

Doping-induced vertical line nodes in the superconducting gap of the iron arsenide $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ from directional thermal conductivity

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The thermal conductivity κ of the iron-arsenide superconductor $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ was measured down to 50 mK in a magnetic field up to 15 T, for a heat current parallel and perpendicular to the tetragonal c axis. In the range from optimal doping ($x \simeq 0.4$) down to $x = 0.16$, there is no residual linear term in $\kappa(T)$ as $T \rightarrow 0$, showing that there are no nodes in the superconducting gap anywhere on the Fermi surface. Upon crossing below $x = 0.16$, a large residual linear term suddenly appears, signaling the onset of nodes in the superconducting gap, most likely vertical line nodes running along the c axis. We discuss two scenarios: (1) accidental nodes in an s -wave gap, resulting from a strong modulation of the gap around the Fermi surface, in which minima deepen rapidly with underdoping; (2) a phase transition from a nodeless s -wave state to a d -wave state, in which nodes are imposed by symmetry.

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To elucidate the pairing mechanism in iron-based superconductors, a key question is whether the underlying interaction is anisotropic. This will typically show up as anisotropy in the gap, as in cuprate superconductors whose d -wave gap goes to zero along lines of nodes imposed by symmetry. The superconducting gap of iron pnictides has been investigated with several probes, but a clear overall picture has yet to emerge [1]. In some materials, like LaFePO [2], KFe_2As_2 [3] and P-doped BaFe_2As_2 [4], nodes have been detected. In other materials, like LiFeAs [5, 6] and Co-doped BaFe_2As_2 (Co-Ba122) [7], an isotropic gap without nodes is seen. However, the gap in Co-Ba122 is only isotropic at optimal doping, where T_c is maximal, and it acquires nodes when doping is either reduced or increased [8, 9]. While several calculations favour a nodeless sign-changing s_{\pm} state, the gap can have strong modulations and even nodes [10–13], and a d -wave state has comparable energy [11, 12].

In this Letter, we investigate the gap structure of K-doped BaFe_2As_2 using low-temperature thermal conductivity, a bulk directional probe of the superconducting gap [14]. Our main finding is the sudden appearance of a finite residual linear term, κ_0/T , upon crossing below the critical concentration $x = 0.16$, the signature that nodes have appeared in the superconducting gap. We discuss whether these are accidental, the result of deep minima in a strongly-modulated s -wave gap, or imposed by symmetry in a doping-tuned transition to a d -wave state. In either case, our findings reveal how easily the full isotropic gap observed at maximal T_c can be destabilized by tuning away from optimal conditions, pointing

to anisotropic and competing interactions.

Experimental.— Single crystals of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ (K-Ba122) were grown from self flux [15]. The doping level x was determined from the superconducting transition temperature T_c , defined by the point of zero resistance, using an empirical curve of T_c vs x [15, 16] (see Fig. 1). T_c values for a -axis samples are: 9.4, 10.0, 14.5, 16.4, 25.5, 29.5, 36.7 K; T_c values for c -axis samples are: 10.7, 11.3, 17.1, 18.8, 28.6, 28.9, 38.1 K. Our samples cover a wide range, both inside and outside the region of co-existence with antiferromagnetic order (below $x \simeq 0.24$ [16]). Thermal conductivity was measured in a standard one-heater two-thermometer technique for two directions of heat flow: parallel ($J||c$; κ_c) and perpendicular ($J||a$; κ_a) to the [001] tetragonal c axis. Sample geometry, contact technique and measurement protocol are described elsewhere [8]. The magnetic field H was applied along the c axis.

Results: $H = 0$.— The thermal conductivity of K-Ba122 is displayed in Fig. 2. The fits show that the data below 0.3 K are well described by the function $\kappa/T = a + bT^\alpha$. The first term, $a \equiv \kappa_0/T$, is the residual linear term, entirely due to electronic excitations [14]. The second term is due to phonons, which at low temperature are scattered by the sample boundaries, with $1 < \alpha < 2$ [17, 18]. For $x > 0.16$ ($T_c > 13$ K), $\kappa_0/T = 0$ within error bars, for both current directions. This is consistent with our previous data for κ_a at $T_c=26$ K (Ref. 19). By contrast, for both directions a large residual linear term is found when $x < 0.16$ ($T_c \simeq 10$ K).

