

## Universal Heat Conduction in the Iron Arsenide Superconductor $\text{KFe}_2\text{As}_2$ : Evidence of a $d$ -Wave State

J.-Ph. Reid,<sup>1</sup> M. A. Tanatar,<sup>2</sup> A. Juneau-Fecteau,<sup>1</sup> R. T. Gordon,<sup>1</sup> S. René de Cotret,<sup>1</sup> N. Doiron-Leyraud,<sup>1</sup> T. Saito,<sup>3</sup> H. Fukazawa,<sup>3</sup> Y. Kohori,<sup>3</sup> K. Kihou,<sup>4</sup> C. H. Lee,<sup>4</sup> A. Iyo,<sup>4</sup> H. Eisaki,<sup>4</sup> R. Prozorov,<sup>2,5</sup> and Louis Taillefer<sup>1,6,\*</sup>

<sup>1</sup>Département de physique and RQMP, Université de Sherbrooke, Sherbrooke, Québec, Canada J1K 2R1

<sup>2</sup>Ames Laboratory, Ames, Iowa 50011, USA

<sup>3</sup>Chiba University and JST-TRIP, Chiba, Japan, 263-8522

<sup>4</sup>AIST and JST-TRIP, Tsukuba, Ibaraki, Japan, 305-8568

<sup>5</sup>Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011, USA

<sup>6</sup>Canadian Institute for Advanced Research, Toronto, Ontario, Canada M5G 1Z8

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The thermal conductivity  $\kappa$  of the iron arsenide superconductor  $\text{KFe}_2\text{As}_2$  was measured down to 50 mK for a heat current parallel and perpendicular to the tetragonal  $c$  axis. A residual linear term at  $T \rightarrow 0$ ,  $\kappa_0/T$  is observed for both current directions, confirming the presence of nodes in the superconducting gap. Our value of  $\kappa_0/T$  in the plane is equal to that reported by Dong *et al.* [*Phys. Rev. Lett.* **104**, 087005 (2010)] for a sample whose residual resistivity  $\rho_0$  was 10 times larger. This independence of  $\kappa_0/T$  on impurity scattering is the signature of universal heat transport, a property of superconducting states with symmetry-imposed line nodes. This argues against an  $s$ -wave state with accidental nodes. It favors instead a  $d$ -wave state, an assignment consistent with five additional properties: the magnitude of the critical scattering rate  $\Gamma_c$  for suppressing  $T_c$  to zero; the magnitude of  $\kappa_0/T$ , and its dependence on current direction and on magnetic field; the temperature dependence of  $\kappa(T)$ .

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The pairing mechanism in a superconductor is intimately related to the pairing symmetry, which in turn is related to the gap structure  $\Delta(\mathbf{k})$ . In a  $d$ -wave state with  $d_{x^2-y^2}$  symmetry, the order parameter changes sign with angle in the  $x$ - $y$  plane, forcing the gap to go to zero along diagonal directions ( $k_y = \pm k_x$ ). Those zeros (or nodes) in the gap are imposed by symmetry. The gap in states with  $s$ -wave symmetry will in general not have nodes, although accidental nodes can occur, depending on the anisotropy of the pairing interaction. In iron-based superconductors, the gap shows nodes in some materials, as in  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  [1] and  $\text{Ba}(\text{Fe}_{1-x}\text{Ru}_x)_2\text{As}_2$  [2], and not in others, as in  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  [3,4] and  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  [5,6] at optimal doping.

In  $\text{KFe}_2\text{As}_2$ , the end member of the  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  series (with  $x = 1$ ), the presence of nodes was detected by thermal conductivity [7], penetration depth [8], and NMR [9,10]. The question is whether those nodes are imposed by symmetry or accidental. Calculations differ in their predictions [11–13]. Some favor a  $d$ -wave state [14], others an  $s$ -wave state with accidental line nodes that run either parallel to the  $c$  axis [15] or perpendicular [11]. One can distinguish a  $d$ -wave state from an extended  $s$ -wave state with accidental nodes by looking at the effect of impurity scattering [16]. Nodes are robust in the former, but impurity scattering will eventually remove them in the latter, as it makes  $\Delta(\mathbf{k})$  less anisotropic.

