Evidence of $d$-wave Superconductivity in $K_{1-x}$Na$_x$Fe$_2$As$_2$ ($x = 0, 0.1$) Single Crystals from Low-Temperature Specific Heat Measurements

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From the measurement and analysis of the specific heat of high-quality $K_{1-x}$Na$_x$Fe$_2$As$_2$ single crystals we establish the presence of large $T^2$ contributions with coefficients $\alpha_{sc} \approx 30 \text{ mJ/mol K}^3$ at low-$T$ for both $x = 0$ and 0.1. Together with the observed $\sqrt{T}$ behavior of the specific heat in the superconducting state both findings evidence $d$-wave superconductivity on almost all Fermi surface sheets with an average gap amplitude of $\Delta_0$ in the range of 0.4 - 0.8 meV. The derived $\Delta_0$ and observed $T_c$ agree well with the values calculated within Eliashberg theory, adopting a spin fluctuation mediated pairing in the intermediate coupling regime.

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In spite of the substantial experimental and theoretical research efforts to elucidate the symmetry and magnitude of the superconducting order parameter for the Fe pnictides $\text{1, 2}$, important questions concerning the doping evolution of the superconducting gap remain unsolved $\text{3, 4}$. In the stoichiometric parent compounds nesting usually occurs between electron (el) and hole (h) Fermi surface sheets (FSS) which is responsible for the presence of long-range spin density wave (SDW) order. Superconductivity (SC) emerges when the SDW order is suppressed by doping or external pressure $\text{2}$. An $s_{\pm}$ gap symmetry (nodeless gap function with opposite signs of the order parameters for el and h pockets) is believed to be realized in under- and optimally doped compounds, since the antiferromagnetic spin fluctuations (SF) on the vector $Q = (\pi, \pi)$ connecting the el and h pockets remain strong in the vicinity of the SDW phase. The situation in the overdoped regime is not so clear. With further doping, el (h) bands disappear. Therefore, the paradigm of the SF glue at the vector $Q = (\pi, \pi)$ does not work. However, SF have been found at some incommensurate propagation vectors $\text{5}$. This has led to several proposals for the order parameters in heavily doped compounds: extended $s, d, s + id$ wave $\text{2, 3, 5}$. Thus, even from the theoretical perspective the situation is still controversial.

One of the most interesting families from this point of view is Ba$_{1-x}$K$_x$Fe$_2$As$_2$. Superconductivity in this compound occurs at $x \approx 0.15$ $\text{2}$. $T_c$ increases with K doping up to $T_c \approx 38$ K for $x=0.4$ (optimally doped regime). In this region experiments such as angle-resolved photoemission spectroscopy (ARPES) $\text{10}$ and thermal conductivity $\text{11}$ show the absence of nodes in the gap, confirming an $s$-wave character of the order parameter. The evolution of the SC order parameter at high hole-doping levels is not well studied, so far. The measurements on polycrystalline samples show that $T_c$ monotonically decreases with doping, reaching $T_c = 3.5$ K for KFe$_2$As$_2$ (K122) $\text{4}$. Whereas the el pockets are completely gone in K122, it was theoretically proposed that a change of the order-parameter symmetry to $d, s_{\pm}$ wave with accidental nodes, or $s + id$ should occur $\text{3, 5, 6}$.

Until now, there is no consensus in the interpretation of available experimental data in favor of one of the proposed order parameters. Indeed, measurements of thermal conductivity $\text{12, 13}$ and of the London penetration depth $\text{14}$ have been interpreted in terms of $d$-wave SC with line-nodes on each FSS. In contrast, recent ultrahigh-resolution laser ARPES data for K122 have been interpreted in terms of a rather specific partial nodal $s$-wave SC having an unusual gap with “octet line-nodes” on the middle FSS, an almost-zero gap on the outer FSS, and a nodeless gap on the inner FSS $\text{15}$. The specific-heat (SH) data for K122 above 0.4 K, only, were interpreted within a weak-coupling BCS-like multiple SC gap scenario with line nodes $\text{16, 17}$, adopting multiband- $d$- or $s$-wave nodal SC. But at the same time an extremely strong coupling and/or correlated regimes with heavy quasiparticles were suggested for the normal state. Moreover, the authors of a subsequent SH study...
in magnetic fields \[18\] doubted such an interpretation and denied the presence of a smaller second gap based on the observation of a non-intrinsic magnetic transition at low-\(T\), probably due to unknown magnetic impurities. Naturally, under such circumstances a \(T^2\) and/or a \(\sqrt{T}\) behavior, generic for line-nodes in the SC gap \[13, 21\], could not be observed. Hence, further studies are necessary to clarify the symmetry of the SC order parameter and the magnitude of the coupling strength. These two issues are the main points of the present letter.

