

Doping evolution of the quasiparticle excitations in heavily hole-doped $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$: A possible superconducting gap with sign-reversal between hole pockets

D. Watanabe,¹ T. Yamashita,¹ Y. Kawamoto,¹ S. Kurata,¹ Y. Mizukami,¹ T. Ohta,¹ S. Kasahara,¹ M. Yamashita,^{1,2}
T. Saito,³ H. Fukazawa,³ Y. Kohori,³ S. Ishida,⁴ K. Kihou,⁴ C. H. Lee,⁴ A. Iyo,⁴ H. Eisaki,⁴ A. B. Vorontsov,⁵
T. Shibauchi,^{1,6} and Y. Matsuda¹

¹Department of Physics, Kyoto University, Kyoto 606-8502, Japan

²Institute for Solid State Physics, University of Tokyo, Chiba 277-8581, Japan

³Department of Physics, Chiba University, Chiba 263-8522, Japan

⁴National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Ibaraki 305-8568, Japan

⁵Department of Physics, Montana State University, Bozeman, Montana 59717, USA

⁶Department of Advanced Materials Science, University of Tokyo, Chiba 277-8561, Japan

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To gain insight into the unconventional superconductivity of Fe pnictides with no electron pockets, we measure the thermal conductivity κ and penetration depth λ in the heavily hole-doped regime of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$. The residual thermal conductivity $(\kappa/T)_{T \rightarrow 0\text{ K}}$ and T dependence of λ consistently indicate the fully gapped superconductivity at $x = 0.76$ and the (line) nodal superconductivity at higher hole concentrations. The magnitude of $\frac{\kappa}{T} T_c|_{T \rightarrow 0\text{ K}}$ and the slope of $\lambda(T)$ at low temperatures, both of which are determined by the properties of the low-energy excitations, exhibit a highly unusual nonmonotonic x dependence. These results indicate a dramatic change of the nodal characteristics in a narrow doping range. We suggest that the observed x dependence is naturally explained by a doping crossover of the gap function between the s -wave states with and without sign reversal between Γ -centered hole pockets.

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I. INTRODUCTION

In most iron-based high- T_c superconductors, the interband interaction between the disconnected hole and electron Fermi-surface pockets is believed to be responsible for electron pairing and superconductivity [1–3]. Exceptional cases are heavily hole-doped iron-pnictide $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ and heavily electron-doped iron-chalcogenide $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$, which have only hole pockets and electron pockets, respectively, but still exhibit superconductivity. Since the superconducting (SC) gap structure reflects the structure of the pairing interaction, the gap structure of this class of iron-based compounds has attracted great interest and various kinds of SC gap functions have been proposed theoretically [4–7].

In $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$, the electron pockets disappear at $x \gtrsim 0.6$ (Ref. [8]) or $x > 0.7$ (Ref. [9]) and the Fermi surface consists of three main two dimensional hole pockets at the center of the Brillouin zone and small hole pockets near the zone corner [10]. In contrast to nodeless extended s -wave superconductivity in optimally doped $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ ($T_c = 38\text{ K}$, $x \approx 0.45$) [8,11–13], line nodes appear in highly hole-doped KFe_2As_2 [14–17], prompting a proposal of a d -wave SC gap function based on the thermal conductivity measurements [17]. However, subsequent laser angle resolved photoemission spectroscopy (ARPES) has revealed a nodal s -wave gap with a peculiar structure with a nodeless gap on the inner hole pocket, “octet-line nodes” on the middle pocket and an almost-zero gap on the outer pocket [18]. Moreover, it has been shown that the specific heat is incompatible with the d -wave gap [19]. These indicate that these line nodes are not symmetry protected but accidental.

It has been extensively discussed that the interband hopping plays a major role in iron-pnictides, and in materials having both hole (h) and electron (e) pockets, it gives rise to S_{\pm}^{he} gap,

where the SC order parameter has opposite signs on hole and electron pockets. Then the question arises: what is the pairing mechanism of superconductivity in pnictides with *no* electron pockets? In this respect, it is of particular interest to find out whether the sign change of the SC order parameter occurs between the *hole* pockets.

Here, we show that the presence or absence of the sign reversal between the hole pockets can be inferred from doping variations of low-energy quasiparticle (QP) density of states (DOS). We have measured the thermal conductivity and penetration depth, both of which are very sensitive bulk probe for the low-energy QP excitations, down to very low temperature in the heavily hole-doped regime of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$. The results suggest that the sign change of SC gap between the hole pockets occurs very close to $x = 1$, implying the importance of the inter-hole-pocket scattering for the electron pairing.