In this Letter, we investigate the pairing symmetry of  $\text{KFe}_2\text{As}_2$  using thermal conductivity, a bulk directional

probe of the superconducting gap [17]. All aspects of heat transport are found to be in agreement with the theoretical expectation for a  $d$ -wave gap [18,19], and inconsistent with accidental line nodes, whether vertical or horizontal. Moreover, the critical scattering rate  $\Gamma_c$  for suppressing  $T_c$  to zero is of order  $T_{c0}$ , as expected for a  $d$ -wave superconductor, while it is 50 times  $T_{c0}$  in optimally doped  $\text{BaFe}_2\text{As}_2$  [20].

*Methods.*—Single crystals of  $\text{KFe}_2\text{As}_2$  were grown from self flux [21]. Two samples were measured: one for currents along the  $a$  axis and one for currents along the  $c$  axis. Their superconducting temperature, defined by the point of zero resistance, is  $T_c = 3.80 \pm 0.05$  K and  $3.65 \pm 0.05$  K, respectively. Since the contacts were soldered with a superconducting alloy, a small magnetic field of 0.05 T was applied to make the contacts normal and thus ensure good thermalization. For more information on sample geometry, contact technique and measurement protocol, see Ref. [6].

*Resistivity.*—To study the effect of impurity scattering in  $\text{KFe}_2\text{As}_2$ , we performed measurements on a single crystal whose residual resistivity ratio (RRR) is 10 times larger than that of the sample studied by Dong *et al.* [7] [Fig. 1(a)]. To remove the uncertainty associated with geometric factors, we normalize the data of Dong *et al.* to our value at  $T = 300$  K. A power-law fit below 16 K yields a residual resistivity  $\rho_0 = 0.21 \pm 0.02 \mu\Omega \text{ cm}$  ( $2.24 \pm 0.05 \mu\Omega \text{ cm}$ ) for our (their) sample, so that  $\rho(300 \text{ K})/\rho_0 = 1180$  (110).

We attribute the lower  $\rho_0$  in our sample to a lower concentration of impurities or defects. Note that except

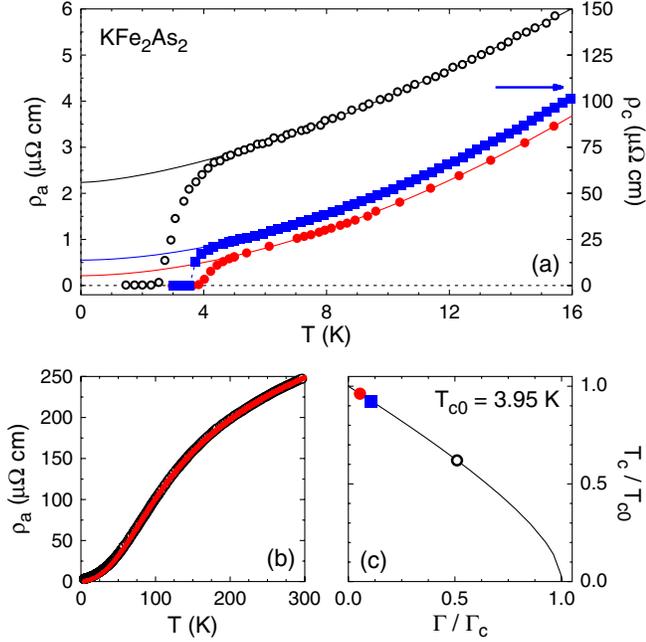


FIG. 1 (color online). (a) Electrical resistivity of the two samples of  $\text{KFe}_2\text{As}_2$  studied here, with  $J \parallel a$  (full red circles, left axis) and  $J \parallel c$  (full blue squares, right axis). Our  $a$ -axis data are compared to those of Dong *et al.* [7] (open circles, left axis), normalized here to have the same value at  $T = 300$  K (see text). The lines are a fit to  $\rho = \rho_0 + aT^\alpha$  from which we extrapolate  $\rho_0$  at  $T = 0$ . (b) Same data for the two  $a$ -axis samples, up to 300 K. (c) Abrikosov-Gorkov formula for the decrease of  $T_c$  with scattering rate  $\Gamma$  (line), used to obtain a value of  $\Gamma/\Gamma_c$  for the three samples of  $\text{KFe}_2\text{As}_2$ , given their  $T_c$  values and the factor 10 in  $\rho_0$  between the two  $a$ -axis samples (circles), assuming a disorder-free value of  $T_{c0} = 3.95$  K.