It is convenient to compare κ_0/T to the normal-state

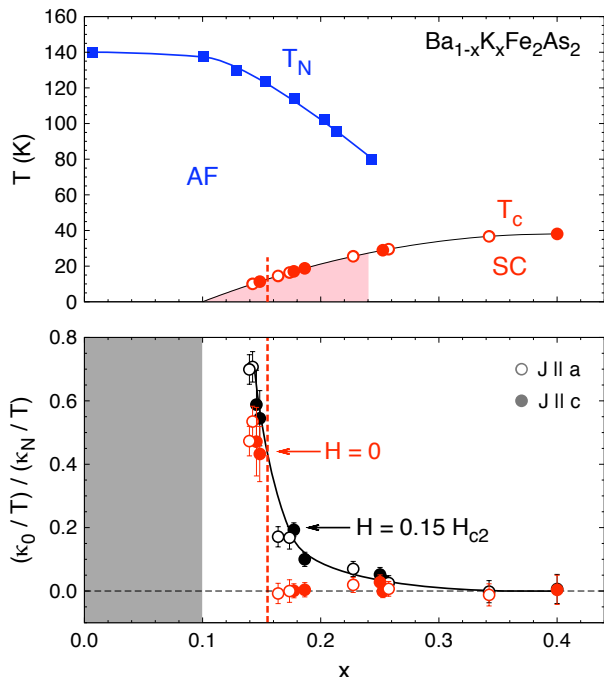


FIG. 1: Top: Doping phase diagram of K-Ba122, showing the antiferromagnetic (AF) phase below T_N (blue squares, from ref. 16) and the superconducting (SC) phase below T_c (open (closed) red circles, for a -axis (c -axis) samples), which coexist in the shaded region. Bottom: Residual linear term in the thermal conductivity κ as $T \rightarrow 0$, κ_0/T , plotted as a fraction of the normal-state conductivity, κ_N/T , for both κ_a (open symbols) and κ_c (closed symbols), for magnetic fields $H=0$ (red) and $H = 0.15 H_{c2}$ (black). Error bars reflect the combined uncertainties in the extrapolation of κ/T and ρ to $T = 0$. The vertical dashed line at $x = 0.16$ separates a region in which the superconducting gap has nodes ($x < 0.16$) from one in which it doesn't ($x > 0.16$). The solid black line is a guide to the eye. There is no superconductivity in the shaded region.

conductivity κ_N/T , estimated using the Wiedemann-Franz law, namely $\kappa_N/T = L_0/\rho_0$ where $L_0 \equiv (\pi^2/3)(k_B/e)^2$, applied to the extrapolated residual resistivity ρ_0 , as discussed in Ref. 8. In Fig. 1, the ratio $(\kappa_0/T)/(\kappa_N/T)$ is plotted vs x . It jumps suddenly from 0 to ~ 0.5 upon crossing below $x = 0.16$. This signals the onset of nodes in the superconducting gap of K-Ba122. The fact that $(\kappa_0/T)/(\kappa_N/T)$ is of the same magnitude for both current directions indicates that the nodes preserve the $a-c$ anisotropy of normal-state transport, consistent with line nodes that run vertically along the quasi-2D Fermi surface of K-Ba122.

Results: $H > 0$.— In Fig. 3, we plot the residual linear term κ_0/T as a function of magnetic field H . As in Fig. 1, κ_0/T is normalized to κ_N/T , and H is normalized to H_{c2} , the upper critical field at $T \rightarrow 0$, given approximately by $H_{c2} = 100$ T ($T_c/40$ K) [20, 21]. At high x , the field