II. EXPERIMENT

Single crystals of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ have been grown by the KAs flux method [20]. For $x = 1, 0.93, 0.88$ and 0.76 , the T_c s are 3.7, 7.3, 11, and 22 K, residual resistivity ratios RRR are 1906, 175, 206, and 49, and the upper critical field $H_{c2}(\parallel c \text{ axis})$ at $T = 0\text{ K}$ are 1.8, 6.5, 17.3, and 28 T, respectively. For $x = 0.88$ and 0.76 , $H_{c2}(0)$ are estimated by assuming Werthamer-Helfand-Hohenberg relation [23]. Thermal conductivity κ is measured by the steady-state method and the penetration depth λ is determined by using tunnel diode oscillators [21]. The x dependence of T_c is determined from the zero resistivity for samples used in the κ measurements, and this $T_c(x)$ relation is used to estimate the x values for the samples used in the λ measurements from their T_c values [22].

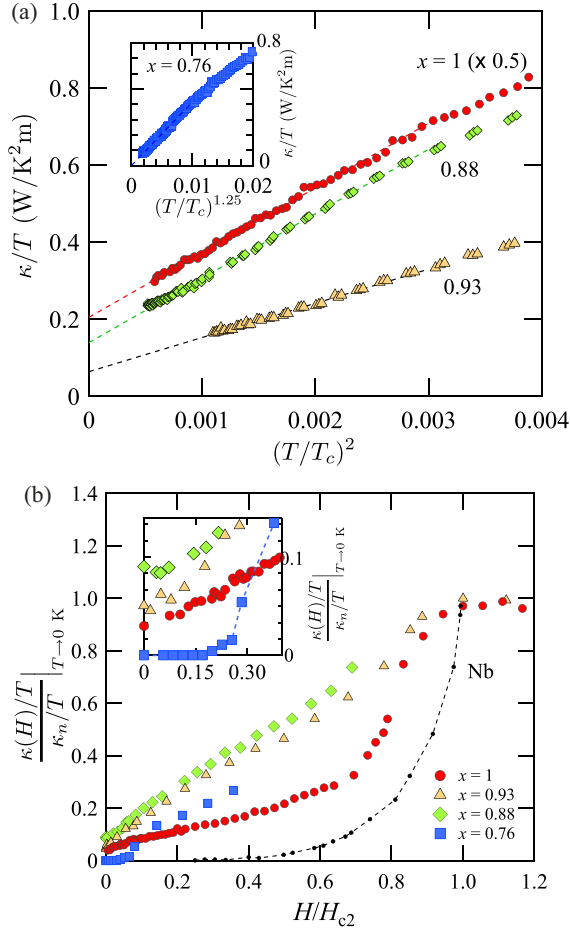


FIG. 1. (Color online) Low-temperature thermal conductivity in $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ single crystals. (a) κ/T vs $(T/T_c)^2$ for $x = 1.0, 0.93, 0.88$. Inset shows κ/T plotted against $(T/T_c)^{1.25}$ for $x = 0.76$. (b) Field dependence of κ/T normalized to the normal-state values κ_n/T . Each point is determined from the extrapolation of the temperature dependence to $T \rightarrow 0$ K at each field applied along the c axis. For comparison, the data for Nb is also plotted [24]. Inset is an expanded view at low fields. Dashed lines are the guide to the eyes.

III. RESULTS

A. Thermal conductivity

There is a fundamental difference in the low-temperature thermal transport between superconductors with isotropic and nodal gaps. In the latter, the QP heat conduction is entirely governed by the low-energy nodal excitations. Figure 1(a) shows the zero-field thermal conductivity divided by temperature κ/T well below T_c plotted as a function of $(T/T_c)^2$ for $x = 0.88, 0.93$, and 1.0 . The total thermal conductivity is a sum of the electron and phonon contributions. The former is represented by $\kappa_e/T \approx N(0)v_F^2\tau_e$, where $N(0)$, v_F , and τ_e are the QP DOS, Fermi velocity, and QP scattering time, respectively. At low temperatures, κ/T is well fitted by $\kappa/T = \tilde{\kappa}_0 + bT^2$, where $\tilde{\kappa}_0$ and b are constants. The finite residual value in κ/T at $T \rightarrow 0$ K, $\tilde{\kappa}_0$, is clearly resolved, indicating the finite residual QP DOS, which is attributed to be the presence of line nodes in the SC gap function [25,26]. In

sharp contrast, as depicted in the inset of Fig. 1(a), κ/T for $x = 0.76$ does not show T^2 dependence but is well fitted by $\kappa/T \propto T^{1.25}$ with no residual value, indicating a full SC gap.

