Theory of Multi-Band Superconductivity in Iron Pnictides

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The precise nature of unconventional superconductivity in Iron Pnictides is presently a hotly debated issue. Here, using insights from normal state electronic structure and symmetry arguments, we show how an unconventional SC emerges from the bad metal “normal” state. Short-ranged, multi-band spin- and charge correlations generates nodeless SC in the active planar $d_{xz,yz}$ bands, and an inter-band proximity effect induces out-of-plane gap nodes in the passive $d_{xy}$ band. While very good quantitative agreement with various key observations in the SC state and reconciliation with NMR and penetration depth data in the same picture are particularly attractive features of our proposal, clinching evidence would be an experimental confirmation of c-axis nodes in future work.

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High Temperature Superconductivity (HTSC) in the recently discovered Iron pnictides (FePn) is the latest surprise among a host of others in d- and f band materials [1]. HTSC arises upon doping (x) a spin-density-wave (SDW) metal with x > x_c, SDW order melts, giving way to a HTSC. In the “normal” phase, with no SDW/SC order for $T > T_{SDW/SC}(x)$, various probes reveal an incoherent metallic state, presumably not too far from a Mott insulator [2]. HTSC in FePn arises from an incoherent normal state, at least in the FeAs-based materials, where a quasi-linear-in-T resistivity is seen immediately above $T_c$: this implies a correlated normal state. Local density approximation plus dynamical mean-field (LDA+DMFT) calculations in the intermediate-to-strong correlation limit [3,4] indeed find very good quantitative agreement with a range of basic physical properties of FePn, attesting to their sizably correlated nature.

If SC is an instability of this incoherent metal, one expects pronounced deviations from weak-coupling BCS-like theories in the SC state, as in the cuprates: this is indeed born out in experiments. Specifically, (i) the pair coherence length, $\xi_{ab}$ is short (< 20 Å), the upper critical field, $H_{c2}$ is high [5], with small superfluid density, (ii) the NMR relaxation rate, $T_1^{-1}(T) \approx T^3$ [6], and the penetration depth in the 1111-FePn, $\lambda(T) \approx T^n$ with n = 2 − 2.5 [3], indicating unconventional SC (U-SC). However, the 122-FePn seem to show conventional s-wave-like behavior, though, even here, the situation is controversial [7]. Moreover, µSR studies reveal Uemura scaling and $T_c$ vs $\epsilon_F$ scaling characteristic of other, well-known, correlated SC [8]. (iii) Optical measurements [9] show a large-scale spectral weight transfer (SWT) across $T_c$ over an energy scale $> O(1.0)$ eV, while ARPES data show a sharpening of the low energy quasiparticle kink [10] below $T_c$. Taken together, as they must be, (i)-(iii) imply a strong coupling SC arising from an incoherent non-FL metal. Given (ii), the existence of gap nodes is an open, controversial issue [11]. In particular, (i)-(iii) resemble observations in cuprates [12], implying an U-SC closer to the Bose condensed, rather than the BCS limit [10]. How can such a U-SC arise as an instability of an incoherent normal state [3,4]? How may nodes appear in the gap function? Does SC pairing involve all, or a subset of the Fe-d orbitals? These issues are of great import for FePn, and call for a systematic resolution. Here, motivated by the above, we focus on the 1111-FePn, specifically on $LaO_1-xFeAsF_x$. Guided by group-theoretical analyses, and by the renormalized multi-band structure, we propose a specific, strong correlation based, interband pairing mechanism for U-SC as an instability of the incoherent metal found in earlier LDA+DMFT studies [3,4].

LDA+DMFT studies for the 1111-FePn find an incoherent, pseudogapped “normal” state, implying blocked coherent one-electron propagation: only collective motion of the spin and charge fluid is possible [13]. This opens the door to coherent two-particle propagation, i.e., to two-particle instabilities, to relieve the finite entropy at lower $T$. At x = 0, the electron- and hole-like Fermi surface (FS) sheets are nearly nested in LDA calculations, and DMFT will not alter this fact, since the self-energies are local; this favors the $q = (\pi,0)$ SDW instability, also within LDA+DMFT [3] (in fact, no nesting is needed at strong coupling [2,3]), in agreement with neutron data [3,14]. Doping weakens the SDW and facilitates the U-SC instability.

