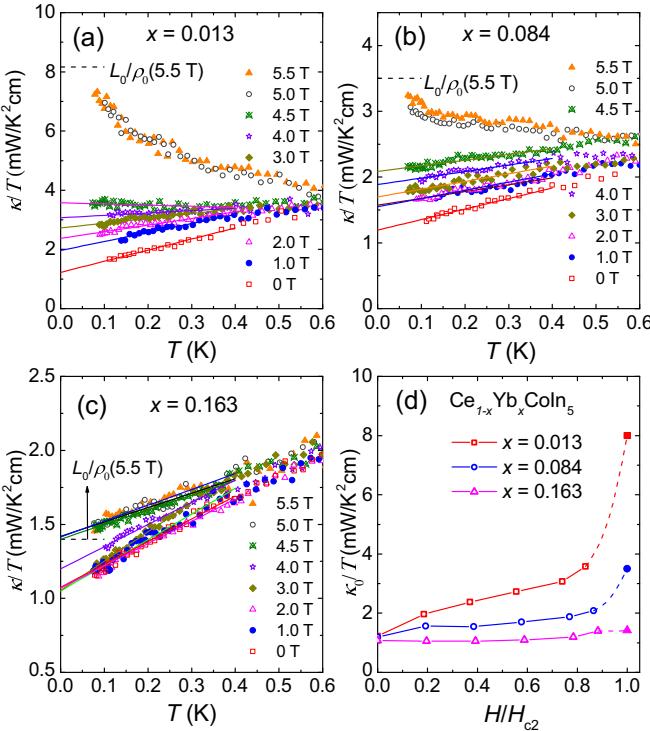


FIG. 3. (a)–(c) Temperature dependence of the thermal conductivity divided by temperature  $\kappa(T)/T$  of  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  under various magnetic fields. The solid lines are linear fits to extrapolate the residual linear term. The dashed lines indicate the normal-state Wiedemann-Franz law expectations  $L_0/\rho_0(H_{c2})$ , with the Lorenz number  $L_0 = 2.45 \times 10^{-8} \text{ W}\Omega\text{K}^{-2}$ . (d) Field dependence of  $\kappa_0/T$ . The field is normalized by the  $H_{c2}$  of the three samples

normal-state Wiedemann-Franz law expectations  $L_0/\rho_0(H_{c2})$ . The field dependence of  $\kappa_0/T$  is shown in Fig. 3(d). For  $x = 0.013$ ,  $\kappa/T$  increases gradually with field, followed by a jump to the normal-state value at  $H_{c2}$ . Such a jump of  $\kappa_0/T$  near  $H_{c2}$  was previously observed in pure  $\text{CeCoIn}_5$ , which was interpreted as the sign of a first-order superconducting transition [10]. For  $x = 0.084$  and  $0.163$ , this jump becomes less and less pronounced.

The major result of this work is that we observe a finite  $\kappa_0/T$  with comparable values at zero field for all three  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  ( $x = 0.013$ ,  $0.084$ , and  $0.163$ ) samples. To

TABLE I. The properties of  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  and  $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$  single crystals. The actual Yb and Cd concentrations  $x_{\text{act}}$  and  $y_{\text{act}}$  were determined from the WDS analysis.  $T_c$  is defined as the midpoint of the resistive transition.

$x_{\text{nom}}$	$x_{\text{act}}$	$T_c$ (K)	$\kappa_0/T$ (mW/K <sup>2</sup> cm)
0.05	0.013	2.13	1.22
0.2	0.084	1.84	1.19
0.4	0.163	1.40	1.08
$y_{\text{nom}}$	$y_{\text{act}}$	$T_c$ (K)	$\kappa_0/T$ (mW/K <sup>2</sup> cm)
0.05	0.004	2.14	1.03
0.075	0.008	2.05	0.90
0.1	0.011	1.92	0.93

