Evolution of superconductivity in Fe-based systems with doping

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We study the symmetry and the structure of the gap in Fe-based superconductors by decomposing the pairing interaction obtained in the RPA into s- and d-wave components and into contributions from scattering between different Fermi surfaces. We show that each interaction is well approximated by the lowest angular harmonics and use this simplification to analyze the origin of the attraction in s± and dxy-y2 channels, the competition between s- and d-wave solutions, and the origin of superconductivity in heavily doped systems, when only electron or only hole pockets are present.

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Introduction. The symmetry and structure of the superconducting gap in Fe-based superconductors (FeSC), and their evolution and possible change with doping are currently subjects of intensive debates in the condensed matter community. The vast majority of researchers believe that superconductivity in FeSC is of electronic origin and results from the screened Coulomb interaction, enhanced at particular momenta due to strong magnetic fluctuations. In FeSC, the Fermi surface (FS) has multiple sheets due to hybridization of all five d-orbitals of Fe, and the interactions between low-energy fermions are a complex mixture of contributions from intra- and inter-orbital interactions. In this situation, both s-wave and non-s-wave pairing are possible, and can be either conventional or extended, with π phase shifts between FSs 1

Previous theoretical works on FeSCs with hole and electron pockets have shown 2,13 that the s-wave pairing channel (for sign-changing s± gap) is generally the most attractive, although the d-wave channel is a strong competitor. s-wave gap symmetry is consistent with ARPES data, which detected only a small variation of the gap along the hole FSs, centered at (0,0), and as such ruled out d-wave gap symmetry However, for the recently discovered heavily electron-doped AFe2Se2(A=K, Rb, Cs) 14, in which only electron FSs remain according to ARPES 15, RPA and functional RG (fRG) studies found that the leading pairing instability is now in the d-wave channel. 16,17 d-wave pairing was also found in an fRG study of heavily hole-doped KFe2As2 18, in which only hole FSs are present 19. For this material, various experimental probes 20 indicate the presence of gap nodes, consistent with a d-wave gap symmetry.

In this communication, we analyze the competition between s- and d-wave pairings in doped FeSCs, the origin of attraction at small and large dopings, and the structure of s- and d-wave gaps at various dopings. We argue that the pairing mechanisms at small and large dopings are qualitatively different and that the d-wave state at large hole doping is a different eigenstate from the one that competes with s-wave at smaller dopings.

We assume, as in earlier works, that FeSCs can be treated as itinerant systems, and that the pairing interaction is enhanced by spin fluctuations (SF). In the band description adopted here, the electronic structure at low energies is obtained by hybridization of all five Fe d-orbitals and in electron-doped FeSCs consists of two cylindrical hole FSs h1 and h2, centered at (0,0), and two cylindrical electron FSs e1 and e2, centered at (π,0) and (0,π), respectively, in the 1-Fe zone. For hole-doped FeSCs, there exists an additional cylindrical hole FS h3 centered at (π,π). In such a description, interactions are dressed by matrix elements associated with the hybridization of orbitals, and depend on the angles along the FSs.

The method. The input for our analysis is the band model with the interactions between the particles on the FSs Γ(kF,−k′F; kF,−k′F) ≡ Γ(kF,k′F). The interactions are obtained numerically in the RPA SF formalism starting from the 5-orbital model 6 with intra- and inter-orbital hoppings and local density-density and exchange interactions U, U′, J, and J′. We show that, in the band basis, each interaction component Γij(kF,k′F) is well approximated by the leading angular harmonics (LAH) in s-wave and dxy-y2-wave channels (similar to the approximation of the dx2-y2 gap by cos 2θ in the cuprates), and use the LAH approximation (LAHA) to reduce s-wave and d-wave gap equations to either 4 × 4 or 5 × 5 sets which can be easily solved and analyzed. This allows us to go a step further than previous works and understand the pairing mechanism at different dopings, the origin of the transition from s-wave to d-wave instability, the role of the SF component of the interaction, and the stability of s-wave and d-wave gap structures with respect to the variation of parameters in the gap equations. For simplicity, we assume in LAHA that all FSs are circular, with the same density of states NF. The results change only
a little if we use the actual lattice fermionic dispersion. The application of LAHA for FeSCs requires some care, as electron FSs are centered at (0, \pi) and (\pi, 0) points, which are not \(k_x \leftrightarrow \pm k_y\) symmetric. As a result, some of the \(s\)-wave gap functions, like \(\cos k_x + \cos k_y\) behave as \(\pm \cos 2\theta\) along the electron FSs, while some of the \(d\)-wave gap functions like \(\cos k_x - \cos k_y\) are approximated on these FSs by constants of opposite sign. With this in mind, we treated the angle-independent and \(\cos 2\phi\) terms on equal footings in both \(s\)-wave and \(d\)-wave components of the interactions. A simple analysis then shows that LAHA consistent with the FS geometry of FeSCs approximates the \(s\) and \(d_{x^2-y^2}\) components of \(\tilde{\Gamma}_{ij} = N_F \Gamma_{ij}\) as