for the different  $\rho_0$ , the two resistivity curves  $\rho(T)$  are essentially identical [Fig. 1(b)]. Supporting evidence for a difference in impurity or defect concentration is the difference in critical temperature:  $T_c = 3.80 \pm 0.05$  K ( $2.45 \pm 0.10$  K) for our (their) sample. Assuming that the impurity scattering rate  $\Gamma \propto \rho_0$ , we can use the Abrikosov-Gorkov formula for the drop in  $T_c$  vs  $\Gamma$  to extract a value of  $\Gamma/\Gamma_c$  for the two samples, where  $\Gamma_c$  is the critical scattering rate needed to suppress  $T_c$  to zero [Fig. 1(c)]. We get  $\Gamma/\Gamma_c = 0.05$  (0.5) for our (their) sample.

The  $c$ -axis resistivity  $\rho_c(T)$  has the same temperature dependence as  $\rho_a(T)$  below  $T \approx 40$  K [Fig. 1(a)], with an intrinsic anisotropy  $\Delta\rho_c/\Delta\rho_a = 25 \pm 1$ , where  $\Delta\rho \equiv \rho(T) - \rho_0$ , with  $\rho_{c0} = 13 \pm 1 \mu\Omega$  cm. We attribute the larger anisotropy at  $T \rightarrow 0$ ,  $\rho_{c0}/\rho_{a0} = 60 \pm 10$ , to a larger  $\Gamma$  in our  $c$ -axis sample, consistent with the lower value of  $T_c$ , from which we deduce  $\Gamma/\Gamma_c = 0.1$  [Fig. 1(c)].

**Universal heat transport.**—The thermal conductivity is shown in Fig. 2. The residual linear term  $\kappa_0/T$  is obtained from a fit to  $\kappa/T = a + bT^\alpha$  below 0.3 K, where  $a \equiv \kappa_0/T$ . The dependence of  $\kappa_0/T$  on magnetic field  $H$  is shown in Fig. 3. Extrapolation to  $H = 0$  yields  $\kappa_{a0}/T = 3.6 \pm 0.5$  mW/K<sup>2</sup> cm and  $\kappa_{c0}/T = 0.18 \pm 0.03$  mW/K<sup>2</sup> cm.

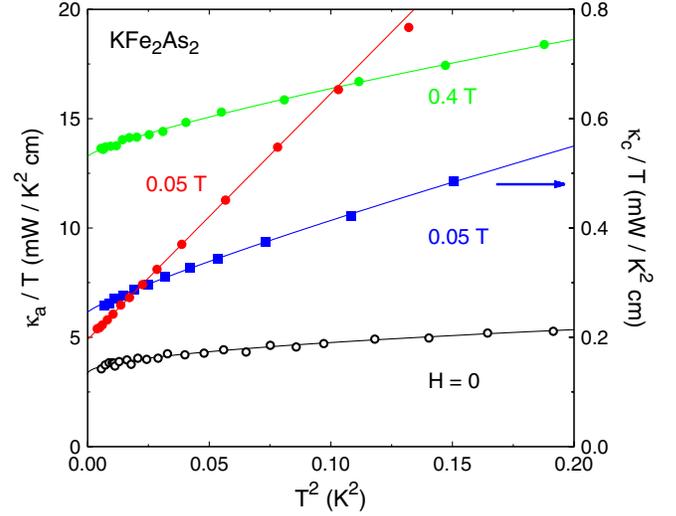


FIG. 2 (color online). Thermal conductivity of  $\text{KFe}_2\text{As}_2$ , plotted as  $\kappa/T$  vs  $T^2$ , for  $J \parallel a$  ( $\kappa_a$ , circles, left axis) and  $J \parallel c$  ( $\kappa_c$ , squares, right axis), for a magnetic field  $H \parallel c$  as indicated. Our  $a$ -axis data are compared to those of Dong *et al.* [7] (open circles, left axis), normalized by the same factor as in Fig. 1 (see text). Lines are a fit to  $\kappa/T = a + bT^\alpha$ , used to extrapolate the residual linear term  $a \equiv \kappa_0/T$  at  $T = 0$ . For our  $a$ -axis sample (full red circles),  $\alpha = 2.0$ , while for others  $\alpha < 2$ .

