Multigap nodeless superconductivity in $FeSe_x$: Evidence from quasiparticle heat transport

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The in-plane thermal conductivity κ of the iron selenide superconductor $FeSe_x$ (T_c=8.8 K) was measured down to 120 mK and up to 14.5 T ($\simeq 3/4H_{c_2}$). In zero field, the residual linear term κ_0/T at $T\rightarrow 0$ is only about 16 μW K^{-2} cm^{-1}, less than 4% of its normal-state value. Such a small κ_0/T does not support the existence of nodes in the superconducting gap. More importantly, the field dependence of κ_0/T in $FeSe_x$ is very similar to that in $NbSe_2$, a typical multigap *s*-wave superconductor. We consider our data as strong evidence for multigap nodeless (at least in *ab* plane) superconductivity in $FeSe_x$. This kind of superconducting gap structure may be generic for all Fe-based superconductors.

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I. INTRODUCTION

Just as CuO plane is the basic building block of high-T_c cuprate superconductors, FeAs layer is the basic structure of the newly discovered FeAs-based high-T_c superconductors.[1–7] 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.[8–12] In this sense, the discovery of superconductivity in binary $FeSe_x$ ($T_c\simeq 8$ K) is of great interests since it only contains the superconducting FeSe layer which has identical structure as FeAs layer, and the Se deficiency may cause the superconductivity.[13] More remarkably, the onset T_c can be enhanced to as high as 37 K for $FeSe_x$ under high pressure,[14–16] 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 multigap nodeless superconductivity, possibly an unconventional $s^{\pm}$ paring mediated by antiferromagnetic fluctuations.[19] 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.[23] The field dependence of the residual thermal conductivity κ_0/T for $BaNi_2As_2$ (T_c=0.7 K) is consistent with a dirty fully gapped superconductivity.[24] For $Ba_{1-x}K_xFe_2As_2$ ($T_c\simeq 30$ K) and $BaFe_{1.9}Ni_{0.1}As_2$ (T_c=20.3 K), a negligible κ_0/T was found in zero field, indicating a full superconducting gap.[25,26] However, $\kappa(T)$ was only measured in magnetic fields up to $\sim H_{c_2}/4$ (H=15 T), thus, cannot show clearly whether the superconductivity has multigap character in FeAs-based superconductors.[25,26]

In this paper, we measure the in-plane thermal conductivity κ of a $FeSe_x$ single crystal with T_c=8.8 K down to 120 mK and up to 14.5 T ($\simeq 3/4H_{c_2}$) to probe its superconducting gap structure. In zero field, κ_0/T is about 16 μW K^{-2} cm^{-1}, less than 4% of its normal-state value. Such a small κ_0/T should not come from the nodal quasiparticle contribution. It may simply result from the slight overestimation when doing extrapolation, due to the lack of lower temperature data. The field dependence of κ_0/T is very similar to that in multigap *s*-wave superconductor $NbSe_2$. Based on our data, it is evident that $FeSe_x$ is a multigap nodeless (at least in *ab* plane) superconductor.

II. EXPERIMENT

$FeSe_x$ 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 μm 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.*[13] However, the actual superconducting phase was later determined to be $FeSe_{0.99\pm 0.02}$ in Ref. 27 and $FeSe_{0.974\pm 0.005}$ in Ref. 28. Therefore the EDX result of our $FeSe_x$ single crystals is consistent with these two later reports.

The ac susceptibility was measured in a Quantum Design Physical Property Measurement System (PPMS) with a modulation field of 10 Oe at 10 kHz. An as-grown single crystal with dimensions 1.0×0.40 mm^2 in the plane and 190 μm thickness along the *c* axis was selected for transport study. Contacts were made directly on the sample surfaces with silver paint, which were used for both resistivity and thermal-conductivity measurements. The resistivity was measured by the standard four-probe method, using a low-