Since coherent one-electron transfer between different orbitals is blocked in the incoherent metal, we propose, in analogy with the situation encountered in coupled Luttinger liquids (LL) [15], that small, residual, inter-site and inter-orbital interactions mediate two-particle instabilities in FePn at low $T$. In principle, various types of instabilities, competing with U-SC [16], may be generated, as studied more extensively for cuprates [17]. Here, we focus on the U-SC alone. We further assume that pair formation primarily involves the active $d_{xz,yz}$ orbitals, and show that, due to the inter-orbital coupling $U'$, an interband (IB) proximity effect will induce SC on all FS sheets, as in $Sr_2RuO_4$ [18].
Excluding spin-triplet pairing, the general interaction in the cooper channel is

\[ H_{\text{pair}} = \frac{1}{2} \sum_{a,b,k,k'} V_{ab}(k,k') c_{a,k,l}^\dagger c_{b,-k,-l}^\dagger c_{b,-k',l} c_{a,k',l}, \]

where \( a, b = xz, yz \) and the scattering vertex is \( V_{ab}(k,k',\omega) = g^2 \chi_{ab}(k-k',\omega) \) with \( \chi_{ab}(k-k',\omega) \) being the inter-orbital susceptibility. Such a term naturally arises at second order from a one-electron inter-band term, \( t_{ab} \sum_{i,j,\sigma} \langle \epsilon_{i\sigma}^a c_{j\sigma}^b + h.c. \rangle \), when the one-electron spectral function is incoherent, much like in coupled LL \([19]\). This is an important feature of our work: normal state one-particle incoherence, arising from the Anderson orthogonality catastrophe \([3, 4]\) due to strong multi-orbital correlations, favors two-particle coherence. The static, nearest- and next-nearest neighbor parts of \( V_{ab}(k,k') \) are \( V_{ab}^{\text{tr}}(k,k',0) \approx \frac{\epsilon^{c}_{i\sigma} \epsilon^{c}_{j\sigma}}{\epsilon^{c}_{i\sigma} + \epsilon^{c}_{j\sigma}} \approx O(40-50) \text{ meV} \) and \( V_{ab}^{\text{ex}} \approx \frac{\epsilon^{c}_{i\sigma}}{\epsilon^{c}_{i\sigma}} \approx O(15-20) \text{ meV}, \) close to the superexchange scale estimated from inelastic neutron scattering (INS) studies \([20]\). Notice that the effective interaction contains coupled inter-orbital charge and spin fluctuations involving nearest (n.n) and next-nearest neighbor (n.n.n) Fe sites, and is explicitly \( \mathbf{k} \)-dependent, favoring U-SC from the outset. \( H_{\text{pair}} \) can be decoupled a la Gorkov \([21]\) to yield

\[ H_{\text{pair}}^\text{MF} = \sum_{a,b,k} \sum_{\mathbf{k},\mathbf{k}'} \Delta_{ab}(k) c_{k,a,l}^\dagger c_{-k,b,-l} + h.c., \]

where \( \Delta_{ab}(k) = \frac{1}{2} V_{ab}(k,k') \langle c_{-k,b,-l} c_{k,a,l} \rangle \) is the SC gap function. Consistent with lattice symmetry, and including the n.n and n.n.n contributions, \( V_{ab}(k,k') = \sum_{\mathbf{k},\mathbf{k}'} V_{ab}(\mathbf{k},\mathbf{k}') \eta(k'), \) where \( \eta(k) \) are the irreducible representations of \( D_{4h} \) point group. So we can expand \( \Delta_{ab}(k) = \sum \Delta_{ab}^{ij}(k) \eta(k) \) \([21]\). Explicitly, \( \Delta_{ab}(k) = \Delta_{ij}(c_{a} + c_{b}) \) or \( \Delta_{ij} c_{i} c_{j} \), with \( c_{a} = \cos(k_{a}) \). The case \( \Delta_{0} = 0 \) \((B_{2g} \text{ representation of } D_{4h})\) is favored in the numerical study \([22]\), however, \( \Delta_{2} \neq 0 \) is rigorously required \([23]\). With well-separated electron- and hole-like FS sheets as in LDA, no in-plane nodes exist in the SC gap for \( \Delta_{2}/\Delta_{1} \) in the chosen range. A \( \Delta_{2} \neq 0 \) is also favored by the observation of appreciable geometric frustration in FePn, where \( J_{2}/J_{1} \approx O(0.7-1.0) \) have been deduced from \textit{ab initio} studies, and consistent with INS data \([20]\).