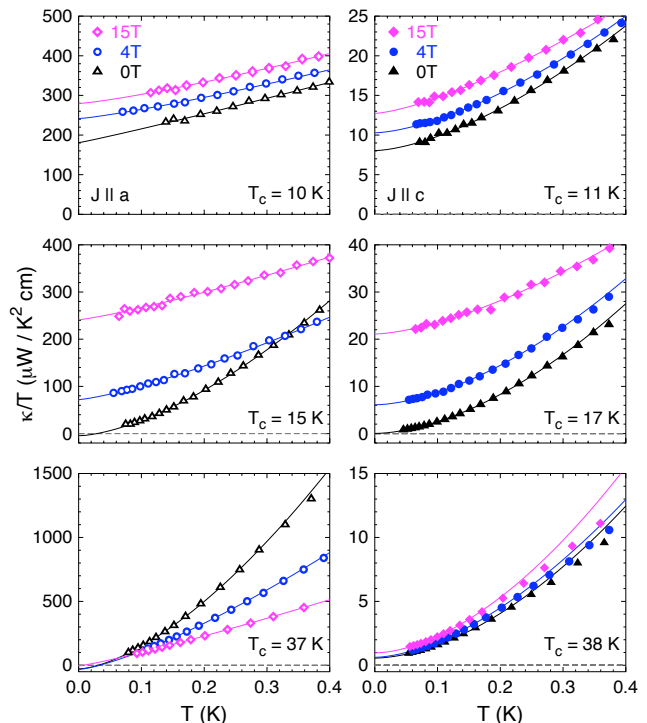


FIG. 2: Thermal conductivity $\kappa(T)$ of K-Ba122 at three representative K concentrations, indicated by their T_c values, plotted as κ/T vs T for a current parallel ($J||c$, right panels) and perpendicular ($J||a$, left panels) to the tetragonal c axis. Data are shown for three values of the magnetic field, as indicated (data for other fields are not shown for clarity). Lines are a fit to $\kappa/T = a + bT^\alpha$, used to extract the residual linear term $a \equiv \kappa_0/T$, plotted vs x in Fig. 1 and vs H in Fig. 3.

dependence is very weak, either flat at $T_c = 38$ K or with a slight upward curvature at $T_c = 30$ K. This is typical of s -wave superconductors, in which a full gap is present on the entire Fermi surface. Quasiparticle transport is caused by tunneling between states localized in adjacent vortex cores and thus grows exponentially as the intervortex separation shrinks with increasing field [22]. By contrast, when $x < 0.16$, κ_0/T increases rapidly at low H , with the downward curvature characteristic of a d -wave superconductor such as the overdoped cuprate Tl-2201 [23] (Fig. 3). In this case, delocalized nodal quasiparticles outside the vortex cores are excited by the Doppler shift in their energies caused by the field [24].

In the range $x = 0.16$, throughout which the gap has no nodes, the field dependence varies significantly, from flat at $x \simeq 0.4$ to strong at $x \simeq 0.18$. In samples with $T_c \simeq 15$ K, the H dependence is very similar to that of a multi-band s -wave superconductor like NbSe₂ [22] (Fig. 3), in which the small value of the superconducting gap on one part of the Fermi surface leads to enhanced excitation. In Fig. 1, we plot the x dependence of κ_0/T measured at $H = 0.15 H_{c2}$. We see that the sudden

