Unconventional pairing originating from disconnected Fermi surfaces in superconducting LaFeAsO$_{1-x}$F$_x$

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Given this background, the purpose of the present Letter is to first construct a microscopic electronic model for LaFeAsO$_{1-x}$F$_x$, which then serves as the basis for identifying the possible mechanisms why this material favors high-$T_c$. The minimal model has turned out to contain all the five Fe $d$ orbitals, to which we have applied the random-phase approximation (RPA) to solve the Eliashberg equation. We shall conclude that a peculiar Fermi surface consisting of multiple pockets and ensuing multiple spin-fluctuation modes realize an unconventional $s$-wave pairing, while $d$-wave pairing can also be another candidate.

LaFeAsO has a tetragonal layered structure, in which Fe atoms are arrayed on a square lattice. Due to the tetrahedral coordination of As, there are two Fe atoms per unit cell. Each Fe layer is then sandwiched between LaO layers. The experimentally determined lattice constants are $a = 4.03552\AA$ and $c = 8.7393\AA$, with two internal coordinates $z_{La} = 0.1415$ and $z_{As} = 0.6512$. We have obtained the band structure (Fig.1(a) inset) with the Quantum-ESPRESSO package, and then construct the maximally localized Wannier functions (MLWFs) of these MLWFs, centered at the two Fe sites in the unit cell, have five orbital symmetries (orbital $1: d_{3z^2-r^2}$, $2: d_{xz}, 3: dy^2$, $4: dx^2-y^2$, $5: dxY$, where $X, Y, Z$ refer to those for the original unit cell). We can note that the two Wannier orbitals in each unit cell are equivalent in that each Fe atom has the same local arrangement of other atoms. We can thus take a unit cell that contains only one orbital per symmetry by unfolding the...
TABLE I: Hopping integrals $t(\Delta x, \Delta y; \mu, \nu)$ in units of 0.1eV. $[\Delta x, \Delta y]$ denotes the in-plain hopping vector, and $(\mu, \nu)$ the orbitals. $\sigma_y$, $I$, and $\sigma_d$ corresponds to $t(\Delta x, -\Delta y; \mu, \nu)$, $t(-\Delta x, -\Delta y; \mu, \nu)$, and $t(\Delta x, \Delta y; \mu, \nu)$, respectively, where '+' and '-' in the row of $(\mu, \nu)$ mean that the corresponding hopping is equal to $t(\Delta x, \Delta y; \mu, \nu)$ and $t(\Delta x, -\Delta y; \mu, \nu)$, respectively. This table, combined with the relation $t(\Delta x, \Delta y; \mu, \nu) = t(-\Delta x, -\Delta y; \nu, \mu)$, gives all the in-plain hoppings $\geq 0.01$eV up to fifth neighbors.

Brillouin zone (BZ) and we end up with an effective five-band model on a square lattice, where $x$ and $y$ axes are rotated by 45 degrees from $X$-$Y$ (Fig.1(b) inset), to which we refer for all the wave vectors hereafter. The in-plain hopping integrals $t(\Delta x, \Delta y, \mu, \nu)$ are displayed in Table I where $[\Delta x, \Delta y]$ is the hopping vector, and $\mu, \nu$ label the Wannier orbitals. The on-site energies for the five orbitals are $(\varepsilon_1, \varepsilon_2, \varepsilon_3, \varepsilon_4, \varepsilon_5) = (10.75, 10.96, 10.96, 11.12, 10.62)$ eV. With these effective hoppings and on-site energies, the in-plane tight-binding Hamiltonian is given in the form

$$H_0 = \sum_{ij} \sum_{\mu \nu} \sum_{\sigma} \left[ t(x_i - x_j, y_i - y_j; \mu, \nu) c^\dagger_{i\mu\sigma} c_{j\nu\sigma} + t(x_i - x_j, y_i - y_j; \nu, \mu) c^\dagger_{i\nu\sigma} c_{j\mu\sigma} + \sum_{i\mu\sigma} \varepsilon_{\mu} n_{i\mu\sigma} \right], \quad (1)$$

where $c^\dagger_{i\mu\sigma}$ creates an electron with spin $\sigma$ on the $\mu$-th orbital at site $i$, and $n_{i\mu\sigma} = c^\dagger_{i\mu\sigma} c_{i\mu\sigma}$. 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 $n = 6 + x$.

