Multi-gap nodeless superconductivity in iron selenide FeSe$_x$ : evident from quasiparticle heat transport

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The in-plane thermal conductivity $\kappa$ of the iron selenide superconductor FeSe$_x$ ($T_c = 8.8$ K) were measured down to 120 mK and up to 14.5 T ($\gtrsim 3/4 H_{c2}$). In zero field, the residual linear term $\kappa_0/T$ at $T \rightarrow 0$ is only about 16 $\mu$W K$^{-2}$ cm$^{-1}$, less than 4% of its normal state value. Such a small $\kappa_0/T$ does not support the existence of nodes in the superconducting gap. More importantly, the field dependence of $\kappa_0/T$ in FeSe$_x$ is very similar to that in NbSe$_2$, a typical multi-gap s-wave superconductor. We consider our data as strong evidence for multi-gap nodeless superconductivity in FeSe$_x$. This kind of superconducting gap structure may be generic for all Fe-based superconductors.

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Just as CuO-plane is the basic building block of high-$T_c$ cuprate superconductors, the FeAs-layer is the basic structure of the newly discovered FeAs-based high-$T_c$ superconductors.$^{1-5,6,7}$ The FeAs-layer consists of a Fe square planar sheet tetrahedrally coordinated by As. However, unlike the rigid CuO-plane in cuprates, partial substitution of Fe by Co or Ni, or As by P within the FeAs-layer can effectively induce superconductivity.$^{8,9,10,11}$ In this sense, the discovery of superconductivity in binary FeSe$_x$ ($T_c \approx 8$ K) is of great interests, since it only contains the superconducting FeAs-layer which has identical structure as FeAs-layer, and the Se deficiency may cause the superconductivity.$^{11}$

More remarkably, the onset $T_c$ can be enhanced to as high as 37 K for FeSe$_x$ under high pressure.$^{14,15,16}$ which further implies that superconductivity in FeSe$_x$ may have the same mechanism as in FeAs-based superconductors.

For this new family of high-$T_c$ superconductors, the pairing symmetry of its superconducting gap is a key to understand the mechanism of superconductivity. Extensive experimental and theoretical work have been done to address this important issue for FeAs-based superconductors (for a theoretical review, see Ref. 17; for an experimental review, see Ref. 18). Although there is still no consensus, more and more evidences point to multi-gap nodeless superconductivity, possibly an unconventional $s^\pm$-paring mediated by antiferromagnetic fluctuations.$^{20}$ For the prototype FeSe$_x$ superconductor, however, there were very few experiments to study the superconducting gap structure. This is due to its relatively lower $T_c$ and lack of sizable high-quality single crystals.$^{20,21}$ The measurements of in-plane magnetic penetration depth for polycrystal FeSe$_{0.85}$ are in favor of anisotropic s-wave superconducting gap or two gaps ($s + s$)$^{22}$ To clarify this important issue, more experimental work are needed for FeSe$_x$ superconductor.

Low-temperature thermal conductivity measurement is a powerful tool to study the superconducting gap structure.$^{22}$ The field dependence of the residual thermal conductivity $\kappa_0/T$ for BaNi$_2$As$_2$ ($T_c = 0.7$ K) is consistent with a dirty fully gapped superconductivity.$^{23}$ For Ba$_{1-x}$K$_x$Fe$_2$As$_2$ ($T_c \simeq 30$ K) and BaFe$_{1.9}$Ni$_{0.1}$As$_2$ ($T_c = 20.3$ K), a negligible $\kappa_0/T$ was found in zero field, indicating a full superconducting gap.$^{23,24}$ However, $\kappa(T)$ was only measured in magnetic fields up to $H = 15$ T ($\simeq 1/4 H_{c2}$), thus cannot show clearly whether the superconductivity has multi-gap character in FeAs-based superconductors.$^{25,26}$

