

Doping Dependence of Heat Transport in the Iron-Arsenide Superconductor $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$: From Isotropic to a Strongly k -Dependent Gap Structure

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The temperature and magnetic field dependence of the in-plane thermal conductivity κ of the iron-arsenide superconductor $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ was measured down to $T \approx 50$ mK and up to $H = 15$ T as a function of Co concentration x in the range $0.048 \leq x \leq 0.114$. At $H = 0$, a negligible residual linear term in κ/T as $T \rightarrow 0$ at all x shows that the superconducting gap has no nodes in the ab plane anywhere in the phase diagram. However, while the slow H dependence of $\kappa(H)$ at $T \rightarrow 0$ in the underdoped regime is consistent with a superconducting gap that is large everywhere on the Fermi surface, the rapid increase in $\kappa(H)$ observed in the overdoped regime shows that the gap acquires a deep minimum somewhere on the Fermi surface. Outside the antiferromagnetic-orthorhombic phase, the superconducting gap structure has a strongly k -dependent amplitude.

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The pairing symmetry of the order parameter in iron-arsenide superconductors remains a subject of debate as different experimental probes point to different conclusions about the gap structure [1]. For example, while the power-law dependence of the penetration depth [2] and rapid increase of thermal conductivity with magnetic field [3] in $(\text{Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$ (K-Ba122) can only be explained if the gap is very small on some portion of the Fermi surface, ARPES studies on the same material find a large gap amplitude everywhere [4,5], in the experimentally accessible portions of the Fermi surface close to $k_z = 0$. Some of the apparent contradictions may come from the fact that most studies so far have been carried out in a region of the phase diagram close to the antiferromagnetic (AFM) phase, introducing an additional level of complexity. One of the few iron-arsenide systems which at present allows a systematic study over the whole doping range, including the strongly overdoped regime, is $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ (Co-Ba122) [6,7].

Here we report measurements of the thermal conductivity κ of Co-Ba122 throughout the superconducting region. By going down to 50 mK we are able to detect any zero-energy quasiparticles that would exist if there are nodes in the gap. We find a negligible residual linear term κ_0/T in κ/T as $T \rightarrow 0$ for all doping levels studied, which is strong evidence that there are no nodes in the superconducting gap of Co-Ba122, for \mathbf{k} directions in the ab plane. However, the ease with which a magnetic field H applied along the c axis induces a finite κ_0/T varies dramatically across the phase diagram. Our results show that the superconducting gap evolves from being uniformly large everywhere on the Fermi surface at low doping to having a very

small value somewhere on the Fermi surface at high doping. This reveals a nodeless yet highly k dependent in-plane gap outside the AFM or orthorhombic phase.

Single crystals of Co-Ba122 were grown from FeAs:CoAs flux, as described elsewhere [7]. The doping level in the crystals was determined by wavelength dispersive electron probe microanalysis, which gave a Co concentration x roughly 0.7 times the flux load composition (or nominal content). We studied four compositions: $x = 0.048$ (underdoped), 0.074 (optimally doped), 0.108 (overdoped), and 0.114 (overdoped). The superconducting transition temperature (where $\rho = 0$) for each sample is $T_c = 17.1, 22.2, 14.6,$ and 10.1 K, respectively. Samples were cleaved into rectangular bars with typical size $2\text{--}3 \times 0.3 \times 0.05$ mm³. Silver wires were attached to the samples with a silver-based alloy, producing contacts with ultralow resistance (< 100 $\mu\Omega$). Thermal conductivity was measured along the [100] direction in the orthorhombic or tetragonal basal (ab) plane in a standard “one-heater-two-thermometer” technique. The magnetic field H was applied along the [001] direction (c axis). All measurements were done on warming after cooling in constant H from above T_c to ensure a homogeneous field distribution.

In Fig. 1, we show the temperature-dependent electrical resistivity $\rho(T)$ of our samples. In the $x = 0.048$ sample, the upturn below 60 K signals the onset of tetragonal-to-orthorhombic structural transition at T_S followed by the AFM order at a temperature T_M [8]. No such feature is seen in the other samples. A smooth extrapolation of $\rho(T)$ to $T = 0$ yields the residual resistivity ρ_0 , equal to 203, 62, 59, and 59 $\mu\Omega$ cm for $x = 0.048, 0.074, 0.108,$ and $0.114,$