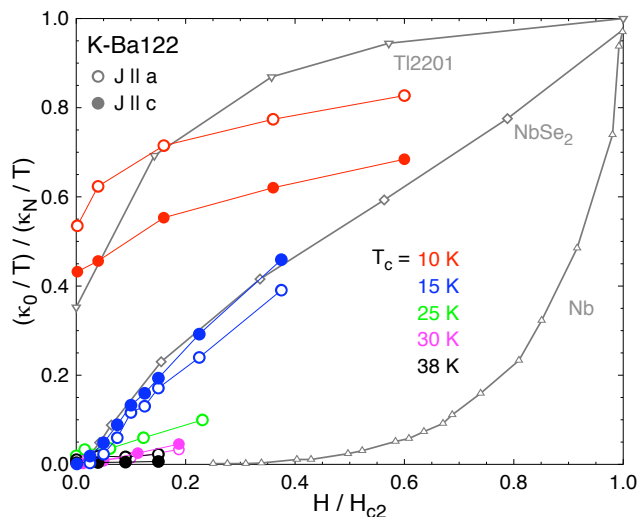


FIG. 3: Residual linear term κ_0/T in K-Ba122 normalized by the normal-state conductivity, κ_N/T , as a function of magnetic field H normalized by the upper critical field H_{c2} . The data for in-plane ($J||a$, open symbols) and inter-plane ($J||c$, closed symbols) transport are shown for five K concentrations, indicated by their T_c values. For comparison, we reproduce corresponding data for the isotropic s -wave superconductor Nb (see ref. 14), the multi-band s -wave superconductor NbSe₂ [22], and the d -wave superconductor Tl-2201 [23].

jump in κ_0/T at $H = 0$, when only zero-energy nodal quasiparticles contribute, turns into a more gradual rise at finite H , when finite-energy quasiparticles are excited across a small gap. Note that there could still be a small jump across $x = 0.16$ in finite H .

Coexisting AF order.— Neutron studies show that antiferromagnetic (AF) order is present down to at least $T \simeq 1$ K from $x = 0$ up to at least $x \simeq 0.24$ ($T_c = 26$ K), and it occupies at least 95 % of the sample volume [16]. The fact that $\kappa_0/T = 0$ (at $H = 0$) for $T_c > 13$ K implies that putative normal-state regions (with $\kappa_0/T = \kappa_N/T > 0$) must occupy a volume fraction below the percolation threshold. These two constraints rule out a scenario of inhomogeneous phase separation in the coexistence region, whereby the sample would consist of mutually exclusive regions of pure AF order and pure superconductivity. Therefore, there is a substantial region of the phase diagram where nodeless superconductivity coexists homogeneously with AF order. It has been argued that this is consistent with a s_{\pm} state but inconsistent with a standard s_{++} state [25, 26].

Scenario I: extended s -wave.— Given that the field-induced enhancement of κ_0/T increases as x approaches the critical value ($x = 0.16$) below which nodes appear in the gap (Fig. 1), it is natural to view the in-field behaviour as a precursor to the nodes, namely as minima in the gap that become gradually deeper until they touch the Fermi surface. This is the so-called ‘extended s -wave’ scenario, illustrated in Fig. 4.

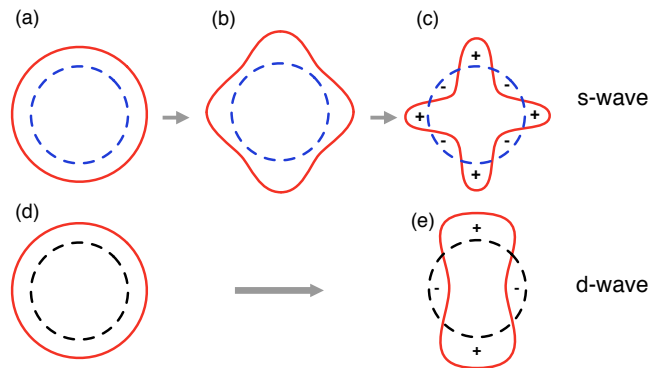


FIG. 4: Two scenarios for the doping evolution of the superconducting gap structure in K-Ba122 (solid red line), sketched for a single 2D Fermi surface (dashed circle). In the extended s -wave scenario (top): a) the gap is isotropic at high x ; b) it develops minima in certain directions at intermediate x ; c) those minima deepen to become negative at low x , producing nodes where $\Delta = 0$ at 8 points on the Fermi surface. In the d -wave scenario (bottom), a phase transition occurs between a superconducting state with s -wave symmetry at high x (d) and one with d -wave symmetry, and associated nodes, at low x (e).

The isotropic gap at high x develops minima in certain directions as x decreases, until this modulation as a function of azimuthal angle becomes strong enough that the minima become negative, thereby producing nodes below some critical x value. These nodes are called ‘accidental’, because they are not imposed by symmetry. There is no transition across the critical doping where nodes appear - the entire evolution takes place within one phase, with no change of symmetry. The fact that the in-field behaviour at $x > 0.16$ is just as isotropic as the zero-field behaviour at $x < 0.16$ reinforces the connection between minima and nodes. It is possible that the reconstruction of the Fermi surface caused by the onset of AF order (below $x \simeq 0.24$) either triggers or favours the gap modulation we observe, first as minima then as nodes.