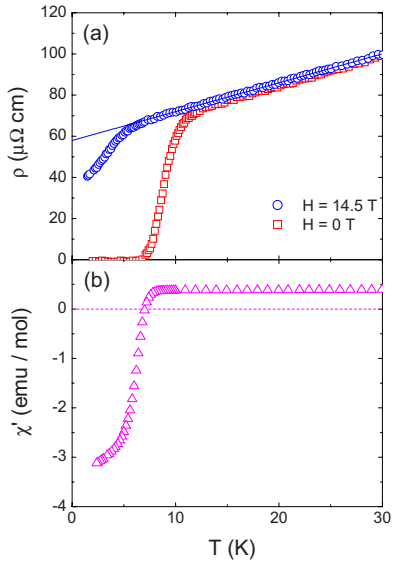

FIG. 1. (Color online) (a) In-plane resistivity $\rho(T)$ of $FeSe_x$ single crystal in $H=0$ and 14.5 T magnetic fields along the c axis. The solid line is a linear fit of $\rho(T)$ from 8 to 30 K, which gives the residual resistivity $\rho_0=57.9\ \mu\Omega$ cm in $H=14.5$ T. (b) The real part of ac susceptibility.

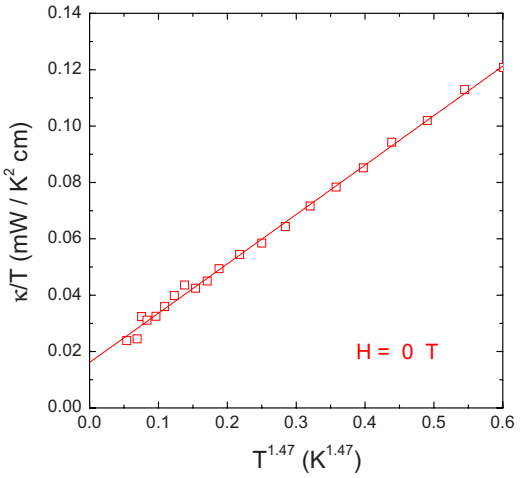

FIG. 2. (Color online) Temperature dependence of the in-plane thermal conductivity for $FeSe_x$ single crystal in zero field. The solid line represents a fit of the data to $\kappa/T=a+bT^{\alpha-1}$. This gives the residual linear term $\kappa_0/T=16\pm2\ \mu$W K^{-2} cm^{-1}.

frequency ac resistance bridge (Lakeshore 370) with an excitation current $I=1$ mA. The typical contact resistance is a few ohms at room temperature and 1.5 K, which is not as good as that on $Ba_{1-x}K_xFe_2As_2$ and $BaFe_{1.9}Ni_{0.1}As_2$ single crystals.[25,26] In-plane thermal conductivity was measured in a dilution refrigerator using a standard one-heater-two-thermometer steady-state technique.[29] Due to the small size of the sample and the nonideal 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.

III. RESULTS AND DISCUSSION

Figure 1(a) 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 $H_{c_2}(0)$ which completely suppresses the resistive transition, we define T_c(onset) at the temperature where $\rho(T)$ deviates from the linear dependence, and get T_c(onset)=11.9 and 6.3 K for $H=0$ and 14.5 T, respectively. Using the relationship $H_{c_2}/H_{c_2}(0)=1-[T/T_c(0)]^2$, we get $H_{c_2}(0)=20.1$ T. Note that $H_{c_2}(0)=16.3$ T was estimated for the powder sample, in which T_c was defined at the middle point of the transition.[13]

In Fig. 1(b), the real part of ac susceptibility 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.[30]

Figure 2 shows the temperature dependence of the in-plane thermal conductivity for $FeSe_x$ in zero field. To extrapolate the residual linear term κ_0/T, we fit the data to $\kappa/T=a+bT^{\alpha-1}$,[29,31] where aT and bT^{α} 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 $\kappa_0/T=16\pm2\ \mu$W K^{-2} cm^{-1}, with $\alpha=2.47$.