\(K_{1-x}Na_xFe_2As_2\) single crystals with \(x = 0\) [K122] and \(x = 0.1\) [(K,Na)122] were grown using KAs as a flux. SH data in various fields were obtained by a relaxation technique in a Physical Properties Measurement System (PPMS, Quantum Design). The magnetic ac susceptibility as a function of temperature was also measured by use of the PPMS. The \(T\)-dependence of the electrical resistivity was measured using a standard 4-probe DC technique. Two single crystals, K122 and (K,Na)122, were selected for SH measurements below 0.4 K using the heat-pulse technique within a dilution refrigerator.

The \(T\)-dependence of the volume ac susceptibilities (\(\chi'\) and \(\chi''\)) of K122 and (K,Na)122 are shown in Fig. 1. The sharp transition with \(4\pi\chi' \sim 1\) confirms the bulk nature of superconductivity and the high quality of our crystals. The \(T\)-dependence of the in-plane electrical resistivity evidences a drop to zero at 3.6 K for K122 and at 2.9 K for (K,Na)122 in agreement with the diamagnetic onset seen in the \(\chi_{AC}\) data. The resistivity ratio \(RR = \rho(300K)/\rho(5K)\) is found to be \(\approx 120\) for K(Na)122 with \(\rho(5K) \approx 3.82\mu\Omega\text{cm}\) which is smaller than \(RR \approx 380\) for K122 with \(\rho(5K) \approx 1.44\mu\Omega\text{cm}\). Therefore, the reduced \(T_c\) of (K,Na)122 can be attributed to the pair-breaking effects due to disorder induced by Na doping.

The zero-field specific-heat, \(C\), of K122 and K(Na)122 between 0.1 and 10 K is shown in Fig. 2. A clear sharp SC anomaly is observed at \(T_c \approx 2.75\text{K}\) for (K,Na)122 and at \(T_c \approx 3.5\text{K}\) for K122, in line with the resistivity and ac susceptibility data. The jump height of \(C\) at \(T_c\) is found to be \(\Delta C/T_c \approx 40\text{mJ/mol K}^2\) for (K,Na)122. This value is a bit reduced from \(\approx 46\text{mJ/mol K}^2\) for K122. A zoom into the low-\(T\) range is shown in the inset of Fig. 2. We observe a pronounced \(T^2\)-behavior in the SH for both crystals. Notice that no kinks and/or small hump anomalies, predicted for the low-\(T\) region by weakly coupled two-band approaches \[16, 17\], have been observed in our measurements. The field dependence of \(C/T\) at \(T = 0.5\text{K}\) obtained from SH measurements in magnetic fields \[22\] are shown in Fig. 3. The observed \(\sqrt{B}\) dependence together with the \(T^2\) behavior provides a direct evidence for a \(d\)-wave SC order parameter of \(K_{1-x}Na_xFe_2As_2\), at least for \(x \leq 0.1\).

The \(T\)-dependence of the SH in the SC state allows one to determine whether the gap function possesses nodes.

**FIG. 1:** (Color online) Temperature dependence of the complex volume ac susceptibility \(\chi_{AC} = \chi' + i\chi''\) measured with an amplitude of 5 Oe and a frequency of \(\nu = 1\text{kHz}\). Inset: the normalized in-plane resistivity \(\rho\) measured in zero-field.

**FIG. 2:** (Color online) Specific heat, \(C\), plotted as \(C/T\) vs. \(T\). Dashed lines: Fit in the normal state (see text). Inset: zoom into the low-\(T\) region, solid lines: Fits using Eq. 3. The data for K122 above 0.4 K are taken from Ref. 24.

**FIG. 3:** (Color online) Field dependence of the specific heat, \(C/T\), at \(0.5\text{K}\). Solid lines: Linear fits to define \(\Delta C_{SC}\) (see Eq. 3). Inset: The scaled upper critical fields for \(KFe_2As_2\) \[22\] and \(K_{0.5}Na_{0.1}Fe_2As_2\) vs. \(T/T_c\).
For instance, the exponential vanishing of $C$ as in conventional $s$-wave SC is caused by the finite gap in the quasiparticle spectrum. In contrast, in the case of gap nodes quasiparticles are generated to the largest extent in the vicinity of the gap nodes. In the case of line-nodes the quasiparticle excitation spectrum takes the form $E_k = \hbar \sqrt{\nu^2 k^2 + v^2 k^2}$, where $\nu$ and $v$ are wavevectors perpendicular and parallel to the FSS, respectively, $v_F$ is the renormalized in-plane Fermi velocity at the position of the node, while $v_\Delta \approx \partial \Delta / \hbar k$ is the slope of the gap at the node associated with the dispersion of the quasiparticles along the FS. This leads to a density of states (DOS) linear in energy. For the two-dimensional (2D) case it reads:

