Tightly-bound Cooper pair, quasiparticle kinks and clues on the pairing potential in a high $T_c$ FeAs Superconductor

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Recent discovery of superconductivity ($T_c$ up to 55K) in iron-based layered compounds promises a new route to high temperature superconductivity. This is quite remarkable in the view that the $T_c$ in the pnictides is already larger than that observed in the single-layer cuprates. These superconductors belong to a comprehensive class of materials where many chemical substitutions are possible. Preliminary studies suggest that the superconducting state in these materials competes with a magneto-ordered state, and the proper description of the magnetically ordered state lies somewhat in between a strong correlation mediated interacting local moment magnetism and quasi-itineracy with a high degree of stripe-like frustration.

Angle-resolved photoemission spectroscopy (ARPES) is a powerful tool for investigating the microscopic electronic behavior of layered superconductors. In this work we report single-particle electronic structure results focusing on the details of the low-lying quasiparticle dispersions and the pair gap formation on very high quality ($\delta T_c \lesssim 1K$ and surface-RMS $\lesssim 2\AA$) single domain single crystal samples, which allow us to gain insight into connections between the superconductivity and magnetism not addressed by other spectroscopic work. Besides a magnitude-oscillating superconducting gap in crossing from the zone center to the zone corner, we observe that the quasiparticles are strongly scattered by collective processes around 15 to 50 meV binding energy range depending on the Fermi surface sheet. Our high resolution and systematic quasiparticle data suggest that a Cooper pair in this superconductor is tightly bound ($\lesssim 4a_s$), reflecting a non-phononic character of the underlying pairing potential in light of retardation or screening effects. Overall results can be self-consistently interpreted in a phase-shifting order parameter scenario.

FIG. 1: Phase transition, magnetization profiles and surface quality. (a-b) Bulk $T_c$ of crystalline (Ba, K)Fe$_2$As$_2$ and (Sr, K)Fe$_2$As$_2$ was determined based on the resistivity and magnetization profiles. (Ba, K)Fe$_2$As$_2$ samples exhibited $T_c \approx 37K$ and $\delta T_c \sim 1K$ whereas (Sr, K)Fe$_2$As$_2$ samples exhibited a broad ($\sim 10K$) transition with a $T_c \sim 26K$. (c) Surface quality was studied by atomic-resolution STM measurements which exhibited a high degree of flatness and confirmed the suitability for spectroscopic measurements. The derivative of an STM image is shown which was taken on a 500Å×500Å patch. The inset shows low-temperature electronic gap on the order of 2$\Delta\approx 30$ meV in superconducting samples. Sample batches with $\delta T_c \sim 1K$ and smooth STM images were selected for UHV cleaves in our ARPES studies.

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ARPES measurements were performed using 18 to 60 eV photons with better than 8 to 15 meV energy resolution respectively and overall angular resolution better than 1% of the Brillouin zone. Most of the data were taken at the Advanced Light Source beamline 12.0.1 and a limited data set was taken at SSRL beamline 5-4 for cross-checking, using a Scienta analyzer with chamber pressures lower than $5 \times 10^{-11}$ torr. Linearly polarized photons were used for all the study. The angle between the $\overrightarrow{E}$-field of the incident light and the normal direction of the cleaved surface was set to about 45 degrees (at 12.0.1). Single crystalline samples of Ba$_{1-x}$K$_x$Fe$_2$As$_2$ ($T_c=37$K) and Sr$_{1-x}$K$_x$Fe$_2$As$_2$ ($T_c=26$K) were used for this systematic study. Cleaving the samples in situ at 15K resulted in shiny flat surfaces. Cleavage properties were characterized by atomic resolution STM measurements and the surface was found to be flat with an RMS deviation of 1Å(Fig.1(c)). Rarely observed steps of size 6Å were seen on the otherwise flat surface. The utilization of a unique scattering geometry and specific photon energy range allowed us to suppress one of the FS sheets so that the other can be studied in full detail.

Quasiparticle behavior around the $\Gamma$-FS sheets is shown in Figure 2. Two square-like FS sheets were clearly resolved near the center of the BZ. An azimuthal variation of ARPES intensity around the FS pockets was observed. This variation is most pronounced while the data are taken at the particular photon-electron scattering geometry described above. A comparison of quasiparticle dispersion measured along the various $\overrightarrow{k}$-space cuts suggests that roughly along a cut 45-degrees to the $\Gamma$ to $(\pi,0)$-line provides a clear spectroscopic look at the quasiparticle dispersion and lineshape behavior on the inner-FS. A bend in the dispersion (E vs. $\overrightarrow{k}$) could be observed in the data which is not resolved in cut-1 or 3 due to the spectral overlap with the outer-FS. The measured quasiparticles in Sr$_{1-x}$K$_x$Fe$_2$As$_2$ (k,l) Wide k-range coarse-step scans are shown which was used for locating the Fermi crossings. (m) Electron distribution map, n(k), within 15 meV of Fermi energy over the complete Brillouin Zone.

