Nodeless superconducting gap in electron-doped BaFe$_{1.9}$Ni$_{0.1}$As$_2$ probed by quasiparticle heat transport

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Abstract. The in-plane thermal conductivity $\kappa$ of electron-doped iron–arsenide superconductor BaFe$_{1.9}$Ni$_{0.1}$As$_2$ ($T_c = 20.3$ K) single crystal was measured down to 70 mK. In zero field, the absence of a residual linear term $\kappa_0/T$ at $T \rightarrow 0$ is strong evidence for a nodeless superconducting gap. In magnetic field, $\kappa_0/T$ shows a slow field dependence up to $H = 14.5$ T ($\approx 30\% H_{c2}$). This is consistent with the superconducting gap structure demonstrated by angle-resolved photoemission spectroscopy experiments in BaFe$_{1.85}$Co$_{0.15}$As$_2$ ($T_c = 25.5$ K), where isotropic superconducting gaps with similar size on hole and electron pockets were observed.

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The recent discovery of iron-based superconductors with $T_c$ as high as 55 K [1]–[5] has attracted great attention. As a second family of high-temperature superconductors after cuprates, the pairing symmetry of its superconducting gap is one of the most important issues to address. The spin triplet pairing was first ruled out in BaFe$_{1.8}$Co$_{0.2}$As$_2$ ($T_c = 22$ K) single crystal by the nuclear magnetic resonance (NMR) Knight shift measurements [6]. This leaves three possible singlet pairing candidates: conventional s, d and s$_\pm$, a superconducting state with order parameters of opposite signs on the electron and hole pockets [7]. While Andreev spectroscopy [8], angle-resolved photoemission spectroscopy (ARPES) [9]–[14] and latest specific heat [15] experiments on FeAs-superconductors support full superconducting gaps without nodes, NMR data [16]–[18] and extensive penetration depth studies [19]–[22] reveal a contradictory picture of either nodeless or nodal superconductivity. Even if the nodeless superconducting gap is eventually confirmed, clear-cut experiments to distinguish s$_\pm$ from conventional s-wave have to be done. Therefore, the pairing symmetry in iron–arsenide superconductors is still far from consensus.

Low-temperature thermal conductivity measurement is a powerful bulk tool to probe the superconducting gap structure [23]. For unconventional superconductors with nodes in the superconducting gap, like d-wave cuprates and p-wave ruthenate, the nodal quasiparticles will contribute a finite $\kappa_0/T$ in zero field [24, 25]. So far, only one heat transport study was reported for FeAs-based superconductors [26]. For the hole-doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ ($T_c \simeq 30$ K) single crystal, a negligible $\kappa_0/T$ was found in zero field, indicating a full superconducting gap. However, $\kappa_0/T$ increases rapidly with magnetic field even for $H \ll H_c$, which was inferred that the gap must be very small on some portion of the Fermi surface, whether from strong anisotropy or band dependence, or both. To clarify this important issue, more heat transport experiments on other FeAs-based superconductors are needed.

In this paper, we probe the superconducting gap of electron-doped BaFe$_{1.9}$Ni$_{0.1}$As$_2$ by measuring the thermal conductivity $\kappa$ of a single crystal with $T_c = 20.3$ K down to 70 mK. In zero field, the residual linear term $\kappa_0/T$ is negligible, a clear indication that BaFe$_{1.9}$Ni$_{0.1}$As$_2$ has a nodeless superconducting gap. In magnetic field, $\kappa_0/T (H)$ shows a slow field dependence, different from the case of hole-doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$. This difference is discussed on the basis of superconducting gap structure in these two systems measured by ARPES.

2. Experimental details

Single crystals with nominal formula BaFe$_{1.9}$Ni$_{0.1}$As$_2$ were prepared by the self-flux method [27]. Energy dispersive of x-ray (EDX) microanalysis shows that the actual Ni content
3. Results and discussion

Figure 1(a) shows the in-plane resistivity of our BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal in zero field. The middle point of the resistive transition is at $T_c = 20.3$ K, in good agreement with the previous study [27]. The 10–90% width of the resistive transition is less than 0.3 K, indicating the high homogeneity of our crystal. The residual resistivity $\rho_0 = 132 \, \mu\Omega\text{cm}$ is extrapolated from the data above $T_c$ by using the Fermi liquid form $\rho = \rho_0 + AT^2$. In figure 1(b), the normalized ac magnetization also shows a sharp superconducting transition similar to figure 1(a).

