Nodal gap in iron-based superconductor $CsFe_2As_2$ probed by quasiparticle heat transport

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The thermal conductivity of iron-based superconductor $CsFe_2As_2$ single crystal ($T_c = 1.81$ K) was measured down to 50 mK. A significant residual linear term $\kappa_0/T = 1.27$ mW K^{-2} cm^{-1} is observed in zero magnetic field, which is about 1/10 of the normal-state value in upper critical field H_{c2}. In low magnetic field, κ_0/T increases rapidly with field. The normalized $\kappa_0/T(H)$ curve for our $CsFe_2As_2$ (with residual resistivity $\rho_0 = 1.80$ $\mu\Omega$ cm) lies between the dirty KFe_2As_2 (with $\rho_0 = 3.32$ $\mu\Omega$ cm) and the clean KFe_2As_2 (with $\rho_0 = 0.21$ $\mu\Omega$ cm), which is consistent with its impurity level. These results strongly suggest a nodal superconducting gap in $CsFe_2As_2$, similar to its sister compound KFe_2As_2.

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I. INTRODUCTION

For the iron-based superconductors,[1,2] one very important issue is the symmetry and structure of their superconducting gap,[3] which is crucial for understanding the mechanism of high-temperature superconductivity.[4] However, after 5 years of extensive studies, it is still a complex situation, mainly due to their multiple electronic bands.[5,6]

Most of the iron-based superconductors have both hole and electron Fermi surfaces, for example, $Ba_{0.6}K_{0.4}Fe_2As_2$.[6] While many of these superconductors show nodeless superconducting gaps, such as optimally doped $BaFe_2As_2$,[6–10] LiFeAs,[11–14] $NaFe_{1-x}Co_xAs$,[15,16] and $FeTe_{1-x}Se_x$,[17,18] some of them manifest nodal superconducting gap, such as $BaFe_2(As_{1-x}P_x)_2$,[19–21] $Ba(Fe_{1-x}Ru_x)_2As_2$,[22] LiFeP,[23] and LaFePO.[24,25] So far, it is not conclusive that the nodeless gaps on different Fermi surfaces are $s_\pm$ wave.[3] The nodal gap in those isovalently P- or Ru-substituted compounds could be accidental nodal s wave on some Fermi surface,[21] but its origin is still not very clear.[23]

More intriguingly, while nodeless superconducting gaps were observed in the extremely electron-doped $A_xFe_{2-y}Se_2$ (A = K, Rb, Cs, ...) with only electron pockets,[26–28] a nodal superconducting gap was found in the extremely hole-doped KFe_2As_2 with only hole pockets.[29,30] It is now under hot debate whether the superconducting gap in KFe_2As_2 is d wave or accidental nodal s wave.[31–33] Thermal conductivity measurements gave compelling evidence for a d-wave gap,[31,32] but recent low-temperature angle-resolved photoemission spectroscopy (ARPES) measurements showed octet-line node structure, suggesting accidental nodal s-wave gap.[33]

To clarify this situation in KFe_2As_2, it will be helpful to investigate the superconducting gap structure of its two sister compounds $RbFe_2As_2$ and $CsFe_2As_2$, both with $T_c = 2.6$ K from the measurements of polycrystalline samples.[34,35] Unexpectedly, recent muon-spin spectroscopy measurements of $RbFe_2As_2$ polycrystal claimed that the temperature dependence of the superfluid density n_s is best described by a two-gap s-wave model,[36,37] which is quite different from KFe_2As_2. In this context, more experiments are highly desired, especially on the single crystals of $RbFe_2As_2$ and $CsFe_2As_2$.

In this paper we present the thermal conductivity measurements of $CsFe_2As_2$ single crystals down to 50 mK. We find clear evidence for superconducting gap nodes from the significant residual linear term κ_0/T in zero field and the field dependence of κ_0/T. Our results suggest common nodal gap structure in $CsFe_2As_2$ and KFe_2As_2.

