

## Quasiparticle heat transport in single-crystalline $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ : Evidence for a $k$ -dependent superconducting gap without nodes

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The thermal conductivity  $\kappa$  of the iron-arsenide superconductor  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  ( $T_c \approx 30$  K) was measured in single crystals at temperatures down to  $T \approx 50$  mK ( $\approx T_c/600$ ) and in magnetic fields up to  $H = 15$  T ( $\approx H_{c2}/4$ ). A negligible residual linear term in  $\kappa/T$  as  $T \rightarrow 0$  shows that there are no zero-energy quasiparticles in the superconducting state. This rules out the existence of line and in-plane point nodes in the superconducting gap, imposing strong constraints on the symmetry of the order parameter. It excludes  $d$ -wave symmetry, drawing a clear distinction between these superconductors and the high- $T_c$  cuprates. However, the fact that a magnetic field much smaller than  $H_{c2}$  can induce a residual linear term indicates that the gap must be very small on part of the Fermi surface, whether from strong anisotropy or band dependence, or both.

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Several experiments have been performed to address the symmetry of the order parameter in iron-arsenide superconductors. Early studies on mostly polycrystalline samples of  $R\text{FeAs}(\text{O},\text{F})$ , where  $R = \text{La}, \text{Nd},$  or  $\text{Sm}$ , have led to indications that appear contradictory. While point-contact spectroscopy,<sup>1</sup> angle resolved photoemission spectroscopy (ARPES),<sup>2</sup> and penetration depth<sup>3-5</sup> point to a full superconducting gap without nodes, specific heat,<sup>6</sup> and nuclear magnetic resonance<sup>7</sup> data were interpreted in terms of a nodal superconducting gap. Reports on single crystals of doped  $\text{BaFe}_2\text{As}_2$ , also appear contradictory. In  $(\text{Ba},\text{K})\text{Fe}_2\text{As}_2$ , ARPES studies have found an isotropic superconducting gap with a magnitude of 12 meV on one Fermi surface and 6 meV on another,<sup>8-10</sup> specific-heat measurements are broadly consistent with an  $s$ -wave gap of 6 meV,<sup>11</sup> as are muon measurements of the superfluid density.<sup>12</sup> By contrast, penetration depth studies find a power-law variation,<sup>13</sup> as in Co-doped  $\text{BaFe}_2\text{As}_2$ ,<sup>14</sup> as opposed to the exponential temperature dependence expected of an isotropic  $s$ -wave gap.

In an attempt to shed further light on the structure of the superconducting gap, we have measured the thermal conductivity  $\kappa$  of  $K$ -doped  $\text{BaFe}_2\text{As}_2$ . Heat transport is a powerful probe of symmetry-imposed nodes in the superconducting gap.<sup>15</sup> We find a negligible residual linear term  $\kappa_0/T$  in  $\kappa/T$  as  $T \rightarrow 0$ , strong evidence that there are no nodes in the gap of this superconductor. Indeed, a line of nodes would have given a sizable and universal (i.e., impurity independent)  $\kappa_0/T$ ,<sup>15</sup> as in cuprates,<sup>16</sup> ruthenates,<sup>17</sup> and some heavy-fermion superconductors.<sup>18</sup> Given the large impurity scattering rate in our samples, point nodes would also have given a sizable (albeit nonuniversal)  $\kappa_0/T$  (Ref. 15)—unless they happen to lie along the  $c$  axis, perpendicular to the direction of heat flow in our measurements. However, a magnetic field  $H$  applied along the  $c$  axis induces a finite  $\kappa_0/T$  even for  $H \ll H_{c2}$ . This shows that the superconducting gap must be very small on some part of the Fermi surface, either because

of a pronounced anisotropy on one Fermi surface (whereby the gap has a deep minimum in some direction) or because of pronounced band dependence, causing one Fermi-surface sheet to have a very small gap.

Single crystals of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  were grown from FeAs flux.<sup>19</sup> The doping level of the two samples used in the present study, labeled A and B, was determined from the  $c$ -axis lattice parameter, giving  $x = 0.25$  for sample A ( $T_c = 26$  K) and  $x = 0.28$  for sample B ( $T_c = 30$  K), both on the underdoped side, i.e., below optimal doping ( $x \approx 0.4$ ).<sup>20</sup>

Samples were cleaved into rectangular bars with typical size  $1.5 \times 0.3 \times 0.05$  mm<sup>3</sup>. Silver wires were attached to the samples with a silver-based alloy, providing ultralow contact resistance of the order of 100  $\mu\Omega$ . Thermal conductivity was measured along the [100] direction in the tetragonal crystallographic plane in a standard one-heater-two thermometer technique.<sup>21</sup> The magnetic field  $H$  was applied along the [001] tetragonal axis. All measurements were done on warming after cooling in constant  $H$  from above  $T_c$  to ensure a homogeneous field distribution in the sample. All aspects of the charge and heat transport are qualitatively the same in both samples, with minor quantitative differences. For simplicity, only the data for sample A are displayed here. Whenever quoted, we provide quantitative values for both samples.