is 0.096, close to the nominal composition. The ac magnetization was measured in a quantum design physical property measurement system (PPMS). The sample was cleaved to a rectangular shape of dimensions $1.5 \times 0.88$ mm$^2$ in the plane, with $55 \, \mu$m 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 $50 \, \Omega$ at 1.5 K. In-plane thermal conductivity was measured in a dilution refrigerator down to 70 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-axis and perpendicular to the heat current. To ensure a homogeneous field distribution in the sample, all fields were applied at temperature above $T_c$. 
In figure 2, the temperature dependence of the in-plane thermal conductivity for BaFe$_{1.9}$Ni$_{0.1}$As$_2$ single crystal in zero field is plotted as κ/T versus T. Since both electrons and phonons contribute to the measured conductivity, we fit the data to κ/T = a + bT$^{\alpha-1}$ [28, 29], where aT and bT$^\alpha$ represent electronic and phonon contributions, respectively. For phonon scattering off the crystal boundary at low temperature, one usually gets $\alpha = 3$, but specular reflection of phonons at the smooth crystal surfaces can result in a lower power $\alpha < 3$ [28, 29]. For BaFe$_{1.9}$Ni$_{0.1}$As$_2$, it is found that the data below 0.8 K can be well fitted (the solid line in figure 2) and gives $\kappa_0/T = -3 \pm 2 \mu$W K$^{-2}$ cm$^{-1}$, with $\alpha = 2.02 \pm 0.01$.

In the non-superconducting parent BaFe$_2$As$_2$ single crystal, the Wiedemann–Franz law, which relates charge and thermal conductivities by $\kappa/T = L_0/\rho$ with $L_0$ the Lorenz number $2.45 \times 10^{-8}$ W Ω K$^{-2}$, was found to be satisfied as $T \to 0$ [30]. For BaFe$_{1.9}$Ni$_{0.1}$As$_2$ with $T_c = 20.3$ K, the normal-state Wiedemann–Franz law expectation is $\kappa_0/T = L_0/\rho_0 = 0.186$ mW K$^{-2}$ cm$^{-1}$ with $\rho_0 = 132 \mu$Ω cm, the dashed line in figure 2.

Since the residual linear term $\kappa_0/T$ is within the experimental error bar $\pm 5 \mu$W K$^{-2}$ cm$^{-1}$ [29], which is less than 3% of the normal-state value, the electronic contribution to the thermal conductivity is negligible in zero field. This is consistent with previous results on hole-doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ single crystals [26] and the low-$T_c$ superconductor BaNi$_2$As$_2$ ($T_c = 0.7$ K) [31], suggesting a nodeless (at least in the $ab$-plane) superconducting gap. However, the power $\alpha = 2.02$ of the phonon conductivity $bT^\alpha$ is much lower than $\alpha = 2.65$ found in Ba$_{1-x}$K$_x$Fe$_2$As$_2$ [26]. We note that in the parent compound BaFe$_2$As$_2$
Figure 3. Low-temperature thermal conductivity of BaFe$_{1.9}$Ni$_{0.1}$As$_2$ in magnetic fields applied along the c-axis ($H = 0, 4, 9$ and $14.5\, \text{T}$). The solid lines are $\kappa/T = a + bT^{\alpha - 1}$ fits (see text). The dashed line is the normal-state Wiedemann–Franz law expectation $L_0/\rho_0$.

ing single crystal [31], the power $\alpha = 2.22$ is closer to our value. Whether specular reflections of the phonon boundary scattering [28, 29] can give such a low $\alpha$ is not clear to us. In fact, phonons scattering off either electrons or grain boundaries give $\alpha = 2$ [32]. Therefore, more experimental results are needed to clarify the temperature dependence of phonon thermal conductivity in FeAs-compound single crystals.

Figure 3 shows the low-temperature thermal conductivity of BaFe$_{1.9}$Ni$_{0.1}$As$_2$ in magnetic fields applied along the c-axis ($H = 0, 9, 13$ and $14.5\, \text{T}$). The data of $\kappa/T$ in high fields below $0.25\, \text{K}$ manifest similar temperature dependence to the zero field data. We fit the $H = 9, 13$ and $14.5\, \text{T}$ curves by using the same equation $\kappa/T = a + bT^{\alpha - 1}$, with fixed $\alpha = 2.02$, due to the slightly increasing noise level of the in-field data. The solid lines are the fitting curves, which give $\kappa_0/T = 4, 15$ and $20\, \mu\text{W K}^{-2}\, \text{cm}^{-1}$ for $H = 9, 13$ and $14.5\, \text{T}$, respectively.

The upper critical field $H_{c2}$ of BaFe$_{1.9}$Ni$_{0.1}$As$_2$ ($T_c = 20.3\, \text{K}$) single crystal has not been determined yet. For BaFe$_{1.8}$Co$_{0.2}$As$_2$ ($T_c = 22\, \text{K}$) single crystal, the $H_{c2}$ was estimated $\sim 50\, \text{T}$ [33]. Taking this value as the $H_{c2}$ of our BaFe$_{1.9}$Ni$_{0.1}$As$_2$ sample, $H = 14.5\, \text{T}$ is just about $30\%$ of $H_{c2}$.