Such a value of κ_0/T is slightly larger than the experimental error bar $\pm5\ \mu$W K^{-2} cm^{-1}.[31] However, it is still fairly small, less than 4% of the normal-state Wiedemann-Franz law expectation $\kappa_{N0}/T=L_0/\rho_0=0.423$ mW K^{-2} cm^{-1}, with $L_0=2.45\times10^{-8}$ W Ω K^{-2} and $\rho_0=57.9\ \mu\Omega$ cm. For unconventional superconductors with nodes in the superconducting gap, a substantial κ_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% κ_{N0}/T.[32] For p-wave superconductor Sr_2RuO_4 with $T_c=1.5$ K, $\kappa_0/T=17$ mW K^{-2} cm^{-1}, more than 9% κ_{N0}/T for the best sample.[33] We also note that κ_0/T in zero field are all negligible in closely related superconductors $BaNi_2As_2$, $Ba_{1-x}K_xFe_2As_2$, and $BaFe_{1.9}Ni_{0.1}As_2$.[24–26] Therefore, it is unlikely that $\kappa_0/T=16\pm2\ \mu$W K^{-2} cm^{-1} in $FeSe_x$ single crystal comes from the nodal quasiparticle contribution. Since no impurity phases were detected, such a small κ_0/T may simply result 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 κ_0/T in $FeSe_x$. Figure 3 shows the low-temperature thermal conductivity of $FeSe_x$ in magnetic fields applied along the c axis ($H=0$, 1, 4,

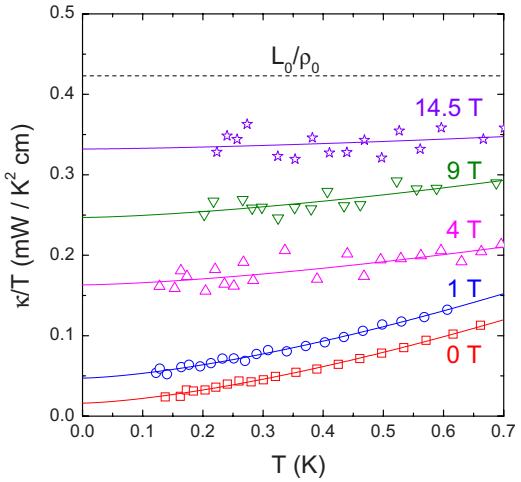

FIG. 3. (Color online) Low-temperature thermal conductivity of $FeSe_x$ in magnetic fields applied along the c axis (H=0, 1, 4, 9, and 14.5 T). The solid lines are $\kappa/T=a+bT^{\alpha-1}$ fits. For H=4, 9, and 14.5 T, the electronic contribution becomes more and more dominant and the data get less smooth, therefore α is fixed to 2.47. The dashed line is the normal-state Wiedemann-Franz law expectation at $T\rightarrow 0$, namely, L_0/ρ_0, with L_0 the Lorenz number 2.45×10^{-8} W Ω K^{-2}.

9, and 14.5 T). For H=1 T, the data is also fitted to $\kappa/T=a+bT^{\alpha-1}$, and gives $\kappa_0/T=47\pm 2$ μW K^{-2} cm^{-1}, with α=2.47. For H=4, 9, and 14.5 T, the electronic contribution becomes more and more dominant and the data get less smooth, therefore α is fixed to 2.47 in the phonon term bT^{α}. From Fig. 3, even higher magnetic field is needed to increase κ/T to its normal-state value.

In Fig. 4, we put the normalized $\kappa_0/T(H)$ of $FeSe_x$ together with the clean s-wave superconductor Nb,[34] the dirty s-wave superconducting alloy InBi,[35] the multiband s-wave superconductor $NbSe_2$,[36] and an overdoped sample of the