II. EXPERIMENT

The $CsFe_2As_2$ single crystals were grown by self-flux method.[38] The sample was cleaved to a rectangular shape of dimensions 3.5 × 1.0 mm^2 in the ab plane, with 30 μm thickness along the c axis. Contacts were made directly on the sample surfaces with silver paint, which were used for both resistivity and thermal conductivity measurements. To avoid degradation, the sample was exposed in air less than 2 h. The contacts are metallic with typical resistance 100 mΩ at 2 K. In-plane thermal conductivity was measured in a dilution refrigerator, 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.

III. RESULTS AND DISCUSSION

Figure 1(a) shows the in-plane resistivity $\rho(T)$ of a $CsFe_2As_2$ single crystal. The shape of the $\rho(T)$ curve mimics that of KFe_2As_2 single crystal.[29,31] From the inset of Fig. 1(a), the T_c defined by $\rho = 0$ is 1.81 K. For the $CsFe_2As_2$ polycrystal in Ref. 34, the T_c defined by the sharp drop of susceptibility is about 2.2 K.[34] The origin of the 0.4 K difference between the T_c of $CsFe_2As_2$ single crystal and polycrystal is not clear. One possible reason is that the polycrystal in Ref. 34 is purer than our single crystal since it has been shown that for KFe_2As_2 the purer sample has higher T_c.[31] In any case, the T_c of (K, Rb, Cs)Fe_2As_2 series (3.8, 2.6, and 1.8–2.2 K, respectively) seems to decrease with the increase of the ionic radius of alkali metal.

In Fig. 1(b) the low-temperature resistivity is plotted as ρ vs $T^{1.5}$. It is found that ρ obeys $T^{1.5}$ dependence nicely above

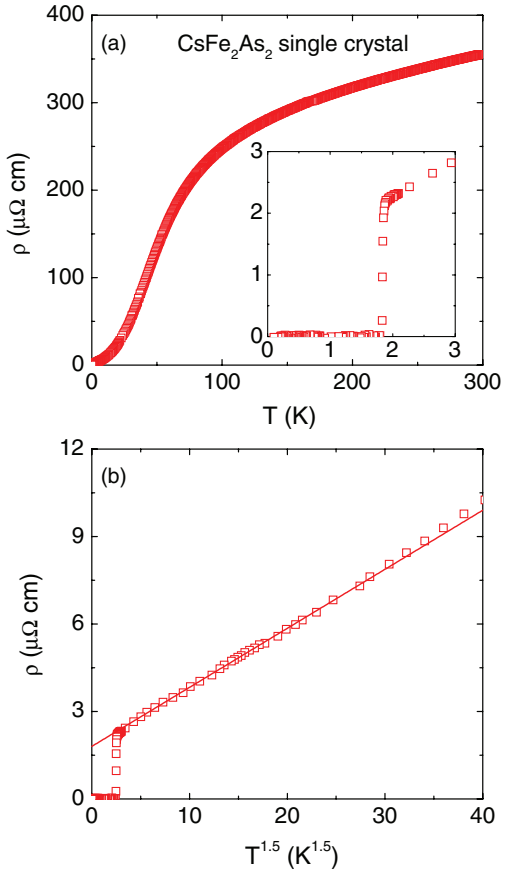

FIG. 1. (Color online) (a) In-plane resistivity of $CsFe_2As_2$ single crystal. The inset shows the resistive superconducting transition with $T_c = 1.81$ K defined by $\rho = 0$. (b) Low-temperature resistivity plotted as ρ vs $T^{1.5}$. The solid line is a fit of the data between 2.6 and 9 K to $\rho = \rho_0 + AT^{1.5}$, which gives residual resistivity $\rho_0 = 1.80\ \mu\Omega$ cm.