The temperature evolution of low-lying quasiparticles through the superconducting transition is shown in Figure 3. Quasiparticles gain spectral weight upon cooling below the transition temperature ($\sim 37$K). The low-temperature quasiparticle peak width is about 10 meV. The opening of the superconducting gap is best viewed upon symmetrization of the near-$E_F$ data and a gap magnitude of about 12±2 meV is quite evident at low temperatures. This value is consistent with the average gap ($< 15$ meV, see Fig.1(c)) we observe with STM on the same batch of samples. The observed gap value is found to be largest in the inner-most central FS ($\Gamma$1 band), then decreases on the next FS ($\Gamma$2 band) moving outward toward the M-point, and then increases again on the corner FS location. This oscillating gap structure is consistent with (but not a unique fit to) an order-parameter that takes the in-plane form of $\Delta_o \cos(k_y) \cos(k_y)$. The reduction of the gap value on the outer central FS is consistent with a $\cos \times \cos$ form. We caution that the ARPES data do not rule out the possibility of an out-of-plane ($k_z$)
FIG. 3: Multi-gap pairing structure: (a) Temperature dependence of quasiparticles (cut-2) near the Fermi level through the superconducting transition. Below $T_c$, samples exhibit coherence-peak-like behavior similar to what is observed in some cuprates. Temperature dependences of the gap at the $k$-space location of the inner-most ($\Gamma$1) FS (b) and the outer central ($\Gamma$2) FS (c) are estimated by symmetrized spectral weight around the Fermi level. (d) The temperature dependences of the gaps measured at different FS locations ($\Gamma$1-FS, blue; $\Gamma$2-FS, red; and near-$M$, black) are plotted along with the bulk resistivity curve (green). A fluctuation regime above $T_c$ is observed. (e) The azimuthal $k$-dependences of gaps, $\Delta(k)$, are shown for different FS sheet locations ($\Gamma$1, $\Gamma$2, $M$). Selected EDCs are shown for the $k$-dependence of gaps on the $\Gamma$1 (f) and outer Fermi surfaces (g). The cuts and pts refer to Fig. 2.

A closer look at the quasiparticle dispersion behavior is presented in Figure 4. A bend in dispersion is evident in the momentum distribution curves (MDC) taken on a crossing near the $\Gamma$1-FS (cut-4). Each MDC could be fitted with a single Lorentzian over a wide binding energy range and, as in the raw data sets, the fitted peak positions trace a kink around $40\pm 15$ meV. This is furtherconfirmed by examining the peak position of the real part of the self-energy which is also observed to be around $40\pm 15$ meV. Although it is less clear, the MDC width plotted as a function of the electron binding energy is found to exhibit a drop below 30 meV that is consistent with a kink at higher energy seen in the raw data. At temperatures above $T_c$, the kink shifts to somewhat lower energies. As the temperature is raised further the MDCs are broadened making its identification or analytic extraction from our experimental data difficult and unreliable. In the MDC widths (Fig.4(g)) an increase is observed at very low energies which was found to be unrelated to the existence of the kink but rather related to some residual signal from the tail of the quasiparticles on the outer FS. The STM data in Fig.1(c) also exhibit a satellite structure around 40-50 meV loss-energy range (with respect to the quasiparticle peak position) roughly consistent with the observed ARPES kink. Assuming that the kink reflects coupling to some bosonic-like modes one can estimate the coupling strength: $\lambda_{eff}^s \gtrsim (0.7/0.45 - 1)\sim 0.6$. This coupling is about a factor of two to three larger than the electron-phonon coupling ($\lambda_{ph} \sim 0.2$) calculated for the Fe-As phonons near 20-40 meV [7]. A careful look at the outer central FS ($\Gamma$2 band, cut-8) also reveals a kink around $18 \pm 5$ meV. This kink is revealed when the band associated with the inner-FS sheet is suppressed by a choice of incident photon energy such as in the data taken near 18 eV. The observed kink energies seems to scale (40 meV and 18 meV) with the superconducting gap energies (12 meV and 6 meV) on the two central Fermi surfaces (Fig.4).

