Theory for superconductivity in iron pnictides at large coulomb $U$ limit

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Superconductivity in iron pnictides is studied by using a two-orbital Hubbard model in the large $U$ limit. The Coulomb repulsion induces an orbital-dependent pairing between charge carriers. The pairing is found mainly from the scattering within the same Fermi pocket where usually one single orbital dominates. The inter-pocket pair scatterings determine the symmetry of the singlet superconductivity, which is an extended $s$-wave at small Hund’s coupling, and $d$-wave at large Hund’s coupling and large $U$. The former is consistent with recent experiments of ARPES and Andreev reflection spectroscopy. Spin triplet states only become important at large exchange interaction $J$.

Keywords iron-based superconductor, strong coupling theory

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Superconducting (SC) iron pnictides have the highest transition temperature next to the cuprates [1−7]. The parent compounds are metallic spin density wave (SDW) state [8−14]. Superconductivity occurs when part of Fe$^{2+}$ ions are replaced by Fe$^+$ ions. Because of its high transition temperature and the unusual physical properties [15, 16], the conventional phonon mechanism seems unlikely to explain the observed superconductivity. A multi-orbital Hubbard model may be a starting point to study its superconductivity; lots of works have been devoted to this direction [17, 19–32]. Since the parent compound is metallic, most theories examine the SC instability from the weak Coulomb interaction point of view [19, 20, 22, 23, 26, 29]. On the other hand, the observed magnetic moment in the SDW phase is large [33], indicating importance of spin couplings. The dynamic mean field theory [17] also suggests its closeness to a Mott insulator. This calls for an alternative approach from the viewpoint of large Coulomb repulsion $U$, which will be the purpose of the present paper, serving as a supplemental study to the weak coupling approach. Our main results were published in a short letter [18], and here we include more details.

The electronic states of the compound are predominantly Fe-3d orbitals near the Fermi surface (FS) [10−12, 34], which is comprised of two hole pockets centered at $I = (0,0)$ and two electron pockets at $X = (\pi,0)$ and $Y = (0,\pi)$, in the unfolded Brillouin zone (BZ), corresponding to 1 Fe atom per unit cell. Note that the buckling of As-atoms reduces the BZ to the square enclosed by the dashed lines in Fig. 1. The FS structure can be reproduced by a 5-orbital model [20]. The bands near the FS are mainly $d_{xz}$ and $d_{yz}$ orbitals [34]. Raghu et al. [24, 25] used a 2-orbital model by combining $d_{xz}$ and $d_{yz}$ orbitals to reproduce the FS in the reduced BZ. Note that the 2-orbital model shifts a hole Fermi pocket from the $\Gamma$- to the $M = (\pi,\pi)$-points in the unfolded BZ, in comparison with the first principle calculations as pointed out by Lee and Wen [29]. However, since the experimental observables are in the reduced BZ, some essential physics may be obtained from the simplified model. In this paper, we also started from the 2-orbital model and study the superconductivity at large $U$ limit which is required by both the strong spin coupling as observed experimentally [33] and Mott physics from DMFT study [17]. We will argue that our qualitative results will remain unchanged by the simplification of the 2-orbital model. We find that the virtual hopping induces orbital
dependent pairings of charge carriers. The intra Fermi pocket pair scattering is strongest, and the the pairing symmetry is determined by inter pocket pair scatterings and is extended s-wave ($s_\pm$) for small Hund’s coupling and d-wave for large Hund’s coupling and large $U$. The $s_\pm$ state was proposed by Mazin et al. [19] based on the analysis of the small Fermi pockets and spin fluctuations, and was found in weak coupling or small $U$ approaches [20, 22]. Our result at large $U$ limit further supports the $s_\pm$ symmetry. This pairing state appears consistent with the APRES [26] and Andreev reflection spectroscopy [16].

