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Universal Heat Conduction in the Iron-Arsenide Superconductor KFe_2As_2 : Evidence of a d-wave State

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The thermal conductivity κ of the iron-arsenide superconductor KFe_2As_2 was measured down to 50 mK for a heat current parallel and perpendicular to the tetragonal c axis. A residual linear term at $T \to 0$, κ_0/T, is observed for both current directions, confirming the presence of nodes in the superconducting gap. Our value of κ_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 ρ_0 was ten times larger. This independence of κ_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 Γ_c for suppressing T_c to zero; the magnitude of κ_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 ($\pm k_x = \pm k_y$). 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 $BaFe_2(As_{1-x}P_x)_2$ [1] and $Ba(Fe_{1-x}Ru_x)_2As_2$ [2], and not in others, as in $Ba_{1-x}K_xFe_2As_2$ [3, 4] and $Ba(Fe_{1-x}Co_x)_2As_2$ [5, 6] at optimal doping.

In KFe_2As_2, the end-member of the $Ba_{1-x}K_xFe_2As_2$ 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 KFe_2As_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 theoretical expectation for a d-wave gap [18, 19], and inconsistent with accidental line nodes, whether vertical or horizontal. Moreover, the critical scattering rate Γ_c for suppressing T_c to zero is of order T_{c0}, as expected for d-wave, while it is 50 times T_{c0} in optimally-doped $BaFe_2As_2$ [20].

Methods.– Single crystals of KFe_2As_2 were grown from self flux [21]. Two samples were measured: one for currents along the a axis, 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 ± 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 KFe_2As_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. 1a). 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 $\rho_0 = 0.21 \pm 0.02$ $\mu\Omega$ cm (2.24 ± 0.05 $\mu\Omega$ cm) for our (their) sample, so that $\rho(300\text{ K})/\rho_0 = 1180$ and 110, respectively.

We attribute the lower ρ_0 in our sample to a lower concentration of impurities or defects. Note that except for the different ρ_0, the two resistivity curves $\rho(T)$ are essentially identical (Fig. 1b). Supporting evidence for a difference in impurity/defect concentration is the difference

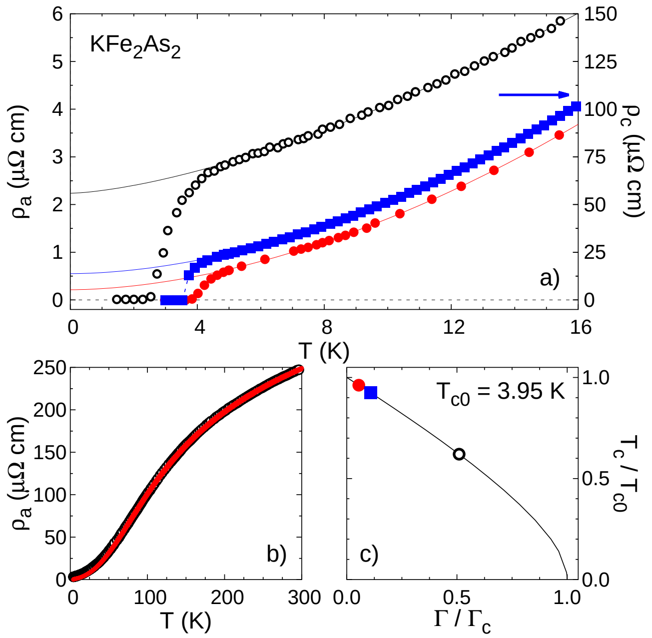

FIG. 1: (a) Electrical resistivity of the two samples of KFe_2As_2 studied here, with $J \parallel a$ (full red circles, left axis) and $J \parallel c$ (full blue squares, right axis). Our a-axis data is compared to that of Dong *et al.* [7] (open circles, left axis), normalized here to have the same value at $T = 300$ K (see text). The lines are a fit to $\rho = \rho_0 + aT^\alpha$ from which we extrapolate ρ_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 Γ (line), used to obtain a value of Γ/Γ_c for the three samples of KFe_2As_2, given their T_c values and the factor 10 in ρ_0 between the two a-axis samples (circles), assuming a disorder-free value of $T_{c0} = 3.95$ K.