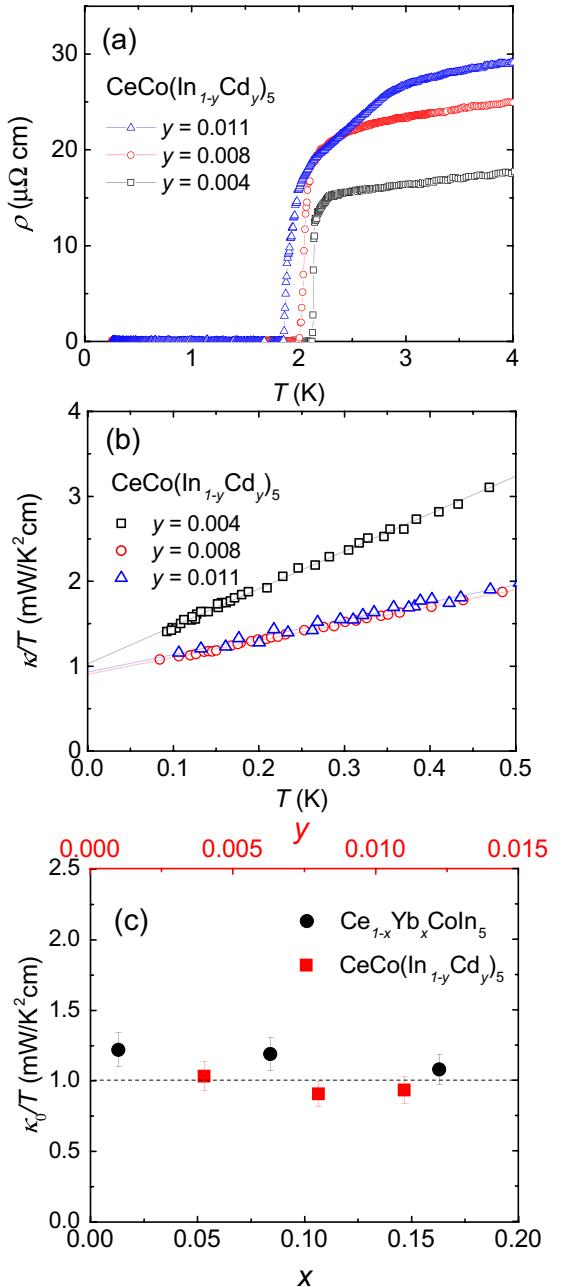


FIG. 4. (a) Low-temperature resistivity of  $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$  single crystals. Here  $y$  is the actual Cd concentration. (b) Temperature dependence of zero-field thermal conductivity divided by temperature  $\kappa(T)/T$  for  $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$ . The solid lines are linear fits to extrapolate  $\kappa_0/T$ . (c)  $\kappa_0/T$  vs doping for all  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  and  $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$  samples at zero field. The error bar is determined from uncertainties in the geometric factor and the fit. The horizontal dashed line is the theoretical universal value for  $\text{CeCoIn}_5$  with a 2D nodal  $d$ -wave superconducting gap [8].

check whether this finite  $\kappa_0/T$  is also present in  $\text{CeCoIn}_5$  containing other dopants, we measured the thermal conductivity of  $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$  ( $y = 0.004$ ,  $0.008$ , and  $0.011$ ) single crystals. The low-temperature resistivity of  $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$  single crystals is shown in Fig. 4(a), from which the values  $T_c = 2.14$ ,  $2.05$ , and  $1.92$  K are obtained, respectively. Figure 4(b)

plots  $\kappa/T$  vs  $T$  at zero field for all three samples, where a linear fit is used to obtain  $\kappa_0/T$ . The value of  $\kappa_0/T$  is 1.03, 0.90, and 0.93 mW K $^{-2}$  cm $^{-1}$  for  $y = 0.004, 0.008$ , and 0.011, respectively, as listed in Table I.

The values for  $\kappa_0/T$  vs dopant concentrations  $x$  and  $y$  for all  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  and  $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$  samples at zero field are plotted in Fig. 4(c). Usually, the presence of a finite  $\kappa_0/T$  is strong evidence for a nodal superconducting gap [27]. For example,  $\kappa_0/T = 1.41$  mW K $^{-2}$  cm $^{-1}$  for the overdoped  $d$ -wave cuprate superconductor  $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+\delta}$  ( $\text{TL}-2201$ ,  $T_c = 15$  K) [31], and  $\kappa_0/T = 17$  mW K $^{-2}$  cm $^{-1}$  for the  $p$ -wave superconductor  $\text{Sr}_2\text{RuO}_4$  ( $T_c = 1.5$  K) [32]. Although there is some uncertainty in the accurate value of  $\kappa_0/T$  for pure  $\text{CeCoIn}_5$  [8,10], the significant  $\kappa_0/T$  observed here for all Yb- and Cd-doped  $\text{CeCoIn}_5$  samples is quite reliable due to the moderate slope. Therefore, the nodal gap in the  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  system persists at least up to  $x = 0.163$ . This is at odds with the earlier penetration depth study [22]. In Ref. [22], Yb doping leads to  $n > 3$  ( $\Delta\lambda(T) \sim T^n$ ) for  $x_{\text{nom}} = 0.2$ , which suggests a nodeless superconducting gap [22]. The reason for this discrepancy is not clear to us.

