Unconventional pairing originating from disconnected Fermi surfaces in the iron-based superconductor

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Abstract. For an iron-based high $T_c$ superconductor LaFeAsO$_{1-x}$F$_x$, we construct a minimal model where all of the five Fe d bands turn out to be involved. We then investigate the origin of superconductivity with a five-band random-phase approximation by solving the Eliashberg equation. We conclude that the spin fluctuation modes arising from the nesting between the disconnected Fermi pockets realize basically an extended s-wave pairing, where the gap changes sign across the nesting vector.

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1. Introduction

The superconductivity in the iron-based pnictide, LaFeAsO, doped with fluorine discovered by Hosono’s group [1] and the subsequent increase of the transition temperature ($T_c$) in the same family of compounds are remarkable as the first non-copper compound that has $T_c$ values exceeding 50 K [2]. This immediately stimulates renewed interest in the electronic mechanism of high $T_c$ superconductivity. In order to investigate the pairing mechanism, here we first construct an electronic model for LaFeAsO$_{1-x}$F$_x$ using maximally localized Wannier orbitals obtained from first-principles calculation. The minimal model turns out to involve all five Fe $d$ orbitals [3]. We then apply the random-phase approximation (RPA) to solve the Eliashberg equation. We conclude that a nesting between multiple Fermi surfaces (pockets) results in the development of a peculiar spin fluctuation mode, which in turn realizes an unconventional pairing, which is basically an extended s-wave but the gap function changes sign across the nesting vector [3, 4]. The result is intriguing as a realization of the general idea that the way in which electron correlation effects appear is very sensitive to the underlying band structure and the shape of the Fermi surface [5].

2. Band structure

LaFeAsO has a tetragonal layered structure, where Fe atoms form a square lattice in each layer, which is sandwiched by As atoms. Due to the tetrahedral coordination of As, there are two Fe atoms per unit cell. The experimentally determined lattice constants are $a = 4.03$ Å and $c = 8.74$ Å, with two internal coordinates $z_{La} = 0.142$ and $z_{As} = 0.651$ [7]. We obtained the band structure (figure 1) with the density-functional approximation with plane-wave basis [8], and then constructed the maximally localized Wannier functions (MLWFs) [9]. These MLWFs, centered at the two Fe sites in the unit cell, have five orbital symmetries ($d_{3z^2-r^2}$, $d_{xy}$, $d_{yz}$, $d_{xz}$, $d_{x^2-y^2}$, $d_{xy}$), where $X$, $Y$, $Z$ refer to those for this unit cell with two Fe sites as shown in the bottom panel of figure 1). The two Wannier orbitals in each unit cell are equivalent in that each

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8 Here we adopt the exchange correlation functional introduced by Perdew et al [8] and the wavefunctions are expanded by plane waves up to a cutoff energy of 40 Ryd. $10^3$ k-point meshes are used with the special points technique of Monkhorst and Pack [8].

9 The Wannier functions are generated by the code developed by Mostofi et al [10] for the energy window $-2 \text{eV} < \epsilon_k - E_F < 2.6 \text{eV}$, where $\epsilon_k$ is the eigenenergy of the Bloch states and $E_F$ is the Fermi energy.
Figure 1. Left panel: the band structure of the ten-band model in the original folded Brillouin zone; the symbols are the present local density approximation results. The Brillouin zone is for the original unit cell. Right panel: the original (dashed) and the reduced (solid) unit cell with • (Fe), ▽ (As below the Fe plane) and △ (above).

Fe atom has the same local arrangement of other atoms. We can then take a unit cell that contains only one orbital per symmetry by unfolding the Brillouin zone and we end up with an effective five-band model on a square lattice, where the $x$- and $y$- axes are rotated by 45° from $X$ to $Y$, to which we refer for all the wavevectors hereafter. We define the band filling $n$ as the number of electrons/number of sites (e.g. $n = 10$ for full filling). The doping level $x$ in LaFeAsO$_{1-x}$F$_x$ is related to the band filling as $n = 6 + x$.

The five bands are heavily entangled as shown in figure 2 (left panel), reflecting strong hybridization of the five 3d orbitals, which is physically due to the tetrahedral coordination of As atoms around Fe. Hence we conclude that the minimal electronic model requires all the five bands. In figure 2 (right panel), the Fermi surface for $n = 6.1$ (corresponding to $x = 0.1$) obtained by ignoring the inter-layer hoppings is shown in the two-dimensional unfolded Brillouin zone.