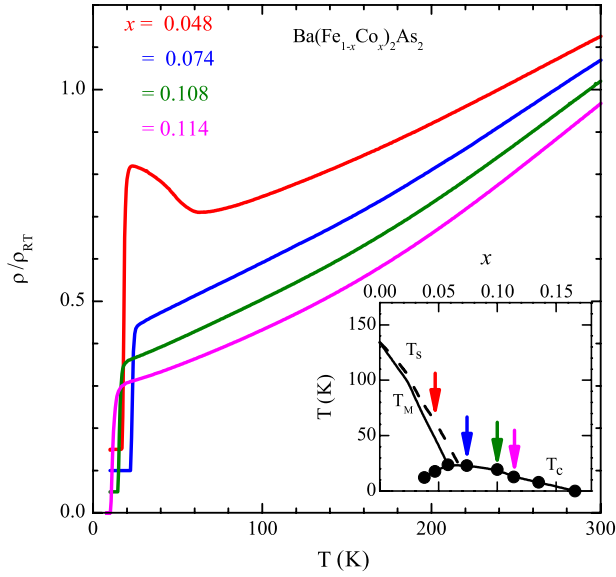


FIG. 1 (color online). Temperature dependence of the in-plane resistivity $\rho(T)$ of the four single crystals of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ studied in this work, with Co concentrations x as indicated. The curves are normalized to their room-temperature value ρ_{RT} and rigidly offset by 0.05 for clarity. Inset: phase diagram showing the orthorhombic phase below T_S , the antiferromagnetic phase below T_M , and the superconducting phase below T_c (from [7,8]), with arrows indicating the composition of the four samples.

respectively. ρ_0 is used to determine the normal-state thermal conductivity κ_N/T in the $T \rightarrow 0$ limit via the Wiedemann-Franz law, $\kappa_N/T = L_0/\rho_0$, where $L_0 = 2.45 \times 10^{-8} \text{ W } \Omega/\text{K}^2$. Because the same contacts are used for electrical and thermal measurements, there is no relative uncertainty between the measured κ and this electrically determined κ_N , other than the small uncertainty in extrapolating $\rho(T)$ to $T = 0$.

The thermal conductivity $\kappa(T)$ of the underdoped, optimally doped, and most overdoped samples is shown in the left panels of Fig. 2, for a magnetic field ranging from $H = 0$ to 15 T. The data below 0.25 K are well described by the function $\kappa/T = a + bT^\alpha$, with $\alpha \approx 1-1.5$. The first term, $a \equiv \kappa_0/T$, is the electronic residual linear term of interest here [9]. The second term is due to phonons, scattered mostly by crystal boundaries at those low temperatures [10]. We attribute the fact that this second term is much smaller in the $x = 0.048$ sample, even though all samples have comparable size, to the presence of structural domains produced by the tetragonal-to-orthorhombic transition at this doping [11]. The size of domains decreases to $\sim 1 \mu\text{m}$ close to the compositional boundary $x_S = 0.073 \pm 0.03$ [at which $T_S(x) \rightarrow 0$] [12], and reduces the phonon mean free path. The optimally doped sample $x = 0.074$ is located on the border of the orthorhombic phase and its phonon conductivity is still notably suppressed.

The magnitude of the residual linear terms extracted from the fits to the $H = 0$ data in the left panels of Fig. 2