In Fig. 1, we show the electrical resistivity  $\rho(T)$  of sample A as a function of temperature. Below  $\sim 150$  K,  $\rho(T)$  shows a notable downturn, followed by a range where  $\rho(T)$  is well described either by a power-law dependence,  $\rho(T) = \rho_0 + AT^n$ , with  $n$  between 1.6 to 1.8, or by  $\rho(T) = \rho_0 + AT^2$ , below 70 K or so. Using the latter fit, we get  $\rho_0 = 47$   $\mu\Omega$  cm (sample A) and 28  $\mu\Omega$  cm (sample B). Since the same contacts are used for electrical and thermal transport, this allows us to accurately estimate the normal-state thermal conductivity  $\kappa_N/T$  in the  $T \rightarrow 0$  limit, via the Wiedemann-Franz law,  $\kappa_N/T = L_0/\rho_0$  where  $L_0 \equiv \frac{\pi^2}{3} \left(\frac{k_B}{e}\right)^2$ , giving  $\kappa_N/T = 520$   $\mu\text{W}/\text{K}^2$  cm

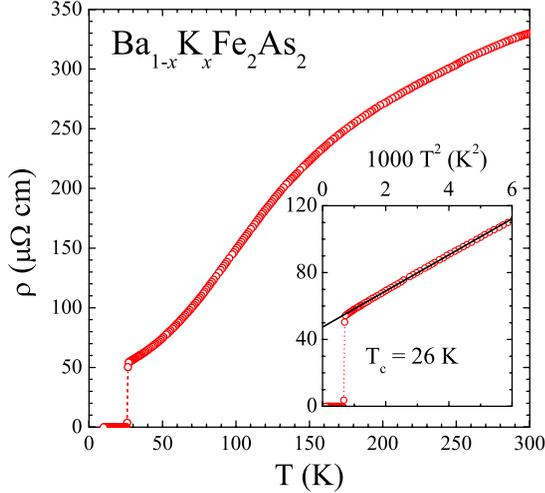


FIG. 1. (Color online) Temperature dependence of the in-plane resistivity  $\rho(T)$  of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ , with  $x \approx 0.25$  and  $T_c = 26$  K. Inset: same data plotted as a function of  $T^2$ . The line is a linear fit to  $\rho(T) = \rho_0 + AT^2$  below 70 K.

(sample A) and  $875 \mu\text{W}/\text{K}^2 \text{ cm}$  (sample B).

**Residual linear term in zero magnetic field.** The thermal conductivity  $\kappa(T)$  of sample A, measured for  $H=0$ , is shown in Fig. 2. The data are plotted as  $\kappa/T$  vs  $T^{1.65}$ . The linear fit displayed in Fig. 2 shows that the data below 0.4 K are well described by the function  $\kappa/T = a + bT^\alpha$ , with  $a \equiv \kappa_0/T = 5 \mu\text{W}/\text{K}^2 \text{ cm}$  and  $\alpha = 1.65$ . This same function describes the data of sample B equally well, with  $a \equiv \kappa_0/T = 7 \mu\text{W}/\text{K}^2 \text{ cm}$  and  $\alpha = 1.5$ . The first term is the residual linear term of interest here.<sup>15</sup> The second term is due to phonons, which at low temperatures are scattered by the sample boundaries. Although the latter term would be expected to give  $\kappa_p \propto T^3$ , i.e.,  $\alpha = 2$ , measurements on single crystals with smooth surfaces rarely show this textbook behavior because of a specular reflection off the surface, and in practice  $1 < \alpha < 2$ .<sup>22-24</sup>

The magnitude of the residual linear term extracted from the fits in Fig. 2 is extremely small. In similar measurements on samples where no residual linear term is expected,  $\kappa_0/T$  was indeed found to be zero, within an error bar of

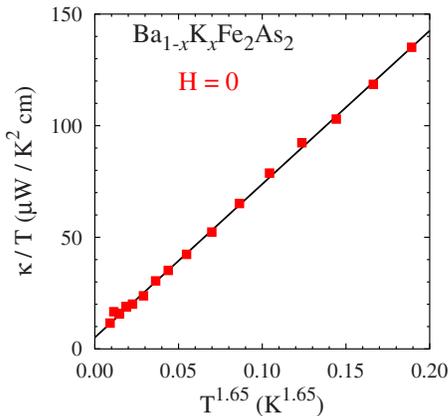


FIG. 2. (Color online) Temperature dependence of the thermal conductivity  $\kappa(T)$ , plotted as  $\kappa/T$  vs  $T^{1.65}$ , in zero magnetic field. The line is a linear fit over the temperature range shown.