The 2-orbital model reads [24, 25]

$$H = H_0 + H_I$$

where $H_I$ is an on-site Coulomb term, and $H_0$ is a tight-binding model on a square lattice of Fe-atoms,

$$H_0 = \sum_{k,m,\sigma} (\epsilon_{k}^{nm} - \mu) c_{k,m,\sigma}^\dagger c_{k,m,\sigma} - \sum_{k,\sigma} \xi_{k,\sigma} c_{k,\sigma}^\dagger c_{k,\sigma}$$

$$\xi_{k,\sigma} = \frac{\epsilon_{k,\uparrow} + \epsilon_{k,\downarrow}}{2} - \mu + \alpha \sqrt{\frac{\epsilon_{k,\uparrow} - \epsilon_{k,\downarrow}}{2} + (\frac{\epsilon_k}{2})^2}$$

where $\epsilon_k^{nm}$ is the hopping matrix between different orbitals in $k$-space, $n = 1$ or (2) denote orbitals $d_{xz}$ (or $d_{yz}$). $\mu$ is the chemical potential. $\alpha = \pm$ represents the electron or upper (+) band and the hole or lower (−) band, corresponding to the diagonalized energy $\epsilon_k^{\pm}$. The band and orbital representations are related by a unitary transformation, $\hat{c}_{k,\sigma} = \sum_{\alpha=\pm} w_{\alpha}(k) \hat{c}_{k,\alpha \sigma}$. Here we follow Refs. [24, 25] and parameterize $H_0$ by hopping integrals $t_{ij}^\alpha$ (or denoted as $t_{ij}^{nm}$ in the latter context) between two sites $i$ and $j = i + \tau$, which is the Fourier transform of $\epsilon_{nm}(k)$. We set

$$t_{1}^{11} = t_{1}^{22} = t_1, \quad t_{1}^{12} = t_{1}^{21} = t_2, \quad t_{1}^{12} = t_{1}^{21} = t_3, \quad t_{1}^{14} = t_{1}^{41} = t_4$$

by lattice and orbital symmetry. Then $\epsilon_{nm}(k)$ reads

$$\epsilon_{k,11} = -2t_1 \cos k_x - 2t_2 \cos k_y - 4t_3 \cos k_x \cos k_y$$

$$\epsilon_{k,12} = -2t_2 \cos k_x - 2t_1 \cos k_y - 4t_3 \cos k_x \cos k_y$$

$$\epsilon_{k,14} = -4t_4 \sin k_x \sin k_y$$

By choosing $t_1 = -t_2 = 1.3t, t_3 = t_4 = -0.85t$, the calculated FS with electron density per site $\approx 2.10$ is reproduced in Fig. 1, which is similar to the results from the first principle calculations [19, 24, 25] for LaFeAsO. The weight contributed from each orbital at the FS are illustrated in the figure. The state on the electron pocket around the $X$ ($Y$) is mainly from $d_{yz}$ ($d_{xz}$) orbital. The state on the hole pocket around the $\Gamma$ consists of $d_{yz}$ and $d_{xz}$ orbitals equally if $k$ is along the diagonals, and mainly from $d_{xz}$ (or $d_{yz}$) orbital if along the $x$ or $y$ axis. This weight distribution affects greatly the pairing strength which varies substantially among different pair scattering channels.

The on-site interaction resulted from the general Hamiltonian,

$$H_I = \frac{1}{2} \sum_{\sigma,\sigma} \int d\mathbf{x}d\mathbf{x}' \psi^\dagger_{\sigma}(\mathbf{x}) \psi_{\sigma}(\mathbf{x}')$$

$$\mathbf{v}(\mathbf{x} - \mathbf{x}') \psi_{\sigma}(\mathbf{x}') \psi_{\sigma}(\mathbf{x})$$

has the following form:

$$H_I = \sum_{i,m=1,2} (U \hat{n}_{i,m\uparrow} \hat{n}_{i,m\downarrow} + J_{i,m\uparrow} c_{i,m\uparrow}^\dagger c_{i,m \downarrow})$$

$$+ \sum_{i,\sigma,\sigma'} (U_2 \hat{n}_{i,\sigma\uparrow} \hat{n}_{i,\sigma'\downarrow} + J_{i,\sigma\uparrow} c_{i,\sigma\uparrow}^\dagger c_{i,\sigma'\downarrow})$$

