Quantum criticality and nodal superconductivity in the FeAs-based superconductor KFe$_2$As$_2$

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The in-plane resistivity $\rho$ and thermal conductivity $\kappa$ of FeAs-based superconductor KFe$_2$As$_2$ single crystal were measured down to 50 mK. We observe non-Fermi-liquid behavior $\rho(T) \sim T^{1.5}$ at $H_{c2} = 5$ T, and the development of a Fermi liquid state with $\rho(T) \sim T^2$ when further increasing field. This suggests a field-induced quantum critical point, occurring at the superconducting upper critical field $H_{c2}$. In zero field there is a large residual linear term $\kappa_0/T$, and the field dependence of $\kappa_0/T$ mimics that in $d$-wave cuprate superconductors. This indicates that the superconducting gaps in KFe$_2$As$_2$ have nodes, likely $d$-wave symmetry. Such a nodal superconductivity is attributed to the antiferromagnetic spin fluctuations near the quantum critical point.

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When superconductivity emerges with the suppression of magnetism, for example in heavy-fermion and high-temperature cuprate superconductors, the spin fluctuations associated with a magnetic quantum critical point is usually considered as the pairing glue. This is also the case for the recently discovered FeAs-based high-temperature superconductors [1–3]. The parent compounds of the FeAs-based superconductors, for example LaFeAsO and BaFe$_2$As$_2$, are not superconducting and manifest antiferromagnetic (AF) order [3, 4]. With electron or hole doping, the AF order is suppressed and superconductivity emerges [1, 4].

Spin fluctuations usually result in superconducting gaps with nodes, but it can also give nodeless superconducting gaps through interband interaction, termed $s_{\pm}$-wave [5, 8]. In the $s_{\pm}$-wave pairing theory for FeAs-based superconductors [2], the interband interaction happens between the hole pockets at $\Gamma$ point and the electron pockets at $M$ point, via the antiferromagnetic spin fluctuations (AFSF) with wave vector $Q_{AF} = (\pi, \pi)$. This gives full superconducting gaps on both electron and hole pockets, but with the opposite signs of the order parameters between the two [7]. While there are accumulating experimental and theoretical works in favor of nodeless gaps, conclusive phase-sensitive experiments are needed to confirm the $s_{\pm}$-wave pairing in FeAs-based superconductors [3]. Moreover, in LaFePO and BaFe$_2$(As$_{1-x}$P$_x$)$_2$, two compounds containing phosphorus, there are evidences for nodal superconductivity [10, 13]. Therefore, the pairing symmetry and superconducting mechanism in iron pnictides are still far from consensus.

Recently, the ARPES experiments on the extremely hole-doped KFe$_2$As$_2$ ($T_c = 3$ K) showed that the electron pockets at $M$ point completely disappear due to the hole doping [14]. This result immediately raises a very important question: what is the superconducting state in heavily overdoped FeAs-based superconductors where the interband interaction is suppressed? While ARPES experiment was unable to study the superconducting gap structure in KFe$_2$As$_2$ due to its low $T_c$, the low-temperature thermal conductivity technique is particularly useful for studying exotic superconductors with low $T_c$ [17].

In this Letter, we report the demonstration of a clear field-induced antiferromagnetic quantum critical point and nodal superconductivity in KFe$_2$As$_2$ by resistivity and thermal conductivity measurements. Our findings not only confirm the spin-fluctuation-mediated pairing mechanism, but also clarify the pairing symmetry when interband interaction is suppressed in heavily overdoped regime, thus complete our understanding of the superconducting state in the FeAs-based superconductors.

Single crystals of KFe$_2$As$_2$ were grown by self-flux method [16]. The dc magnetic susceptibility was measured by a SQUID (Quantum Design). The sample was cleaved to a rectangular shape of dimensions $1.5 \times 1.0$ mm$^2$ in the $ab$-plane, with 40 $\mu$m 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 exposure in air less than 2 hours. The contacts are metallic with typical resistance 100 m$\Omega$ at 1.5 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 $T_c$.

Fig. 1 shows the in-plane resistivity of our KFe$_2$As$_2$ single crystal. The residual resistivity ratio (RRR) $\rho$(297
K)/ρ(5 K) = 86, which is very close to that reported previously, RRR = 87 [17]. The upper and lower insets of Fig. 1 show the resistive and magnetic superconducting transitions at low temperature. The middle point of the resistivity transition is at Tc = 3.0 K.