A strong-coupling kink phenomenology is observed in the electron dynamics of high $T_c$ cuprates which occurs around $60\pm 20$ meV and is often attributed to phonons or magnetism or polarons with $\lambda^s_{eff} \sim 1$ to 1.5 [16]. In cuprates the superexchange coupling is on the order of 130 meV, whereas the optical phonons are in the range of 40 to 80 meV overlapping with the kink. In the pnictides, although a $T_c$ value of 37K is not outside the phonon-induced strong-coupling pairing regime, the vibrational modes of the the FeAs plane are rather soft ($\leq 35$ meV) making electron-phonon interaction [7] an unlikely source of the majority part of the quasiparticle’s self-energy beyond 40 meV, considering the observed coupling $\lambda_{eff} \gtrsim 0.6$ for the FeAs compounds here. The parent compounds of superconducting FeAs exhibit a robust SDW groundstate [5] due to a $Q=(\pi, \pi)$ inter-band instability or due to the interaction of quasi-localized moments. The SDW short range order seems to survive well into the superconducting doping regime [23]. The doping evolution of the Fermi surface lacks robust nesting conditions for purely band-magnetism to be operative at these high dopings and the relevant magnetism likely comes from the local exchange energy scales in a doping induced frustrated background. Therefore, quite naturally, strong spin fluctuations in the presence of electron-electron interaction are important contributors to the electron’s self-energy and a natural candidate for the constituent of the kinks. In accounting for the parent SDW groundstates of these materials the known values of $J_1$ and $J_2$ are on the order of 20 to 50 meV [14, 15]. This is the energy range where we have observed the quasiparticle kinks (Fig.4). In an itinerant picture, there exists a Stoner continuum whose energy scales are parameterized by $J_1$ and $J_2$ whereas in a local picture, $J_1$ and $J_2$ reflect Fe-Fe and Fe-As-Fe superexchange paths and the groundstate is a highly frustrated doped Heisenberg magnet [15]. The proper description of the experimentally observed magnetism in these systems lies somewhere in between.
In the photoemission process removal of an electron from the crystal excites the modes the electron is coupled to, so the observed quasiparticle breaks the locally frustrated magnetic bonds associated with $J_1$ and $J_2$ which then contributes a characteristic energy scale in the electron's self-energy. Since these characteristic magnetic scales are quite large ($\sim 400$K) it is expected that the kink behavior in the electron’s dispersion relation would survive above $T_c$ consistent with our experimental observation. Despite the high signal-to-noise quality of our data, it is premature to draw an intimate connection between the kinks and the resonant spin mode ($\sim 15$ meV) observed in neutron scattering.

High-resolution (Fig.-4) dispersion measurements further allow us to estimate the Fermi velocity of the normal state which is about 0.7 eVÅ. Using the observed superconducting gap (ARPES or STM data in Fig.1) we can estimate the average size of the Cooper pair wave-function $\xi = \frac{\hbar}{MV} \Delta$ by invoking the uncertainty relation [24]. Taking $v_F$(Fig.4) $\sim 0.7 \pm 0.1$ eVÅ and a gap (Fig.3) value of $\Delta \sim 12 \pm 2$ meV, this gives $\xi \lesssim 20 \AA$. This value is remarkably consistent with the high magnitude of $\xi$ in $\sim 70$ reported in the same materials. The ARPES based Cooper pair scale and unusually high $\xi$ clearly suggest that the Cooper pairs in this class of FeAs superconductors are tightly bound which is in contrast to the point-contact Andreev spectroscopy results on Sm-based FeAs superconductors exhibiting a conventional BCS ratio [27]. The agreement between ARPES, bulk $\xi$ and the bulk resistivity profile (Fig.3(d)) provides further support for our identification of the superconducting gap and its bulk-representative value through a surface-sensitive measurement such as ARPES. This also confirms that the ARPES observed gaps in superconducting materials [17, 18, 19] are not the SDW gaps as theoretically claimed by some authors. More importantly, such a small Cooper pair size scale ($\sim 4\xi$) is not known in any phonon-based BCS superconductor [28] but has only been observed in unconventional correlated superconductors. Our observed value is much smaller than that in the multi-band s-wave BCS-phonon superconductors such as MgB$_2$ [28]. In fact a combination of small Cooper pair size and in-plane nodeless superconductivity is consistent with an unconventional $\Delta_{c2}\cos(k_x)\cos(k_y)$ (in the unfolded BZ with one iron atom per unit cell)-type or $s_{2}+is_{3}$ or $s_{\pm}$ wave states $8, 9, 11, 12, 13$ since such an order parameter has a nearest-neighbor structure in real space and thus a reduction of the Coulomb interaction within the pair is naturally possible, so the electrons can come closer to each other leading to a short coherence scale. In cuprates pairing electrons come close to each other, and the short coherence length is achieved by introducing a node in the order parameter ($d$-wave) leading to a reduction of the Coulomb interaction within the pair. This is often the only choice in a single band system such as the cuprates or the organics. In pnictides, multiband structure can accommodate a phase change without the need for introducing a ”node” [29] on the Fermi surface, therefore an isotropic gap and short pairing scale can coexist with a phase shifted order-parameter structure.

In summary, we have presented a single-particle study of high $T_c$ superconductor class (Sr/Ba)$_{1-x}$K$_x$Fe$_2$As$_2$. Our systematic ARPES data suggest an unusually small dimension of the Cooper pair, complex kink phenomena and an oscillating gap function all of which collectively point to an unconventional pairing potential. We have...
presented arguments that in the presence of Coulomb interaction and magnetism, the observed short pairing scale and a nearly-isotropic in-plane gap can be self-consistently realized if the order parameter contains a phase factor. Our results thus provide important clues to a novel route to high temperature superconductivity.

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[29] The $\cos \times \cos$ or $s_{\pm}$ gap does have a node in it but the node does not intersect the Fermi surface as we measure here (Figure 3). However, it is clear that nearest or next nearest neighbour structure of the gap lowers the kinetic energy as opposed to a constant on-site $s$-wave gap.
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FIG. 5: Enlarged View of Figure 1.

FIG. 6: Enlarged View of Figure 2.

FIG. 7: Enlarged View of Figure 3
FIG. 8: Enlarged View of Figure 4.