Observation of Strong-Coupling Pairing with Weakened Fermi-Surface Nesting at Optimal Hole Doping in Ca$_{0.33}$Na$_{0.67}$Fe$_2$As$_2$ *

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We report an angle-resolved photoemission investigation of optimally doped Ca$_{0.33}$Na$_{0.67}$Fe$_2$As$_2$. The Fermi surface topology of this compound is similar to that of the well-studied Ba$_2$K$_4$Fe$_7$As$_2$ material, except for larger hole pockets resulting from a higher hole concentration per Fe atoms. We find that the quasi-nesting conditions are weakened in this compound compared to Ba$_2$K$_4$Fe$_7$As$_2$. Similar to Ba$_2$K$_4$Fe$_7$As$_2$, we observe nearly isotropic superconducting gaps with Fermi surface-dependent magnitudes for Ca$_{0.33}$Na$_{0.67}$Fe$_2$As$_2$. A small variation in the gap size along the momentum direction perpendicular to the surface is found for one of the Fermi surfaces. Our superconducting gap results on all Fermi surface sheets fit simultaneously very well to a global gap function derived from a strong coupling approach, which contains only 2 global parameters.

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Although there is a broad consensus on existence of non-conventional superconductivity in Fe-based superconductors, intense debates persist on the precise nature of the pairing mechanism in these compounds. Arguably, the candidate models can be divided into two main categories. On the one hand, the Fermi surface (FS)-driven pairing mechanisms or weak-coupling approaches are considered as the primordial interactions between various FSs.\(^3,4,5\) On the other hand, short-range pairing mechanisms, which may include approaches from the intermediate to the strong coupling,\(^6,7,8\) are more naturally described in the real space. These various models necessarily have fingerprints in the size and symmetry of the superconducting (SC) order parameter, which are accessible directly in the momentum space by angle-resolved photoemission spectroscopy (ARPES).

The easiest way to address this debate using a momentum-resolved probe such as ARPES is to tune the size of various FSs, which is carried out by varying the carrier concentration in the Fe-As planes. The 122 family of ferropnictides is ideal for this purpose since it counts several members with relatively high SC critical temperatures ($T_c$'s), in addition to leaving nice and shiny cleaved surfaces. In this Letter, we focus on optimally-doped Ca$_{0.33}$Na$_{0.67}$Fe$_2$As$_2$, which has a $T_c$ nearly as high as the well-studied Ba$_2$K$_4$Fe$_7$As$_2$ material, despite a much larger hole concentration per Fe at optimum concentration.\(^9\) Consistently, we observe very large hole FSs centered at the Brillouin zone (BZ) center ($\Gamma$ point) showing weaker quasi-nesting conditions to the M-centered electron FSs than in Ba$_2$K$_4$Fe$_7$As$_2$.\(^10,11\) We determine a rather isotropic SC gap that is FS-dependent, with SC gap sizes ranging from 5.7 to 10.2 meV. Interestingly, we show that the SC gap on one of these bands is slightly modulated as a function of the out-of-plane momentum $k_z$. More importantly, we demonstrate that the SC gap data on all the FSs can all fit together by using the same global gap function derived from a strong coupling, with the same 2 global gap parameters, thus suggesting that local interactions play a major role in the Cooper pairing in the Fe-based superconductors.

Single crystals of Ca$_{0.33}$Na$_{0.67}$Fe$_2$As$_2$ showing bulk superconductivity at $T_c = 33$ K were grown by the self-flux method.\(^12\) Most of the synchrotron ARPES measurements were performed at beamline SIS of the Swiss Light Source. Synchrotron data were also...
recorded at 1 K by using the 1-cubed ARPES end-
station of BESSY, and additional SC gap measure-
ments were performed in our own facilities at the In-
stitute of Physics, Chinese Academy of Sciences, by
using the He α line of a helium discharge lamp. All
these systems are equipped with VG-Scienta R4000
electron analyzers and the angular resolution was set
to 0.2° while the energy resolution for the SC gap
measurements ranged from 4 to 7 meV. All samples
were cleaved in situ and measured in a working vac-
uum better than 5 × 10⁻¹¹ torr. In the following, we
label the in-plane momentum values with respect to
the 1 Fe/unit cell Brillouin zone (BZ) and use \( c' = c/2 \)
as the distance between two Fe planes.

Based on previous reports on similar materials,[14] we
conclude that two of them, the \( \alpha \) and \( \alpha' \) FSs, are
nearly degenerate near \( k_z = 0 \).

