Thermal conductivity of overdoped $BaFe_{1.73}Co_{0.27}As_2$ single crystal: Evidence for nodeless multiple superconducting gaps and interband interactions

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The in-plane thermal conductivity κ of overdoped FeAs-based superconductor $BaFe_{1.73}Co_{0.27}As_2$ (T_c =8.1 K) single crystal was measured down to 80 mK. In zero field, the residual linear term κ_0/T is negligible, suggesting a nodeless superconducting gap in the *ab* plane. In magnetic field, κ_0/T increases rapidly, very different from that of conventional *s*-wave superconductors. This anomalous $\kappa_0/T(H)$ may reveal an exotic superconducting gap structure in overdoped $BaFe_{1.73}Co_{0.27}As_2$: the vanishing hole (β) pocket has a much larger gap than the electron (γ and δ) pockets which contain most of the carriers. Such an exotic gap structure is an evidence for superconducting state induced by interband interactions, in which the band with the *smaller* density of states has a *larger* gap.

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I. INTRODUCTION

For the recently discovered FeAs-based high-T_c superconductors,[1–5] the pairing symmetry of its superconducting gap remains the most important issue to resolve. Although extensive experimental and theoretical work have been done, there is still no consensus.[6,7] While angle-resolved photoemission spectroscopy (ARPES) experiments clearly demonstrated nearly isotropic multigaps,[8–13] the Andreev spectroscopy,[14–16] NMR,[17–20] and penetration depth experiments[21–26] gave conflicting claims on whether there are nodes in the superconducting gaps.

Low-temperature thermal conductivity measurement is a powerful bulk tool to probe the superconducting gap structure.[27] The residual linear term κ_0/T is very sensitive to the existence of gap nodes and the field dependence of κ_0/T can give useful information on multigaps. Very recently, several heat transport studies have been done on this new family of FeAs-based and related superconductors. For the hole-doped $Ba_{1-x}K_xFe_2As_2$ ($T_c \simeq 30$ K) (Ref. 28) and electron-doped $BaFe_{1.9}Ni_{0.1}As_2$ (T_c=20.3 K),[29] a negligible κ_0/T was found in zero field, indicating a full superconducting gap. By contrast, a large κ_0/T was observed in $BaFe_{1.86}Co_{0.14}As_2$.[30] For the prototype $FeSe_x$ (T_c=8.8 K) superconductor, the thermal conductivity shows clear behavior of multiple nodeless superconducting gaps.[31] In two superconductors with lower T_c, κ_0/T of $BaNi_2As_2$ (T_c=0.7 K) is consistent with a dirty fully gapped superconductivity,[32] while LaFePO (T_c=7.4 K) appears to have a finite κ_0/T, suggesting the gap on some band may have nodes.[33]

For the most interested hole- and electron-doped $BaFe_2As_2$ superconductors, all samples studied by heat transport so far are near optimal doping.[28–30] It will be very interesting to study highly underdoped and overdoped samples to demonstrate its superconducting gap structure over the whole doping range. Furthermore, due to the high T_c and H_{c2} of optimally doped samples, magnetic field can only be applied up to about 30% of their H_{c2}. While for highly underdoped and overdoped samples with relatively lower T_c, one may get a complete $\kappa_0/T(H)$ behavior to see if it has the multigap character, as in $FeSe_x$.[31]

In this paper, we measure the thermal conductivity κ of a highly overdoped $BaFe_{1.73}Co_{0.27}As_2$ single crystal with T_c=8.1 K down to 80 mK. In zero field, the residual linear term κ_0/T is negligible, suggesting a nodeless superconducting gap, at least in *ab* plane. In magnetic field, $\kappa_0/T(H)$ increases sharply, very different from the $Ba_{1-x}K_xFe_2As_2$ and $BaFe_{1.9}Ni_{0.1}As_2$ samples near optimal doping. Such an anomalous $\kappa_0/T(H)$ likely results from an exotic superconducting gap structure in overdoped $BaFe_{1.73}Co_{0.27}As_2$: the vanishing hole (β) pocket has a much larger gap than the electron (γ and δ) pockets which contain most of the carriers. Our finding of this exotic gap structure supports the $s_{\pm}$-wave superconducting state induced by interband interactions in FeAs-based superconductors.