We compare these to Dong *et al.*'s [7] data, normalized by the same factor as for electrical transport, giving  $\kappa_{0a}/T = 3.32 \pm 0.03$  mW/K<sup>2</sup> cm. At  $H \rightarrow 0$ ,  $\kappa_{a0}/T$  is the same in the two samples (inset of Fig. 3), within error bars.

This universal heat transport, whereby  $\kappa_0/T$  is independent of the impurity scattering rate, is a classic signature of line nodes imposed by symmetry [18,19]. Calculations show the residual linear term to be independent of scattering rate and phase shift [18], and free of Fermi-liquid and vertex corrections [19]. For a quasi-2D  $d$ -wave superconductor [18,19],

$$\frac{\kappa_0}{T} \simeq \frac{\kappa_{00}}{T} \equiv \frac{\hbar}{2\pi} \frac{\gamma_N v_F^2}{\Delta_0}, \quad (1)$$

where  $\gamma_N$  is the residual linear term in the normal-state electronic specific heat,  $v_F$  is the Fermi velocity, and the superconducting gap  $\Delta = \Delta_0 \cos(2\phi)$  [22].

ARPES measurements on  $\text{KFe}_2\text{As}_2$  reveal a Fermi surface with three concentric holelike cylinders centered on the  $\Gamma$  point of the Brillouin zone, labeled  $\alpha$ ,  $\beta$ , and  $\gamma$ , and a 4th cylinder near the  $X$  point [23,24]. dHvA measurements detect all of these surfaces except the  $\beta$ , and obtain Fermi velocities in reasonable agreement with ARPES dispersions, with an average value of  $v_F \approx 4 \times 10^6$  cm/s [25]. The measured effective masses account for approximately 80% of the measured  $\gamma_N = 85 \pm 10$  mJ/K<sup>2</sup> mol [26,27]. In  $d$ -wave symmetry, the gap in  $\text{KFe}_2\text{As}_2$  will necessarily have nodes on all  $\Gamma$ -centered Fermi surfaces, and possibly on the  $X$ -centered surface as well [14]. The total  $\kappa_0/T$  may be estimated from Eq. (1) by using the average  $v_F$

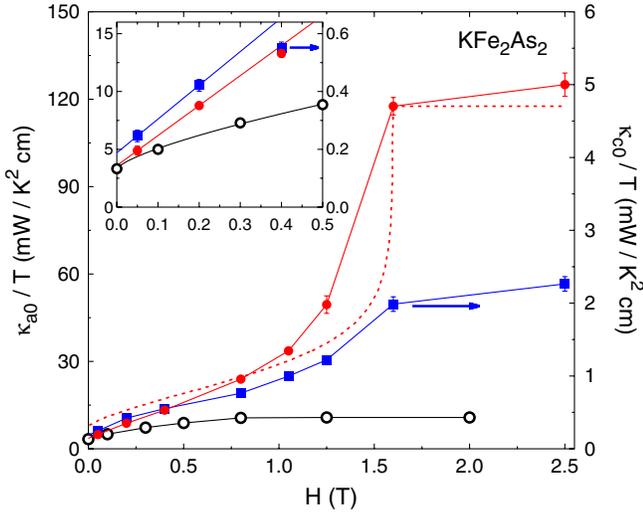


FIG. 3 (color online). Field dependence of  $\kappa_0/T$  obtained as in Fig. 2 (with corresponding symbols). The dashed line is a theoretical calculation for a  $d$ -wave superconductor with  $\hbar\Gamma/\Delta_0 = 0.1$  [38]. Inset: Zoom at low field. Lines are a power-law fit to extract the value of  $\kappa_0/T$  at  $H = 0$ .

and the measured (total)  $\gamma_N$ , which yields  $\kappa_{00}/T = 3.3 \pm 0.5$  mW/K<sup>2</sup> cm, assuming  $\Delta_0 = 2.14k_B T_{c0}$ , with  $T_{c0} = 3.95$  K. This is in excellent agreement with the experimental value of  $\kappa_0/T = 3.6 \pm 0.5$  mW/K<sup>2</sup> cm.