$\pm 5 \mu\text{W}/\text{K}^2 \text{ cm}$  or so.<sup>22,25</sup> Within those error bars, our two samples of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  exhibit negligible residual linear terms. Let us put these minute  $\kappa_0/T$  values into perspective. Comparison with the normal-state conductivity, gives the ratio  $(\kappa_0/T)/(\kappa_N/T) \approx 1\%$  in both samples. Second, these  $\kappa_0/T$  values are much smaller than theoretical expectations for a nodal superconductor. (For a gap without nodes,  $\kappa_0/T$  should be zero.<sup>15</sup>) For a quasi-two-dimensional  $d$ -wave superconductor, with four line nodes along the  $c$ -axis, the residual linear term is given, in the clean limit ( $\hbar\Gamma_0 \ll \Delta_0$ ), by  $\kappa_0/T = (k_B^2/6d)(k_F v_F/\Delta_0)$ ,<sup>15,21,26,27</sup> where  $d$  is the interlayer separation,  $k_F$  and  $v_F$  the Fermi wavevector and velocity at the node, respectively, and  $\Delta_0$  the gap maximum. We can estimate the expected  $\kappa_0/T$  using parameters specific to the measured Fermi surface of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ .<sup>28</sup> For the  $\alpha$  and  $\beta$  sheets centered on the  $\Gamma$  point of the Brillouin zone, both of which would inevitably have nodes imposed by  $d$ -wave symmetry (whether  $d_{xy}$  or  $d_{x^2-y^2}$ ), the relevant parameters are:  $v_F = 0.5 \pm 0.1 \text{ eV \AA}$  and  $k_F = 0.16 \pm 0.03 \text{ \AA}^{-1}$  for the  $\alpha$  sheet and  $v_F = 0.22 \pm 0.04 \text{ eV \AA}$  and  $k_F = 0.32 \pm 0.05 \text{ \AA}^{-1}$  for the  $\beta$  sheet.<sup>28</sup> Taking  $d = 6.6 \text{ \AA}$  and assuming a weak coupling  $\Delta_0 = 2.14 k_B T_c$ , the theoretical estimate works out to be  $\kappa_0/T \approx 70 \mu\text{W}/\text{K}^2 \text{ cm}$  for each Fermi surface (the product  $k_F v_F$  is the same for both within error bars), for a total of  $\kappa_0/T \approx 140 \mu\text{W}/\text{K}^2 \text{ cm}$ . This is at least 20 times larger than the measured residual linear term. Note that away from the clean limit, when the impurity scattering rate  $\Gamma_0$  becomes comparable to the gap maximum  $\Delta_0$  (see below), the residual linear term  $\kappa_0/T$  is expected to be even larger.<sup>26</sup> In those materials where universal heat transport has been verified, the measured value of  $\kappa_0/T$  is in good quantitative agreement with this theoretical expectation,<sup>17,18,21</sup> e.g., in the overdoped cuprate Tl-2201, a well-established  $d$ -wave superconductor, measurements give  $\kappa_0/T \approx 300 \mu\text{W}/\text{K}^2 \text{ cm}$  for samples with the same  $T_c$  as our pnictide samples ( $T_c \approx 26 \text{ K}$ ).<sup>21</sup> Thus we can safely conclude that the gap in  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  does not contain a line of nodes anywhere on the Fermi surface. In particular, this rules out  $d$ -wave symmetry, whether  $d_{x^2-y^2}$  or  $d_{xy}$ . This result sets cuprates and iron arsenides apart as two distinct types of high-temperature superconductors.