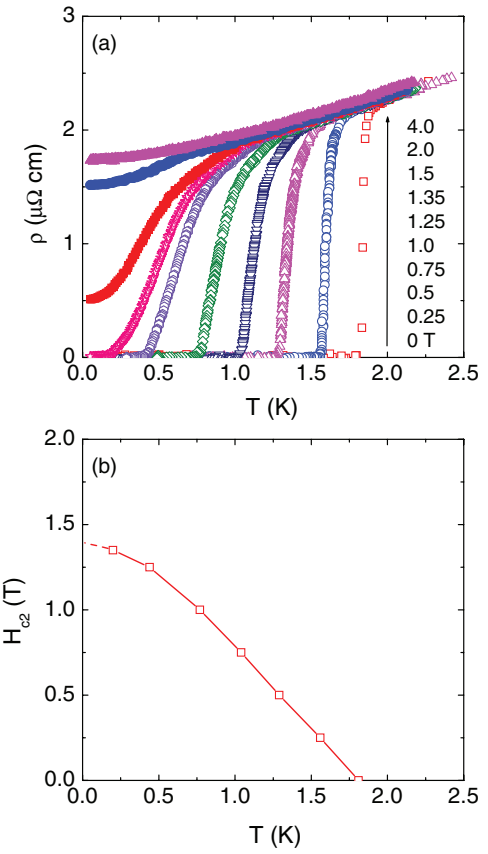

FIG. 2. (Color online) (a) Low-temperature resistivity of $CsFe_2As_2$ single crystal in magnetic field up to 4 T. (b) Temperature dependence of the upper critical field $H_{c2}(T)$, defined by $\rho = 0$. The dashed line is a guide to the eye, which points to $H_{c2}(0) \approx 1.4$ T.

T_c, up to about 9 K. The fit of the data between 2.6 and 9 K gives residual resistivity $\rho_0 = 1.80\ \mu\Omega$ cm, thus the residual resistivity ratio (RRR) $= \rho(300\text{ K})/\rho_0 \approx 200$ is obtained. For the dirty KFe_2As_2 single crystal with $\rho_0 = 3.32\ \mu\Omega$ cm and RRR ≈ 110, $\rho \sim T^{1.5}$ has already been noticed.[29] For the clean KFe_2As_2 single crystal with $\rho_0 = 0.21\ \mu\Omega$ cm and RRR ≈ 1180, $\rho \sim T^{1.8}$ was found.[31] Such a non-Fermi-liquid behavior of $\rho(T)$ in KFe_2As_2 and $CsFe_2As_2$ may result from the antiferromagnetic spin fluctuations.[39] In $BaFe_2(As_{1-x}P_x)_2$, the non-Fermi-liquid linear behavior of $\rho(T)$ near optimal doping, and the increase of power n in the overdoped regime have been considered as the signature of a quantum critical point.[40]

In order to estimate the upper critical field $H_{c2}(0)$ of $CsFe_2As_2$, the resistivity was also measured in magnetic fields up to $H = 4$ T, as shown in Fig. 2(a). Figure 2(b) plots the temperature dependence of $H_{c2}(T)$, defined by $\rho = 0$. This definition usually corresponds to the bulk H_{c2}. From Fig. 2(b) we estimate $H_{c2}(0) \approx 1.4$ T. To choose a slightly different H_{c2} does not affect our discussion on the field dependence of κ_0/T below.

The ultra-low-temperature heat transport measurement is a bulk technique to probe the gap structure of superconductors.[41] In Fig. 3(a) we present the temperature dependence of in-plane thermal conductivity for $CsFe_2As_2$ single crystal in zero and applied magnetic fields, plotted as κ/T vs T. All the curves are roughly linear, as previously observed in dirty KFe_2As_2,[29] $BaFe_{1.9}Ni_{0.1}As_2$,[9] and $Ba(Fe_{1-x}Ru_x)_2As_2$ single crystals.[22] Therefore we fit all the curves to $\kappa/T = a + bT^{\alpha-1}$ with α fixed to 2. The two terms aT and bT^{α} represent contributions from electrons and phonons, respectively. Here we only focus on the electronic term.