In this paper, we measure the thermal conductivity $\kappa$ of a FeSe$_x$ single crystal with $T_c = 8.8$ K down to 120 mK and up to 14.5 T ($\simeq 3/4 H_{c2}$) to probe its superconducting gap structure. In zero field, $\kappa_0/T$ is about 16 $\mu$W / K$^2$ cm, less than 4% of its normal-state value. Such a small $\kappa_0/T$ should not come from the nodal quasiparticle contribution. It may simply come from the slight overestimation when doing extrapolation, due to the lack of lower temperature data. The field-dependence of $\kappa_0/T$ is very similar to that in multi-gap s-wave superconductor NbSe$_2$. Based on our data, it is evident that FeSe$_x$ is a multi-gap nodeless superconductor.

FeSe$_x$ single crystals with nominal formula FeSe$_{0.82}$ were grown via a vapor self-transport method.$^{21}$ The ab-plane dimensions of as-grown crystals ranges from a few hundred $\mu$m to 1 mm. Energy Dispersive of X-ray (EDX) microanalysis (Hitachi S-4800) show that the actual Fe:Se ratio is very close to 1:1 in our FeSe$_x$ single crystals. The nominal formula FeSe$_{0.82}$ was used in the initial work by Hsu et al.$^{21}$ However, the actual superconducting phase was later determined to be FeSe$_{0.99\pm0.02}$ in Ref. 27 and FeSe$_{0.974\pm0.005}$ in Ref. 28. Therefore the EDX result of our FeSe$_x$ single crystals is consistent with these two later reports.

The ac magnetization was measured in a Quantum Design Physical Property Measurement System (PPMS). An as-grown single crystal with dimensions $1.0 \times 0.40$ mm$^2$ in the plane and 190 $\mu$m thickness along the c-axis was selected for transport study. Contacts were made directly on the sample surfaces with silver paint, which
completely suppresses the resistive transition, we define

\[ \rho(T) \sim T^a \]

single crystals. To ensure a homogeneous field distribution in the sample, all fields were applied at perpendicular to the heat current. To estimate the upper critical field \( H_c(0) \) at the temperature where \( \rho(T) \) deviates from the linear dependence, and get \( T_c(\text{onset}) = 11.9 \) and 6.3 K for \( H = 0 \) and 14.5 T, respectively. Using the relationship \( H_{c2}/H_{c2}(0) = 1 - (T_c/T_c(0))^2 \), we get \( H_{c2}(0) = 20.1 \) T. Note that \( H_{c2}(0) = 16.3 \) T was estimated for the powder sample, in which \( T_c \) was defined at the middle point of the transition.\(^{13}\)

In Fig. 1b, the normalized ac magnetization for FeSe\(_x\) single crystal is plotted. The positive ferromagnetic background has been attributed to the existence of Fe impurity in the FeSe\(_x\) powder sample.\(^{13}\) However, no iron, iron oxide, or iron silicide impurities were detected in our crystals,\(^{21}\) therefore the ferromagnetic background likely results from the magnetic Fe cluster promoted by Se vacancies.\(^{20}\)

Fig. 2 shows the temperature dependence of the in-plane thermal conductivity for FeSe\(_x\) in zero field. To extrapolate the residual linear term \( \kappa_0/T \), we fit the data to \( \kappa/T = a + bT^{\alpha-1} \), where \( aT \) and \( bT^{\alpha} \) represent electronic and phonon contributions, respectively. In Fig. 2, the data from 120 mK to 0.7 K can be fitted (the solid line) and gives \( \kappa_0/T = 16 \pm 2 \mu W K^{-2} cm^{-1} \), with \( \alpha = 2.47 \).