Another strong indication for the SC gap with line nodes for $x \geq 0.88$ and the full gap for $x = 0.76$ is provided by the field dependence of the thermal conductivity. In magnetic fields, the thermal transport of superconductors with line nodes is dominated by contributions from delocalized QP states outside the vortex cores. Then $N(0)$ increases in proportion to \sqrt{H} (Volovik effect), which gives rise to a steep increase of $\kappa(H)$ well below upper critical field H_{c2} [27]. On the other hand, in fully gapped superconductors where all QPs are essentially localized within the vortex cores, $\kappa(H)$ exhibits an exponential behavior with a very small growth with H at low fields [see the data of Nb [24] in Fig. 1(b)]. Figure 1(b) shows the field dependence of κ/T normalized by the normal-state value κ_n/T at $T \rightarrow 0$ K for $H \parallel c$. For $x = 0.88$ and 0.76 , κ_n is determined by the normal state resistivity extrapolated to $T \rightarrow 0$ K assuming the Wiedemann-Franz law. The field dependence of κ/T for $x = 0.76$ is fundamentally different from that for other systems: at low fields, κ/T for $x = 1.0, 0.93, 0.88$, displays a convex field dependence, while concave one is observed for $x = 0.76$. This can be seen more clearly in the inset of Fig. 1(b). For $x = 1.0, 0.93$, and 0.88 , $\kappa(H)/T (\propto N(H))$ increases in proportional to \sqrt{H} , indicating the presence of line nodes. In contrast, $\kappa(H)/T$ for $x = 0.76$ stays nearly zero up to $\sqrt{H/H_{c2}} \sim 0.16$, followed by a rapid increase at around $\sqrt{H/H_{c2}} \sim 0.3$, indicating the fully gapped superconductivity.

B. Penetration depth

A further test of the absence or presence of line nodes in the SC gap is provided by the penetration depth λ . In the inset of Fig. 2, the temperature dependence of the relative change of $\lambda(T)$, $\delta\lambda(T) = \lambda(T) - \lambda(0)$, at low temperatures is plotted against T/T_c for $x = 0.76$ and 0.86 . The data for $x = 0.76$ exhibits exponential behavior at low temperatures and is completely flat within the experimental error of ~ 0.3 nm below $T/T_c < 0.05$, indicating negligible QP excitations, i.e., a fully gapped SC state. In sharp contrast, $\delta\lambda(T)$ for $x = 0.86$ increases with finite temperature slope even at $T \rightarrow 0$ K, indicating the presence of significant QP excitations. Generally, in nodal superconductors one expects that $\delta\lambda(T)$ at low temperatures can be written in analogy to the d -wave case [28] as

$$\delta\lambda = \alpha(T/T_c)^2/(T/T_c + T_{\text{imp}}/T_c). \quad (1)$$

This formula interpolates the low-temperature T^2 dependence arising from the impurity band with width $\sim T_{\text{imp}} \ll T_c$, and the T -linear dependence at $T \gtrsim T_{\text{imp}}$ associated with the nodal excitations. In a wide temperature range below $T/T_c < 0.2$, the data for $x \geq 0.86$ can be fitted well by Eq. (1) (Fig. 2). These results provide a strong support of the presence of line nodes for $x \geq 0.86$.

Thus the T and H dependencies of the thermal conductivity and the penetration depth, all consistently indicate the full SC gap for $x = 0.76$ and SC gap with line nodes for higher concentration $x \geq 0.86$. Compared to the conventional superconductor Nb, $\kappa(H)/T$ for $x = 0.76$ exhibits a rapid