$$N_{SC}(E) = \sum_i \frac{E}{\pi \hbar^2 \nu_F^2 \nu_\Delta},$$

where $i$ denotes the sum over all nodes. Here, we would like to note that both materials considered are highly anisotropic, justifying the 2D approximation. The ratio of $H_{c2}$ (in-plane vs. out-of-plane) according to inset Fig. 3 suggests a large mass ratio $T^2 \approx 25$. This value agrees with DFT predictions for the ratio of the corresponding squared plasma frequencies $22$.

Eq. (1) results in a quadratic power-law dependence $C_n \propto \alpha_{sc} T^2$, with

$$\alpha_{sc} (\text{mJ/mol K}^3) \approx 0.283 \frac{\nu_F \text{(mJ/mol K}^2)}{\Delta_0 (\text{meV})}$$

(2)

taken from Refs. 14, 20, 28 and valid for SC if both the impurity scattering rate and magnetic field scales are smaller than the temperature. Therefore, the observed $T^2$ behavior suggests that this condition is fulfilled for both crystals studied. Using Eq. (1) we also assumed that the energy gap at $T = 0$, $\Delta_0$, is equal for all bands, while $\gamma_{el}$ is a renormalized normal-state Sommerfeld coefficient, which is an average value over all FSS (see the Supplement for details 22). In this context, $\nu$ is a dimensionless factor taking into account the anisotropy of $1/v_F$ at the position of the nodes with respect to the averaged isotropic value. According to our preliminary ab initio calculations for the $k$-dependence of the DOS near the Fermi energy in the nodal direction, and the expected amount of the anisotropic mass renormalization due to anisotropic SF, $\nu \sim 1.1$ to 1.15. This range of values arises from the dominant contribution of the two inner FSS around the $\Gamma$-point in the Brillouine zone, which is somewhat reduced by a different antinodal anisotropy of the third FSS. The DOS of these three FSS is about 90% to the total DOS. The resulting anisotropy is also in accord with the slightly larger gap value obtained in our Eliashberg-theory based calculations (see below) than the value derived from Eq. (2).

To analyze the low-$T$ SH data we use the ansatz:

$$C = \gamma_T T + \alpha T^2 + \beta_3 T^3,$$

(3)

where the cubic term $\beta_3$ comes from phonons, which is defined from the data in the normal state. The remaining terms stem from a magnetic cluster-glass contribution reported previously in Ref. 26 and from electronic degrees of freedom, which is the focus here. The best fit using Eq. (3) at $T \leq 0.4K$ yields $\gamma_T = 44.8 \text{ mJ/mol K}^2$ and $\alpha = 33.7 \text{ mJ/mol K}^2$ for K122 and $\gamma_T = 55.2 \text{ mJ/mol K}^2$ and $\alpha = 35.4 \text{ mJ/mol K}^2$ for (K,Na)122. It has been shown in Ref. 26 that the residual "Sommerfeld" coefficient $\gamma_T$ of K122 mainly stems from a cluster-glass phase. At variance with K122, for (K,Na)122 an additional contribution to $\gamma_T$, exceeding 10 mJ/mol K$^2$, caused by a disorder-induced pair-breaking effect 19 in accord with the enhanced residual resistivity of this crystal. However, we emphasize that the $T^2$ contribution measured by $\alpha$ is not an extrinsic "glassy" contribution but stems from the generic electronic contribution in the superconducting state only. The estimated contribution of the cluster glass amounts $\alpha_{CG} \approx 2 \text{ mJ/mol K}^3$ for K122 and $\alpha_{CG} \approx 0$ for K(Na)122 22, 26. Hence, we argue that our observed large $T^2$ contribution to the low $T$ SH provides evidence of the $d$-wave character of the gap function.