For $CsFe_2As_2$ in zero field, the fitting gives $\kappa_0/T = a = 1.27 \pm 0.04$ mW K^{-2} cm^{-1}. This value is about 1/10 of the normal-state Wiedemann-Franz law expectation $\kappa_{N0}/T = L_0/\rho_0 = 13.6$ mW K^{-2} cm^{-1}, with L_0 the Lorenz number 2.45×10^{-8} W Ω K^{-2} and normal-state $\rho_0 = 1.80\ \mu\Omega$ cm. For a high-quality superconductor with no impure phase, such a significant κ_0/T in zero field is usually contributed by nodal quasiparticles, thus considered as strong evidence for nodes in the superconducting gap.[41] For example, $\kappa_0/T = 1.41$ mW K^{-2} cm^{-1} for the overdoped cuprate $Tl_2Ba_2CuO_{6+\delta}$ (Tl-2201), a d-wave superconductor with $T_c = 15$ K.[42] Previously, $\kappa_0/T = 2.27$ and 3.6 mW K^{-2} cm^{-1} were observed for dirty and clean KFe_2As_2, respectively.[29,31]

For the clean KFe_2As_2, an additional large electronic term $\kappa/T \sim T^2$ was also observed.[31] Reid *et al.* considered this term as the leading-order finite-temperature correction to κ/T for d-wave superconductor, which will be rapidly suppressed by impurity scattering and magnetic field.[31] From Fig. 3(a),

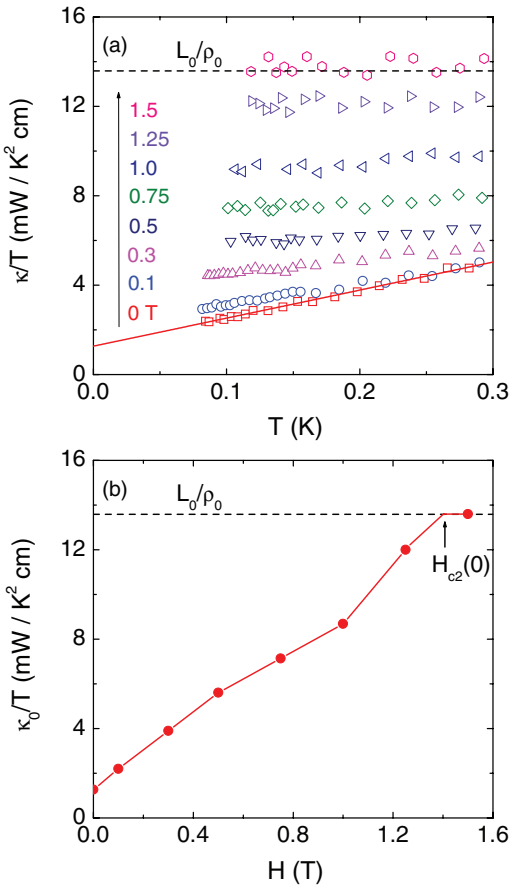

FIG. 3. (Color online) (a) Low-temperature in-plane thermal conductivity of $CsFe_2As_2$ single crystal in zero and magnetic fields applied along the c axis. The solid line is a fit of the zero-field data to $\kappa/T = a + bT$, which gives a residual linear term $\kappa_0/T = 1.27$ mW K^{-2} cm^{-1}. The dashed lines are 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} and $\rho_0 = 1.80\ \mu\Omega$ cm. (b) Field dependence of κ_0/T. In $H = 1.5$ T, slightly above $H_{c2}(0) = 1.4$ T, the Wiedemann-Franz law $\kappa_0/T = L_0/\rho_0$ is satisfied.

such an electronic term is absent in our $CsFe_2As_2$ single crystal, which is not very clean.

The field dependence of κ_0/T can provide further support for the gap nodes.[41] For a nodal superconductor, κ_0/T increases rapidly in low field due to the Volovik effect,[43] as in Tl-2201.[42] In contrast, for a single-gap s-wave superconductor, κ_0/T displays a very slow field dependence at low field, as in Nb.[44] In Fig. 3(b) we plot the field dependence of κ_0/T for $CsFe_2As_2$. At low field, κ_0/T indeed increases rapidly. Then it shows slight downward curvature before reaching the upper critical field $H_{c2}(0)$. In $H = 1.5$ T slightly above $H_{c2}(0) = 1.4$ T, the fitting gives $\kappa_0/T = 13.6 \pm 0.3$ mW K^{-2} cm^{-1}, satisfying the Wiedemann-Franz law perfectly.