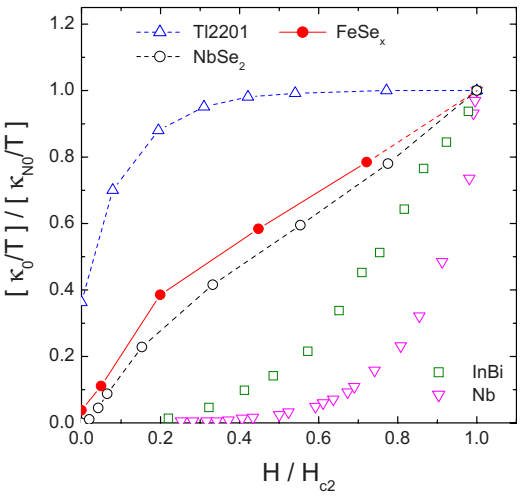

FIG. 4. (Color online) Normalized residual linear term κ_0/T of $FeSe_x$ plotted as a function of H/H_{c_2}. Similar data of the clean s-wave superconductor Nb (Ref. 34), the dirty s-wave superconducting alloy InBi (Ref. 35), the multiband s-wave superconductor $NbSe_2$ (Ref. 36), and an overdoped sample of the d-wave superconductor Tl-2201 (Ref. 32) are also shown for comparison.

d-wave superconductor Tl-2201,[32] plotted as a function of H/H_{c_2}. For a clean (like Nb) or dirty (like InBi) type-II s-wave superconductor with isotropic gap, κ_0/T should grow exponentially with field (above H_{c_1}). This usually gives negligible κ_0/T for field lower than $H_{c_2}/4$. For the d-wave superconductor Tl-2201, κ_0/T increases roughly proportional to $\sqrt{H}$ at low field due to the Volovik effect.[37] By contrast, for multigap superconductors $NbSe_2$ and MgB_2,[36,38] 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 $FeSe_x$ manifests almost identical behavior as that of multigap s-wave superconductor $NbSe_2$. For $NbSe_2$, the shape of $\kappa_0/T(H)$ has been quantitatively explained by multiband superconductivity, whereby the gap on the Γ band is approximately one third of the gap on the other two Fermi surfaces.[36] Therefore, we consider our data as strong evidence for multigap nodeless superconductivity in $FeSe_x$, with the ratio of the large gap to small gap close to 3. 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.[22] The ratio of these two gaps is about 4, in agreement with the value estimated from our thermal-conductivity results. Since point nodes along c axis do not contribute to in-plane transport, measurement along c axis has to be done to rule out this possibility in $FeSe_x$.

So far, there is still no experiment to directly measure the superconducting gap in $Fe_{1+y}Te_{1-x}Se_x$ system. Density functional calculations show that the electronic band structure of FeS, FeSe, and FeTe are very similar to the FeAs-based superconductors.[39] In doped $BaFe_2As_2$, multigap nodeless superconductivity has been clearly demonstrated by angle-resolved photoemission spectroscopy (ARPES) experiments.[40–42] For hole-doped $Ba_{0.6}K_{0.4}Fe_2As_2$ (T_c=37 K), the average gap values Δ(0) for the two hole pockets (α and β) are 12.5 and 5.5 meV, respectively, while for the electron (γ and δ) pockets, the gap value is similar, about 12.5 meV.[40,41] For electron-doped $BaFe_{1.85}Co_{0.15}As_2$ (T_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.[42] The ratio of the large gap to small gap is 2.3 for $Ba_{0.6}K_{0.4}Fe_2As_2$. This may explain the linear increase of $\kappa_0/T(H)$ at low field in $Ba_{1-x}K_xFe_2As_2$, with a smaller slope than that in $NbSe_2$.[25] However, magnetic field was only applied up to $H_{c_2}/4$ for $Ba_{1-x}K_xFe_2As_2$ and the multigap character of $\kappa_0/T(H)$ was not as clear as in $NbSe_2$ and our $FeSe_x$ single crystal.

IV. SUMMARY

In summary, we have measured the low-temperature thermal conductivity of iron selenide superconductor $FeSe_x$ to investigate its superconducting gap structure. A fairly small κ_0/T at zero field and the dramatic field dependence of κ_0/T give strong evidence for multigap nodeless (at least in ab plane) superconductivity in $FeSe_x$. Such a gap structure may be generic for all Fe-based superconductors. More experiments are needed to distinguish unconventional $s^{\pm}$ wave

from the conventional *s*-wave superconductivity in this new family of high-T_c superconductors.

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