In the case of a $d$-wave SC order parameter $C(B)/T = P_{SC}(B/C(0)/T$ holds, if the energy associated with the Doppler shift $E_H = h \nu_F (\pi B/\Phi_0)^{0.5}$ is large as compared to $E_T = k_B T$ 28, where $\Phi_0$ is the flux quantum and $\nu_F = 4 \cdot 10^6 \text{ cm/s}$ 24. According to our data shown in Fig. 3 for $T = 0.5 \text{ K}$ the field dependence of $C/T$ follows a $\sqrt{B}$-law at $B > 0.06 \text{ T}$ which corresponds to $E_H/E_T > 3$ in accord with the theoretical prediction 28. Using the expressions (66, 67) in Ref. 28 for $P_{SC}$ and $\alpha_{sc}$ we get:

$$P_{SC}/\alpha_{sc} \approx \frac{\pi^{5/2} h M_1 \nu_F}{54 \zeta(3) k_B \Phi_0^{0.5}} \approx 1.8 M_1 (K/T^{0.5}),$$

(4)

where $M_1 \approx 1$ for a liquid vortex state and $M_1 = 1/\pi^{0.5} \approx 0.56$ for the disordered distribution 28. This gives $P_{SC}$ in the range of $30 - 60 \text{ mJ/mol K}^2 T^{0.5}$ in accord with the experimental values $\approx 48 \text{ mJ/mol K}^2 T^{0.5}$ for K122 and $\approx 42 \text{ mJ/mol K}^2 T^{0.5}$ for (K,Na)122.

Now, we will check whether the value of $\alpha_{sc}$ can be satisfactorily described by the standard $d$-wave mechanism on all FSS. Our analysis is based on Eq. (4), which was obtained within a BCS-like theory. But it works very well also in the case of strongly coupled cuprates 24. Therefore, we expect its validity for pnictides, too. In addition, we analyse the relevant coupling constants with the help of the strong-coupling Eliashberg equations. The coupling constants $\lambda_{sf}$ of the el-SF and $\lambda_{ph}$ of the electron-phonon interaction together with the two bosonic spectral densities $\alpha^2 F(\omega)$ determine the gap value and $T_c$ as well. A broad spectral density for intraband SF as
obtained by recent inelastic neutron scattering measurements \(^3\) can be described as:

\[
\alpha^2 F(\omega) = \frac{\lambda_{sf} \Gamma_{sf}}{\pi} \frac{\omega}{\omega^2 + \Gamma_{sf}^2},
\]

where \(\Gamma_{sf} = 7.9 \text{ meV}\). For the phonons we assume a sharp Lorentzian spectral density with a width of 0.5 meV centered at \(\omega_{ph} \sim 20 \text{ meV}\). Notice that in our simple isotropic effective single-band model considered here for a \(d\)-wave superconducting order parameter, the el-ph coupling (EPC) drops out from the gap equation by symmetry and it enters only the equation for the \(Z\) function which describes the bosonic mass renormalization of the \(h\)-like quasiparticles \(^{2,22}\). Thus, here the EPC results in a slight suppression of \(T_c\) and \(\Delta_0\) in contrast to the \(s_{\pm}\) case, where the superconductivity becomes stronger taking into account an intraband EPC. The SC gap and \(T_c\) calculated within Eliashberg-theory and their dependence on the SF coupling constant \(\lambda_{sf}\) for \(\lambda_{ph} = 0\) for the sake of simplicity and also for the more realistic case, with a weak el-ph coupling constant \(\lambda_{ph} \sim 0.2\) (according to density functional theory based calculations \(^{24}\)) included, are shown in Fig. S2 of Ref. \(^{22}\). In order to reproduce \(T_c\), \(\lambda_{ph, sf} \sim 0.64 = 0.8 \lambda_{z, sf}\) should be adopted for K122, where the former describes the strength of the pairing interaction and the latter stands for the mass renormalization. (In the isotropic \(s\)-wave case \(\lambda_{ph, sf} = \lambda_{z, sf}\) holds.) These values for the coupling constants together with the calculated EPC constant \(\lambda_{ph} \approx 0.2\) \(^{24}\) yield a total coupling constant of \(\lambda_{z, tot} \approx 1\). As a result we get \(\Delta(0)^{ET} \approx 0.75 \text{ meV}\), which is close to that in BCS theory \(\Delta_{0}^{BCS} \approx 0.65 \text{ meV}\) obtained from the \(2\Delta_0/T_c\) ratio for a \(d\)-wave superconductor in the 2D case:

\[
\frac{2\Delta_0^{BCS}}{k_B T_c} = \frac{4\pi}{\gamma_E \nu_E} \approx 4.28,
\]