The "normal" state is modelled by a five-band Hubbard model, \( H_{n} = H_{0} + H_{1} \), treated earlier with LDA+DMFT \([2, 4]\), and reads \( H_{0} = \sum_{\mathbf{k},\alpha} \epsilon_{\mathbf{k},\alpha} c_{\mathbf{k},\alpha}^\dagger c_{\mathbf{k},\alpha} \) while \( H_{1} = U \sum_{\mathbf{i},\alpha,\alpha'} n_{\mathbf{i},\alpha} n_{\mathbf{i},\alpha'} + U' \sum_{\mathbf{i},\alpha,\alpha'} n_{\mathbf{i},\alpha} n_{\mathbf{i},\alpha'} - J_{H} \sum_{\mathbf{i},\alpha,\beta} \mathbf{S}_{\alpha} \cdot \mathbf{S}_{\beta}, \) where \( \alpha, \beta = xy, yz, zy, x^2-y^2, 3z^2-r^2 \). In the U-SC phase, we have to solve \( H = H_{n} + H_{\text{pair}}^\text{MF} \) within LDA+DMFT. Fortunately, the \textit{intersite} nature of \( H_{\text{pair}} \) adds simply a term bilinear in fermions to \( H \). The LDA+DMFT equations are now readily extendable to the U-SC regime: the \( G_{ab} \) and \( \Sigma_{ab} \) now have normal and anomalous components, and a closed set of DMFT equations, yielding both \( G_{aa}, F_{ab} \) selfconsistently, is solved by extending earlier LDA+DMFT \([4]\) to include an explicit pair-field term. Including the pair-field, the propagators are \( G_{aa}(k,\omega) = \left[ \omega - \epsilon_{ka} - \Sigma_{aa}(\omega) - \Delta_{ab}^{\text{tr}}(k) \right]^{-1} \) and \( F_{ab}(k,\omega) = G_{aa}(k,\omega) \Sigma_{ab}(\omega) \), where the * denotes complex conjugation. Given \( \text{intersite} \) pairing, \( F_{ij,ab}(\omega) \) falls off at least as \( 1/\omega_{ij} \), (here, \( D \) is the lattice dimension) the \textit{dynamical} effects of these non-local pair fluctuations do not enter the DMFT self-energies. Obviously, however, they do affect the \( G_{aa} \). Our treatment is thus different from that of Garg et al. \([24]\), where the dynamical effects of \textit{local} pair fluctuations must be kept in DMFT. Anomalous contributions like \( \Sigma_{ij}^{\text{tr}}(t) = -V_{ij}^{\text{tr}} G_{i,aa}(t) F_{j,ab}(-t) G_{j,bb}(t) \), etc, to the \textit{dynamical} self-energy would thus enter in a cluster-DMFT approach, but drop out in \( D = \infty \). It then suffices to use the DMFT with the modified matrix propagators above. Finally, since these equations couple all Fe-\( d \)-orbitals, the opening up of a SC gap in the \( d_{xz,yz} \) bands could induce secondary gaps in the remaining dip orbitals, in a way reminiscent of the inter-band proximity effect in \( \text{Sr}_{2}\text{RuO}_{4} \) \([18]\) (see below). Further, given sizable \( U = 4.0 \text{ eV}, U' = 2.6 \text{ eV} \) for FePn \([3, 4]\), large spectral changes within DMFT-like approaches should accompany the U-SC instability: these are seen in spectral probes \([3, 4]\). Is such a scenario also consistent with other signatures of a strong coupling, U-SC?

Aiming to shine light on these issues, we now describe our results. Upon convergence of the DMFT equations, the one-electron- and pair spectral functions can be read off and used for direct comparison with observables in the U-SC state. In Fig \( \text{a} \) we show the changes induced by U-SC in the \textit{total} one-electron DOS. Clear sharpening of the low-energy kink at \( \Omega \approx 20.0 \text{ meV} \) \([10, 25]\), already somewhat visible in the "normal" state \([29]\), is seen across the U-SC instability. Strong normal state incoherence \( \Sigma_{ij}^{\text{tr}}(\omega) \) the equation for \( G_{aa}(k,\omega) \) prevents opening up of a clean SC gap in the DOS, as indeed observed in PES. Remarkably, the \textit{position} of this kink and the detailed PES lineshape up to a binding energy of \( -0.5 \text{ eV} \) are both quantitatively reproduced within our theory.