In the obtained band structure in Fig.1(a), we notice that the five bands are heavily entangled, reflecting strong hybridization (see Table I) 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 Fig.1(b), 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 BZ. The Fermi surface consists of four pieces (pockets in 2D): two concentric hole pockets (denoted as $\alpha_1, \alpha_2$) centered around $(k_x, k_y) = (0, 0)$, 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 $\Gamma Z$ (MA) line in the original BZ in the first-principles band calculation. [14] [15] [17]

Having constructed the model, we now move on to the
the self-energy correction in the Green’s function is ne-
hances tendency towards magnetism. In the RPA (where
\( \sim \) those in models studied by Bulut
hole and outer electron Fermi pockets is analogous to
velop. Such a sign change of the gap between inner
in the original BZ) at which the spin fluctuations de-
π, \( \pi/2 \)) to \( (\pi/2, \pi) \). This in fact reflects
the Fermi surface of each band. At this temperature
bands 3 and 4 (as counted from below), together with
range considered.

magnetic ordering does not take place in the temperature
as small as 1.2eV to ensure
U > U’, so we focus on
the spin susceptibility. We denote the largest eigenvalue
of the spin susceptibility matrix for \( \omega_n = 0 \) as \( \chi_s(k) \).
The gap function matrix at the lowest Matsubara fre-
cency is transformed into the band representation by
a unitary transformation, and its diagonal element for
band i is denoted as \( \phi_i(k) \).

Let us first look at the result for \( \chi_s \) for \( U = 1.2, U’ = 0.9, J = J’ = 0.15, \) and \( T = 0.02 \) (all in units
of eV) in Fig.2(a). The spin susceptibility has peaks
around \((k_x, k_y) = (\pi, 0), (0, \pi)\) and also a ridge-like
structure from \((\pi, \pi/2)\) to \((\pi/2, \pi)\). This in fact reflects
the Fermi surface in Fig.1(b), where we have two kinds
of nesting vector: \( \sim (\pi, 0), (0, \pi) \) across \( \alpha \) and \( \beta \), and
\( \sim (\pi, \pi/2), (\pi/2, \pi) \) across \( \beta_1 \) and \( \beta_2 \). A good nesting en-
hances tendency towards magnetism. In the RPA (where
the self-energy correction in the Green’s function is ne-
eglected), we have to take \( U \) as small as 1.2eV to ensure
magnetic ordering does not take place in the temperature
range considered.

For SC, we show in Fig.2(c)(d) the gap function for
bands 3 and 4 (as counted from below), together with
the Fermi surface of each band. At this temperature
\( T = 0.02 \), the eigenvalue of the Eliashberg equation
is \( \lambda = 0.96. \) The gap is basically s-wave, but changes
sign between the Fermi surface of band 3 (\( \alpha_2 \)) (and also
band 2; \( \alpha_1 \), not shown) and those of band 4 (\( \beta_1, \beta_2 \)),
namely, across the nesting vector \( \sim (\pi, 0), (0, \pi) \) (M point
in the original BZ) at which the spin fluctuations de-
velop. Such a sign change of the gap between inner
hole and outer electron Fermi pockets is analogous to
those in models studied by Bulut et al.,\(^{26} \) and also
by two of the present authors.\(^{27, 28} \) It is also remi-
niscent of the unconventional s-wave pairing mechanism
for Na\(_x\)CoO\(_2\).\(^{29} \) proposed by four of the present
authors.\(^{30} \) After completion of the present study, we
have come to notice that a recent preprint by Mazin et al.
also concludes an s-wave pairing in which the gap changes
sign between \( \alpha \) and \( \beta \) Fermi surfaces,\(^{31} \) as schematically
shown in the upper panel of Fig.2(b). For the present set
of parameter values, in addition to this sign change, we
find that the nodes of the gap intersect the \( \beta \) Fermi sur-
face. This is because the spin fluctuations due to the
\( \beta_1 - \beta_2 \) nesting favor a sign change in the gap between \( \beta_1 \)
and \( \beta_2 \) Fermi surfaces. In fact, we have found that this
nodal line moves out of the \( \beta \) Fermi surface for the pa-
rameter values for which the spin fluctuations due to the
\( \beta_1 - \beta_2 \) nesting become less effective, e.g., for \( U = U’ \). In
that case, the gap becomes closer to the upper panel of
Fig.2(b).\(^{32} \)