where \(\gamma_E\) is the Euler constant \(\gamma_E = 1.781\). The obtained \(\lambda_{z, tot}\) values suggest that we are in the regime of intermediate coupling and we may apply Eq. \(^2\) to extract \(\Delta_0\). Considering the isotropic case \((\chi = 1)\) of Eq.\(^{2}\), the same gap function for all bands and using the experimental electronic linear coefficient \(\gamma_{el} = 52-68 \text{ mJ/mol K}^2\) \(^{20}\), we estimate \(\Delta_0^{exp}\) in the range of 0.4 - 0.7 meV, which is close to the values obtained above theoretically. It is also close to the larger gap values \(\Delta_1 \approx 0.7\) to 0.8 meV obtained within effective weak-coupling two-band models for the electronically weakly connected bands \(^{16,17}\). However, the eight times smaller second gap \(\Delta_2\) \(^{16,17}\) with a comparable partial DOS for the second effective band would produce too large \(\alpha_{sc}\) values exceeding our experimental value by a factor of three or more. Hence, such a multiband scenario can be excluded on the basis of our study \(^3\). In this context, we note that the SH jump \(\Delta C/T_c\gamma_{el} \approx 0.7 - 0.9\) is close to the theoretical weak coupling value of 0.95 for a \(d\)-wave superconductor \(^{20}\). Thus, the proposed effective single-band \(d\)-wave scenario is in a good agreement with experimental SH data.

Now, we compare the experimental value of \(\alpha_{sc}\) with that for the “octet line-nodes” scenario proposed in Ref. \(^{15}\). In this case, the SH exhibits a \(T^2\) behavior at low \(T\), too, however as shown in Ref. \(^{22}\), the experimental value of \(\alpha_{sc}\) is too large to be described by a single gap with “octet line-nodes”on the middle \((\zeta)\) FSS, only, as suggested in Ref. \(^{15}\). Thus, we argue that the recently proposed interpocket \(s_{\pm}\) scenario \(^{8}\) with accidental line-nodes on a particular FSS, only, is rather unlikely to be realized in the bulk as probed by the SH. Therefore, despite the multi-band topology of \(K_{1-x}Na_xFe_2As_2\), according to our SH data it behaves more or less like a single-band \(d\)-wave SC with corresponding line-nodes on all FSS as suggested by Reid et al. based on a thermal-conductivity study and universal scaling arguments valid for a single-band \(d\)-wave SC \(^{11,12}\). It was predicted even before by theoretical calculations based on microscopic weak-coupling theory \(^3\). However, quantitatively, in the interpretation of the thermal conductivity in Refs. \(^{11,12}\) the nominal Sommerfeld coefficient \(\gamma_n\) was used and a possible magnetic contribution was not subtracted.

In summary, high-quality \(K_{1-x}Na_xFe_2As_2\) \((x=0.1)\) single crystals were studied by specific-heat. The large \(T^2\) contributions and the \(\sqrt{B}\) behavior at low-\(T\) evidence the presence of line-nodes in the superconducting gap. From the experimental data, an effective gap amplitude \(\Delta_0^{exp} \sim 0.4 - 0.7 \text{ meV}\) was estimated. This \(\Delta_0^{exp}\) and \(T_c\) agree well with the gap value calculated within Eliashberg-theory for a one-band \(d\)-wave superconductor implying a moderately strong electron-boson coupling constant \(\lambda_{z, tot} \approx 1\). This suggests that almost all, i.e. at least the three large Fermi-surface sheets with about 90\% of the total DOS (according to the LDA) have comparable gap amplitudes. Our data provide the first direct and quantitative evidence for \(d\)-wave superconductivity in a Fe-pnictide system. A detailed comparison with the cuprates might be helpful to deepen our understanding of the pairing mechanism in both unconventional and still challenging superconducting families.

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SUPPLEMENT

We present details of the specific heat (SH) measurements in external magnetic fields and of the analysis of data in the normal state. We also briefly sketch some aspects of the renormalization of the normal-state Sommerfeld coefficient and details of our Eliashberg-theory based calculations for $T_c$ and the superconducting order parameter $\Delta_0$ discussed in the main text. For the clean limit case an estimate of the coefficient $\alpha_{sc}$ which enters the $T^2$ term in the low-$T$ SH expansion is provided for the $s_\pm$-scenario with accidental line nodes proposed recently, too.

Specific-heat of $K_{0.9}Na_{0.1}Fe_2As_2$ (K(Na)122) in magnetic fields

The temperature dependence of the SH of a K(Na)122 single crystal in magnetic fields $B \parallel c$ (a) and $B \parallel ab$ (b) is summarized in Fig. S1. The SC jump shifts and broadens systematically to lower temperature with increasing field. The shift is more pronounced for $B \parallel c$ than for $B \perp c$ reflecting the large upper critical field anisotropy of this compound. The extracted temperature dependence of the upper critical fields for both field orientations are shown in the main text in Fig. 3. For the measurements in fields $B$ parallel $ab$ a small copper block was used to orient the sample. The SH of the copper block was determined in a separate measurement and subtracted from the data. However, we observed a small discrepancy between the data measured with that copper block (but subtracting its contribution afterwards) and without a copper block as can be seen in Fig. S1 which originates in the larger experimental error when using the copper block. Therefore, for the quantitative analysis of the SH we used only data measured without a copper block.
Analysis of the specific-heat in the normal state