More microscopic insight is obtained by investigating the orbital resolved DOS. As seen from Fig \( \text{b} \) only the \( d_{xz,yz} \) and \( d_{3z^2-r^2} \) DOS show the sharpening of the kink feature. Moreover, dominant SWT occurs from the \( d_{xz} \) band to the \( d_{xz,yz,3z^2-r^2} \) bands, as clearly seen in Fig \( \text{b} \). The first implies an orbital \textit{selective} coupling of the carrier propagators to multi-orbital, overdamped and short-ranged, charge- and spin correlations, and fully agrees with indications from ARPES studies \([25]\), which show clear evidence thereof. Large SWT, over an energy scale \( O(2.0) \text{ eV} \), also accompanies the U-SC instability. This is also in qualitative accord with findings in optical stud-
conflict with extant ARPES results, as seen in

explain the NMR and penetration depth data, being ca-

possible line nodes in the SC gap function along

k

one-particle mixing between the

for doped (x = 0.1) LaO1−xFexAsFy, for U = 4.0 eV, U′ =

2.6 eV, and the effective interaction, Vab =max(Vab(1), Vab(2)) =

50 meV. The inset shows how the low-energy kink, already

visible above Tc (dashed curve), sharpens up below Tc (dotted

curve), in very good agreement with ARPES data[10, 25].

ies[8]. Taken together, these findings imply a strong

coupling SC, and fully accord with other observations in

the SC state in FePn, as shown below.

Additional fundamental features of interest are gleaned

from observation of the sharpening of the kink in the

d32−r2 DOS in the U-SC state above. This strongly sug-

gests that an IB-proximity driven, out-of-plane SC gap

component should enter on symmetry grounds. Exami-

nation of the Slater-Koster fit to the LDA shows a sizable

dxy − d32−r2 mixing [27]; this will induce a ∆′cos(kzc)

component, reduced by dipolar reduction factors [18], in

the SC gap function. Strong supportive evidence for this

comes from extant dHvA studies on LaFePO [28]. This

indeed finds sizable

|−dxy|, indicating sizable IB-proximity, and, interest-

ingly, recently, INS work on SmFeAsO1−xFx [32] strongly suggesting line nodes in the SC gap. However, data on SmFeAsO1−xFx [32]

are consistent with a smooth (nodeless) angular variation, Δ(φ) = Δ0(1 + ccos4φ), of the in-plane gap, and λab(T) follows the well-known e−Δ(φ)/κT law. In the FeAs materials, it is conceivable that the multiband coupling does not always induce out-of-plane nodes in the SC gap; this will be sensitive to the detailed topology of the renor-

malized FS. In cases where a non-exponential λ(T) is seen, we

suggest that dHvA results should show significant c-axis warping of the dxy−FS sheet: this should be investigated in more detail. However, the planar (1 + ccos4φ) variation deduced for Sm-based FePn is not inconsistent with our in-plane form factor, as proposed above. Addition-

ally, recently, INS work on Ni-doped BaFe2As2 clearly shows both planar and c-axis variation of Δ(k); interestingly, precisely Δ(kz) ∝ cos(kzc) was found there[11]. It would be very interesting to see if this is also true

FIG. 1: (Color online): Evolution of the total one-particle
density-of-states (DOS) across the superconducting transition
for doped (x = 0.1) LaO1−xFexAsFy, for U = 4.0 eV, U′ =

2.6 eV, and the effective interaction, Vab = max(Vab(1), Vab(2)) =

50 meV. The inset shows how the low-energy kink, already

visible above Tc (dashed curve), sharpens up below Tc (dotted

curve), in very good agreement with ARPES data[10, 25].

pable of giving the observed power-law-in-T behavior for

both the NMR T−1(T) [6] and λab(T) [7] at low T << Tc.

In contrast to the s± gap with disorder scenario [24], exis-
tence of nodes would imply universal power-law depend-

ces in these quantities as a function of x, and along differ-
ing FePn members, as recently seen [7].