The Fermi surface consists of four pieces (pockets in 2D): two concentric hole pockets (denoted by $\alpha_1$, $\alpha_2$) centered around $(k_x, k_y) = (0, 0)$, and two electron pockets around $(\pi, 0)$ ($\beta_1$) or $(0, \pi)$ ($\beta_2$), respectively. $\alpha_i$ ($\beta_i$) corresponds to the Fermi surface around the $\Gamma Z$ line (MA in the original Brillouin zone) in the first-principles band calculation [9]. Besides these pieces of the Fermi surface, there is a portion of the band near $(\pi, \pi)$ that is flat and touches the $E_F$ at $n = 6.1$, so that the portion acts as a ‘quasi-Fermi surface ($\gamma$)’ around $(\pi, \pi)$. As for the orbital character, $\alpha$ and portions of $\beta$ near the Brillouin zone edge have mainly $d_{xz}$ and $d_{yz}$ character, whereas the portions of $\beta$ away from the Brillouin zone edge and $\gamma$ have mainly $d_{x^2-y^2}$ orbital character.

An interesting feature of the band structure is the presence of Dirac cones, i.e. places where the upper and the lower bands make a conical contact [11, 12]. The ones closest to the Fermi

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10 While ambiguity exists in unfolding the Brillouin zone (if the sign of the hoppings $t(\Delta x, \Delta y; \mu, \nu)$ with $\Delta x + \Delta y = \text{odd}$ (i.e. hoppings between A and B sites in a bipartite lattice) is changed, a band structure is reflected with respect to $|k_x| + |k_y| = \pi$), this is just a unitary transformation, and either way the five-band structure gives the same ten bands in the folded Brillouin zone as well as the same RPA results.
Figure 2. Left: the band structure of the five-band model in the unfolded Brillouin zone. Orbital characters of the bands are also shown. The short solid arrow denotes the position of the Dirac cone shown in figure 3. Right: Fermi surface for $n = 6.1$. The blue (red) portions have strong $X^2-Y^2$ character. Dashed blue curves represent the portions where the band lies very close to the Fermi level at $n = 6.1$ and actually becomes a Fermi surface for smaller doping. The original (dashed lines) and the unfolded (solid) Brillouin zone are shown.

level lie at positions where the $d_{x^2-y^2}$ and the $d_{xz}/d_{yz}$ bands cross, just below the $\beta$ Fermi surface. A three-dimensional representation of the Dirac cone is shown in figure 3.

3. Many-body Hamiltonian and RPA

We consider a two-dimensional model where the inter-layer hoppings are neglected. For the many-body part of the Hamiltonian, we consider the standard interaction terms that comprise the intra-orbital Coulomb $U$, the inter-orbital Coulomb $U'$, the Hund’s coupling $J$ and the pair-hopping $J'$. The many-body Hamiltonian reads

$$H = \sum_{ij} \sum_{\mu\nu} \sum_{\sigma} t_{ij}^{\mu\nu} c_{i\mu\sigma}^\dagger c_{j\nu\sigma} + \sum_{\mu} \sum_{\sigma} \varepsilon_{\mu} n_{i\mu\sigma} + \sum_{\mu} \left[ U \sum_{\mu} n_{i\mu\uparrow} n_{i\mu\downarrow} + U' \sum_{\mu>\nu} n_{i\mu\sigma} n_{i\mu\sigma'} + J \sum_{\mu\neq\nu} \mathbf{S}_{i\mu} \cdot \mathbf{S}_{i\nu} + J' \sum_{\mu\neq\nu} c_{i\mu\uparrow}^\dagger c_{i\mu\downarrow}^\dagger c_{i\nu\downarrow} c_{i\nu\uparrow} \right],$$

where $i$, $j$ and $\mu$, $\nu$ denote the sites and the orbitals, respectively, and $t_{ij}^{\mu\nu}$ is given by the tight-binding model obtained in the previous section.