II. EXPERIMENTAL

Single crystals with nominal formula $BaFe_{1.7}Co_{0.3}As_2$ were prepared by self-flux method.[34] The diamagnetic superconducting transition was measured by a vibrating sample magnetometer based on a physical property measurement system (PPMS of Quantum Design) with the magnetic field perpendicular to the *ab* plane of the crystals. Energy-dispersive of x-ray microanalysis (Hitach S-4800) show that the actual Co content is 0.27. The sample was cleaved to a rectangular shape of dimensions 2.1×1.4 mm^2 in the *ab* plane with 25 μm thickness along the *c* axis. Contacts were made directly on the fresh sample surfaces with silver paint, which were used for both resistivity and thermal conductivity measurements. The contacts are metallic with typical resistance 150 mΩ at 1.5 K. In-plane thermal conductivity was measured in a dilution refrigerator down to 80 mK, 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*

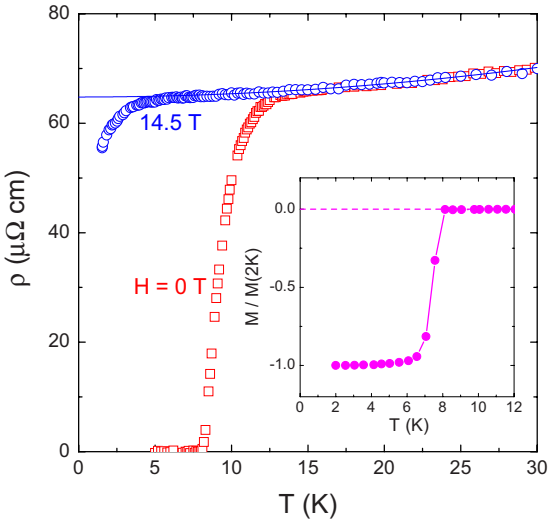

FIG. 1. (Color online) In-plane resistivity $\rho(T)$ of $BaFe_{1.73}Co_{0.27}As_2$ single crystal in $H=0$ and 14.5 T. The zero-resistance point of the resistive transition is at $T_c=8.1$ K in zero field. The solid line is a fit of the $H=14.5$ T data between 10 and 30 K to the Fermi liquid form $\rho=\rho_0+AT^2$, which gives residual resistivity $\rho_0=64.8$ $\mu\Omega$ cm. Inset: normalized magnetization which shows the diamagnetic superconducting transition.

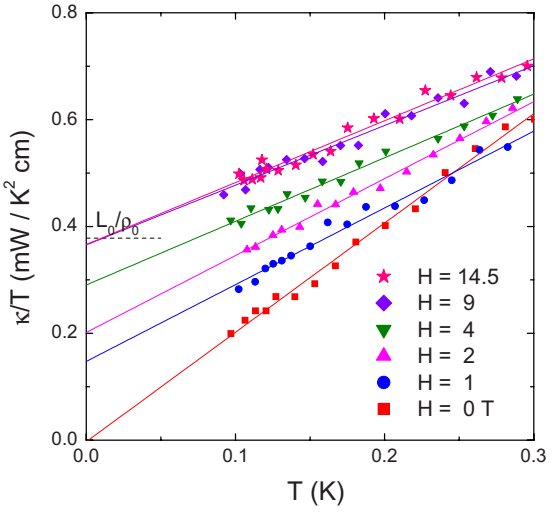

FIG. 2. (Color online) Low-temperature thermal conductivity of $BaFe_{1.73}Co_{0.27}As_2$ in magnetic fields applied along the c axis ($H=0$, 1, 2, 4, 9, and 14.5 T). The solid lines are $\kappa/T=a+bT$ fits (see text). The dashed line is the normal-state Wiedemann-Franz law expectation L_0/ρ_0 with L_0 the Lorenz number 2.45×10^{-8} W Ω K^{-2}.

axis and perpendicular to the heat current. To ensure a homogeneous field distribution in the sample, all fields were applied at temperature above T_c.

III. RESULTS AND DISCUSSION

Figure 1 shows the in-plane resistivity of our $BaFe_{1.73}Co_{0.27}As_2$ single crystal in $H=0$ and 14.5 T. The zero-resistance point of the resistive transition is at $T_c=8.1$ K in zero field, which is consistent with the diamagnetic superconducting transition shown in the inset of Fig. 1. The residual resistivity $\rho_0=64.8$ $\mu\Omega$ cm is extrapolated from the $H=14.5$ T data between 10 and 30 K by using the Fermi-liquid form $\rho=\rho_0+AT^2$.

