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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 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 $x_{nom} = 0.2$). Here we present systematic thermal conductivity measurements on $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$ ($x = 0.013, 0.084, \text{ and } 0.163$) single crystals. The observed finite residual linear term κ_0/T is insensitive to Yb doping, verifying the universal heat conduction of nodal d -wave superconducting gap in $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$. Similar universal heat conduction is also observed in $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$ system. These results reveal robust nodal d -wave gap in CeCoIn_5 upon Yb or Cd doping.

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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 non-Fermi-liquid normal state, the unconventional superconductivity in CeCoIn_5 has long been thought to be due to spin fluctuation pairing [2–5]. The superconducting gap structure $\Delta(k)$ of CeCoIn_5 was widely studied by various experimental probes ever since it was discovered. These include specific heat [6], thermal conductivity [7–10], as well as 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 CeCoIn_5 .

The investigation of impurity effects can give a better understanding of the exotic normal and superconducting state of CeCoIn_5 [14, 15]. Recently, the anomalous phenomena observed in $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$ 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 Kondo-coherence temperature T_{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_{coh} with Yb doping [17]. To solve this discrepancy, Jang *et al.* carefully determined the actual Yb concentration x_{act} in $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$ single crystals, and found that x_{act} is only about 1/3 of the nominal value x_{nom} up to $x_{nom} \approx 0.5$ [18]. With x_{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 $x_{nom} = 0.2$ are still very puzzling, including Fermi surface topology change [19], Yb valence transition [20], significant quasiparticle effective mass reduction as well as 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 $x_{nom} = 0.2$ [22]. To explain it, 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 $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$ 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 κ_0/T in zero field, one may judge whether there exist gap nodes or not. The field dependence of κ_0/T can give further information about nodal gaps, gap anisotropy, or multiple gaps.

In this paper, we report a systematic heat transport study of $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$ ($x_{nom} = 0.05, 0.2, \text{ and } 0.4$) single crystals. Finite κ_0/T is observed in all three samples, which does not support a fully-gapped superconducting state at $x_{nom} \geq 0.2$. Furthermore, κ_0/T manifests a nearly constant value upon doping, *i.e.*, the universal heat conduction, which is an important property

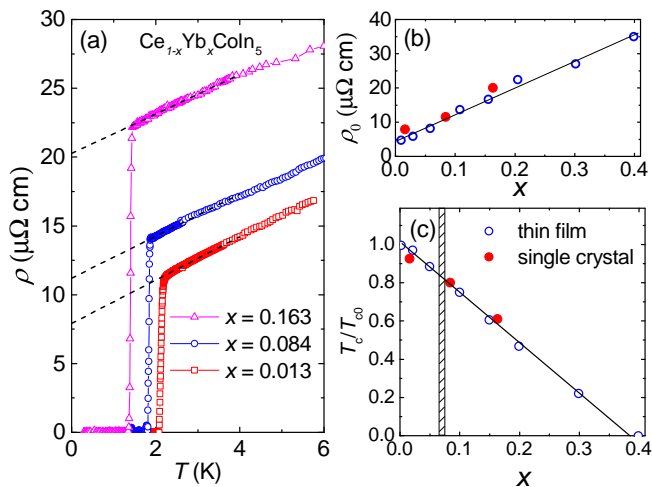


FIG. 1: (Color online). (a) Low-temperature resistivity of $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$ single crystals. Here x is the actual Yb concentration. The dashed lines are linear extrapolation to get the residual resistivity ρ_0 . (b) and (c) Doping dependence of ρ_0 and normalized T_c/T_{c0} ($T_{c0} = 2.3$ K for pure CeCoIn_5) for our single crystals and the thin films (from Ref. [17]). The shadowed narrow bar marks $x \approx 0.07$, corresponding to $x_{nom} \approx 0.2$, where several anomalies were reported, including the Yb valence transition.

of a nodal d -wave superconducting gap. Similar universal heat conduction is also observed in $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$ ($y_{nom} = 0.05, 0.075, \text{ and } 0.1$) system. These results demonstrate that the nodal d -wave gap in CeCoIn_5 is robust against Yb or Cd doping.