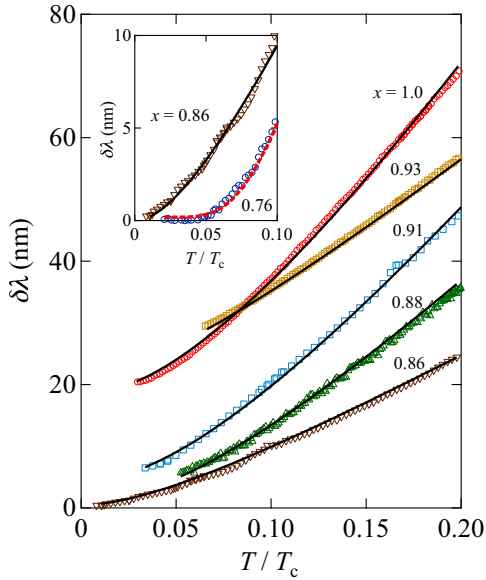


FIG. 2. (Color online) Change in the penetration depth as a function of T/T_c in $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$. Each curve is vertically shifted for clarity. The solid lines are fits to Eq. (1) with $T_{\text{imp}}/T_c = 0.17, 0.08, 0.15, 0.18, 0.08$ for $x = 1.0, 0.93, 0.91, 0.88, 0.86$, respectively. Inset shows a comparison between the data for $x = 0.86$ and 0.76 , the latter of which is fitted to the temperature dependence of a fully gapped state (dashed line).

increase at much lower H/H_{c2} . Moreover, a flat region of $\delta\lambda(T)$ is achieved only below $T/T_c < 0.05$. These indicate that the SC gap function for $x = 0.76$ is strongly modulated and the minimum gap value is very small, which can be estimated as $\lesssim 0.4k_B T_c$ [inset of Fig. 2]. This implies that the line node appears slightly above $x = 0.76$.

C. Doping dependence of low-energy excitations

We discuss the issue of sign reversal of the SC gap between the hole pockets. Figure 3(a) depicts the doping evolution of $\frac{\kappa}{T} T_c|_{T \rightarrow 0\text{K}}$ and the coefficient α in $\delta\lambda(T)$, which corresponds to $\frac{d\lambda}{d(T/T_c)}$ in the clean limit $T_{\text{imp}}/T_c \rightarrow 0$ [see Eq. (1)]. Some intuition about their behavior can be obtained based on the clean limit results for d -wave [25,29] and anisotropic s -wave [26] that look similar:

$$(\kappa/T) T_c|_{T \rightarrow 0\text{K}} = a N_F v_F^2 \mu^{-1}, \quad (d\lambda/dT) T_c \propto \mu^{-1}. \quad (2)$$

Here, a is a constant of order unity and N_F is the normal state DOS, and the transport time, $1/\mu$, is inverse “gap velocity” $\mu = \frac{1}{\Delta} \frac{d|\Delta(\phi)|}{d\phi}|_{\text{node}}$ that describes opening of the SC gap at the node [here, Δ is the T_c -related gap scale and $|\Delta(\phi)|$ is the impurity renormalized gap] [25,26,29].

As reported by the NMR Knight shift measurements [30], N_F is nearly x -independent in the present x range. From this also follows that it is unlikely that v_F strongly depends on x . The nodal parameter μ is, therefore, the main factor responsible for the x dependence of $\frac{\kappa}{T} T_c|_{T \rightarrow 0\text{K}}$ and $\frac{d\lambda}{d(T/T_c)}$. However, to determine this μ parameter, a comparison with theory is needed, as it is strongly affected by the impurity renormalized gap $|\tilde{\Delta}(\phi)|$ in case of accidental nodes [26].