in critical temperature: $T_c = 3.80 \pm 0.05$ K (2.45 ± 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 Γ to extract a value of Γ/Γ_c for the two samples, where Γ_c is the critical scattering rate needed to suppress T_c to zero (Fig. 1c). 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 $T \simeq 40$ K (Fig. 1a), 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 $T \to 0$, $\rho_{c0}/\rho_{a0} = 60 \pm 10$, to a larger Γ in our c-axis sample, consistent with the lower value of T_c, from which we deduce $\Gamma/\Gamma_c = 0.1$ (Fig. 1c).

Universal heat transport.– The thermal conductivity is shown in Fig. 2. The residual linear term κ_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 κ_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^2 cm and $\kappa_{c0}/T = 0.18 \pm 0.03$ mW/K^2 cm. We compare to the data by Dong *et al.* [7], normalized by the same factor as for elec-

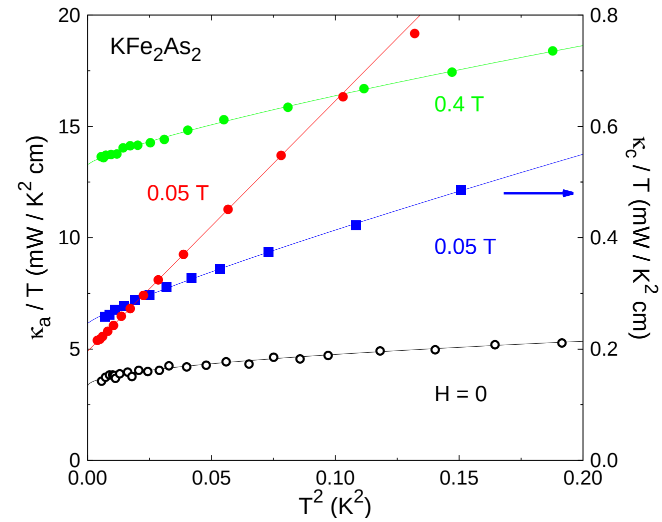

FIG. 2: Thermal conductivity of KFe_2As_2, plotted as κ/T vs T^2, for $J \parallel a$ (κ_a, circles, left axis) and $J \parallel c$ (κ_c, squares, right axis), for a magnetic field $H \parallel c$ as indicated. Our a-axis data is compared to that of Dong *et al.* [7] (open circles, left axis), normalized 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 $\alpha < 2$.

trical transport, giving $\kappa_{0a}/T = 3.32 \pm 0.03$ mW/K^2 cm. At $H \to 0$, κ_{a0}/T is the same in the two samples (inset of Fig. 3), within error bars.

This universal heat transport, whereby κ_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]:

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

where γ_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 KFe_2As_2 reveal a Fermi surface with three concentric hole-like cylinders centered on the Γ point of the Brillouin zone, labeled α, β and γ, and a 4th cylinder near the X point [23, 24]. dHvA measurements detect all of these surfaces except the β, and obtain Fermi velocities in reasonable agreement with ARPES dispersions, with an average value of $v_F \simeq 4 \times 10^6$ cm/s [25]. The measured effective masses account for approximately 80% of the measured $\gamma_N = 85 \pm 10$ mJ/K^2 mol [26, 27]. In d-wave symmetry, the gap in KFe_2As_2 will necessarily have nodes on all Γ-centered Fermi surfaces, and possibly on the X-centered surface as well [14]. The total κ_0/T may be estimated