In figure 4, the normalized $\kappa_0/T$ of BaFe$_{1.9}$Ni$_{0.1}$As$_2$ is plotted as a function of $H/H_{c2}$, together with the clean s-wave superconductor Nb [34], the dirty s-wave superconducting alloy InBi [35], the multi-band s-wave superconductor NbSe$_2$ [36], an overdoped sample of the d-wave superconductor Tl-2201 [24] and Ba$_{0.78}$K$_{0.22}$Fe$_2$As$_2$ [26]. For a clean (like Nb) or dirty (like InBi) type-II s-wave superconductor with isotropic gap, $\kappa_0/T$ should grow exponentially with field (above $H_{c1}$). This usually gives negligible $\kappa_0/T$ for field lower than $H_{c2}/4$, as seen in figure 4. For NbSe$_2$, $\kappa_0/T$ increases much rapid at low field. This can be explained by its multi-gap structure, whereby the gap on the $\Gamma$ band is approximately one-third of the gap on the
other two Fermi surfaces, and the magnetic field will first suppress the superconductivity on the Fermi surface with smaller gap (given that $H_c(0) \propto \Delta_0^2$) [36].

As seen in figure 4, the $\kappa_0/T(H)$ of BaFe$_{1.9}$Ni$_{0.1}$As$_2$ more likely follows the behavior of the isotropic s-wave gap. This field dependence is different from that of the hole-doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ ($T_c \simeq 30$ K) sample [26], where $\kappa_0/T$ increases almost linearly with $H$ up to 15 T. Such a rapid increase of $\kappa_0/T(H)$ in Ba$_{1-x}$K$_x$Fe$_2$As$_2$ has been interpreted as evidence for a $k$-dependent gap magnitude, coming from angle (i.e. anisotropic) or band (i.e. isotropic but with different magnetitudes on different bands) dependence or both [26].

In order to explain this difference, let us examine the gap values on all Fermi surface sheets for both hole- and electron-doped BaFe$_2$As$_2$ measured by ARPES [9, 13, 14]. For hole-doped Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ ($T_c = 37$ K), the average gap values $\Delta(0)$ for the two hole pockets ($\alpha$ and $\beta$) are 12.5 and 5.5 meV, respectively, while for the electron ($\gamma$ and $\delta$) pockets, the gap value is about 12.5 meV [9, 13]. For electron-doped BaFe$_{1.85}$Co$_{0.15}$As$_2$ ($T_c = 25.5$ K), the average gap values $\Delta(0)$ of hole ($\beta$) and electron ($\gamma$ and $\delta$) pockets are 6.6 and 5.0 meV, respectively [14].

Since the doping level and $T_c$ of our BaFe$_{1.9}$Ni$_{0.1}$As$_2$ sample are close to those of BaFe$_{1.85}$Co$_{0.15}$As$_2$, their superconducting gap structure should also be similar. Therefore, due to the similar sizes (6.6 versus 5.0 meV) of these isotropic superconducting gaps, the $\kappa_0/T(H)$ of our BaFe$_{1.9}$Ni$_{0.1}$As$_2$ sample behaves more like a conventional single-gap s-wave superconductor. For hole-doped Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$, the sizes of these gaps are quite different (12.5 versus 5.5 meV), which gives a ratio $R = 12.5/5.5 = 2.3$ [9, 13]. Taking this ratio for the slightly underdoped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ [26], it is smaller than that in NbSe$_2$ ($R \approx 3$). This may explain the nearly linear increase of $\kappa_0/T(H)$ in Ba$_{1-x}$K$_x$Fe$_2$As$_2$ with the slope smaller than...
that in NbSe$_2$ [26], given $H_{c2}(0) \propto \Delta_0^2$ and magnetic field first suppresses the superconductivity on the Fermi surface with smallest superconducting gap.

4. Conclusion

In summary, we have used low-temperature thermal conductivity to clearly demonstrate the nodeless superconducting gap in electron-doped iron–arsenide superconductor BaFe$_{1.9}$Ni$_{0.1}$As$_2$. Furthermore, the $k_0/T(H)$ shows a slow $H$ dependence at low field, different from the rapid, linear $k_0/T(H)$ in hole-doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$. This difference can be explained by the different ratio of the band-dependent superconducting gaps. Our results are consistent with nodeless multi-gaps in iron–arsenide superconductors, as revealed by ARPES experiments.

Note: After our present work first appeared on arXiv (0906.0138), two similar works on BaFe$_{2-x}$Co$_x$As$_2$ have also been put on arXiv [37, 38]. In [37], a large $k_0/T$ was observed in BaFe$_{1.86}$Co$_{0.14}$As$_2$ single crystal at zero field, apparently contradicting our results. In [38], the results of BaFe$_{2-x}$Co$_x$As$_2$ with $x = 0.148$ are consistent with ours.

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