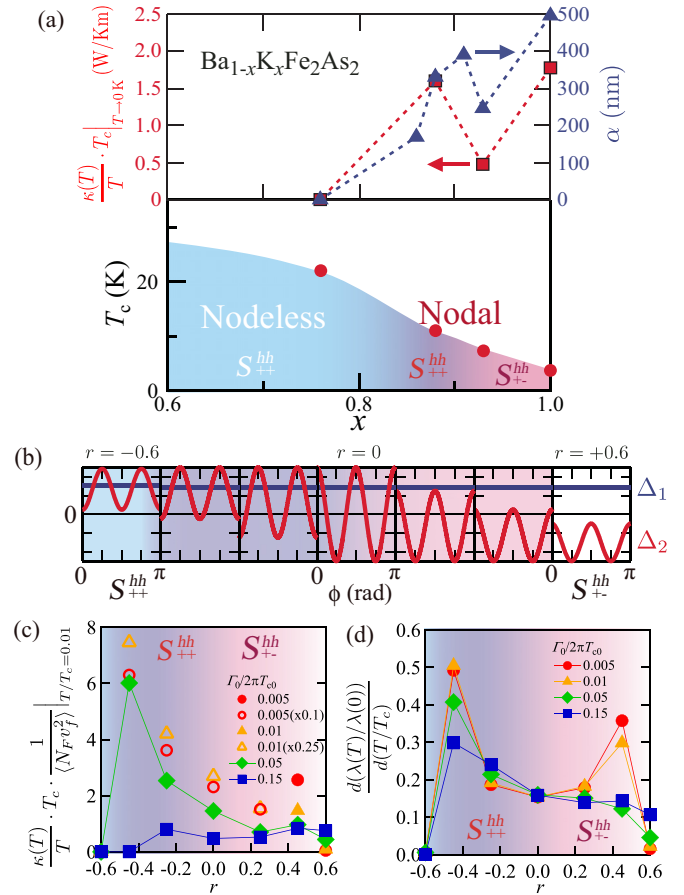


FIG. 3. (Color online) Doping evolution of low-energy QP excitations in $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$. (a) (Top) x dependence of $\frac{\kappa(T)}{T} T_c|_{T \rightarrow 0\text{K}}$ (red squares, left axis) and the coefficient α in $\delta\lambda(T)$ [see Eq. (1)] (blue triangles, right axis). (Bottom) x dependence of T_c . (b) We associate the above nonmonotonic behavior with the gap evolution with the parameter r from predominantly S_{++}^{hh} ($r = 0.6$) to predominantly S_{+-}^{hh} ($r = -0.6$) through intermediate regime with accidental nodes in $\Delta_2(\phi)$ [see Eq. (3)]. (c) and (d) Theoretically determined residual $\kappa(T)/T T_c$ (c) and $d\lambda(T)/d(T/T_c)$ (d) as functions of gap parameter r with several values of impurity scattering Γ_0 .

The most remarkable feature in Fig. 3(a) is that both $\frac{\kappa}{T} T_c|_{T \rightarrow 0\text{K}}$ and the penetration depth coefficient α exhibit similar doping dependence. As x is decreased from 1, both of them first decrease, go through a minimum at $x \sim 0.93$, increase to a maximum at $x \sim 0.9$, and then decrease again. Such a nonmonotonic behavior can also be readily seen in the raw data of $\delta\lambda(T)$ (Fig. 2) as a change in the slope in a region of $0.1 < T/T_c < 0.2$ where almost T -linear dependence is found. Both the residual thermal conductivity and the $\lambda(T)$ slope in the zero-temperature limit are expected to vanish at x slightly above 0.76 where nodes disappear. The observed nonmonotonic x dependence in a narrow doping range is directly connected with unusual evolution of the low-energy QP excitations. We first point out that these results provide a strong support for the nodal s -wave symmetry for $x \geq 0.86$, rather than d wave. This is because in symmetry-protected d -wave superconductors μ and hence both $\frac{\kappa}{T} T_c|_{T \rightarrow 0\text{K}}$ and

$\frac{d\lambda}{d(T/T_c)}$, are expected to be mostly independent of doping and impurity level. On the other hand, gap with accidental nodes has nothing special about its structure and is expected to depend strongly on both [3,26].

IV. DISCUSSION

We stress that the observed nonmonotonic doping evolution of $\frac{\kappa}{T} T_c|_{T \rightarrow 0 \text{ K}}$ and α provides an important hint for the SC order parameter in the heavily hole-doped regime, and suggests a crossover from an s -wave state with sign reversed order parameter between the hole bands (S_{+-}^{hh}) to s -wave state without sign reversal (S_{++}^{hh}). Here, we consider the SC gap evolution shown in Fig. 3(b), assuming a model with different gap functions, isotropic in band 1 and anisotropic in band 2 [31]. Since the Fermi surface and transition temperature evolve gradually with doping, it is natural to assume that the line nodes are present in the same hole pocket in the very narrow doping range $0.88 \leq x \leq 1.0$:

$$\begin{aligned} \Delta_1(\phi) &= \Delta_{10}, \\ \Delta_2(\phi) &= -\text{sgn}(r)\Delta_{20}[|r| - (1 - |r|)\cos 4\phi]. \end{aligned} \quad (3)$$