High-quality $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$ ($x_{nom} = 0.05, 0.2, \text{ and } 0.4$) and $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$ ($y_{nom} = 0.05, 0.075, \text{ and } 0.1$) 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 Yb concentration $x_{act} = 0.013, 0.084, \text{ and } 0.163$, and the actual Cd concentration $y_{act} = 0.004, 0.008, \text{ and } 0.011$ were determined by wavelength-dispersive spectroscopy (WDS), utilizing an electron probe microanalyzer (Shimadzu EPMA-1720H). Hereafter, x and y represent x_{act} and y_{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 ^3He cryostat. The thermal conductivity was measured in a dilution refrigerator, using a standard four-wire steady-state method with two RuO_2 chip thermometers, calibrated *in situ* against a reference 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 resis-

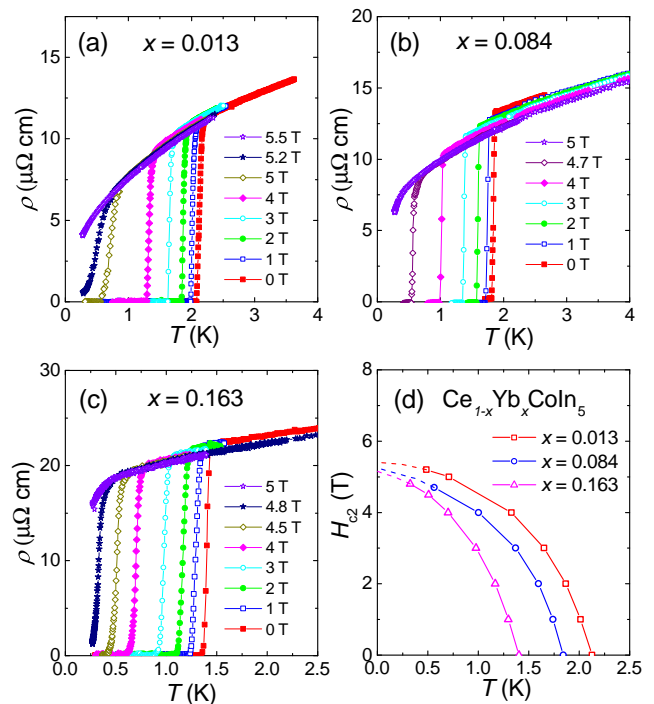


FIG. 2: (Color online). (a)-(c) Temperature dependence of resistivity for $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$ single crystals at various magnetic fields $H \parallel c$ up to 5.5 T. (d) Temperature dependence of the upper critical field $H_{c2}(T)$ for all three samples.

tivity $\rho(T)$ of $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$. For $x = 0.013, 0.084, \text{ and } 0.163$, the normal-state resistivity $\rho(T)$ below 4 K is roughly linear, and their residual resistivity $\rho_0 = 7.9, 11.2, \text{ and } 20.2 \mu\Omega \text{ cm}$ 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. Figure 1(b) and 1(c) plot the doping dependence of ρ_0 and normalized T_c/T_{c0} ($T_{c0} = 2.3$ K for pure 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 AG pair breaking curve, suggesting that Yb ions act as impurity centers with unitary scattering, regardless of the Yb valence.

Figure 2(a)-(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 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, $H_{c2}(0) \approx 5.4, 5.2, \text{ and } 5.1$ T are obtained for $x = 0.013, 0.084, \text{ and } 0.163$, respectively. $H_{c2}(0)$ only exhibits a slight decrease