To estimate the upper critical field $H_{c2}(0)$ which completely suppresses the resistive transition, we define $T_c(onset)$ at the temperature where $\rho(T)$ deviates from the T^2 dependence, and get $T_c(onset)=13.0$ and 5.0 K for $H=0$ and 14.5 T, respectively. Using the relationship $H_{c2}/H_{c2}(0)=1-[T/T_c(0)]^2$, $H_{c2}(0)=17.0$ T is obtained.

In Fig. 2, the temperature dependence of the in-plane thermal conductivity for $BaFe_{1.73}Co_{0.27}As_2$ in $H=0$, 1, 2, 4, 9, and 14.5 T magnetic fields are plotted as κ/T vs T. All the curves are roughly linear, therefore we fit the data to $\kappa/T=a+bT^{\alpha-1}$ (Refs. 35 and 36) with α fixed to 2. The two terms aT and bT^{α} represent electronic and phonon contributions, respectively. In the phonon term, the value of α is usually between 2 and 3, due to specular reflection of phonons at the smooth crystal surfaces in the boundary scattering limit at low temperature.[35,36] Previously, $\alpha=2.22$ and 2.02 were observed in $BaFe_2As_2$ (Ref. 37) and $BaFe_{1.9}Ni_{0.1}As_2$ (Ref. 29) single crystals, respectively. Here we only focus on the electronic term.

In zero field, the fitting gives residual linear term $\kappa_0/T=-3\pm9$ μW K^{-2} cm^{-1}. This value of κ_0/T is within the experimental error bar ±5 μW K^{-2} cm^{-1},[36] although the fitting error bar is a little high due to the slight noise of the data. Even after considering these error bars, the κ_0/T is still less than 3% of the normal-state Wiedemann-Franz law expectation $L_0/\rho_0=0.378$ mW K^{-2} cm^{-1}, with L_0 the Lorenz number 2.45×10^{-8} W Ω K^{-2}. Such a negligible κ_0/T in zero field suggests a nodeless (at least in *ab* plane) superconducting gap in overdoped $BaFe_{1.73}Co_{0.27}As_2$, which is consistent with previous results on $Ba_{1-x}K_xFe_2As_2$ (Ref. 28) and $BaFe_{1.9}Ni_{0.1}As_2$,[29] and different from $BaFe_{1.86}Co_{0.14}As_2$.[30] It has been noted that the large κ_0/T in zero field observed in $BaFe_{1.86}Co_{0.14}As_2$ by Machida *et al.*[30] may be extrinsic.[38]

In $H=9$ and 14.5 T magnetic fields, $\kappa_0/T=0.365\pm0.009$ mW K^{-2} cm^{-1} and 0.366 ± 0.009 mW K^{-2} cm^{-1} were obtained from the fittings, respectively. For both values, one gets the Lorenz ratio $L=\rho_0\kappa_0/T=0.97\pm0.03L_0$, which shows that Wiedemann-Franz law is roughly satisfied within the experimental error bar. Note that in the nonsuperconducting parent $BaFe_2As_2$ single crystal, the Wiedemann-Franz law was found to be satisfied as $T\to0$.[37]

The saturation of thermal conductivity in $H>9$ T suggests that the bulk H_{c2} has been reached at $H=9$ T, although the resistive transition is not completely suppressed until $H_{c2}=17$ T. Similar situation happened in overdoped cuprate superconductor Tl-2201 with $T_c=15$ K, in which $H_{c2}=13$ T was obtained from the resistivity data, while bulk $H_{c2}=7$ T was determined from the thermal conductivity data.[39] Here we take bulk $H_{c2}=9$ T for overdoped $BaFe_{1.73}Co_{0.27}As_2$. To choose a slightly different bulk H_{c2} does not affect our discussions below.