For comparison, the normalized $(\kappa_0/T)/(\kappa_{N0}/T)$ of $CsFe_2As_2$ is plotted as a function of H/H_{c2} in Fig. 4, together with Nb,[44] Tl-2201,[42] the dirty and clean KFe_2As_2.[29,31] Clearly the curve of $CsFe_2As_2$ lies between the dirty and clean KFe_2As_2. The dirty KFe_2As_2 shows similar field dependence of κ_0/T to that of Tl-2201,[29] which should also be dirty, with $\rho_0 = 5.6\ \mu\Omega$ cm and RRR ≈ 30.[42] For the clean KFe_2As_2,

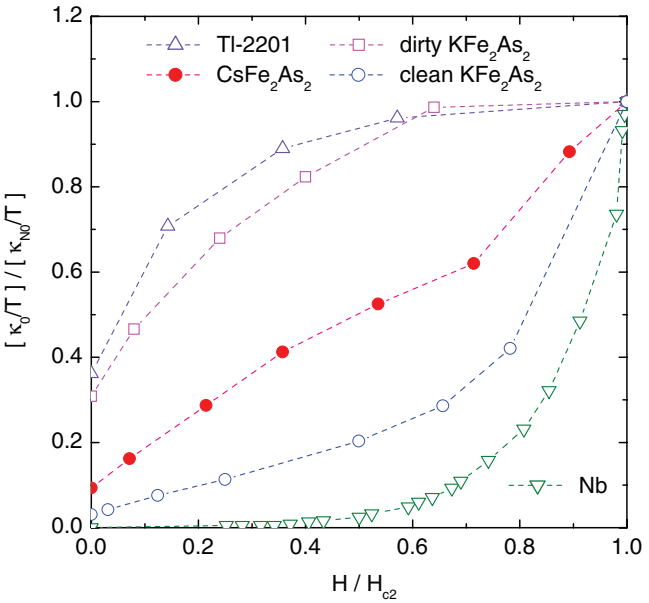

FIG. 4. (Color online) Normalized residual linear term κ_0/T of $CsFe_2As_2$ as a function of H/H_{c2}. For comparison, similar data are shown for the clean s-wave superconductor Nb,[44] an overdoped d-wave cuprate superconductor Tl-2201,[42] the dirty and clean KFe_2As_2.[29,31]

Reid *et al.* argued that the field dependence of κ_0/T is compelling evidence for a d-wave gap since the experimental $\kappa_0/T(H)$ curve is close to the calculated curve of a d-wave superconductor in the clean limit ($\hbar\Gamma/\Delta_0 = 0.1$).[31] The ρ_0 and RRR of our $CsFe_2As_2$ lie between the dirty and clean KFe_2As_2, indicating that its impurity level lies between the dirty and clean KFe_2As_2. This may reasonably explain the position and shape of its normalized $\kappa_0/T(H)$ curve in Fig. 4. Such a result suggests that $CsFe_2As_2$ has a nodal superconducting gap structure similar to that of KFe_2As_2, and shows how the field dependence of κ_0/T evolves with the impurity level.

IV. SUMMARY

In summary, we have measured the thermal conductivity of $CsFe_2As_2$ single crystal, the sister compound of KFe_2As_2, down to 50 mK. Both the significant κ_0/T in zero field and the field dependence of κ_0/T provide clear evidence for nodal superconducting gap in $CsFe_2As_2$. Our results suggest that the extremely hole-doped (K, Rb, Cs)Fe_2As_2 series of iron-based superconductors should have a common nodal gap structure. More experiments on these compounds are needed to get the consensus on their exact gap symmetry (d wave or accidental nodal s wave).

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