To extract correctly $\alpha_{\text{ac}}$ from the experimental data shown in Fig. 2 in the main text, at first, the normal state SH for both samples has been fitted by the expression accounting for the electronic, lattice, and magnetic (due to a cluster glass (CG) phase) contributions as proposed in Ref. [S1]

$$C_p = \gamma_n T + \varepsilon_{\text{CG}} T^2 + \beta_3 T^3 + \beta_5 T^5,$$

(S1)

where $\gamma_n = 95(2)$ mJ/mol-K$^2$ for K(Na)$_{122}$ and 89(3) mJ/mol-K$^2$ for KFe$_2$As$_2$ (K122) are linear contributions to the SH (denoted as nominal Sommerfeld coefficients) which are the sum of the standard intrinsic electronic contributions $\gamma_{\text{el}}$ related to the itinerant charge carriers (quasiparticles) (the so-called Sommerfeld coefficient) and another linear, somewhat unusual glassy magnetic contribution $\gamma_{\text{CG}}$ related to the CG-phase [S1], and $\varepsilon_{\text{CG}}$ is a quadratic contribution due to the CG-phase. It was found that $\varepsilon_{\text{CG}} \approx 0$ for K(Na)$_{122}$ and $\varepsilon_{\text{CG}} = 2$ mJ/mol-K$^3$ for K122 [S1]. Finally, the lattice contributions are $\beta_3 = 0.556$ mJ/mol-K$^4$ and $\beta_5 = 1.16 \cdot 10^{-3}$ mJ/mol-K$^6$ for K(Na)$_{122}$ and $\beta_3 = 0.589$ mJ/mol-K$^4$ and $\beta_5 = 1.2 \cdot 10^{-3}$ mJ/mol-K$^6$ for K122.

Now, we briefly comment on the renormalization of the Sommerfeld coefficient due to the electron-boson interaction. The Eliashberg-theory, which we use to calculate $T_c$ and $\Delta$, considers the low-energy sector of characteristic bosonic energies. Within this approach the relation between the SH of the "bare" electrons $\gamma_{\text{el}}$ and the renormalized quasiparticles due to electron-boson interaction $\gamma_{\text{el}}$ reads:

$$\gamma_{\text{el}} = \gamma_{\text{el}}^0 (1 + \lambda_{z,\text{tot}}),$$

where $\lambda_{z,\text{tot}} = \lambda_{z,\text{sf}} + \lambda_{z,\text{ph}}$ consists of two contributions: the electron-spin-fluctuation interaction $\lambda_{z,\text{sf}}$ and the electron-phonon interaction $\lambda_{z,\text{ph}}$ (see also the next subsection.) This renormalization gives a factor of about two for the enhancement of the Sommerfeld coefficient and similarly for the mass in the plasma frequency which determines the penetration depth and the superconducting condensate density for $T \to 0$. The enhancement factor of the order $2.5 - 3$ of the Sommerfeld coefficient as compared to the LDA calculations $\gamma_{\text{el}}^{LDA} = 10.5$ mJ/mol K$^3$ and $\gamma_{\text{el}}^0 \sim 30$ mJ/mol K$^3$ is due to a high-energy renormalization of the bands by self-energy effects beyond the LDA based approach seen also in the ARPES data and similarly in the optical mass which enters the Drude weight, i.e., the intraband plasma frequency at high temperature and energies as compared to the energies of the low-energy bosons included in our Eliashberg-theory based approach (for details see Refs. S2-S5).

Details for the $d$-wave Eliashberg-theory based calculations

We consider here, for the sake of simplicity, a two-dimensional (2D) single-band $d$-wave superconductor with a cylindrical Fermi surface parameterized by the angle $\theta$. In general, the momentum dependence of the electron-boson

![Graph](image-url)  
FIG. S1: Temperature dependence of the specific heat of K$_{0.9}$Na$_{0.1}$Fe$_2$As$_2$ in various applied magnetic fields parallel to the c axis (a) and parallel to the ab plane (b).
spectral function for a given mode can be expanded in terms of Fermi surface harmonics

\[ \alpha^2 F(\omega, \theta, \theta') = \sum_{J, J'} Y_J(\theta) \alpha^2 F(\omega, J, J') Y_{J'}(\theta'), \]  