\[
\hat{\Gamma}_{h,h} = u_{h,h} + \hat{u}_{h,h} \cos 2\phi_h \cos 2\phi_i \\
\hat{\Gamma}_{h,e} = u_{h,e} (1 + 2\alpha_{h,e} \cos 2\theta_i) + \hat{u}_{h,e} (1 + 2\beta_{h,e} \cos 2\theta_1) \cos 2\phi_i \\
\hat{\Gamma}_{e,e} = u_{e,e} (1 + 2\alpha_{e,e} (\cos 2\theta_1 + \cos 2\theta_2) + 4\beta_{e,e} \cos 2\theta_1 \cos 2\theta_2) + \hat{u}_{e,e} (1 + 2\beta_{e,e} (\cos 2\theta_1 + \cos 2\theta_2) + 4\beta_{e,e} \cos 2\theta_1 \cos 2\theta_2)
\]

where \(u_{ij}\) and \(\hat{u}_{ij}\) are dimensionless interactions in \(s\)-wave and \(d\)-wave channels, respectively, and \(\phi_i\) and \(\theta_i\) label the angles along the hole and electron FSs, measured from the \(k_x\)-axis. Interactions involving other electron FSs are obtained by transformations consistent with \(s\)-wave or \(d\)-wave symmetry.

We use Eq. \((1)\) to fit the RPA interaction \(\Gamma_{ij}\) by LAHA and substitute the parameters extracted from the fit into \(s\)-wave and \(d\)-wave BCS gap equations, which within LAHA are \(4 \times 4\) matrix equations for two hole and two electron FSs and \(5 \times 5\) when the additional hole FS is present. We find the gap structure for the largest positive eigenvalue \(\lambda_{s,d}\) (if it exists) and then vary the parameters \(u_{ij}\) by hand to understand what is the mechanism for the attraction. For two hole and two electron FSs the generic gap structure is

\[
\Delta_{h_1}^s(\phi) = \Delta_{h_2}^s(\phi) = \Delta_{h_2}^d(\phi) = \Delta_{h_1}^d(\phi) \\
\Delta_{h_1}^d(\theta) = \Delta_{h_2}^d(\theta) = \Delta_{h_2}^s(\theta) = \Delta_{h_1}^s(\theta) = \Delta_e^s - \Delta_e^d \cos 2\theta \\
\Delta_{e_1}^d(\theta) = \Delta_{e_1}^d(\theta) = \Delta_{e_2}^d(\theta) = \Delta_{e_2}^d(\theta) = -\Delta_e^d + \Delta_e^s \cos 2\theta
\]

and for five FSs we add one more \(\Delta_{h_2}^{s,d}(\phi) = \Delta_{h_2}^{s,d}(\phi)\).

In Figs. 1 and 2 we compare LAHA with the full RPA \(\Gamma_{ij}(k_F, k_F')\). The agreement is remarkably good. We analyzed eight different sets of \(U, U'\) and \(J\), and the agreement is equally good for all sets [21]. A very few disagreements are cured by adding \(\cos \theta\) harmonics to LAHA. Some of the LAHA parameters extracted from the fit, which we will need for comparisons, are shown in Tables I and II.

The cases of weak electron and hole dopings were solved numerically within RPA in earlier works, and we verified that LAHA results are very close to the full solutions. For brevity, we present only the results for larger dopings, when one type of pockets either almost or completely disappears. We will see that there are quite abrupt changes between the two regimes.