We have so far focused on the diagonal elements of
the gap matrix in the band representation, but to be more
precise, we have to consider the off-diagonal (interband)
elements in order to make accurate comparison with ex-
periments. The off-diagonal elements in the present case

![FIG. 2: (color online) RPA result for the spin susceptibility \( \chi_s \)
(a), the gap functions \( \phi_3 \) (c) and \( \phi_4 \) (d), \( \sqrt{\langle \delta \phi^\dagger \delta \phi \rangle_{M \text{M}} \text{c}} \) (e) for \( U = 1.2, U’ = 0.9, J = J’ = 0.15, n = 6.1 \) and \( T = 0.02 \) (in eV). In
(c) and (d), the black (green) solid lines represent the Fermi
surfaces (gap nodes). In (b), the fully gapped extended s-
(upper panel) and \( d_{x^2-y^2} \)-wave gaps are schematically shown.]
turn out to be not negligibly small due to the heavy entanglement of the bands. One way to look at this effect is to calculate the quantity $\sqrt{(\hat{\phi}^\dagger \hat{\phi})_{14}}$, where $\hat{\phi}$ is the gap matrix and 44 denotes the diagonal element of band 4. As shown in Fig.2(e), we find that this quantity is finite over the entire BZ, but a remnant of the nodal lines of the diagonal element still appears as a valley that intersects the $\beta$ Fermi surface. In this sense, we can say that the magnitude of the gap varies along the $\beta$ Fermi surface (becomes large at points far from the BZ edge) if the spin fluctuations arising from $\alpha$-$\beta$ and $\beta_1$-$\beta_2$ interactions have competing strength. The degree of the variation of the gap in the actual materials may be determined experimentally from the density of states, e.g., tunneling spectroscopy, or directly by angle resolved photoemission studies.

In the above, we mainly considered the possibility of unconventional s-wave pairing. On the other hand, if the $\alpha$ Fermi surfaces are absent (or less effective), the simplest form of the gap would be the $d_{x^2-y^2}$-wave pairing ($d_{xy}$ in the original BZ), where the gap changes sign between $\beta_1$ and $\beta_2$ Fermi surfaces as shown in the lower panel of Fig.2(b). To check this, we have performed an RPA calculation on (i) the present model with $n = 6.3$ and (ii) a model where we artificially shift the crystal field splitting to let the $\alpha$ Fermi surfaces disappear for $n = 6.1$. In both cases, we indeed obtain the $d_{x^2-y^2}$-wave. Since the band structure generally changes from the LDA result due to correlation effects [1] or a band filling different from the formally expected value, we leave, at the present stage, this $d$-wave state as another candidate for the pairing symmetry in this material.

Many other interesting problems remain for future studies. Spin fluctuations and SC should be studied by taking into account the self-energy correction, for which a fluctuation exchange study is underway [22]. It is also intriguing to investigate whether the present unconventional gap can quantitatively account for the specific heat [2] and point-contact conductance [3] results. Also, further insight into the origin of the high $T_c$ SC in LaFeAsO$_{1-x}$F$_x$ may be obtained by performing similar microscopic studies on LaFePO$_{1-x}$F$_x$ [4] or LaNiPO [5].

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