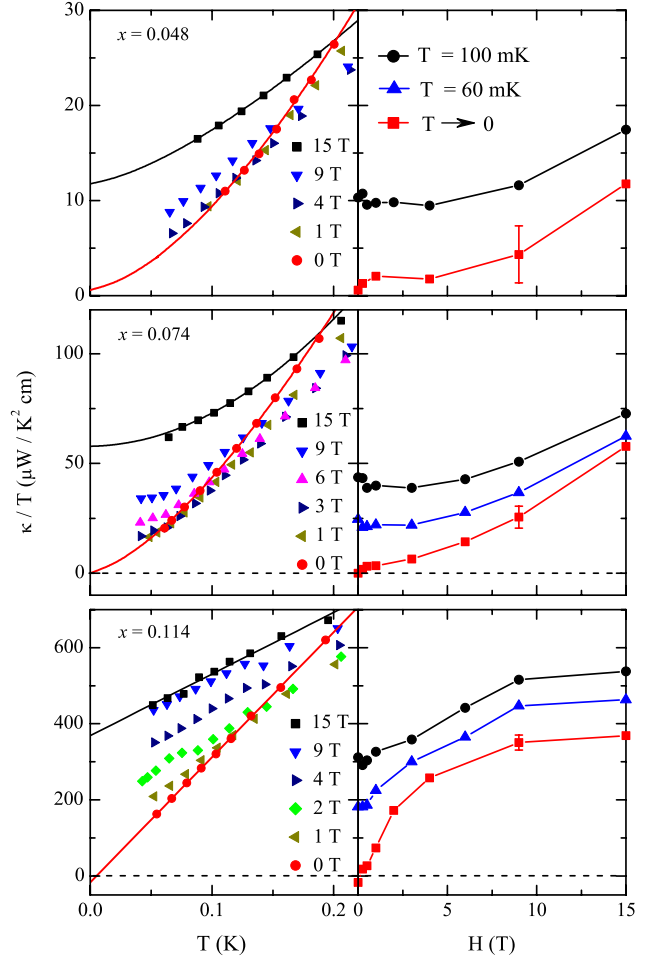


FIG. 2 (color online). Left panels: Temperature dependence of the thermal conductivity, plotted as κ/T vs T , for three Co concentrations, each measured in magnetic fields as indicated. The lines are a power-law fit of the form $\kappa/T = a + bT^\alpha$ to the $H = 0$ and $H = 15$ T data, used to extract the residual linear term $\kappa_0/T \equiv a$ in the $T \rightarrow 0$ limit (see text). Right panels: field dependence of κ/T plotted for three temperatures, as indicated. The $T \rightarrow 0$ data (red squares) are obtained from the power-law extrapolations, with a typical error bar as shown.

is extremely small, below $5 \mu\text{W}/\text{K}^2 \text{ cm}$ in all cases. This is within the absolute error bar of $\pm 5 \mu\text{W}/\text{K}^2 \text{ cm}$ on κ_0/T obtained in similar measurements on samples where no residual linear term is expected [10]. Within those error bars, all Co-Ba122 samples exhibit a negligible residual linear term, irrespective of the doping level. Similarly negligible values of κ_0/T were observed in K-Ba122 [3] and, as we argued there, this allows us to safely conclude that the superconducting gap of Co-Ba122 does not contain a line of nodes anywhere on the Fermi surface, at any doping. By comparison, in a d -wave superconductor like the cuprate $\text{Ti}_2\text{Ba}_2\text{CuO}_{6+\delta}$ (Ti-2201) with $T_c \approx 15$ K, $\kappa_0/T = 35\%$ of κ_N/T [13], while in all four Co-Ba122 samples, $\kappa_0/T < 1-2\%$ of κ_N/T . This rules out d -wave symmetry even for the overdoped samples of Co-Ba122

and further confirms that cuprates and iron arsenides have distinct superconducting states.

The possibility of point nodes in the gap of Co-Ba122 was raised in the context of penetration depth measurements which gave $\Delta\lambda \propto T^\beta$ with $\beta \approx 2$ [14]. Zero-energy quasiparticles associated with symmetry-imposed point nodes give a residual linear term in κ which grows with impurity scattering (if the gap grows linearly from the node) and for high scattering rates κ_0/T should become a substantial fraction of κ_N/T [15]. (If the gap grows quadratically from the node, κ_0/T is independent of scattering rate and comparable to the case of a line node [15].) The normal-state impurity scattering rate Γ_0 of Co-Ba122 can be estimated from ρ_0 and the plasma frequency $\omega_p = c/\lambda_0$ via $\Gamma_0 = (\omega_p^2/8\pi)\rho_0$, where λ_0 is the penetration depth at $T \rightarrow 0$ and c is the speed of light. At optimal doping, $\lambda_0 \approx 200$ nm [14], so that $\hbar\Gamma_0/k_B T_c \approx 1-2$ in the samples $x = 0.074, 0.108,$ and 0.114 . This is very substantial, and would give a large κ_0/T from point nodes, comparable to the case of a line node. We therefore conclude that symmetry-imposed point nodes in the gap of Co-Ba122 are also ruled out, unless they are located in the k_z direction (along the c axis).

In a recent study, a large κ_0/T was reported [16] in a Co-Ba122 sample with T_c and residual resistivity ratio similar to our $x = 0.074$ sample, amounting to 60% of κ_N/T , compared with less than 1%–2% in our sample. This stands in contradiction not only with our results on more than 10 crystals of Co-Ba122 at various dopings, but also with previous results on K-Ba122 [3] and Ni-doped BaFe₂As₂ (Ni-Ba122) [17], where in all cases $\kappa_0/T \approx 0$ at $H = 0$.