In a number of calculations applied to pnictides, a superconducting state with s_{\pm} symmetry is found to be the most stable, but often the associated gap function has strong modulations, possibly leading to accidental nodes [11–13]. The gap modulation comes from a strongly anisotropic pairing interaction, which is also band-dependent. In 2D models, it is typically the gap on the electron Fermi surface centered at the M point of the Brillouin zone which shows a strong angular dependence within the basal plane [11, 12], leading to what would be vertical line nodes in 3D. In 3D calculations, nodes have been found on the hole Fermi surface at the Γ point [27]. This raises the question of which sheet of the Fermi surface of K-Ba122 has gap nodes. (Note that the nodes occur inside the phase of coexisting AF order, on a Fermi surface that is reconstructed relative to the paramagnetic Fermi surface assumed in the calculations.)

Because nodal conduction is $\sim 50\%$ of the normal-state conductivity (Fig. 3), which is dominated by hole-like carriers (Hall [28] and Seebeck [29] coefficients are positive), the dominant hole-like sheets must have nodes (and possibly other sheets as well).

Comparison with Co-Ba122.— There are clear similarities in the behavior of κ_0/T in Co-Ba122 and K-Ba122. At optimal doping, the gap is nodeless in both cases, with $\kappa_0/T = 0$ in both directions. The field dependence is more rapid in Co-Ba122, indicative of gap minima already present at maximal T_c , while they are absent in optimal K-Ba122. This points to a correlation between gap modulation and T_c weakness, as $T_c^{\text{max}} = 25$ K in Co-Ba122 and $T_c^{\text{max}} = 38$ K in K-Ba122. Away from optimal x , the gap minima deepen for both materials. In Co-Ba122, this is seen not only on the underdoped side (with the coexisting AF order), but also on the overdoped side, where there is no AF order. In both cases, the gap minima lead to isotropic in-field transport (relative to κ_N), and so are consistent with vertical line minima along the c axis.

The difference lies in the nodal character. While K-Ba122 has vertical line nodes, Co-Ba122 only has nodes on regions of the Fermi surface that contribute strongly to c -axis conduction. Recent calculations can account for this in terms of a Fermi surface with strong c -axis dispersion [30]. In a scenario where nodes are the result of minima that gradually deepen to become negative, this deepening would be uniform in K-Ba122, independent of k_z location on the Fermi surface, but k_z -dependent in Co-Ba122. It remains to be seen whether the nodes detected in KFe_2As_2 via a large in-plane κ_0/T [3] can be viewed as the end point of a gradual deepening of line minima in the overdoped regime of K-Ba122.

Scenario II: transition from s -wave to d -wave.— In a second scenario, the nodes appear below $x = 0.16$ because of a phase transition from an s -wave state (e.g. s_{\pm}) to a d -wave state. The nodes would no longer be accidental, but imposed by the d -wave symmetry, and they would necessarily appear on the Fermi-surface sheets centered on Γ , presumably hole-like. This would be compatible with the large value of κ_0/T and the dominance of hole-like carriers in the normal-state conductivity. The strong field dependence of κ_0/T for x above the would-be transition at $x = 0.16$ requires that the s -wave gap develops deep minima before the transition to d -wave.

In summary, the thermal conductivity of K-Ba122 in the $T = 0$ limit reveals that nodes appear in the superconducting gap when the K concentration is reduced below a critical value. Because the nodal quasiparticles are as effective in conducting heat parallel and perpendicular

to the c axis, the nodes are likely to be vertical lines on the quasi-2D Fermi surface, running parallel to the c axis. The large value of the nodal conductivity implies that nodes are present on those hole-like sheets of the Fermi surface that dominate the normal-state transport. The fact that the isotropic gap observed in K-Ba122 when T_c is maximal becomes anisotropic as superconductivity weakens suggests that pairing in iron-arsenide superconductors involves competing interactions.

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