We emphasize that whether nodes along kz exist or not is a delicate matter, and that a careful examination

of the details of the band structure should provide

invaluable clues. For example, dHvA study [25] on

LaFePO indeed finds sizable c-axis corrugation of the
dxy band, indicating sizable IB-proximity, and, interest-

ingly, λab(T) ≃ T1.2 [28], strongly suggesting line nodes in the SC gap. However, data on SmFeAsO1−xFx [32]

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suggest that dHvA results should show significant c-axis warping of the dxy−FS sheet: this should be investigated in more detail. However, the planar (1 + ccos4φ) variation deduced for Sm-based FePn is not inconsistent with our in-plane form factor, as proposed above. Addition-

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FIG. 2: (Color online): Orbital resolved evolution of the

one-particle DOS across Tc, for the same parameter set as

in Fig. 1. The orbital-selective gap structure, arising from

multi-band correlations, is manifest. This is manifested in the

orbital-selective sharpening of the low-energy kink, in quali-

tative accord with ARPES[10, 25].

FIG. 2: (Color online): Evolution of the one-particle density-of-states (DOS) across the superconducting transition for doped (x = 0.1) LaO1−xFexAsFy, for U = 4.0 eV, U′ = 2.6 eV, and the effective interaction, Vab = max(Vab(1), Vab(2)) = 50 meV. The inset shows how the low-energy kink, already visible above Tc (dashed curve), sharpens up below Tc (dotted curve), in very good agreement with ARPES data[10, 25].

612.0x792.0
of the 1111-FePn. We then predict that this should be correlated with the c-axis warping of planar $d_{xy}$ band in dHvA work. This would constitute a non-trivial check of our proposal.

From LDA+DMFT, we estimate the gap magnitude to be $\Delta_{ab} \simeq 4.5$ meV, close to that found in PES [33]. Using the carrier Fermi velocity from LDA (note that this is not changed by LDA+DMFT), $v_F \approx 0.7$ eV A, the relation $\xi = v_F h/\pi \Delta$ gives $\xi \simeq 60 - 80$ A, close to observed values. The upper critical field is now estimable as $H_{c2} = \phi_0/2\pi \xi^2 \simeq 50 - 70$ T, ($\phi_0$ is the usual flux quantum) again consistent with experiment [3]. These are classic signatures of a strong coupling SC [34], and require an instability of a strongly correlated "normal" state, as proposed here.

Our proposal is distinct from other, extant ones [11, 16, 20, 22, 30]. The $s_\pm$ proposal [16, 30] is derived from a weak-coupling instability of an itinerant FL. Wu et al. [31] derive a similar state from the strong coupling limit of a reduced two-band model. The alternative, extended-s wave idea gives eight nodes on the electron-FS sheets [11, 20, 22]. Our proposal is a generalization of the $s_\pm$ proposal [30, 31]. All earlier proposals, however, cannot access possible c-axis nodal structure, which arises via an IB proximity effect involving $d_{xy} - d_{3z^2-r^2}$ bands in our work. Moreover, rigorous analysis and the geometrically frustrated superexchanges [2, 20] in FePn require both $\Delta_1, \Delta_2$ to be finite. This naturally leads to $k$-dependent $\Delta_{ab}(k)$, as deduced experimentally [11, 32]. Finally, the U-SC derived here arises as an instability of the correlated, incoherent metal [3, 4] "normal" state seen in doped FePn, and naturally explains large spectral changes [4] and orbital selective sharpening of the QP kink [10] across $T_c$ as manifestations of proximity to a Mott insulator [2, 3, 4].

In conclusion, we have derived the instability of the incoherent "normal" state of FePn [3, 4] to an U-SC. Blocking of inter-band one-particle coherence in the normal state clears the way for inter-band two-particle coherence to emerge, much like in coupled LLs, giving a multi-orbital SC with nodes. The latter are shown to arise from a multiband proximity effect between the $d_{xy}$ and the $d_{3z^2-r^2}$ bands. Extending earlier LDA+DMFT calculations for the incoherent metal, we show how good quantitative agreement with the sharpening of the low-energy kink in the PES spectrum in the U-SC phase, as well as strong spectral weight transfer across the U-SC transition, is obtained. Further, induction of line nodes due to the interband proximity effect offers a reconciliation of these attractive features with NMR and $\mu$SR data, which suggest existence of line nodes in the U-SC gap. Taken together, these strongly support our proposal of an U-SC with a planar $\Delta_{ab}(k) \simeq (c_x + c_y) + c_cc_d$ form factor; i.e. a SC with co-existent, inter-site (and inter-orbital) $s^a$ and $s_{xy}$ pair symmetries, and out-of-plane nodes $[\Delta_c(k) \simeq c_z]$, at least in some of the 1111-FePn family. Our proposal calls for careful experimental search for the out-of-plane nodes in the 1111 Iron Pnictides.