To compare with cuprates, the archetypal  $d$ -wave superconductors, we use Eq. (1) expressed directly in terms of  $v_\Delta$ , the slope of the gap at the node, namely,  $\kappa_{00}/T \simeq (k_B^2/3\hbar c)(v_F/v_\Delta)$ , with  $c$  the interlayer separation [18,19]. The ratio  $v_F/v_\Delta$  was measured by ARPES on Ba<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+ $\delta$</sub>  [28], giving  $v_F/v_\Delta \simeq 16$  at optimal doping, so that  $\kappa_{00}/T \simeq 0.16$  mW/K<sup>2</sup> cm. This is in excellent agreement with the experimental value of  $\kappa_0/T = 0.15 \pm 0.01$  mW/K<sup>2</sup> cm measured in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub> at optimal doping [29].

In Fig. 4(a), we plot  $\kappa_0/T$  vs  $\Gamma$  for both KFe<sub>2</sub>As<sub>2</sub> and YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>, the superconductor in which universal heat transport was first demonstrated [30]. We see that  $\kappa_0/T$  remains approximately constant up to at least  $\hbar\Gamma \simeq 0.5k_B T_{c0}$  in both cases. We conclude that both the magnitude of  $\kappa_0/T$  in KFe<sub>2</sub>As<sub>2</sub> and its insensitivity to impurity scattering are precisely those expected of a  $d$ -wave superconductor. By contrast, in an extended  $s$ -wave superconductor, there is no direct relation between  $\kappa_0/T$  and  $\Delta_0$ , and a strong nonmonotonic dependence on  $\Gamma$  is expected, since impurity scattering will inevitably make  $\Delta_0$  less anisotropic [16]. This is confirmed by calculations applied to pnictides, which typically find that  $\kappa_0/T$  vs  $\Gamma$  first rises and then plummets to zero when nodes are lifted by strong scattering [31] [see Fig. 4(a)].

**Critical scattering rate.**—In a  $d$ -wave superconductor, the critical scattering rate  $\Gamma_c$  is such that  $\hbar\Gamma_c \simeq k_B T_{c0}$  [32]. We can estimate  $\Gamma_c$  for KFe<sub>2</sub>As<sub>2</sub> from the critical value

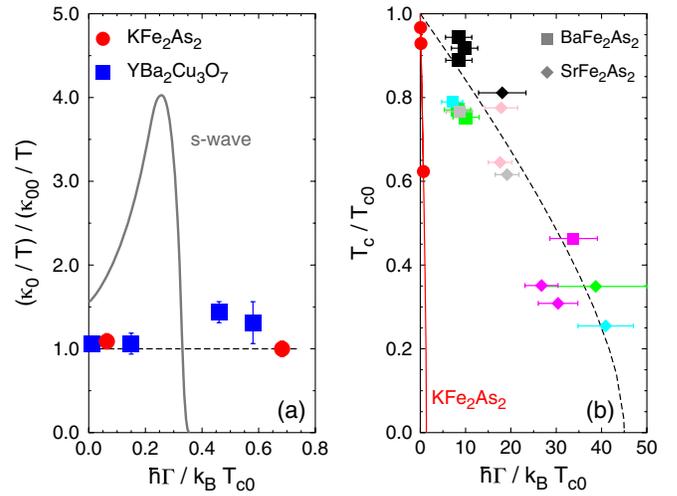


FIG. 4 (color online). Dependence of  $\kappa_0/T$  (a) and  $T_c$  (b) on impurity scattering rate  $\Gamma$ , normalized by  $T_{c0}$ , the disorder-free superconducting temperature. (a)  $\kappa_0/T$  for KFe<sub>2</sub>As<sub>2</sub> (red circles; see text) and the cuprate YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> (blue squares; from Ref. [30]), normalized by the theoretically expected value for a  $d$ -wave superconductor,  $\kappa_{00}/T = 3.3$  and  $0.16$  mW/K<sup>2</sup> cm, respectively (see text). The typical dependence expected of an  $s$ -wave state with accidental nodes is also shown, from a calculation applied to pnictides (black line; from Ref. [31]). (b)  $T_c$  for KFe<sub>2</sub>As<sub>2</sub> [red circles; from Fig. 1(c)] and for the pnictides BaFe<sub>2</sub>As<sub>2</sub> and SrFe<sub>2</sub>As<sub>2</sub> at optimal doping (from Ref. [20]).