The \( \alpha' \) band, which is formed mainly by the even
combination of the \( d_{xz} \) and \( d_{yz} \) orbitals[15] is the
outermost FS near \( k_z = \pi/c' \). As illustrated in
Fig.1(c), this FS is the only one showing signif-
icant modulation along the \( k_z \) axis, which we access
by converting the probing photon energy into \( k_z \)
using the free-electron approximation[14] and an inner
potential \( V_0 = 12.7 \text{ eV} \). This modulation is partic-
ularly obvious within the \([\pi/2, 3\pi/2]\) range. As with
optimally-doped \( \text{Ba}_0.6\text{K}_{0.4}\text{Fe}_2\text{As}_2 \),[16] the other bands
do not exhibit any noticeable modulation with re-
spect to \( k_z \), and thus the three-dimensional (3D) FS of
\( \text{Ca}_{0.33}\text{Na}_{0.67}\text{Fe}_2\text{As}_2 \) near the zone center, reproduced
schematically in Fig.1(d), is essentially composed of
cylinders, except for one strongly dispersive FS sheet.

To discuss the FS topology of \( \text{Ca}_{0.33}\text{Na}_{0.67}\text{Fe}_2\text{As}_2 \),
we present in Figs.1(a) and 1(b) the ARPES intensity
mappings of this material recorded with 58 eV and
42 eV photons, which correspond approximately to
the \( k_z = 0 \) and \( k_z = \pi/c' \) planes, respectively.
The FS topology, quite similar to that of other 122-
ferropnictides,[14] is composed of \( \Gamma(Z) \)-centered
hole FS pockets and \( M(R) \)-centered electron FS pockets.
Using the momentum distribution curves (MDCs) at
the Fermi level (\( E_F \)), we extracted the Fermi wave
vectors (\( k_F \)) of the hole FSs, as illustrated in Figs.1(a)
and 1(b). While we can distinguish only two hole FSs
for \( k_z \approx 0 \), we clearly observe three for \( k_z \approx \pi/c' \).
electron pockets. To discuss the nesting conditions between the $M$-centered electron pockets and the $I'$-centered hole pockets, which are central to FS-driven pairing mechanisms, we display in Figs. 2(d) and 2(e) the intensity plots of 2D curvatures in the $k_x-k_y$ plane, for data recorded using $\sigma$ and $\pi$ polarizations, respectively. As we can see from the summary of the FSs observed, which is illustrated in Fig. 2(f), and as reported previously,[18] no electron-hole pair of FS pockets has a good nesting. Although quasi-nesting in the sense of Ref. [14] remains possible, it is weaker than that in the Ba$_{1-x}$K$_x$Fe$_2$As$_2$ cousin compounds, despite a similarly high $T_c$. 

To check if the SC gap structure of Ca$_{0.33}$Na$_{0.67}$Fe$_2$As$_2$ is strongly affected by the relatively weaker quasi-nesting conditions compared with Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$, we performed ultra-high energy resolution measurements below the SC transition. The results are shown in Fig. 3. Following a common practice, we symmetrized the EDCs to approximately remove the Fermi–Dirac cut-off. The momentum distribution of the resulting curves are given in Figs. 3(a)–3(c) for the $\alpha$, $\beta$ and $\gamma$ FSs, respectively. As illustrated with the polar plot in Fig. 3(d), rather isotropic SC gaps open on all these FSs, which is similar to observations on other 122-ferropnictides,[14] while contrasts with a recent report on Ca$_{1-x}$Na$_x$Fe$_2$As$_2$, where a noticeable gap anisotropy is found on the $\beta$ FS.[18] Interestingly, the gap found on the large $\beta$ FS is significantly smaller than that on the $\alpha$ and $\gamma$ FSs. Similar observation has been reported from a calorimetric investigation of Ba$_{0.65}$Na$_{0.35}$Fe$_2$As$_2$, where $\Delta/k_BT_c$ ratios of 1.06 and 2.08 were found.[19] The gap sizes we determined directly from ARPES on Ca$_{0.33}$Na$_{0.67}$Fe$_2$As$_2$ are much larger and more consistent with other Fe-based superconductors with comparable $T_c$’s. Indeed, while average SC gaps of $\Delta_{\alpha} = 10.2$ meV ($\Delta_{\alpha}/k_BT_c = 3.4$) and $\Delta_{\gamma} = 9.2$ meV ($\Delta_{\gamma}/k_BT_c = 3.2$) are found for the $\alpha$ and $\gamma$ FSs, the average SC gap size on the $\beta$ band is only $\Delta_{\beta} = 5.7$ meV ($\Delta_{\beta}/k_BT_c = 2.1$), a situation similar to that of Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$, where the 6 meV SC gap found on the $\beta$ FS differs largely from the 12 meV SC gap found on the other bands.[10,11] We note that our data show slightly larger SC gap sizes than that in the study of Evtushinsky et al.[18] which is possibly due to a sample dependence.

![Fig. 3.](image-url)

![Fig. 4.](image-url)
that, after converting the photon energy into $k_z$, the gap size obeys the periodic function $\Delta_\alpha = 8.8 + 1.13 \cos(k_z)$ (meV), as shown in Fig. 4(c). This result suggests the presence of interlayer interactions affecting the SC pairing. This effect is relatively small compared to the average value of the SC gap.