(S2)

where the functions \( Y_J \) denote the spherical harmonics. For simplicity, we assume that \( \alpha^2 F(\omega, \theta, \theta') \) is dominated by contributions from \( J = 0 \) \( (\Omega_0 = \Omega_2 = 1) \) and \( J = 2 \) \( (\Omega_2 = \Omega_0 = \sqrt{2} \cos(2\theta)) \) and that it is diagonal in the basis \( J, J' \). This is the minimal set required to model \( d \)-wave superconductivity. Then the electron-boson spectral function can be parameterized via dimensionless coupling constants \( \lambda_z \) and \( \lambda_\phi \):

\[ \alpha^2 F(\nu, \theta, \theta') = \sum_i [\lambda_{i,z} + 2\lambda_{i,\phi} \cos(2\theta) \cos(2\theta') ] F_i(\nu) \]  

(S3)

where \( i \) is a mode index, \( F_i(\omega) = B_i(\omega) / \int_{-\infty}^{\infty} B_i(\nu) d\nu / \nu \) is a normalized electron-boson spectral function, and \( B_i(\omega) \) is the bosonic density of states. We will consider contributions from two types - a single dispersionless optical-phonon \( (i = \text{ph}) \) branch and a spectrum of antiferromagnetic spin fluctuations \( (i = \text{sf}) \).

It is useful to introduce:

\[ \lambda_{z,\phi}(n - m) = \lambda_{z,\phi} \sum_i \int_{0}^{\infty} \frac{2\nu F_i(\nu) d\nu}{\nu^2 + (\omega_n - \omega_m)^2} \]  

(S4)

where \( \omega_n \) and \( \omega_m \) are fermionic Matsubara frequencies. The set of the imaginary-axis Eliashberg equations for a \( d \)-wave superconductor are then:

\[ \phi(i\omega_n) = \frac{\pi}{\beta} \sum_m 2\lambda_{\phi}(m - n) \left\langle \frac{\phi(i\omega_m) \cos^2(2\theta)}{\sqrt{\omega_m^2 Z^2(i\omega_m) + \phi^2(i\omega_m) \cos^2(2\theta)}} \right\rangle_\theta \]  

(S5)

and

\[ \omega_n Z(i\omega_n) = \omega_n + \frac{\pi}{\beta} \sum_m \lambda_z(m - n) \left\langle \frac{\omega_m Z(i\omega_m)}{\sqrt{\omega_m^2 Z^2(i\omega_m) + \phi^2(i\omega_m) \cos^2(2\theta)}} \right\rangle_\theta \]  

(S6)

where \( \beta = 1/k_B T \) is the inverse temperature, \( \langle \ldots \rangle_\theta \) denotes a Fermi surface average, and the Matsubara frequency sum is cut-off at \( m = 500 \). The gap function is then given by \( \Delta(i\omega_n) = \phi(i\omega_n)/Z(i\omega_n) \). The transition temperature \( T_c \) is determined from the highest temperature at which \( \phi \) has a non-zero solution while the gap magnitude \( \Delta(T = 0) \) is approximated with a low-temperature \( (T \sim 0.2 \text{ K}) \) value of \( \Delta(\omega_0 = i\pi / \beta) \).

To describe the spin fluctuations we adopt the usual form for the bosonic density of states \( B_{\text{sf}}(\nu) = \Gamma_{\text{sf}} / \nu (\nu + \Gamma_{\text{sf}}) \), with \( \Gamma_{\text{sf}} = 7.9 \text{ meV} \). The high-frequency tail of this spectrum was cut at 100 meV. For the optical-phonons we assume a narrow Lorentzian line-shape \( B_{\text{ph}}(\nu) = \Gamma_{\text{ph}} / (\nu - \Omega_{\text{ph}})^2 + \Gamma_{\text{ph}}^2 \), centered at \( \Omega_{\text{ph}} = 20 \text{ meV} \) and \( \Gamma_{\text{ph}} = 0.5 \text{ meV} \). Furthermore, the spectrum for the spin-fluctuations is cut-off at 100 meV. We further assume that the phonons do not contribute to the \( d \)-wave pairing \( (\lambda_{\phi,\text{ph}} = 0) \) and we adopt the so-called unbalanced scenario for \( d \)-wave superconductivity [S7], \( \lambda_{\phi,\text{sf}} = 0.8\lambda_{\text{sf}} \). This ratio is also in good agreement with \( \lambda_{s,d} \) in Ref. [S8].

In Fig. S2 we plot \( 2\Delta(T = 0) / k_B T_c \) extracted by use of this model. For \( \lambda_{\text{sf}} \) and \( \lambda_{\text{ph}} = 0 \), this ratio tends to about 4.28 (not shown in Fig. S2). Thus, the electron-phonon coupling reduces the strong-coupling corrections in this special case.