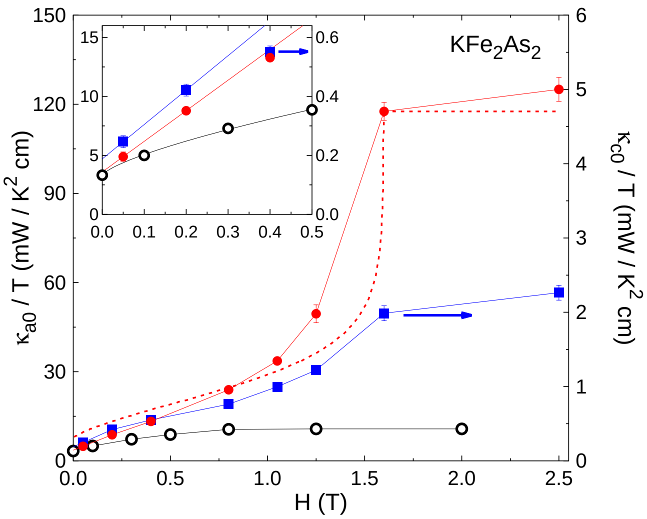

FIG. 3: Field dependence of κ_0/T obtained as in Fig. 2 (with corresponding symbols). The dashed line is a theoretical calculation for a d-wave superconductor with $\hbar\Gamma/\Delta_0 = 0.1$ [38]. *Inset*: Zoom at low field. Lines are a power-law fit to extract the value of κ_0/T at $H = 0$.

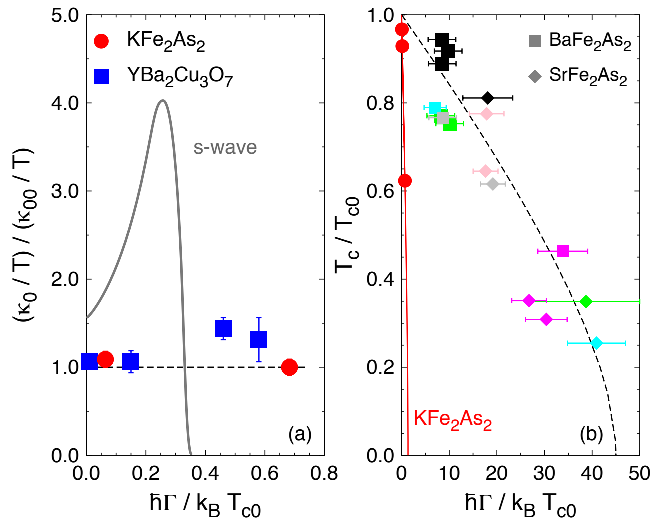

FIG. 4: Dependence of κ_0/T (a) and T_c (b) on impurity scattering rate Γ, normalized by T_{c0}, the disorder-free superconducting temperature. (a) κ_0/T for KFe_2As_2 (red circles; see text) and the cuprate $YBa_2Cu_3O_7$ (blue squares; from ref. 30), normalized by the theoretically expected value for a d-wave superconductor, $\kappa_{00}/T = 3.3$ and 0.16 mW/K^2 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_2As_2 (red circles; from Fig. 1c) and for the pnictides $BaFe_2As_2$ and $SrFe_2As_2$ at optimal doping (from ref. 20).

from Eq. 1 by using the average v_F and the measured (total) γ_N, which yields $\kappa_{00}/T = 3.3 \pm 0.5$ mW/K^2 cm, assuming $\Delta_0 = 2.14\ k_B T_{c0}$, 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^2 cm.

To compare with cuprates, the archetypal d-wave superconductors, we use Eq. 1 expressed directly in terms of v_Δ, 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_Δ was measured by ARPES on $Ba_2Sr_2CaCu_2O_{8+\delta}$ [28], giving $v_F/v_\Delta \simeq 16$ at optimal doping, so that $\kappa_{00}/T \simeq 0.16$ mW/K^2 cm. This is in excellent agreement with the experimental value of $\kappa_0/T = 0.15 \pm 0.01$ mW/K^2 cm measured in $YBa_2Cu_3O_y$ at optimal doping [29].