Furthermore, in Fig. 4(c),  $\kappa_0/T$  manifests a nearly constant value around 1 mW K $^{-2}$  cm $^{-1}$ , irrespective of Yb or Cd concentration, which demonstrates a universal heat conduction in Yb- and Cd-doped  $\text{CeCoIn}_5$ . The universal heat conduction is an important property of a nodal  $d$ -wave superconducting gap, which means that the thermal conductivity is unaffected by change in the bandwidth of impurity bound states  $\gamma$  [33,34]. The universality results from the cancellation between two factors: (i) the density of Andreev bound states, which is proportional to  $\gamma$ , and (ii) the reduction of phase space for scattering of gapless excitations, which is proportional to  $\gamma^{-1}$  [34]. Experimentally, the universal  $\kappa_0/T$  was observed in optimally doped high- $T_c$  cuprates  $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$  and  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  with a  $d$ -wave gap [35,36]. For pure  $\text{CeCoIn}_5$ , in which a 2D nodal  $d$ -wave gap was also well established, the universal  $\kappa_0/T$  was theoretically estimated to be  $\sim 1$  mW K $^{-2}$  cm $^{-1}$  [8]. This agrees very well with our experimental results for the  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  and  $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$  systems, showing that the nodal  $d$ -wave superconducting gap in  $\text{CeCoIn}_5$  is robust upon Yb and Cd doping. In this context, there is no need to propose a fully gapped  $d$ -wave molecular superfluid of composite pairs beyond  $x_{\text{nom}} = 0.2$  [23]. Note that for the  $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$  system, despite the fact that substitution of Cd for In will introduce holes and the samples with  $y = 0.008$  and 0.011 have already entered the region where superconductivity and antiferromagnetism coexist [29,37,38], the universal heat conduction still holds. However, we also notice that in an earlier heat transport study of  $\text{Ce}_{1-x}\text{La}_x\text{CoIn}_5$ ,  $\kappa_0/T \approx 2$  mW K $^{-2}$  cm $^{-1}$  was observed for  $x = 0.05$  and 0.1 [9]. This value is slightly larger than the universal  $\kappa_0/T$  observed here in Yb- and Cd-doped  $\text{CeCoIn}_5$ .

It should be pointed out that this universal heat conduction is only valid in the clean limit where  $\hbar\Gamma \ll \Delta_0$  [39]. Here  $\Gamma$

is the impurity scattering rate, and  $\Delta_0$  is the gap maximum. When  $\Gamma$  is large, the behavior is no longer universal, and the measured  $\kappa_0/T$  may be close to the normal-state value  $\kappa_N/T$ . For the Yb-doped  $x = 0.163$  sample, the  $T_c$  drops by 40%, and its  $\kappa_0/T$  reaches 76% of the  $\kappa_N/T$ . Therefore, the  $x = 0.163$  sample is likely already beyond the universal limit. Since its  $\kappa_N/T$  is not far from the universal value, it may be just an accident that its  $\kappa_0/T$  falls close to the universal value. Nevertheless, the universal heat conduction is at least valid up to  $x = 0.084$ .

The above discussion of universal heat conduction is based on a single  $d$ -wave gap in  $\text{CeCoIn}_5$ . However,  $\text{CeCoIn}_5$  is actually a multigap superconductor [10,12,40–42], which may make the situation slightly complex. The STM study of pure  $\text{CeCoIn}_5$  demonstrated that the superconducting gap on the  $\alpha$  band has the  $d_{x^2-y^2}$  symmetry, with the gap maximum being about 600  $\mu\text{eV}$ , while the gap on the  $\beta$  band is very small, less than their energy resolution of 75  $\mu\text{eV}$  [12]. The small gap is presumably also  $d_{x^2-y^2}$ . Since the  $T_c = 2.3$  K of pure  $\text{CeCoIn}_5$  is determined by the large gap, the “ $T_c$ ” corresponding to the small gap may be only or even smaller than 0.2 K. This means that, by extrapolating the data between 0.1 and 0.4 K, what we get is a sum of the universal  $\kappa_0/T$  of the  $\alpha$  band and nearly the normal-state  $\kappa_0/T$  of the  $\beta$  band. An even more complex situation may happen when Yb doping induces a Fermi surface topology change [19]. To further discuss the universal heat conduction in a scenario of multigaps for  $\text{CeCoIn}_5$ , more angle-resolved photoemission spectroscopy and STM measurements on doped samples are needed.

In summary, the heat transport properties of the  $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$  and  $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$  systems have been systematically studied. We observe a finite value of  $\kappa_0/T$  for  $x$  up to 0.163 and  $y$  up to 0.011. Furthermore,  $\kappa_0/T$  is universal for both systems, with a value around 1 mW K $^{-2}$  cm $^{-1}$  which agrees very well with the theoretical estimation. These results demonstrate that the nodal  $d$ -wave superconducting gap in  $\text{CeCoIn}_5$  is robust against Yb or Cd doping.

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