In Fig. 2a, the resistivity in H = 0 and 2 T are plotted as ρ vs T1.5. It is found that ρ obeys T1.5 dependence nicely above Tc, up to about 20 K. Previously, Terashima et al. claimed that ρ exhibits a T2 dependence below ~45 K [17]. However, we note that their T2 fitting does not look good at low temperature. To elucidate how low the T1.5-dependent ρ(T) can go, we measure the resistivity down to 50 mK in a dilution refrigerator and in higher magnetic fields. Fig. 2b plots ρ vs T1.5 for H = 4, 5, 6, 8, 11, and 14.5 T. The downward deviation of ρ from T1.5 dependence below 1.6 K in H = 4 T is attributed to the onset of superconductivity. In H = 5 T, we find a perfect T1.5-dependent resistivity down to 50 mK. In H > 5 T, there is an upward deviation of ρ from the T1.5 dependence at low temperature. The data of H = 5, 6, 8, 11, and 14.5 T were re-plotted as ρ vs T2 in Fig. 2c. It is clearly seen that a Fermi liquid behavior of resistivity, ρ ~ T2, develops with increasing field.

Based on our experimental determined ranges of T2 behavior at low temperatures, we have constructed a phase diagram of the T − H plane (Fig. 3). The inset of Fig. 3 plots the field dependence of the coefficient A of the T2 term, which tends to diverge towards Hc2 = 5 T. Such a phase diagram of KFe2As2 is strikingly similar to that of the heavy-fermion superconductor CeCoIn5 with Tc = 2.3 K, in which a field-induced AF quantum critical point (QCP) is located at Hc2 [18, 21].

Near an AF QCP, the scattering of electrons by AFSC usually leads to non-Fermi-liquid behavior of resistivity, ρ ~ T1.5 in 3D system and ρ ~ T in 2D system [22]. The observation of ρ ~ T1.5 at Hc2 in quasi-2D CeCoIn5 is explained by the similar character of the magnetic fluctuations in the CeIn3 planes of CeCoIn5 and in bulk 3D CeIn3 itself [20]. As for KFe2As2 in this study, the T1.5 dependence of ρ at Hc2 indicates that the magnetic fluctuations in KFe2As2 also have some 3D character, which may need further investigation.

The similarity between the T − H phase diagrams of KFe2As2 and CeCoIn5 suggests that there is also a field-induced AF QCP at Hc2 in KFe2As2. To our knowledge, this is the first time to demonstrate an AF QCP in FeAs-based superconductors down to low temperature.

A field-induced AF QCP at Hc2 is very unusual, since
it indicates that the superconducting and magnetic orders are tightly coupled [22]. This may be easy to understand in CeCoIn$_5$, since there is a closely related compound CeRhIn$_5$ which is an ambient pressure antiferromagnet with the Néel temperature $T_N = 3.8$ K. For KFe$_2$As$_2$, this is highly unexpected, in the sense that KFe$_2$As$_2$ is far away from the AF parent compound BaFe$_2$As$_2$ in Ba$_1$-xK$_x$Fe$_2$As$_2$ system. However, our finding is strongly supported by recent nuclear magnetic resonance (NMR) experiments on KFe$_2$As$_2$ single crystal, which did claim the existence of strong AFSF [23].

Having demonstrated the field-induced AF QCP in KFe$_2$As$_2$, we continue to investigate its superconducting gap structure. Fig. 4a shows the temperature dependence of the in-plane thermal conductivity for KFe$_2$As$_2$ in $H = 0$, 0.1, 0.3, 0.5, 0.8, 1.25, and 2 T magnetic fields, plotted as $\kappa/T$ vs $T$. All the curves are roughly linear, as previously observed in BaFe$_{1.9}$Ni$_{0.1}$As$_2$ and overdoped BaFe$_{2-x}$Co$_x$As$_2$ single crystals [24–26]. Therefore we fit the data to $\kappa/T = a + bT^{-\alpha}$ with $\alpha$ fixed to 2. The two terms $aT$ and $bT^{-\alpha}$ represent contributions from electrons and phonons, respectively. Here we only focus on the electronic term.

In zero field, the fitting gives a residual linear term $\kappa_0/T = 2.27 \pm 0.02$ mW K$^{-2}$ cm$^{-1}$. Such a large $\kappa_0/T$ in KFe$_2$As$_2$ is really very surprising, since previous thermal conductivity studies of FeAs-based superconductors (without phosphorus), including Ba$_{0.75}$K$_{0.25}$Fe$_2$As$_2$, have given negligible $\kappa_0/T$ in $H = 0$ [24–27]. From Fig. 4a, a very small field $H = 0.1$ T has significantly increased the $\kappa/T$. Above $H = 0.8$ T, $\kappa/T$ tends to saturate. In $H = 1.25$ and 2 T, $\kappa_0/T = 7.39 \pm 0.03$ and 7.36 $\pm 0.04$ mW K$^{-2}$ cm$^{-1}$ were obtained from the fittings, respectively. Both values meet the expected normal state Wiedemann-Franz law expectation $L_0/\rho_0(2$ T$) = 7.38$ mW K$^{-2}$ cm$^{-1}$, within experimental error bar. We take $H = 2$ T as the bulk $H_{c2}$ of KFe$_2$As$_2$, despite that the resistive transition is not completely suppressed until $H_{c2}$ (onset) $= 5$ T. To choose a slightly different bulk $H_{c2}$ does not affect our discussions below.