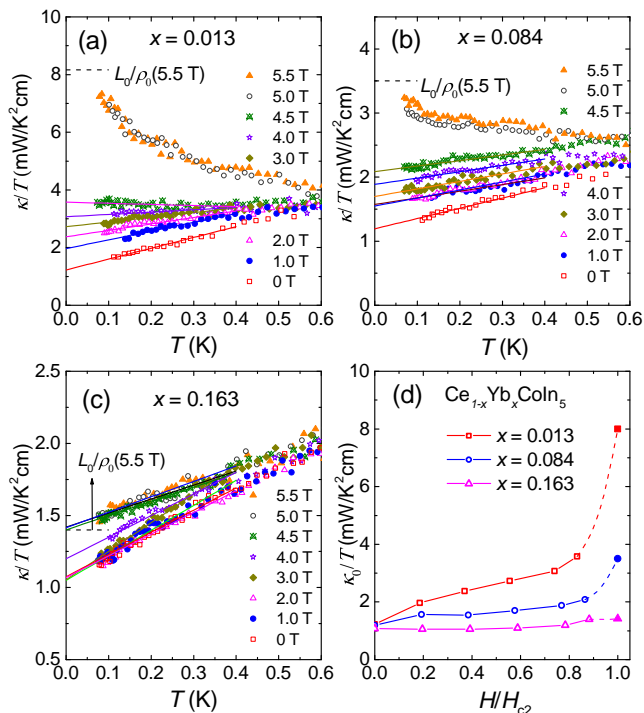


FIG. 3: (Color online). (a)-(c) Temperature dependence of the thermal conductivity divided by temperature $\kappa(T)/T$ of $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$ under various magnetic fields. The solid lines are linear fits to extrapolate the residual linear term. The dashed lines indicate the normal-state 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 κ_0/T . The field is normalized by the H_{c2} of the three samples, respectively.

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 Fig. 3(a)-(c). In zero field, all the curves are roughly linear below 0.4 K, with a moderate slope. Previously for pure 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 κ/T to zero temperature. $\kappa_0/T < 2 \text{ mW K}^{-2} \text{ cm}^{-1}$ and $< 3 \text{ mW K}^{-2} \text{ cm}^{-1}$ was estimated for pure CeCoIn_5 in Refs. [8] and [10], respectively. Here, due to the moderate slope of our doped samples, we linearly extrapolate κ/T to zero temperature to obtain $\kappa_0/T = 1.22, 1.19,$ and $1.08 \text{ mW K}^{-2} \text{ cm}^{-1}$ 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 $H > 4.5 \text{ T}$, as seen in Fig. 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 normal-state Wiedemann-Franz law expectations $L_0/\rho_0(H_{c2})$. The field dependence of κ_0/T is shown in Fig. 3(d). For $x = 0.013$, κ_0/T increases gradually with

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 concentration x_{act} and y_{act} were determined from the WDS analysis. T_c is defined as the midpoint of the resistive transition.

x_{nom}	x_{act}	T_c (K)	κ_0/T ($\text{mW}/\text{K}^2\text{cm}$)
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_{nom}	y_{act}	T_c (K)	κ_0/T ($\text{mW}/\text{K}^2\text{cm}$)
0.05	0.004	2.14	1.03
0.075	0.008	2.05	0.90
0.1	0.011	1.92	0.93

field, followed by a jump to the normal-state value at H_{c2} . Such a jump of κ_0/T near H_{c2} was previously observed in pure 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 κ_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 check whether this finite κ_0/T is also present in 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 κ/T vs T at zero field for all three samples, where a linear fit is used to obtain κ_0/T . The value of κ_0/T is $1.03, 0.90,$ and $0.93 \text{ mW K}^{-2} \text{ cm}^{-1}$ for $y = 0.004, 0.008,$ and 0.011 , respectively, as listed in Table I.

The κ_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 is plotted in Fig. 4(c). Usually, the presence of a finite κ_0/T is strong evidence for a nodal superconducting gap [27]. For example, $\kappa_0/T = 1.41 \text{ mW K}^{-2} \text{ cm}^{-1}$ for the overdoped d -wave cuprate superconductor $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+\delta}$ (Tl-2201, $T_c = 15 \text{ K}$) [31], and $\kappa_0/T = 17 \text{ mW K}^{-2} \text{ cm}^{-1}$ for the p -wave superconductor Sr_2RuO_4 ($T_c = 1.5 \text{ K}$) [32]. Although there is some uncertainty in the accurate value of κ_0/T for pure CeCoIn_5 [8, 10], the significant κ_0/T observed here for all Yb and Cd doped CeCoIn_5 samples is quite reliable due to the moderate slope. Therefore, the nodal gap in $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$ 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 $x_{nom} = 0.2$, which suggests a nodeless superconducting gap [22]. The reason for this discrepancy is not clear to us.