Results and discussion. We varied the magnitudes and angle dependencies of the interactions by hand and checked what most influences the value of \(\lambda\) and the structure of the gap. We found that some system properties are sensitive to the ratios of the parameters, but some are quite universal.
For electron doping, parameter-sensitive properties include the gap symmetry, since $\lambda_\text{s}$ and $\lambda_\text{d}$ remain comparable as long as both hole and electron FSs are present (see Table II, and the presence or absence of accidental nodes in the $s$-wave gap, although for most of parameters the gap does have nodes, as in Fig. I(b)). The universal observation is that the driving force for attraction in both $s$-wave and $d$-wave channels is the inter-pocket electron-hole interaction ($u_{\text{hh}}$ and $u_{\text{he}}$ terms), no matter how small the hole pockets are. When the SF component of the interaction is large, $u_{\text{hh}}$ and $\tilde{u}_{\text{he}}$ exceed the hole-hole and electron-electron interactions. Then $\lambda_{\text{s},\text{d}}$ are positive already if we neglect the $\cos 2\theta$ terms in (I) (for two equal hole FSs the conditions are $u_{\text{ee}}^2 > u_{\text{hh}} u_{\text{ee}}$ and $\tilde{u}_{\text{he}}^2 > u_{\text{hh}} \tilde{u}_{\text{ee}}$). In this case, the $\cos 2\theta$ terms in the $s$-wave and $d$-wave gaps scale with the corresponding $g_{\text{he}}$. For smaller SF component, when $u_{\text{ee}}^2 < u_{\text{hh}} u_{\text{ee}}$ (the case considered in Fig. I and Table II), the electron-hole interaction still generates solutions with $\lambda_{\text{s},\text{d}} > 0$, only this time the gap develops a stronger $\cos 2\theta$ component, which effectively reduces $u_{\text{ee}}$.

The situation changes qualitatively once the hole pockets disappear (Fig. II(d)-(f)). We see from Table II that $\lambda_\text{s}$ is reduced, but $\lambda_\text{d}$ is enhanced, i.e., the $d$-wave $T_\text{c}$ increases. Comparing the LAHA parameters for the two dopings, we see the reason: once the hole pockets disappear, a direct $d$-wave electron-electron interaction $u_{\text{ee}}$ becomes strong and attractive. To understand why this happens, we note that $u_{\text{ee}}$ and $\tilde{u}_{\text{ee}}$ are symmetric and antisymmetric combinations of intra-pocket and inter-pocket electron-electron interactions: $u_{\text{ee}} = u_{\text{intr}A} + u_{\text{inter}}, \tilde{u}_{\text{ee}} = u_{\text{intr}A} - u_{\text{inter}}$. Both $u_{\text{intr}}$ and $u_{\text{inter}}$ are positive (repulsive), hence $u_{\text{ee}} > 0$, but the sign of $\tilde{u}_{\text{ee}}$ depends on the interplay between $u_{\text{intr}}$ and $u_{\text{inter}}$. As long as the hole FS is present, SF are peaked near $q = (0, \pi)$ and $(\pi, 0)$, which are an equal distance from the relevant momenta $q = 0$ for $u_{\text{intr}}$ and $q = (\pi, \pi)$ for $u_{\text{inter}}$. In this situation, $u_{\text{intr}}$ and $u_{\text{inter}}$ remain close in magnitude, and $\tilde{u}_{\text{ee}}$ is small. Once the hole pocket disappears, the peak in the RPA spin susceptibility shifts towards $(\pi, \pi)$ and $u_{\text{intr}}$ increases more due to the SF component than $u_{\text{intr}}$. A negative $u_{\text{intr}} - u_{\text{inter}}$ then gives rise to a “plus-minus” gap on the two electron FSs for the same reason that large $u_{\text{he}}$ gives rise to a sign-changing gap between hole and electron FSs, and (the interaction between hot spots in the cuprates gives rise to a sign-changing gap in the hot regions). Such a gap changes sign between electron pockets, which differ by $k_x \rightarrow k_y$ and therefore has $d_{x^2-y^2}$ symmetry 16, 17. Our gap functions in both $s$- and $d$-wave channels at large electron doping are in good quantitative agreement with the full solution of the RPA gap equation 17 and with RRG results 16.

Next we consider the hole doping of the LAHA fits to the cases when electron FSs are small but still present and when only hole FSs remain are shown in