of  $\rho_0$ , evaluated as twice that for which  $\Gamma/\Gamma_c = 0.5$  in Fig. 1(c), namely,  $\rho_0^{\text{crit}} \simeq 4.5 \mu\Omega$  cm. Using  $L_0/\rho_0^{\text{crit}} = \gamma_N v_F^2 \tau_c/3$ , where  $L_0 \equiv (\pi^2/3)(k_B/e)^2$ , we get  $\hbar\Gamma_c = \hbar/2\tau_c \simeq 1.3 \pm 0.2k_B T_{c0}$ , in excellent agreement with the expectation for a  $d$ -wave state. By contrast,  $\hbar\Gamma_c/k_B T_{c0} \simeq 45$  in BaFe<sub>2</sub>As<sub>2</sub> and SrFe<sub>2</sub>As<sub>2</sub> at optimal Co, Pt, or Ru doping [20] [see Fig. 4(b)]. This factor 30 difference in the sensitivity of  $T_c$  to impurity scattering is proof that the pairing symmetry of KFe<sub>2</sub>As<sub>2</sub> is different from the  $s$ -wave symmetry of Co-doped BaFe<sub>2</sub>As<sub>2</sub> [6].

**Direction dependence.**—In the case of a  $d$ -wave gap on a single quasi-2D cylindrical Fermi surface (at the zone center), the gap would necessarily have four line nodes that run vertically along the  $c$  axis. In such a nodal structure, zero-energy nodal quasiparticles will conduct heat not only in the plane but also along the  $c$  axis by an amount proportional to the  $c$ -axis dispersion of the Fermi surface. In the simplest case,  $c$ -axis conduction will be smaller than  $a$ -axis conduction by a factor equal to the mass tensor anisotropy [ $v_F^2$  in Eq. (1)]. In other words,  $(\kappa_{a0}/T)/(\kappa_{c0}/T) \simeq (\kappa_{aN}/T)/(\kappa_{cN}/T) = (\sigma_{aN})/(\sigma_{cN})$ , the anisotropy in the normal-state thermal and electrical conductivities, respectively. This is confirmed by calculations for a quasi-2D  $d$ -wave superconductor [33], whose vertical line nodes yield an anisotropy ratio in the superconducting state very similar to that of the normal state. This is what we see in KFe<sub>2</sub>As<sub>2</sub> (inset of Fig. 3):  $(\kappa_{a0}/T)/(\kappa_{c0}/T) = 20 \pm 4$ , very close to the intrinsic normal-state anisotropy

$(\sigma_{aN})/(\sigma_{cN}) = (\Delta\rho_c)/(\Delta\rho_a) = 25 \pm 1$ . This shows that the nodes in  $\text{KFe}_2\text{As}_2$  are vertical lines running along the  $c$  axis, ruling out proposals [11] of horizontal line nodes lying in a plane normal to the  $c$  axis.

Moreover, the fact that the Fermi surface of  $\text{KFe}_2\text{As}_2$  contains several sheets with very different  $c$ -axis dispersions [25,34] provides compelling evidence in favor of  $d$ -wave symmetry. In an extended  $s$ -wave scenario, the gap would typically develop vertical line nodes on some but not all zone-centered sheets of the Fermi surface [15], and so the anisotropy in  $\kappa$  would typically be very different in the superconducting and normal states, unlike what is measured. By contrast, in  $d$ -wave symmetry all zone-centered sheets must necessarily have nodes, thereby ensuring automatically that transport anisotropy remains similar in the superconducting and normal states.

*Temperature dependence.*—So far, we have discussed the limits  $T \rightarrow 0$  and  $H \rightarrow 0$ , where nodal quasiparticles are excited only by the pair-breaking effect of impurities. Raising the temperature will further excite nodal quasiparticles. Calculations for a  $d$ -wave superconductor show that the electronic thermal conductivity grows as  $T^2$  [18,22]:

$$\frac{\kappa}{T} \simeq \frac{\kappa_{00}}{T} \left( 1 + a \frac{T^2}{\gamma^2} \right), \quad (2)$$

where  $a$  is a dimensionless number and  $\hbar\gamma$  is the impurity bandwidth, which grows with the scattering rate  $\Gamma$  [18]. A  $T^2$  slope in  $\kappa/T$  was resolved in  $\text{YBa}_2\text{Cu}_3\text{O}_7$  [29].