Such a value of \( \kappa_0/T \) is slightly larger than the experimental error bar \( \pm 5 \mu W K^{-2} cm^{-1} \).\(^{21}\) However, it is still fairly small, less than 4% the normal state Wiedemann-Franz law expectation \( \kappa_{\text{NQ}}/T = L_0/\rho_0 = 0.423 \) mW K\(^{-2}\) cm\(^{-1}\), with \( L_0 \) the Lorenz number \( 2.45 \times 10^{-8} \) W K\(^{-2}\) and \( \rho_0 = 57.9 \mu \Omega \) cm. For unconventional superconductors with nodes in the superconducting gap, a substantial \( \kappa_0/T \) in zero field contributed by the nodal quasiparticles has been found.\(^{32,33}\) For example, for overdoped \( d\)-wave cuprate superconductor Tl2201 with \( T_c = 15 \) K, \( \kappa_0/T = 1.41 \) mW K\(^{-2}\) cm\(^{-1}\), about 36\% \( \kappa_{\text{NQ}}/T \). For \( p\)-wave superconductor Sr\(_2\)RuO\(_4\) with \( T_c = 1.5 \) K, \( \kappa_0/T \) was used for both resistivity and thermal conductivity measurements. The typical contact resistance is a few ohms at room temperature and 1.5 K, which is not as good as that on Ba\(_{1-x}\)K\(_x\)Fe\(_2\)As\(_2\) and BaFe\(_{1.9}\)Ni\(_{0.1}\)As\(_2\) single crystals.\(^{22,23}\) In-plane thermal conductivity was measured in a dilution refrigerator using a standard one-heater-two-thermometer steady-state technique. Due to the small size of the sample and the non-ideal contacts, good thermalization between sample and the two RuO\(_2\) thermometers can only be achieved down to 120 mK. 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 \( T_c \).

Fig. 1a shows the in-plane resistivity of FeSe\(_x\) single crystal in \( H = 0 \) and 14.5 T magnetic fields. The middle point of the resistive transition is at \( T_c = 8.8 \) K in zero field. The 10-90% transition width of our crystal is as broad as the powder sample,\(^{13}\) which has been noticed in Ref. 21. Above \( T_c \), \( \rho(T) \) manifests a very good linear dependence up to 80 K, similar to the powder sample.\(^{13}\) A linear fit of \( \rho(T) \) gives the residual resistivity \( \rho_0 = 57.9 \) \( \mu \Omega \) cm in \( H = 14.5 \) T, which is about 1/4 the value of powder sample.\(^{13}\)

To estimate the upper critical field \( H_{c2}(0) \) which completely suppresses the resistive transition, we define \( T_c(\text{onset}) \) at the temperature where \( \rho(T) \) deviates from
$= 17 \text{ mW K}^{-2} \text{ cm}^{-1}$, more than 9% $\kappa_{N0}/T$ for the best sample.\textsuperscript{23} We also note that $\kappa_0/T$ in zero field are all negligible in closely related superconductors BaNi$_2$As$_2$, Ba$_{1-x}$K$_x$Fe$_2$As$_2$, and BaFe$_{1.9}$Ni$_{0.1}$As$_2$.\textsuperscript{24,25,26} Therefore, it is unlikely that $\kappa_0/T = 16 \pm 2 \mu\text{W K}^{-2} \text{ cm}^{-1}$ in FeSe single crystal comes from the nodal quasiparticles. Since no impurity phases were detected, such a small $\kappa_0/T$ may simply come from the slight overestimation when doing extrapolation, due to the lack of experimental data below 120 mK.

Below we turn to the field dependence of $\kappa_0/T$ in FeSe. Fig. 3 shows the low-temperature thermal conductivity of FeSe in magnetic fields applied along the c-axis ($H = 0, 1, 4, 9, \text{ and } 14.5 \text{ T}$). For $H = 1 \text{ T}$, the data is also fitted to $\kappa/T = a + bT^{-\alpha-1}$, and gives $\kappa_0/T = 47 \pm 2 \mu\text{W K}^{-2} \text{ cm}^{-1}$, with $\alpha = 2.47$. For $H = 4, 9, \text{ and } 14.5 \text{ T}$, the electronic contribution becomes more and more dominant and the data get less smooth, therefore $\alpha$ is fixed to 2.47. The dashed line is the normal state Wiedemann-Franz law expectation at $T \to 0$, namely $L_0/\rho_0$, with $L_0$ the Lorenz number $2.45 \times 10^{-8} \text{ W} \Omega \text{ K}^{-2}$.