The parameter r varies from +1 to -1 and continuously connects S_{+-}^{hh} state with S_{++}^{hh} state through a state with accidental nodes in $\Delta_2(\phi)$ when $|r| < 0.5$ [Fig. 3(b)]. Theoretically, we self-consistently compute the order parameter and, more importantly, impurity self-energies with the quasiclassical two-band approach using t matrix, outlined in Refs. [26,32]. We use unitary scattering limit and for the case of two Γ -centered hole bands we assume uniform scattering, $V_{\text{interband}} = V_{\text{intra-band}}$. Impurity self-energies in two bands are used to calculate the thermal conductivity $\kappa(T) = \kappa_1(T) + \kappa_2(T)$ using Keldysh formalism [25,33]. The results of the theoretical model for various gap structures are presented in Figs. 3(c) and 3(d) for several values of impurity scattering Γ_0 .

In the optimally doped sample ($x = 0.45$), both theory and experiment point to the hole-electron S_{+-}^{he} state with same-sign gap on the hole pockets, S_{++}^{hh} . There is no evidence for another nodal state appearing in the intermediate doping region between $x = 0.45$ and 0.76 [8,9], and thus we consider that $x = 0.76$ sample has an S_{++}^{hh} state as well, but possibly with strongly anisotropic $\Delta_2(\phi)$. The fact that the line nodes disappear slightly above $x = 0.76$ suggests that the gap function of $x = 0.76$ is close to $r = -0.6$ in our model. On the other end $x = 1$, the laser ARPES measurement in KFe_2As_2 [18] points to the gap structure similar to either $r = -0.4$ (S_{+-}^{hh}) or 0.4 (S_{++}^{hh}). So the possible gap evolution with doping between $x = 1$ and 0.76 can either go (I) from $r = -0.4 \rightarrow -0.6$, or (II) from $r = 0.4 \rightarrow -0.6$. In case (I), it is a transition between very similar gap structures $S_{+-}^{hh} \rightarrow S_{++}^{hh}$, where one expects at most a monotonic change in the nodal parameter μ , and consequently monotonic low- T observables.

In case (II), on the other hand, a crossover from S_{+-}^{hh} to S_{++}^{hh} state occurs. If one considers the $x = 1$ order parameter to correspond in our model to $r \approx 0.45$, and the other peak doping $x \sim 0.9$ to $r \approx -0.45$, the theoretical nonmonotonic doping dependence of $\frac{\kappa}{T} T_c|_{T \rightarrow 0 \text{ K}}$ and $\frac{d\lambda}{d(T/T_c)}$ is consistent with experiment, and points to the sign reversal between the hole pockets in KFe_2As_2 .

We also note that the calculation results indicate that the residual thermal conductivity only weakly depends on impurity concentration on the S_{+-}^{hh} side, and very sensitive to it on the S_{++}^{hh} side, c.f. cases $r = \pm 0.45$. This, together with the reported results that a couple of $x = 1$ samples with different residual resistivity show similar values of $\frac{\kappa}{T} T_c|_{T \rightarrow 0 \text{ K}}$ [17,34], gives a fully consistent picture that in the A_{1g} state, inferred from laser ARPES [18], the Γ -centered hole bands have order parameter of predominantly opposite signs.

In iron-pnictides with both electron and hole pockets, the electron-hole interaction is crucially important for the electron pairing. According to the antiferromagnetic spin fluctuation scenario, this interaction gives rise to the sign change between these pockets [3,35]. Without the electron pockets, however, it seems that the hole-hole interaction steps up to the front stage, as revealed by the sign reversal between hole pockets in KFe_2As_2 . The present results imply that several competing interactions (electron-hole and hole-hole, or simply hole-hole in the absence of electron pockets), give rise to a unique and rich variety of SC gap structure of iron pnictides, very different from that of all other superconductors.

V. CONCLUSION

In summary, from the thermal conductivity and penetration depth measurements of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ in the heavily hole-doped regime, we show that the slope of the gap at the nodes exhibit a highly unusual nonmonotonic x dependence. These results imply that the nodal gap function of KFe_2As_2 is not symmetry protected but has extended s -wave structure and reverses sign between the hole pockets, suggesting that the inter-hole-pocket scattering plays an important role in superconductivity.

Note added. Recently, we became aware of very recent ARPES results, which show a change from nodal to nodeless state between dopings corresponding to samples with $T_c \sim 17$ and 22 K [36], consistent with our results.

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[1] J. Paglione and R. L. Greene, *Nat. Phys.* **6**, 645 (2010).