Furthermore, in Fig. 4(c), κ_0/T manifests a nearly constant value around $1 \text{ mW K}^{-2} \text{ cm}^{-1}$, irrespective of Yb or Cd concentration, which demonstrates a univer-

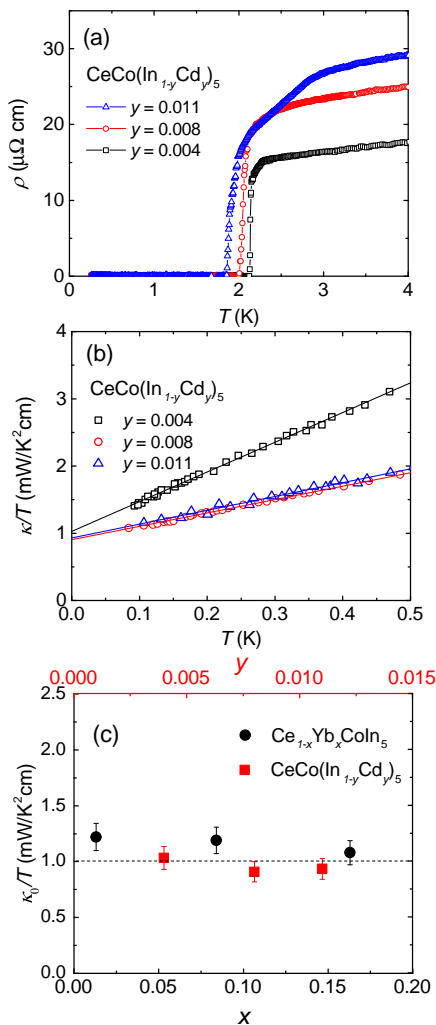


FIG. 4: (Color online). (a) Low-temperature resistivity of $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$ single crystals. Here y is the actual Cd concentration. (b) Temperature dependence of zero-field thermal conductivity divided by temperature $\kappa(T)/T$ for $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$. The solid lines are linear fits to extrapolate κ_0/T . (c) κ_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 CeCoIn_5 with 2D nodal d -wave superconducting gap [8].

sal heat conduction in Yb and Cd doped 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 γ [33, 34]. The universality results from the cancelation between two factors: (i) the density of Andreev bound states, which is proportional to γ , and (ii) the reduction of phase space for scattering of gapless excitations, which is proportional to γ^{-1} [34]. Experimentally, the universal κ_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 d -wave gap [35, 36]. For pure CeCoIn_5 , in which a 2D nodal d -wave gap was also well-established, the universal κ_0/T was theoretically estimated to be $\sim 1 \text{ mW K}^{-2} \text{ cm}^{-1}$ [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 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 $x_{nom} = 0.2$ [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 $\text{Ce}_{1-x}\text{La}_x\text{CoIn}_5$, $\kappa_0/T \approx 2 \text{ mW K}^{-2} \text{ cm}^{-1}$ was observed for $x = 0.05$ and 0.1 [9]. This value is slightly larger than the universal κ_0/T observed here in Yb and Cd-doped 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 Γ is the impurity scattering rate, and Δ_0 is the gap maximum. When Γ is large, the behavior is no longer universal, and the measured κ_0/T may be close to the normal-state value κ_N/T . For the Yb-doped $x = 0.163$ sample, the T_c drops by 40%, and its κ_0/T reaches 76% of the κ_N/T . Therefore, the $x = 0.163$ sample is likely already beyond the universal limit. Since its κ_N/T is not far from the universal value, it may be just an accident for its κ_0/T falling 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 CeCoIn_5 . However, CeCoIn_5 is actually a multigap superconductor [10, 12, 40–42], which may make the situation slightly complex. The STM study of pure CeCoIn_5 demonstrated that the superconducting gap on the α band has the $d_{x^2-y^2}$ symmetry, with the gap maximum about $600 \mu\text{eV}$, while the gap on the β 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 $T_c = 2.3 \text{ K}$ of pure 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 κ_0/T of the α band, and nearly the normal-state κ_0/T of the β band. Even more complex situation may happen when Yb doping induces Fermi surface topology change [19]. To further discuss the universal heat conduction in a scenario of multigaps for CeCoIn_5 , more angle-resolved photoemission spectroscopy (ARPES) and STM measurements on doped samples are needed.

In summary, the heat transport properties of $\text{Ce}_{1-x}\text{Yb}_x\text{CoIn}_5$ and $\text{CeCo}(\text{In}_{1-y}\text{Cd}_y)_5$ systems have

been systematically studied. We observe a finite value of κ_0/T for x up to 0.163 and y up to 0.011. Furthermore, κ_0/T is universal for both systems, with a value around $1 \text{ mW K}^{-2} \text{ cm}^{-1}$ which agrees very well with the theoretical estimation. These results demonstrate that the nodal d -wave superconducting gap in CeCoIn_5 is robust against Yb or Cd doping.

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