Having constructed the model, we move on to the RPA calculation, where the modification of the band structure due to the self-energy correction is not taken into account. Multiorbital RPA is described in, e.g., [13, 14]. In the present case, Green’s function is a $5 \times 5$ matrix, while

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Figure 3. Dirac cones (solid arrows) where the third (red) and the fourth (green) bands make point contact. The black (blue) dashed line is the portion of the bands with strong $X^2-Y^2$ ($XZ/YZ$) character. The brown curve is the $\beta$ Fermi surface.

each of the spin and orbital susceptibilities $\chi_{l_1l_2l_3l_4}$ ($l_i = 1, \ldots, 5$) has $25 \times 25$ components. The Green’s function and the effective pairing interactions, obtained from the susceptibilities, are plugged into the linearized Eliashberg equation, and the gap function as a $5 \times 5$ matrix along with the associated eigenvalue $\lambda$ are obtained. $T_c$ corresponds to the temperature where $\lambda$ reaches unity. $32 \times 32$ $k$-point meshes and 1024 Matsubara frequencies are taken. We find that the spin fluctuations dominate over orbital fluctuations as far as $U > U'$, so we can characterize the system with the spin susceptibility. We denote the largest eigenvalue of the spin susceptibility matrix for $i\omega_n = 0$ as $\chi_s(k)$.

4. Results: spin structure

Let us look at the result for the spin susceptibility $\chi_s$ for $U = 1.2$, $U' = 0.9$, $J = J' = 0.15$ and $T = 0.02$ (all in units of eV) in figure 4. The susceptibility has peaks around $(k_x, k_y) = (\pi, 0)$, $(0, \pi)$ (i.e. a collinear spin density wave), which reflects the Fermi surface nesting around $\sim (\pi, 0)$, $(0, \pi)$ across $\alpha$ and $\beta$ and also $\beta$ and $\gamma$ in the top panels of figure 5. This feature is consistent with the stripe-type antiferromagnetic order for the undoped case, which was first suggested by transport and optical reflectance, [6] and further confirmed by neutron scattering experiments [7].

5. Results: superconductivity

In the bottom panels of figure 5, we show the diagonal elements of the gap function matrix for orbitals $d_{YZ}$, $d_{ZX}$ and $d_{X^2-Y^2}$. The gap is basically $s$-wave, but changes sign across the $\alpha - \beta$ or $\beta - \gamma$ nesting vector $\sim (\pi, 0)$, $(0, \pi)$ at which the spin fluctuations develop [3, 4, 20].

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Figure 4. Left panel: RPA result for (a) the spin susceptibility $\chi_s$ for $U = 1.2$, $U' = 0.9$, $J = J' = 0.15$, $n = 6.1$ and $T = 0.02$ (in eV). Right panel: the spin configuration in real space that corresponds to the wavevector $(0, \pi)$.

Figure 5. Top panels: the portions of the Fermi surface with mainly $d_{xz}$, $d_{yz}$ (left) or $d_{x^2-y^2}$ (right) orbital character. The dashed curve around $(\pi, \pi)$ (at the corners of the Brillouin zone) is the portion where a flat portion of the band lies very close to the Fermi level, although it does not actually form a Fermi surface. The arrows indicate the nesting vector for the Fermi surface. Bottom panels: the gap function for the $d_{xz}$, $d_{yz}$ (left) or $d_{x^2-y^2}$ (right) orbitals.
particular, the magnitude of the $d_{x^2-y^2}$ orbital gap turns out to be larger than that of other orbital components, at least for the present set of parameter values. The sign change in the $d_{x^2-y^2}$ orbital gap is analogous to those in models studied by Bulut et al [15] and also by two of the present authors [16, 17]. It is also reminiscent of the unconventional s-wave pairing mechanism for Na$_x$CoO$_2$·$\gamma$H$_2$O [18] proposed by four of the present authors [19].