The dependence of κ on magnetic field $H \parallel c$ is displayed in the right panels of Fig. 2. The purely electronic component is given by the extrapolations to $T = 0$ ($T \rightarrow 0$; red squares). To confirm that the H dependence is not affected by the extrapolation procedure, we compare the extrapolated data with cuts through the actual data at two fixed temperatures, namely, 60 and 100 mK. We see that the latter cuts produce what is essentially a rigid shift of the data relative to $T \rightarrow 0$, caused by the added phonon contribution. With this validation of the $T \rightarrow 0$ data, we plot it on normalized scales in Fig. 3 as $(\kappa_0/T)/(\kappa_N/T)$ vs H/H_{c2} , where H_{c2} is the upper critical field, obtained from resistivity measurements in high fields [7,18], which give $H_{c2} \approx 45, 60, 30,$ and 20 T for $x = 0.048, 0.074, 0.108,$ and 0.114 , respectively. This plot reveals the ease with which zero-energy quasiparticles are excited by a magnetic field. At $x = 0.048$, Co-Ba122 shows the slow rise with upward curvature typical of isotropic s -wave superconductors like Nb or InBi (see Fig. 3), where the rise in κ_0/T is exponentially slow because it relies on the tunneling of quasiparticles between localized states inside adjacent vortex cores, which at low fields are far apart. By contrast, at $x = 0.114$, Co-Ba122 shows a very rapid rise with downward curvature more typical of d -wave super-

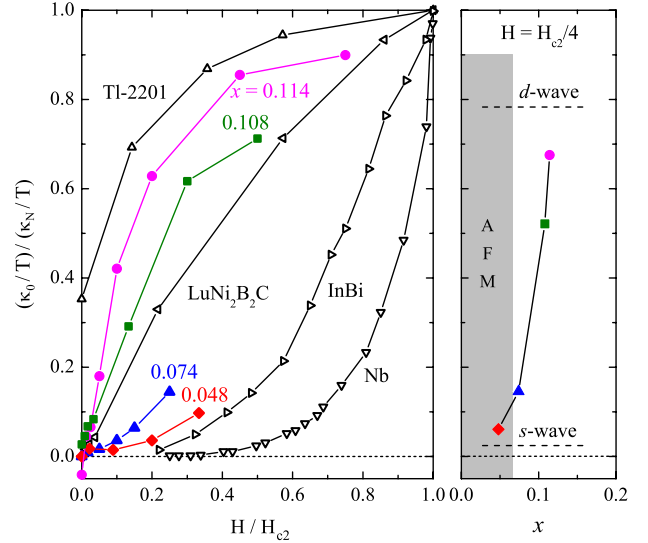


FIG. 3 (color online). Left panel: Residual linear term κ_0/T in the thermal conductivity of Co-Ba122 (full symbols) as a function of magnetic field H , plotted on normalized scales as $(\kappa_0/T)/(\kappa_N/T)$ vs H/H_{c2} , where κ_N is the normal-state thermal conductivity in the $T \rightarrow 0$ limit, obtained from the Wiedemann-Franz law (see text), and H_{c2} is the upper critical field, for the four Co concentrations x as indicated. For comparison, we also show the behavior of pure (Nb) and dirty (InBi) s -wave superconductors (from [30]), and of the d -wave superconductor Tl-2201 ($T_c = 15$ K and $H_{c2} \approx 7$ T) (from [13]). The data for the borocarbide superconductor LuNi₂B₂C ($T_c = 16$ K and $H_{c2} = 7$ T) are also shown (from [19]). Right panel: $(\kappa_0/T)/(\kappa_N/T)$ at $H/H_{c2} = 0.25$ in Co-Ba122 as a function of x . This measures the extent to which zero-energy quasiparticles are excited by a relatively small magnetic field compared to H_{c2} . The Co-Ba122 data show a degree of excitation typical of s -wave superconductors in the region of coexisting antiferromagnetic or orthorhombic order at low doping, rising rapidly to a level typical of a d -wave superconductor at higher doping.

conductors such as Tl-2201 (see Fig. 3). Even though the superconducting gap does not go to zero anywhere, as established by the fact that $\kappa_0/T \approx 0$, it must become very small somewhere on the Fermi surface to account for such a rapid rise with field. We can estimate roughly the minimum value of the gap Δ from the fact that at $x = 0.114$ the rise in κ_0/T from zero starts around $H \approx 0.5$ T (see lower right panel in Fig. 2), i.e., at $H/H_{c2} \approx 1/40$. Given that $H_{c2} \propto \Delta^2$, we obtain for the gap minimum $\Delta_{\min} \approx \Delta_0/6$, in terms of the gap maximum, Δ_0 , which controls H_{c2} .