![Fig. 2](image-url) The parameters extracted from the fit are shown in Table II. We analyzed these and other dopings and again found universal and parameter-sensitive features. The parameter-sensitive property is again the presence or absence of accidental nodes in the $s$-wave gap along the electron FSs. For most of the parameters, the gap does not have nodes (see Fig. II) because $u_{\text{he}}$ increases once it acquires an additional contribution $u_{\text{ee}}$, but for some parameters we still found nodes along the electron FSs. The universal observations are that, as long as both hole and electron pockets are present, (i) the $s$-wave is the leading instability ($\lambda_\text{s} > \lambda_\text{d} > 0$), and (ii) the driving force for the attraction in both $s$- and $d$- channels is again inter-pocket electron-hole interaction ($u_{\text{he}}$ and $\tilde{u}_{\text{he}}$ terms), no matter how small the electron pockets are. In the $d$-wave channel, the electron-hole interaction changes

| Parameter | $s$ | $d$ |
|-----------|-----|-----|
| $u_{\text{hh}}$ | 1.20 | 1.30 |
| $u_{\text{he}}$ | 1.10 | 1.20 |
| $\alpha_{\text{he}}$ | 0.10 | 0.10 |
| $\lambda_\text{s}$ | 1.20 | 1.30 |
| $\lambda_\text{d}$ | 0.90 | 1.00 |
| $\lambda_{\text{s},\text{d}}$ | 1.10 | 1.20 |

Table II: Some LAHA parameters extracted from the fit in Fig. II for hole doping. Block (i) corresponds to panels (a)-(c) (tiny electron pockets), block (ii) corresponds to panels (d)-(f) (no electron pockets).

FIG. 2: The same as in Fig. I but for hole doping (3 hole FSs). Panels (a)-(c) are for the case of tiny electron pockets, (d)-(f) are for stronger hole doping, when there are no electron pockets. The parameters are presented in [21].
sign between the two hole FSs at (0, 0), as a result d-wave gaps on these FS have a π-phase shift (see Fig. 2(c)).

The situation rapidly changes once electron pockets disappear. The d-wave eigenvalue $\lambda_d$ grows relative to $\lambda_s$ and for the doping shown in Fig. 2 almost exceeds it. It is very likely that d-wave becomes the leading instability at even higher dopings, and we therefore focus on the d-wave channel. Comparing $\tilde{u}$ in Table I for the cases with and without electron pockets, we find that the d-wave channel is attractive in the absence of the electron-hole interaction because of two reasons. First, the d-wave intra-pocket interaction $\tilde{u}_{h_1h_3}$ becomes negative (attractive). Second, the inter-pocket interaction $\tilde{u}_{h_1h_2}$ is larger in magnitude than repulsive $\tilde{u}_{h_1h_1}$ and $\tilde{u}_{h_2h_2}$. The solutions with positive $\lambda_d$ then exist separately for FSs $h_{1,2}$ and $h_3$, and the residual inter-pocket interaction just sets the relative magnitudes and phases between the gaps at $h_3$ and $h_{1,2}$. Because $\tilde{u}_{h_1h_2}$ is attractive, the two d-wave gaps at $h_{1,2}$ are now in phase, i.e., this d-wave solution is a different eigenfunction from the one with phase shift $\pi$ at smaller dopings. The difference is clearly seen by comparing panels (c) and (f) in Fig. 2. The d-wave gap symmetry at large doping and in-phase structure of the gaps at $h_{1,2}$ is consistent with the fRG solution [18].

**Conclusions.** The key result of this work is the observation that the mechanism of the pairing in FeSCs with hole and electron FSs is different from the one at strong hole or electron doping, when only one type of FS remains. At small/moderate dopings, the pairing is driven by inter-pocket electron-hole interaction, no matter how small hole or electron FSs are. In hole-doped FeSCs, the leading instability is s-wave, while in electron-doped FeSCs, s- and d-wave channels are strong competitors, and which of the two wins depends on the model parameters. At large electron and hole dopings, d-wave is the leading instability, although the s-wave channel remains attractive. At strong electron doping, the origin of the pairing is a direct d-wave attraction between electron pockets. At strong hole doping, however, the reason for the d-wave pairing is a d-wave attraction within the ($\pi, \pi$) pocket and between the two hole pockets at $(0, 0)$. The d-wave pairing at strong hole doping is consistent with the observation of nodal quasiparticles in the heavily hole doped superconductor KFe$_2$As$_2$ with $T_c = 3K$. Superconductivity at heavy electron doping at a rather high $T_c \sim 30K$ has been recently discovered in AFe$_2$Se$_2$(A=K, Cs, Rb), which only have electron FSs, according to recent ARPES studies [12]. Whether this is a d-wave superconductor remains to be seen.

We have only studied the strictly 2D case thus far, and neglected aspects of the 3D I4/mmm crystal symmetry characteristic of 122 materials and the hybridization of electron pockets in the folded zone [22]. We nevertheless believe that the general evolution of interactions and gap symmetry discussed here will be generic to the FeSCs.

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