In Fig. 3, the normalized κ_0/T of $BaFe_{1.73}Co_{0.27}As_2$ is plotted as a function of H/H_{c2}, together with the clean s-wave superconductor Nb,[40] the dirty s-wave superconduct-

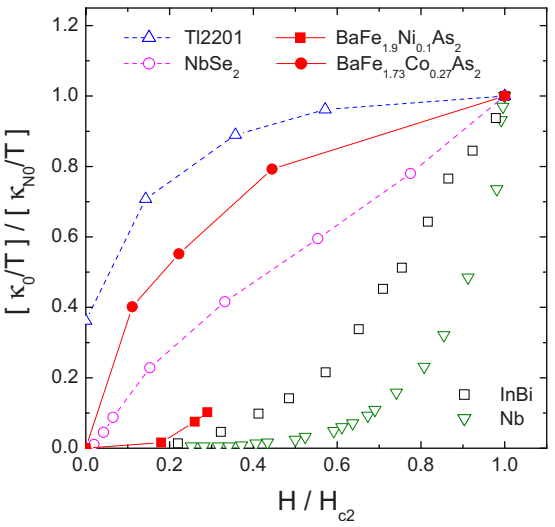

FIG. 3. (Color online) Normalized residual linear term κ_0/T of $BaFe_{1.73}Co_{0.27}As_2$ as a function of H/H_{c2}. Similar data of the clean *s*-wave superconductor Nb (Ref. 40), the dirty *s*-wave superconducting alloy InBi (Ref. 41), the multiband *s*-wave superconductor $NbSe_2$ (Ref. 42), an overdoped sample of the *d*-wave superconductor Tl-2201 (Ref. 39), and $BaFe_{1.9}Ni_{0.1}As_2$ (Ref. 29) are also shown for comparison.

ing alloy InBi,[41] the multiband *s*-wave superconductor $NbSe_2$,[42] an overdoped sample of the *d*-wave superconductor Tl-2201,[39] and $BaFe_{1.9}Ni_{0.1}As_2$.[29] As seen in Fig. 3, the rapid increase in κ_0/T at low field for overdoped $BaFe_{1.73}Co_{0.27}As_2$ is clearly different from the optimally doped $BaFe_{1.9}Ni_{0.1}As_2$. In fact, it looks more like the typical behavior of *d*-wave superconductors, due to the Volovik effect.[43] However, the negligible κ_0/T in zero field, which means nodeless superconducting gap, has excluded *d*-wave gap in $BaFe_{1.73}Co_{0.27}As_2$.

For *s*-wave superconductor $NbSe_2$, κ_0/T is zero in $H=0$, but it also increases rapidly at low field, unlike Nb and InBi. This has be explained by its multigap structure: the gap on the Γ band is approximately one third of the gap on the other two Fermi surfaces and magnetic field first suppresses the superconductivity on the Fermi surface with smaller gap [given that $H_{c2}(0) \propto \Delta_0^2$].[42] Therefore, the even sharper increase in $\kappa_0/T(H)$ in $BaFe_{1.73}Co_{0.27}As_2$ may result from an extreme case of multigap structure, in which the gap of one band is much smaller than others (e.g., 1/4 or 1/5). However, there is an apparent difference between $NbSe_2$ and $BaFe_{1.73}Co_{0.27}As_2$. For $NbSe_2$, after the smaller gap was suppressed, $\kappa_0/T(H)$ shows a slight downward curvature at high field due to the larger gap. Similar curvature of $\kappa_0/T(H)$ was found in MgB_2.[44] In Fig. 3, $\kappa_0/T(H)$ of $BaFe_{1.73}Co_{0.27}As_2$ increases so rapidly and it does not show downward curvature at high field. Such an anomalous $\kappa_0/T(H)$ has never been seen before. If it indeed results from multiple nodeless gaps, the bands with smaller gaps must contain most of the carriers so that $\kappa_0/T(H)$ can increase so rapidly all the way to high field.