In Fig. 4b, the normalized $\kappa_0/T$ of KFe$_2$As$_2$ is plotted as a function of $H/H_{c2}$, together with the clean s-wave superconductor Nb [28], the multi-band s-wave superconductor NbSe$_2$ [29], and an overdoped sample of the d-wave superconductor Tl-2201 [30]. For KFe$_2$As$_2$, the large $\kappa_0/T$ in $H = 0$ and the rapid increase of $\kappa_0/T$ at low field mimic the behavior of Tl-2201, and provide clear evidences for unconventional superconducting gap with nodes [13]. We note that recent 75As nuclear quadrupole resonance (NQR) and specific heat measure-
ments on KFe$_2$As$_2$ polycrystals also suggested multiple nodal gaps [31].

The nodal gap in extremely hole-doped KFe$_2$As$_2$ is distinctly different from the nodeless gaps in FeAs-based superconductors at other doping [24–27, 32, 33]. In optimally hole-doped Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ and electron-doped BaFe$_{1.85}$Co$_{0.15}$As$_2$, the observations of nearly nested Fermi-surface pockets and nodeless gaps suggest that the interband interaction may play a crucial role in superconducting pairing [32, 33], thus support the $s_\pm$-wave pairing mechanism in FeAs-based superconductors [7]. However, in KFe$_2$As$_2$, the electron pockets at M point completely disappear due to hole doping [14]. Although four new small hole (ε) pockets are found around M point, the interband interaction is nevertheless suppressed, therefore the $s_\pm$-wave pairing mechanism does not work in KFe$_2$As$_2$ [14]. Having known that there exist strong AF SF near the AF QCP, one can be sure that the nodal superconductivity in KFe$_2$As$_2$ results from interband pairing via AF SF. Usually, the pairing mediated by AF SF has $d$-wave symmetry, as in CeCoIn$_5$ [34]. Because of the great similarity between the $T-H$ phase diagrams of KFe$_2$As$_2$ and CeCoIn$_5$, and the similar behavior of $\kappa_0(H)/T$ between KFe$_2$As$_2$ and Ti-2201, the nodal gap in KFe$_2$As$_2$ is very likely also $d$-wave.

We note that there is an electron-hole asymmetry in the phase diagram of FeAs-based superconductors at the heavily overdoped regime. For the heavily electron-doped BaFe$_{1.7}$Co$_{0.3}$As$_2$, the hole pocket at T point completely disappears [35], and the interband interaction is also suppressed. But it turns out that BaFe$_{1.7}$Co$_{0.3}$As$_2$ is not superconducting [35]. This electron-hole asymmetry may be explained by the different strength of AF SF on electron- and hole-doped sides measured by NMR [24, 36]. In BaFe$_{2-x}$Co$_x$As$_2$, the spin fluctuations are completely suppressed at $x > 0.3$ [36], while strong AF spin fluctuations were found in KFe$_2$As$_2$ [23].

Finally, it is worth to point out that the possible nodal superconductivity in LaFePO and BaFe$_2$As$_2$ may be different from that in KFe$_2$As$_2$. The main reason is that the isovalent substitution of P for As only slightly modifies the Fermi surface [12], therefore the Fermi-surface nesting and interband interaction still exist in these two compounds. To get nodal superconductivity in LaFePO and BaFe$_2$As$_2$, two needs to consider the competition between $s_\pm$-wave and $d$-wave superconducting states even more carefully.

In summary, we have measured the resistivity and thermal conductivity of extremely hole-doped KFe$_2$As$_2$ single crystal down to 50 mK. A field-induced AF QCP is demonstrated by the observation of $\rho \sim T^{1.5}$ at $H_{c2} = 5$ T, and the development of $\rho \sim T^2$ Fermi liquid behavior at $H > 5$ T. Furthermore, the large $\kappa_0(T)/T$ at zero field and a rapid increase of $\kappa_0(T)/T$ at low field give strong evidences for nodal superconductivity in KFe$_2$As$_2$. Such a nodal superconductivity, very likely $d$-wave, naturally results from the intraband pairing via AFSF near the AF QCP. Our results are consistent with the suppression of interband interaction in KFe$_2$As$_2$, as revealed by ARPES experiments.

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