Comparison of \( \alpha_{\text{sc}} \) for \( d^- \) and \( s_+ \)-wave (with accidental nodes) SC order parameters

The experimental value of \( \alpha_{\text{sc}} \) discussed in the main text is too large to be described by a gap with "octet-line nodes" on the middle (\( \zeta \)) FSS, only, as suggested in Ref. [S7]. In fact, let us consider the ratio between \( \alpha_{\text{sc}}^+ \) for a \( d \)-wave gap with 16 nodes on all FSS having nearly equal gaps \( \Delta_0 \approx 0.6 \text{ meV} \) and \( \alpha_{\text{sc}}^- \) for a single \( s \)-wave gap with 8 nodes on a particular FSS. Using the general expression for the coefficient \( \alpha_{\text{sc}} \) in the \( T^2 \) term entering Eqs. (1) and (2) in the main text, we have:

\[ N_{\text{SC}}(E) = \sum_i \frac{E}{\pi \hbar v_F \partial \Delta / \partial k} \approx \frac{E}{\partial \Delta / \partial \theta} \sum_i \alpha_i N^{i}(\varepsilon_F). \]  

(S7)
FIG. S2: (a) Superconducting transition temperature $T_c$ obtained from our model calculations as a function of $\lambda_{z,sf}$. (b) The same as in (a) for the corresponding gap to $T_c$ ratio $2\Delta/k_B T_c$.

Here, we assumed that $\partial \Delta / \partial k = \partial \Delta / \partial \theta \frac{1}{k_F}$, and then $N^i(\varepsilon_F)$ is the DOS in the normal state corresponding to the $i^{th}$ FSS with a node. According to the considerations given in the main text we have:

$$\alpha_{sc} \approx \frac{9}{2} \zeta(3) k_B^3 \frac{E}{\partial \Delta / \partial \theta} \sum_i \varkappa^i N^i(\varepsilon_F) \approx 0.283 \frac{\varkappa \gamma_{el}}{\Delta_0}. \quad (S8)$$

The last expression is written for the case of a $d$-wave SC gap ($\partial \Delta / \partial \theta = 2\Delta_0$) measured in meV and where $\zeta$ denotes Riemann’s zeta function and $\zeta(3) = 1.2$. We used also that the bare Sommerfeld coefficient $\gamma_{el} = \frac{\pi^2 k_F^2}{3 N_{tot}(\varepsilon_F)}$. For the sake of simplicity, we also assumed that all FSS have similar $\varkappa$ values, therefore, $\sum_i \varkappa^i N^i(\varepsilon_F) = 4 \varkappa N^i(\varepsilon_F) = \varkappa N_{tot}(0)$. Finally, from Eq. (S8) for $T < \Delta_{s,min} \approx 1.3 K$ we estimate:

$$\frac{\alpha_{sc}}{\alpha_{sc}^d} \approx \frac{\varkappa \sum_i \varkappa N^i(\varepsilon_F)(1+\lambda_i)}{2 \varkappa N(\varepsilon_F)(1+\lambda_s)} \approx 4.5, \quad (S9)$$

where we used (according to our calculations and Ref. [S4]) $\frac{2 \varkappa N(\varepsilon_F)(1+\lambda_s)}{\varkappa \sum_i \varkappa N^i(\varepsilon_F)(1+\lambda_i)} \sim 5$ with $\lambda_s = \lambda_{tot}$, $\varkappa / \varkappa^d \sim 1.3$, and $\partial \Delta_s / \partial \theta \approx 1.8 \partial \Delta_d / \partial \theta$, where $\varkappa$ and $\partial \Delta_s / \partial \theta$ have been evaluated at the nodal directions of the $\zeta$ FSS according to the data given in Ref. [S9]. For the physical consequences of this estimated ratio with respect to the symmetry of the superconducting order parameter, see the main text.

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[S1] V. Grinenko et al., Phys. Stat. Sol. B 250, 593 (2013).
[S2] M. Abdel-Hafiez et al., Phys. Rev. B 85, 134533 (2012).
[S3] W. Lee and D. Rainer, Z. Phys. 73, 149 (1976).
[S4] K. Hashimoto et al., Phys. Rev. B 82, 014526 (2010).
[S5] S.-L. Drechsler et al., Phys. Rev. Lett. 101, 257004 (2008); - Physica C 470, Supplement 1, S332-S333 (2010).
[S6] See for example J.P. Carbotte and C. Jiang, Phys. Rev. B 48, 4231 (1993).
[S7] E. Cappellutti and G.A. Ummarino, Phys. Rev. B 76, 104522 (2007).
[S8] H. Ikeda, R. Arita, and J. Kunesh, Phys. Rev. B 81, 054502 (2010).
[S9] K. Okazaki et al., Science 337, 1314 (2012).