[2] G. R. Stewart, *Rev. Mod. Phys.* **83**, 1589 (2011).

[3] P. J. Hirschfeld, M. M. Korshunov, and I. I. Mazin, *Rep. Prog. Phys.* **74**, 124508 (2011).

- [4] R. Thomale, C. Platt, W. Hanke, J. Hu, and B. A. Bernevig, *Phys. Rev. Lett.* **107**, 117001 (2011).
- [5] S. Maiti, M. M. Korshunov, and A. V. Chubukov, *Phys. Rev. B* **85**, 014511 (2012).
- [6] C. Platt, R. Thomale, C. Honerkamp, S.-C. Zhang, and W. Hanke, *Phys. Rev. B* **85**, 180502(R) (2012).
- [7] S. Maiti and A. V. Chubukov, *Phys. Rev. B* **87**, 144511 (2013).
- [8] W. Malaeb, T. Shimojima, Y. Ishida, K. Okazaki, Y. Ota, K. Ohgushi, K. Kihou, T. Saito, C. H. Lee, S. Ishida, M. Nakajima, S. Uchida, H. Fukazawa, Y. Kohori, A. Iyo, H. Eisaki, C.-T. Chen, S. Watanabe, H. Ikeda, and S. Shin, *Phys. Rev. B* **86**, 165117 (2012).
- [9] K. Nakayama, T. Sato, P. Richard, Y.-M. Xu, T. Kawahara, K. Umezawa, T. Qian, M. Neupane, G. F. Chen, H. Ding, and T. Takahashi, *Phys. Rev. B* **83**, 020501(R) (2011).
- [10] T. Terashima, M. Kimata, N. Kurita, H. Satsukawa, A. Harada, K. Hazama, M. Imai, A. Sato, K. Kihou, C. H. Lee, H. Kito, H. Eisaki, A. Iyo, T. Saito, H. Fukazawa, Y. Kohori, H. Harima, and S. Uji, *J. Phys. Soc. Jpn.* **79**, 053702 (2010).
- [11] H. Ding, P. Richard, K. Nakayama, K. Sugawara, T. Arakane, Y. Sekiba, A. Takayama, S. Souma, T. Sato, T. Takahashi, Z. Wang, X. Dai, Z. Fang, G. F. Chen, J. L. Luo, and N. L. Wang, *Europhys. Lett.* **83**, 47001 (2008).
- [12] K. Hashimoto, T. Shibauchi, S. Kasahara, K. Ikada, S. Tonegawa, T. Kato, R. Okazaki, C. J. van der Beek, M. Konczykowski, H. Takeya, K. Hirata, T. Terashima, and Y. Matsuda, *Phys. Rev. Lett.* **102**, 207001 (2009).
- [13] D. V. Evtushinsky, D. S. Inosov, V. B. Zabolotnyy, A. Koitzsch, M. Knupfer, B. Büchner, M. S. Viazovska, G. L. Sun, V. Hinkov, A. V. Boris, C. T. Lin, B. Keimer, A. Varykhalov, A. A. Kordyuk, and S. V. Borisenko, *Phys. Rev. B* **79**, 054517 (2009).
- [14] K. Hashimoto, A. Serafin, S. Tonegawa, R. Katsumata, R. Okazaki, T. Saito, H. Fukazawa, Y. Kohori, K. Kihou, C. H. Lee, A. Iyo, H. Eisaki, H. Ikeda, Y. Matsuda, A. Carrington, and T. Shibauchi, *Phys. Rev. B* **82**, 014526 (2010).
- [15] H. Fukazawa, Y. Yamada, K. Kondo, T. Saito, Y. Kohori, K. Kuga, Y. Matsumoto, S. Nakatsuji, H. Kito, P. M. Shirage, K. Kihou, N. Takeshita, C. H. Lee, A. Iyo, and H. Eisaki, *J. Phys. Soc. Jpn.* **78**, 083712 (2009).
- [16] J. K. Dong, S. Y. Zhou, T. Y. Guan, H. Zhang, Y. F. Dai, X. Qiu, X. F. Wang, Y. He, X. H. Chen, and S. Y. Li, *Phys. Rev. Lett.* **104**, 087005 (2010).
- [17] J.-Ph. Reid, M. A. Tanatar, A. Juneau-Fecteau, R. T. Gordon, S. R. de Cotret, N. Doiron-Leyraud, T. Saito, H. Fukazawa, Y. Kohori, K. Kihou, C. H. Lee, A. Iyo, H. Eisaki, R. Prozorov, and Louis Taillefer, *Phys. Rev. Lett.* **109**, 087001 (2012).