In Fig. 4a, we plot κ_0/T vs Γ for both KFe_2As_2 and $YBa_2Cu_3O_7$, the superconductor in which universal heat transport was first demonstrated [30]. We see that κ_0/T remains approximately constant up to at least $\hbar\Gamma \simeq 0.5\ k_B T_{c0}$ in both cases. We conclude that both the magnitude of κ_0/T in KFe_2As_2 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 κ_0/T and Δ_0, and a strong non-monotonic dependence on Γ is expected, since impurity scattering will inevitably make Δ_0 less anisotropic [16]. This is confirmed by calculations applied to pnictides, which typically find that κ_0/T vs Γ first rises, and then plummets to zero when nodes are lifted by strong scattering [31] (see Fig. 4a).

Critical scattering rate.– In a d-wave superconductor, the critical scattering rate Γ_c is such that $\hbar\Gamma_c \simeq k_B T_{c0}$ [32]. We can estimate Γ_c for KFe_2As_2 from the critical value of ρ_0, evaluated as twice that for which $\Gamma/\Gamma_c = 0.5$ in Fig. 1c, namely $\rho_0^{\rm crit} \simeq 4.5\ \mu\Omega$ cm. Using $L_0/\rho_0^{\rm crit} = \gamma_N v_F^2 \tau_c/3$, where $L_0 \equiv (\pi^2/3)(k_B/e)^2$, we get $\hbar\Gamma_c = \hbar/2\tau_c \simeq 1.3 \pm 0.2\ k_B T_{c0}$, in excellent agreement with expectation for a d-wave state. By contrast, $\hbar\Gamma_c/k_B T_{c0} \simeq 45$ in $BaFe_2As_2$ and $SrFe_2As_2$ at optimal Co, Pt or Ru doping [20] (see Fig. 4b). This factor 30 difference in the sensitivity of T_c to impurity scattering is proof that the pairing symmetry of KFe_2As_2 is different from the s-wave symmetry of Co-doped $BaFe_2As_2$ [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 4 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 [34], 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_2As_2 (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 KFe_2As_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 KFe_2As_2 contains several sheets with very different c-axis dispersions [25, 35] 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 κ 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 limit $T \to 0$ and $H \to 0$, 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]:

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

where a is a dimensionless number and $\hbar\gamma$ is the impurity bandwidth, which grows with the scattering rate Γ [18]. A T^2 slope in κ/T was resolved in $YBa_2Cu_3O_7$ [29].

Our KFe_2As_2 sample shows a clear T^2 dependence below $T \simeq 0.3$ K, with $\kappa_a/T = (\kappa_{a0}/T)(1+23\,T^2)$ (Fig. 2). Comparison with the data by Dong *et al.* [7] 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 [33], the fact that the slope of κ/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 Γ would yield a 10-times smaller T^2 slope [18], consistent with observation. The temperature below which the T^2 dependence of κ_e/T sets in, $T \simeq 0.1\ T_c$, is a measure of γ. It is in excellent agreement with the temperature below which the penetration depth $\lambda_a(T)$ of KFe_2As_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 κ/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 κ_0/T [17, 37, 38], as observed in all three samples of KFe_2As_2 (inset of Fig. 3). Calculations for a d-wave superconductor in the clean limit ($\hbar\Gamma \ll k_B T_c$) yield a non-monotonic increase of κ_0/T vs H [38] in remarkable agreement with data on the clean sample (Fig. 3).

A rapid initial rise in κ_0/T vs H has been observed in the cuprate superconductors $YBa_2Cu_3O_7$ [39] and $Tl_2Ba_2CuO_{6+\delta}$ [40]. In the dirty limit, KFe_2As_2 [7] and $Tl_2Ba_2CuO_{6+\delta}$ [40] show nearly identical curves of κ_0/T vs H/H_{c2} (see ref. 7). Measurements on cuprates in the clean limit, such as optimally-doped $YBa_2Cu_3O_y$, have so far been limited to $H \ll H_{c2}$.

In summary, all aspects of the thermal conductivity of KFe_2As_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 pairing symmetry is believed to be s-wave. This is compelling evidence that the pairing symmetry in this iron-arsenide superconductor is d-wave, in agreement with renormalization-group calculations [14]. Replacing K in KFe_2As_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 provides insight into what boosts T_c in these systems.

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