To investigate this possibility, we have examined the band structure and superconducting gaps in doped $BaFe_2As_2$ system, revealed by ARPES experiments.[8,12,13,45] From the hole-doped side, in $Ba_{0.6}K_{0.4}Fe_2As_2$ ($T_c=37$ K), the average gap values $\Delta(0)$ for the two hole (α and β) pockets at the Γ point are 12.5 meV and 5.5 meV, respectively, while for the electron (γ and δ) pockets at the *M* point, the gap value is about 12.5 meV.[8,12] For electron-doped $BaFe_{1.85}Co_{0.15}As_2$ ($T_c=25.5$ K) at optimal doping, the inner hole (α) pocket disappears, and the average gap values $\Delta(0)$ of hole (β) and electron (γ and δ) pockets are 6.6 meV and 5.0 meV, respectively.[13] Such nearly isotropic multigaps with similar size has been used to explain the slow field dependence of κ_0/T up to 30% H_{c2} in $BaFe_{1.9}Ni_{0.1}As_2$,[29] as seen in Fig. 3. With further electron doping, in heavily doped nonsuperconducting $BaFe_{1.7}Co_{0.30}As_2$, the β hole pocket is absent or very small, while the two electron (γ and δ) pockets at the *M* point significantly expand.[45] This is consistent with the band-structure calculation, which shows that the β band disappears at ~26.5% electron doping.[46] Based on this trend of band-structure evolution, in our superconducting $BaFe_{1.73}Co_{0.27}As_2$ sample there should be a very small hole (β) pocket, together with two large electron (γ and δ) pockets which contain most of the carriers. To explain its anomalous $\kappa_0/T(H)$, the gap on hole (β) pocket must be much larger, 4–5 times, than the gaps on electron (γ and δ) pockets.

In the BCS theory, larger density of states (DOS) usually leads to a larger superconducting gap $\Delta(0)$. Therefore it is counterintuitive that the vanishing β pocket with much smaller DOS ends up with a much larger gap. However, in the theory of interband superconductivity,[47] this is exactly the result of the interband-only pairing, since the pairing amplitude on one band is generated by the DOS on the other.

Right after the discovery of $T_c=26$ K superconductivity in $LaFeAsO_{1-x}F_x$,[1] the importance of interband pairing interaction has been emphasized due to the multiband fermiology.[48,49] The interaction, possibly via antiferromagnetic spin fluctuations, connects the well-separated Fermi-surface pockets located around Γ and around *M*, and gives extended *s*-wave pairing symmetry whose order parameter has opposite sign on the electron and hole pockets.[48–50] Although the relative strength of interband and intraband pairing interactions is still under debate,[47–51] the experimentally observed different $2\Delta/k_BT_c$ values on different Fermi-surface pocket in $Ba_{0.6}K_{0.4}Fe_2As_2$ and $BaFe_{1.85}Co_{0.15}As_2$ prefer the interband pairing mechanism.[13]

Quantitatively, in the interband-only pairing model, the gap ratio $\Delta_2/\Delta_1=\sqrt{N_1/N_2}$, with N_1 and N_2 the Fermi-level density of states.[47] In this sense, our overdoped $BaFe_{1.73}Co_{0.27}As_2$ sample has provided the best testing ground for the theory of interband superconductivity, due to its biggest difference of DOS between the hole and electron pockets in doped $BaFe_2As_2$ superconductors so far. Indeed, the results of our current work have given strong support for the interband superconductivity in FeAs-based superconductors. It will be very interesting to directly measure the superconducting gaps in our overdoped $BaFe_{1.73}Co_{0.27}As_2$ sample with ARPES, which needs to be done at temperature below the $T_c=8.1$ K.

IV. SUMMARY

In summary, we have used low-temperature thermal conductivity to clearly demonstrate nodeless superconducting

gap in overdoped iron-arsenide superconductor $BaFe_{1.73}Co_{0.27}As_2$. Furthermore, the $\kappa_0/T(H)$ increases sharply at low field, very different from $Ba_{1-x}K_xFe_2As_2$ and $BaFe_{1.9}Ni_{0.1}As_2$ near optimal doping. It may reveal an exotic superconducting gap structure in overdoped $BaFe_{1.73}Co_{0.27}As_2$: the vanishing hole (β) pocket has a much larger gap than the electron (γ and δ) pockets, although the electron pockets have much larger density of states. Such an exotic gap structure is an evidence for the theory of interband superconductivity, thus of great importance to understand the superconducing state in FeAs-based superconductors.

Note added. During preparation of this manuscript (arXiv:0908.2209), a similar work on $BaFe_{2-x}Co_xAs_2$ was post online.[52] In Ref. 52, the results of overdoped $BaFe_{1.772}Co_{0.228}As_2$ (T_c=10.1 K) are consistent with ours, but the anomalous increase in $\kappa_0/T(H)$ at low field was explained by highly anisotropic superconducting gap with deep minima. While this debate needs to be resolved by low-temperature ARPES experiments, we note a very recent calculation of $\kappa_0/T(H)$ with unequal size of isotropic $s_{\pm}$-wave gaps has successfully fit the experimental data,[53] thus supports our interpretation.

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