- [18] K. Okazaki, Y. Ota, Y. Kotani, W. Malaeb, Y. Ishida, T. Shimojima, T. Kiss, S. Watanabe, C.-T. Chen, K. Kihou, C. H. Lee, A. Iyo, H. Eisaki, T. Saito, H. Fukazawa, Y. Kohori, and S. Shin, *Science* **337**, 1314 (2012).
- [19] F. Hardy, R. Eder, M. Jackson, D. Aoki, C. Paulsen, T. Wolf, P. Burger, A. Boehmer, P. Schweiss, P. Adelman, R. A. Fisher, and C. Meingast, [arXiv:1309.5654](https://arxiv.org/abs/1309.5654).
- [20] K. Kihou, T. Saito, S. Ishida, M. Nakajima, Y. Tomioka, H. Fukazawa, Y. Kohori, T. Ito, S. Uchida, A. Iyo, C. H. Lee, and H. Eisaki, *J. Phys. Soc. Jpn.* **79**, 124713 (2010).
- [21] K. Hashimoto, M. Yamashita, S. Kasahara, Y. Senshu, N. Nakata, S. Tonegawa, K. Ikada, A. Serafin, A. Carrington, T. Terashima, H. Ikeda, T. Shibauchi, and Y. Matsuda, *Phys. Rev. B* **81**, 220501(R) (2010).
- [22] The absolute values of x have some errors because of the finite transition width, but the monotonic T_c increase with decreasing x as well as similar T_{imp}/T_c values in these samples ensure that the relative order of doping levels should be safely determined.
- [23] N. R. Werthamer, E. Helfand, and P. C. Hohenberg, *Phys. Rev.* **147**, 295 (1966).
- [24] J. Lowell and J. B. Sousa, *J. Low Temp. Phys.* **3**, 65 (1970).
- [25] M. J. Graf, S.-K. Yip, J. A. Sauls, and D. Rainer, *Phys. Rev. B* **53**, 15147 (1996).
- [26] V. Mishra, A. Vorontsov, P. J. Hirschfeld, and I. Vekhter, *Phys. Rev. B* **80**, 224525 (2009).
- [27] Y. Matsuda, K. Izawa, and I. Vekhter, *J. Phys.: Condens. Matter* **18**, R705 (2006).
- [28] P. J. Hirschfeld and N. Goldenfeld, *Phys. Rev. B* **48**, 4219 (1993).
- [29] D. Xu, S. K. Yip, and J. A. Sauls, *Phys. Rev. B* **51**, 16233 (1995).
- [30] M. Hirano *et al.*, *J. Phys. Soc. Jpn.* **81**, 054704 (2012).
- [31] An almost-zero SC gap has been reported in the outer hole pocket in KFe_2As_2 at 2 K [18]. Such a small gap, however, is not observed in the penetration depth and H dependence of thermal conductivity measurements at very low temperatures, implying that this gap has no node and grows rapidly at low temperatures, or this band gives much smaller contribution than other bands to these quantities as it has a smaller Fermi velocity. We therefore simply assume two gap functions given by Eq. (3).
- [32] T. Das, A. B. Vorontsov, I. Vekhter, and M. J. Graf, *Phys. Rev. B* **87**, 174514 (2013).
- [33] A. B. Vorontsov and I. Vekhter, *Phys. Rev. B* **75**, 224502 (2007).
- [34] A. F. Wang, S. Y. Zhou, X. G. Luo, X. C. Hong, Y. J. Yan, J. J. Ying, P. Cheng, G. J. Ye, Z. J. Xiang, S. Y. Li, and X. H. Chen, [arXiv:1206.2030](https://arxiv.org/abs/1206.2030).
- [35] H. Kontani and S. Onari, *Phys. Rev. Lett.* **104**, 157001 (2010).
- [36] Y. Ota, K. Okazaki, Y. Kotani, T. Shimojima, W. Malaeb, S. Watanabe, C.-T. Chen, K. Kihou, C. H. Lee, A. Iyo, H. Eisaki, T. Saito, H. Fukazawa, Y. Kohori, and S. Shin, *Phys. Rev. B* **89**, 081103(R) (2014).