Our  $\text{KFe}_2\text{As}_2$  sample shows a clear  $T^2$  dependence below  $T \simeq 0.3$  K, with  $\kappa_a/T = (\kappa_{a0}/T)(1 + 23T^2)$  (Fig. 2). Comparison with Dong *et al.*'s [7] data reveals that this  $T^2$  term must be due to quasiparticles. Indeed, because phonon conduction at sub-Kelvin temperatures is limited by sample boundaries and not impurities [35], the fact that the slope of  $\kappa/T$  in their sample (of similar dimensions) is at least 10 times smaller (Fig. 2) implies that the larger slope in our data must be electronic.

In the limit of unitary scattering,  $\gamma^2 \propto \Gamma$ , so that a 10 times larger  $\Gamma$  would yield a 10 times smaller  $T^2$  slope [18], consistent with the observation. The temperature below which the  $T^2$  dependence of  $\kappa_e/T$  sets in,  $T \simeq 0.1T_c$ , is a measure of  $\gamma$ . It is in excellent agreement with the temperature below which the penetration depth  $\lambda_a(T)$  of  $\text{KFe}_2\text{As}_2$  (in a sample with similar RRR) deviates from its linear  $T$  dependence [8], as expected of a  $d$ -wave superconductor [36]. Note that the  $T$  dependence of  $\kappa/T$  for an extended  $s$ -wave gap is not  $T^2$  [31].

*Magnetic field dependence.*—Increasing the magnetic field is another way to excite quasiparticles. If the gap has nodes, the field will cause an immediate rise in  $\kappa_0/T$  [17,37,38], as observed in all three samples of  $\text{KFe}_2\text{As}_2$  (inset of Fig. 3). Calculations for a  $d$ -wave superconductor in the clean limit ( $\hbar\Gamma \ll k_B T_c$ ) yield a nonmonotonic

increase of  $\kappa_0/T$  vs  $H$  [38] in remarkable agreement with data on the clean sample (Fig. 3).

A rapid initial rise in  $\kappa_0/T$  vs  $H$  has been observed in the cuprate superconductors  $\text{YBa}_2\text{Cu}_3\text{O}_7$  [39] and  $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+\delta}$  [40]. In the dirty limit,  $\text{KFe}_2\text{As}_2$  [7] and  $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+\delta}$  [40] show nearly identical curves of  $\kappa_0/T$  vs  $H/H_{c2}$  (see Ref. [7]). Measurements on cuprates in the clean limit, such as optimally doped  $\text{YBa}_2\text{Cu}_3\text{O}_y$ , have so far been limited to  $H \ll H_{c2}$ .

In summary, all aspects of the thermal conductivity of  $\text{KFe}_2\text{As}_2$ , including its dependence on impurity scattering, current direction, temperature, and magnetic field, are in detailed and quantitative agreement with theoretical calculations for a  $d$ -wave superconductor. The scattering rate needed to suppress  $T_c$  to zero is exactly as expected of  $d$ -wave symmetry, and vastly smaller than that found in other pnictide superconductors where the gap is believed to have an  $s$ -wave symmetry. This is compelling evidence that, for this iron arsenide superconductor, the gap has a  $d$ -wave symmetry, in agreement with renormalization-group calculations [14]. Replacing K in  $\text{KFe}_2\text{As}_2$  by Ba leads to a superconducting state with a 10 times higher  $T_c$ , but with a full gap without nodes [4], necessarily of a different symmetry. Understanding the relation between this factor 10 and the pairing symmetry is expected to provide insight into what boosts  $T_c$  in these systems.

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*Note added in proof.*—By adding Co impurities in  $\text{KFe}_2\text{As}_2$ , a recent study [41] has confirmed that  $T_c$  falls rapidly to zero with impurity scattering, roughly at  $\rho_{\text{crit}} = 4.5 \mu\Omega \text{ cm}$ , and the residual linear term in the thermal conductivity is indeed universal, remaining approximately constant even when the normal-state conductivity is decreased by a factor of 30.

\*louis.taillefer@physique.usherbrooke.ca

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