It is in principle possible for the superconducting gap in pnictides to have point nodes as opposed to line nodes, one of the scenarios suggested by penetration depth studies.<sup>14</sup> The zero-energy quasiparticles associated with point nodes give a residual linear term which grows with impurity scattering,<sup>26</sup> so that  $\kappa_0/T$  can become a substantial fraction of  $\kappa_N/T$ .<sup>26</sup> Given  $\rho_0$ , we can estimate the normal-state impurity scattering rate  $\Gamma_0$  roughly from the plasma frequency  $\omega_p = c/\lambda_0$ , where  $\lambda_0$  is the penetration depth, approximately equal to 200 nm.<sup>14</sup> This gives  $\hbar\Gamma_0/k_B T_c \approx 1.7$  and 0.9 for samples A and B, respectively. This is very substantial, and would give a large residual linear term, comparable to the case of the line node. We therefore conclude that point nodes in the gap of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  are also unlikely, unless they are located along the  $c$  axis and do not contribute to in-plane transport. Needless to say, our data also rule out the possibility of an entirely gapless (or ungapped) Fermi surface, proposed by some authors,<sup>29</sup> at least down to the 1% level.

The nodes we have discussed so far are imposed by sym-

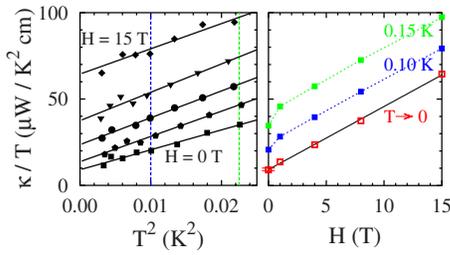


FIG. 3. (Color online) Left panel: temperature dependence of the thermal conductivity measured in magnetic fields of 0, 1, 4, 10, and 15 T (from bottom to top), plotted as  $\kappa/T$  vs  $T^2$ . Solid lines are a linear fit to each curve in the range shown. Vertical dashed lines indicate  $T=0.1$  K (blue) and  $T=0.15$  K (green). The value of  $\kappa/T$  at those two temperatures is plotted vs magnetic field in the right panel. Right panel: isotherms of  $\kappa/T$  as a function of magnetic field  $H$ , for  $T \rightarrow 0$  (obtained by extrapolating the linear fits in the left panel to  $T=0$ ), 0.1 and 0.15 K. In all three cases,  $\kappa/T$  rises approximately linearly with  $H$ , with the same slope. The solid line is a linear fit to the  $T \rightarrow 0$  data, also reproduced in Fig. 4.

metry, the result of a sign change in the order parameter around the Fermi surface. Such symmetry-related nodes are broadened by impurity scattering, giving rise to a sizable  $\kappa_0/T$ . The superconducting gap can also go to zero in certain directions because of a pronounced anisotropy that is not imposed by symmetry. However, such “accidental” nodes in a gap with  $s$ -wave symmetry will be lifted by impurity scattering, making the gap more isotropic.<sup>30</sup> Our zero-field data are consistent with an  $s$ -wave gap, including one with strong anisotropy.

**Field dependence of thermal conductivity.** The effect of a magnetic field  $H(H||c)$  on the thermal conductivity of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  is displayed in Fig. 3. In the panel on the left,  $\kappa/T$  curves are seen to shift upwards almost rigidly with field. In the right panel, the value of  $\kappa/T$  at three temperatures ( $T \rightarrow 0$ ,  $T=0.1$  K, and  $T=0.15$  K) is seen to rise linearly with  $H$  up to our highest field of 15 T, which corresponds roughly to  $H_{c2}/4$  [using  $H_{c2}$  at  $T \rightarrow 0$  as approximately 65 T (Ref. 31)]. In Fig. 4, we compare the field dependence of  $\kappa_0/T$  in  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  with the dependence in various other superconductors, using normalized conductivity and field scales,  $\kappa_s/\kappa_N$  and  $H/H_{c2}$ . In a  $d$ -wave superconductor like the overdoped cuprate Tl-2201 ( $T_c = 15$  K and  $H_{c2} \approx 7$  T),  $\kappa_0/T$  rises very steeply at the lowest fields,<sup>15,21,32</sup> roughly as  $\kappa_0/T \propto \sqrt{H}$ , following the density of delocalized zero-energy quasiparticles outside the vortex cores.<sup>33</sup> By contrast, in an isotropic  $s$ -wave superconductor like Nb, the rise in  $\kappa_0/T$  is exponentially slow at low fields as it relies on the tunneling of quasiparticles between localized states inside adjacent vortex cores, which at low fields are far apart. In many materials, however, the situation is not so clearcut. A good example is the multiband superconductors  $\text{MgB}_2$  (Ref. 34) and  $\text{NbSe}_2$ .<sup>25</sup> Here the magnitude of the  $s$ -wave superconducting gap is very different on two sheets of the Fermi surface. In both materials, the small gap is roughly one third of the large gap, so that a field  $H \approx H_{c2}/9$  is sufficient to kill superconductivity on the small-gap Fermi surface, which can then contribute its full normal-state conductivity even deep inside the vortex state. Specifi-