This is similar to what has been found in the borocarbide superconductor LuNi₂B₂C ($T_c = 16$ K and $H_{c2} = 7$ T), where even though $\kappa_0/T = 0$ at $H = 0$, κ_0/T rises very rapidly at low fields [19] (see Fig. 3). There, an estimate of the gap minimum gave $\Delta_{\min} \approx \Delta_0/10$ [19]. Recent ARPES studies on the closely related material YNi₂B₂C found a sharp superconducting gap minimum at positions on the

Fermi surface which are connected by a nesting wave vector [20].

The fact that the deep minimum in the gap of Co-Ba122 is only present in compositions free of competing phases (in our case orthorhombic or AFM phases) is consistent with previous heat transport studies performed at single dopings on other arsenide superconductors. There is no AFM order in K-Ba122 at $x \approx 0.3$, where a relatively rapid increase of κ_0/T vs H was reported [3], indicative of a gap minimum. In the low- T_c nickel-arsenide superconductor BaNi_2As_2 ($T_c = 0.7$ K) [21], which orders around 132 K with an as yet unknown order parameter [22], the weak field dependence of κ is consistent with a full gap everywhere on the Fermi surface, compatible with the behavior in underdoped Co-Ba122. In $\text{Ba}(\text{Fe}_{1-x}\text{Ni}_x)_2\text{As}_2$ (Ni-Ba122), only a weak H dependence was observed near optimal doping ($T_c = 20$ K) [17]—it will be interesting to see how that evolves at higher doping. Note that in Ni-Ba122, the competing magnetic or structural phase may extend to higher electron doping [23]. In the low- T_c iron-arsenide superconductor LaFePO ($T_c = 7$ K), a line of nodes in the gap has been inferred from the linear T dependence of the penetration depth [24]. Such a line of nodes would necessarily lead to a finite κ_0/T , and indeed it does, as reported recently [25]. This extreme form of anisotropy again occurs in the absence of competing orders.

In summary, zero-field heat transport in the $T \rightarrow 0$ limit shows that the superconducting gap of Co-Ba122 does not go to zero at any point on the Fermi surface, for quasiparticle k vectors in the basal plane. This is true throughout the phase diagram, from underdoped to overdoped, and is consistent with heat transport data in both K-Ba122 [3] and Ni-Ba122 [17], measured previously at a single doping close to optimal. In Co-Ba122, the initial rise of κ_0/T with magnetic field H varies enormously with doping, from slow and typical of an isotropic s -wave gap at low doping, in the region of coexisting AFM and orthorhombic order, to very fast and comparable to a d -wave gap at high doping. This fast initial rise shows that the gap must be very small on some portion of the Fermi surface, roughly a sixth of the gap maximum. One way to understand this pronounced k dependence of a nodeless superconducting gap is in terms of an extended s -wave scenario [26–28], whereby the symmetry of the order parameter is s wave but the pairing interaction is so strongly k dependent that the gap is highly anisotropic, going to zero in certain directions. (In addition, it may also change sign, as in the s_{\pm} state [29].) Because these gap zeros or nodes are “accidental,” i.e., not imposed by symmetry (as they would be for a d -wave order parameter), the effect of impurity scattering is not to broaden them (as in a d -wave gap), but to lift them, making the gap more isotropic [27]. This could then explain why the undoped material LaFePO has a line of nodes, while the doped superconductor Co-Ba122 does not have nodes,

given that the former has a 10 times smaller impurity scattering rate (as measured by the residual resistivity ratio). In other words, as recently proposed [27], the pronounced k dependence of the gap produces nodes in the pure limit which turn into deep minima in the presence of strong impurity or disorder scattering. The fact that no pronounced anisotropy (i.e., field dependence of κ) is observed in Co-Ba122 when there is AFM order leads us to propose that the AFM order in iron arsenides reconstructs the Fermi surface by gapping those regions where the minima in the superconducting gap are located. This is reminiscent of what occurs in the borocarbide superconductors $\text{RNi}_2\text{B}_2\text{C}$ ($R = \text{Y}$ or Lu) [19], where gap minima are observed at k locations connected by a nesting wave vector [20]. This suggests an intimate interplay of magnetism and superconductivity.

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