One of the main consequences of a short-range SC pairing mechanism is that the SC gap on each of the various FS sheets obeys the same global function defined in the entire momentum space. In other words, the SC gap magnitude depends essentially on the absolute $k_F$ position rather than on the relative positions of two FSs, in contrast to FS-driven pairing mechanisms for which inter-FS and intra-FS interactions dominate. Inelastic neutron scattering experiments suggest that the interactions between next-nearest Fe neighbors, characterized by the exchange parameter $J_3$, is the most important anti-ferromagnetic exchange parameter to describe the spin wave dispersion in the magnetically-ordered 122-ferropnictides. Assuming that fluctuations of these interactions persist in the SC materials and are responsible for the SC pairing leads to the conclusion that the SC pairing function must either have the form $\sin(k_x) \sin(k_y)$ or $\cos(k_x) \cos(k_y)$. We discard the former one since it implies the presence of nodes that are not detected in our experiments. In Fig. 4(d), we plot the magnitude of the SC gap determined by ARPES as a function of the absolute value of the latter function, ARPES being directly sensitive only to the magnitude of the SC gap. The agreement is qualitatively pretty good with a linear relationship, with the data spreading over a considerable range of the $[0,1]$ range limiting the $| \cos(k_x) \cos(k_y) |$ function.

The fit of the experimental data to a global gap function derived from short-range interactions by using a single parameter for all the FS sheets pushes towards a local pairing origin in this particular material, similar to the other Fe-based superconductors. The fit is not perfect, suggesting that some parameters have been ignored. For instance, as mentioned above, there must be a finite interlayer interaction responsible for the modulation of the SC gap on the $\alpha$ FS shown in Fig. 4(c). Following previous studies on Ba$_{0.4}$K$_{0.6}$Fe$_2$As$_2$ and BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ and Ba(Fe$_{0.75}$Ru$_{0.25}$)$_2$As$_2$, addressing this issue for ARPES measurements of the SC gap on the Fe-based superconductors, we improve the gap function by including an interlayer coupling that translates into the global gap function

$$\Delta(k) = |\Delta_2 \cos(k_x) \cos(k_y) + (\Delta_3/2) \cos(k_x) \cos(k_y)|.$$ (1)

Here we have two parameters. The result of the fit, displayed in Fig. 4(e), suggests the validity of this approach for Ca$_{0.33}$Na$_{0.67}$Fe$_2$As$_2$ as well, and thus suggests that this behavior is common to most of the 122 ferropnictide compounds. Interestingly, we extract the global parameters $\Delta_2 = 9.9$ meV and $\Delta_3 = 1.2$ meV, which leads to a $\Delta_2/\Delta_3$ ratio of 8.3 that is similar to the $J_2/J_3$ ratio of 7 determined from inelastic neutron scattering on the parent compound CaFe$_2$As$_2$. Although interband and intraband interactions very near $k_F$ certainly play a very important role in the physics of the Fe-based superconductors, our current results adding to previous ones suggest that the SC pairing of electrons in these materials rather originates from short-range interactions better described in the real space.

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References

[1] Graser S, Maier T A, Hirschfeld P J and Scalapino 2009 New J. Phys. 11 025016
[2] Hirschfeld P H, Korshunov M M and Mazin I I 2011 Rep. Prog. Phys. 74 124508
[3] Kontani K and Onari S 2010 Phys. Rev. Lett. 104 157001
[4] Seo K, Bernevig B A and Hu J 2008 Phys. Rev. Lett. 101 206404
[5] Fang C et al 2011 Phys. Rev. X 1 011009
[6] Zhang Y et al 2010 Phys. Rev. Lett. 105 117003
[7] Hu J P and Hao N N 2012 Phys. Rev. X 2 021009
[8] Hu J P and Ding H 2012 Sci. Rep. 2 381
[9] Zhao K et al 2011 Phys. Rev. B 84 184534
[10] Ding H et al 2008 Europhys. Lett. 83 47001
[11] Zhao L et al 2008 Chin. Phys. Lett. 25 4402
[12] Zhao K et al 2010 J. Phys.: Condens. Matter 22 222203
[13] Damascelli A et al 2004 Phys. Scr. T109 61
[14] Richard Pet al 2011 Rep. Prog. Phys. 74 124512
[15] Wang X P et al 2012 Phys. Rev. B 85 214518
[16] Xue Y M et al 2011 Nat. Phys. 7 198
[17] Zhang P et al 2011 Rev. Sci. Instrum. 82 043712
[18]Evtushinsky D V et al 2013 Phys. Rev. B 87 094501
[19] Pramanik A K et al 2011 Phys. Rev. B 84 064525
[20] Huang Y B et al 2012 AIP Adv. 2 041409
[21] Zhang Y et al 2012 Nat. Phys. 8 371
[22] Xu N et al 2013 Phys. Rev. B 87 094513
[23] McQueeney R J et al 2008 Phys. Rev. Lett. 101 227005