In the above, we focused on the diagonal elements of the gap matrix in the orbital representation, but to be more precise, there are also the off-diagonal (interorbital) elements. To take into account the effect of all the elements and also to make direct comparison with experiments possible, we calculate the quasiparticle excitation energy by diagonalizing the $10 \times 10$ Hamiltonian matrix

$$H = \begin{pmatrix} H_0 - \mu I & \hat{\Delta}^\tau \\ \hat{\Delta} & -H_0 + \mu I \end{pmatrix},$$

where $H_0$ is the $5 \times 5$ non-interacting Hamiltonian and $\hat{\Delta}$ is the $5 \times 5$ gap matrix obtained by the calculation described above, $\mu$ is the chemical potential and $I$ is the unit matrix. Since $\hat{\Delta}$ obtained by solving the linearized Eliashberg equation has ambiguity in the overall factor, here we take the maximum value of the gap matrix in the orbital representation to be 0.0035 eV. In figure 6, we show the quasiparticle excitation energy along $\alpha_1$, $\alpha_2$ and $\beta$ Fermi surfaces. We find that the quasiparticle excitation has a finite gap over each of the entire pockets, but there is a difference in the gap magnitude between $\alpha$ and $\beta$ Fermi surfaces. Also, the gap significantly varies along the $\beta$ Fermi surface. These features of the gap reflect the difference in the gap magnitude between the $d_{x^2-y^2}$ and $d_{xz}, d_{yz}$ orbitals shown in figure 5. However, we find, in further calculations, that this gap variance is not universal and depends strongly on the electron density and also on the details of the band structure. This dependence will be published elsewhere.

6. Conclusion

To summarize, we have constructed a five-band electronic model for LaFeAsO$_{1-x}$F$_x$, which we consider to be the minimum microscopic model for this material. Applying a five-band RPA to
this model, we have found that spin fluctuation modes around \((\pi, 0)\) develop due to the nesting between disconnected Fermi surfaces. Based on the linearized Eliashberg equation analysis, we have concluded that spin fluctuation modes realize unconventional, extended s-wave pairing, where the gap changes sign across the nesting vectors.

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References

[1] Kamihara Y, Watanabe T, Hirano M and Hosono H 2008 J. Am. Chem. Soc. 130 3296
[2] Ren Z-A et al 2008 Chin. Phys. Lett. 25 2215
[3] Kuroki K, Onari S, Arita R, Usui H, Tanaka Y, Kontani H and Aoki H 2008 Phys. Rev. Lett. 101 087004
[4] Mazin I I, Singh D J, Johannes M D and Du M H 2008 Phys. Rev. Lett. 101 057003
[5] Aoki H 2006 Physica C 437–38 11
   Aoki H 2007 Condensed Matter Theories vol 21 ed H Akai et al (New York: Nova Science) pp 147–61
[6] Dong J, Zhang H J, Xu G, Li Z, Hu W Z, Wu D, Chen G F, Dai X, Luo J L, Fang Z and Wang N L 2008 Europhys. Lett. 83 27006
[7] de la Cruz C et al 2008 Nature 453 899
[8] Baroni S et al 1996 http://www.pwscf.org/
   Perdew J P, Burke K and Wang Y 1996 Phys. Rev. B 54 16533
   Monkhorst H J and Pack J D 1976 Phys. Rev. B 13 5188
[9] Singh D J and Du M-H 2008 Phys. Rev. Lett. 100 237003
[10] Marzari N and Vanderbilt D 1997 Phys. Rev. B 56 12847
   Souza I, Marzari N and Vanderbilt D 2002 Phys. Rev. B 65 035109
   Mostofi A A, Yates J R, Marzari N, Souza I and Vanderbilt D http://www.wannier.org/
[11] Ishibashi S, Terakura K and Hosono H 2008 J. Phys. Soc. Japan 77 053709
[12] Fukuyama H 2008 J. Phys. Soc. Japan online-news and comments (12 May 2008)
[13] Yada K and Kontani H 2005 J. Phys. Soc. Japan 74 2161
[14] Takimoto T, Hotta T and Ueda K 2004 Phys. Rev. B 69 104504
[15] Bulut N, Scalapino D J and Scalletar R T 1992 Phys. Rev. B 45 5577
[16] Kuroki K and Arita R 2001 Phys. Rev. B 64 024501
[17] Kuroki K, Kimura T and Arita R 2002 Phys. Rev. B 66 184508
[18] Takada K, Sakurai H, Takayama-Muromachi E, Izumi F, Dilanian R and Sasaki T 2003 Nature 422 53
[19] Kuroki K, Onari S, Tanaka Y, Arita R and Nojima T 2006 Phys. Rev. B 73 184503
[20] Aoki H 2008 Proc. µSR, Physica B at press (arXiv:0811.1656)