In Fig. 4, we put the normalized $\kappa_0/T(H)$ of FeSe together with the clean s-wave superconductor Nb\textsuperscript{34} the dirty s-wave superconducting alloy InBi\textsuperscript{35} the multi-band s-wave superconductor NbSe\textsubscript{36} and an overdoped sample of the d-wave superconductor Ti-2201\textsuperscript{32} plotted as a function of $H/H_{c2}$. 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 $H_{c3}/4$. For the d-wave superconductor Ti-2201, $\kappa_0/T$ increases roughly proportional to $\sqrt{H}$ at low field due to the Volovik effect.\textsuperscript{23} By contrast, for multi-gap superconductors NbSe\textsubscript{2} and MgB\textsubscript{2} magnetic field will first suppress the superconductivity on the Fermi surface with smaller gap, and give distinct shape of $\kappa_0/T(H)$ curve, as seen in Fig. 4.

From Fig. 4, the $\kappa_0/T(H)$ of FeSe\textsubscript{x} manifests almost identical behavior as that of multi-gap s-wave superconductor NbSe\textsubscript{2}. For NbSe\textsubscript{2}, the shape of $\kappa_0/T(H)$ has been quantitatively explained by multiband superconductivity, whereby the gap on the $\Gamma$ band is approximately one third of the gap on the other two Fermi surfaces.\textsuperscript{36} Therefore, we consider our data as strong evidence for multi-gap nodeless superconductivity in FeSe\textsubscript{x}. Note that in the two-gap s + s-wave model to describe the in-plane penetration depth data, the magnitude of the two gaps are 1.60 and 0.38 meV, respectively.\textsuperscript{22} The ratio between these two gaps is about 4, close to that in NbSe\textsubscript{2}, thus supports the multi-gap scenario from our thermal conductivity results.

So far, there is still no experiment to directly measure the superconducting gap in Fe$_{1+y}$Te$_{1-x}$Se\textsubscript{x} system. Density functional calculations show that the electronic band structure of FeS, FeSe, and FeTe are very similar to the FeAs-based superconductors.\textsuperscript{39} In doped BaFe$_2$As$_2$, multi-gap nodeless superconductivity has been clearly demonstrated by angle-resolved photoemission spectroscopy (ARPES) experiments.\textsuperscript{40,41,42} 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
δ) pockets, the gap value is similar, about 12.5 meV.\textsuperscript{41,44} For electron-doped BaFe\textsubscript{1−x}Co\textsubscript{x}As\textsubscript{2} (T\textsubscript{c} = 25.5 K), the average gap values Δ(0) of hole (β) and electron (γ and δ) pockets are 6.6 and 5.0 meV, respectively.\textsuperscript{42} The ratio between the large and small gaps is 2.3 for Ba\textsubscript{0.6}K\textsubscript{0.4}Fe\textsubscript{2}As\textsubscript{2}. This may explain the rapid increase of δ0/T(H) at low field in Ba\textsubscript{1−x}K\textsubscript{x}Fe\textsubscript{2}As\textsubscript{2} although magnetic field was only applied up to 1/4 H\textsubscript{c2} thus could not see clear multi-gap character as in our FeSe\textsubscript{2} single crystal.

In summary, we have measured the low-temperature thermal conductivity of iron selenide superconductor FeSe\textsubscript{2} to investigate its superconducting gap structure. A fairly small δ0/T at zero field and the dramatic field dependence of δ0/T give strong evidence for multi-gap nodeless superconductivity in FeSe\textsubscript{2}. Such a gap structure may be generic for all Fe-based superconductors. More experiments are needed to distinguish conventional s-wave from the unconventional s\textsuperscript{±}-wave superconductivity in this new family of high-\textit{Tc} superconductors.

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