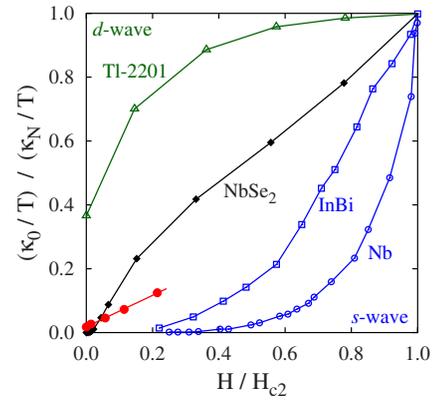


FIG. 4. (Color online) Residual linear term  $\kappa_0/T$  in the thermal conductivity of various superconductors as a function of magnetic field  $H$ , presented on normalized scales  $\kappa_s/\kappa_N$  vs  $H/H_{c2}$ . For  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  (red circles), we use  $\kappa_N/T$  obtained from the Wiedemann-Franz law to the extrapolated residual resistivity  $\rho_0$  (see text) and  $H_{c2}=65$  T (Ref. 31). The data for the clean and dirty isotropic  $s$ -wave superconductors Nb and InBi, respectively, are reproduced from (Ref. 24). The data for the  $d$ -wave superconductor Tl-2201 are for a strongly overdoped sample ( $T_c=15$  K and  $H_{c2} \approx 7$  T) (Ref. 32). The data for multiband superconductor  $\text{NbSe}_2$  ( $T_c=7$  K and  $H_{c2}=4.5$  T) are from Ref. 25.

cally, at  $H=H_{c2}/5$ ,  $\kappa_0/T$  is already half (one-third) of  $\kappa_N/T$  in  $\text{MgB}_2$  ( $\text{NbSe}_2$ ). It is one tenth in  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ . By comparison,  $\kappa_0/T$  is still negligible in a single-gap superconductor such as pure Nb or disordered InBi (see Fig. 4). This shows that the superconducting gap must be small on some part of the Fermi surface of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ , relative to the gap maximum which controls  $H_{c2}$ .

In Fig. 4, we show data for  $\text{NbSe}_2$ ,<sup>25</sup> where we see that  $\kappa_0/T=0$  at  $H=0$  and  $\kappa_0/T$  rises linearly above  $H_{c2}/30$ , with a slope of 1.67 in the normalized units of Fig. 4. In  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ ,  $\kappa_0/T$  also rises linearly, with a normalized slope of 0.46 in sample A and 0.21 in sample B. In a multiband scenario, the magnitude of this slope is roughly proportional to the value of the normal-state conductivity  $\kappa_N/T$  of the small-gap Fermi surface relative to the overall conductivity. The fact that the slope in absolute units is larger in sample A ( $3.7 \mu\text{W}/\text{K}^2 \text{ cm T}$ ) than in sample B ( $2.8 \mu\text{W}/\text{K}^2 \text{ cm T}$ ) even though its total normal-state conductivity  $\kappa_N/T=L_0/\rho_0$  is smaller (520 vs 875  $\mu\text{W}/\text{K}^2 \text{ cm}$ ) is suggestive of a multiband situation with the impurity scattering rate being different on different Fermi surfaces.

A recent heat transport study of the low- $T_c$  nickel-arsenide superconductor  $\text{BaNi}_2\text{As}_2$  ( $T_c=0.7$  K) (Ref. 35) gave  $\kappa_0/T=0$ , as here in  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  ( $T_c=26-30$  K). However, it found a much slower increase of  $\kappa_0/T$  at low  $H/H_{c2}$ , consistent with a gap that is large everywhere on the Fermi surface. This suggests that a strong  $k$  dependence of the gap may be important for achieving a high  $T_c$  value.

We conclude that there are no nodes in the superconducting gap of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ , at least at  $x \approx 0.25-0.28$ , with the possible exception of point nodes along the  $c$ -axis. This excludes  $d$ -wave symmetry, and any other symmetry that requires line nodes on the multisheet Fermi surface of this superconductor. Symmetries consistent with this constraint

include  $s$ -wave and  $s_{\pm}$ , whereby a full gap changes sign from the electron Fermi surface to the hole Fermi surface.<sup>36</sup> From the rapid rise of  $\kappa_0/T$  with magnetic field at very low fields, we infer that the gap must be very small on some portion of the Fermi surface. This  $k$  dependence of the gap magnitude can come from angle dependence or band dependence, or both. In many experiments, the presence of a very small gap could mimic that of a node.

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