where $\hat{n}_{i,\sigma} = c_{i,\sigma\uparrow}^\dagger c_{i,\sigma\downarrow}, U$ and $U_2$ are the intra- and inter-orbital direct Coulomb repulsions, respectively. The terms with $J$ are the exchange interaction. By rotational invariant symmetry, $U = U_{12} + 2J$ [35]. In the limit, $U, J \gg t$, each lattice site is doubly occupied in the parent compound. Upon electron doping, some sites will have 3 electrons (or 1 hole). A single hole at site $i$ may interchange with a two-hole state at site $j$, leading to a metallic phase. The effective interaction between two single holes on neighboring sites ($i, j$) can be derived by using the second-order perturbation theory by considering the virtual hopping processes [35], similar to the super-exchange interaction derived in the single band Hubbard model, and it is given by
By using the singlet and triplet pairing operators between different orbits

\[ H_{\text{eff}} = -\sum_{i,j,\sigma,n,m,n',m'} t_{i,j}^{n,m,n',m'} c_{i,n,\sigma}^\dagger c_{j,n',\sigma} + \frac{U_{12}}{U_{12} - J_{12}^2} \hat{n}_{j,m,\sigma} + \frac{1}{U_{12} - J_{12}} \hat{n}_{j,m,\sigma} \]

\[ -\sum_{i,j,\sigma,n,m,n',m'} t_{i,j}^{n,m,n',m'} c_{i,n,\sigma}^\dagger c_{j,n',\sigma} + \frac{J_{12}}{U_{12} - J_{12}^2} c_{j,m,\sigma} c_{j,m,\sigma} \]

\[ -\sum_{i,j,\sigma,n,m,n',m'} t_{i,j}^{n,m,n',m'} c_{i,n,\sigma}^\dagger c_{j,n',\sigma} \left( -\frac{J_{12}}{U_{12} - J_{12}^2} c_{j,m,\sigma} c_{j,m,\sigma} + \frac{1}{U_{12} - J_{12}} c_{j,m,\sigma} c_{j,m,\sigma} \right) \]

\[ -\sum_{i,j,\sigma,n,m,n',m'} t_{i,j}^{n,m,n',m'} c_{i,n,\sigma}^\dagger c_{j,n',\sigma} \left( -\frac{J_{12}}{U_{12} - J_{12}^2} c_{j,m,\sigma} c_{j,m,\sigma} - \frac{1}{U_{12} - J_{12}} c_{j,m,\sigma} c_{j,m,\sigma} \right) \]

(6)

The effective pairing interaction between carriers derived in the large limit should be relevant to the intermediate coupling region [36, 37].

The effective Hamiltonian is then \( H_{\text{eff}} = H_0 + H_2 \), subject to the constraint of no more than 2 holes per site. This can formally be represented by a Gutzwiller projection operator to project all the unphysical states, similar to that in the \( t-J \) model [38]. Mathematically, \( H_{\text{eff}} \) may be studied by using a renormalized Hamiltonian approach to take into account the projection [39] by introducing renormalization factors, \( g_0 \) for \( H_0 \) and \( g_2 \) for \( H_2 \), both are doping dependent. For a given doping, the effect of the renormalization is to scale all the \( t \)'s to \( g_0 t \)'s, and \( (U, J) \) to \( (g_0^2/g_2)(U, J) \). Below we will absorb these renormalization factors into the parameters \( (t \text{'s and } U) \) and effectively set \( g_0 = g_2 = 1 \) in our calculations.

\[ H_{\text{eff}} \] can then be solved using a mean field theory by introducing mean fields for the spin–singlet pairing with even parity and symmetric orbitals [40–43],

\[ \Delta_{mnm}(\tau) = \Delta_{mnm}(\tau) \left( \frac{1}{\sqrt{2}} \hat{n}_{mnm}(i, i + \tau) \right) \]

(10)

with \( \tau = \pm \hat{x}, \pm \hat{y}, \pm (\hat{x} \pm \hat{y}) \). By symmetry analysis, we have \( \Delta_{11}(\hat{x}) = \pm \Delta_{22}(\hat{y}) \), \( \Delta_{11}(\hat{y}) = \pm \Delta_{22}(\hat{x}) \), \( \Delta_{12}(\hat{x}) = \Delta_{12}(\hat{y}) = 0 \), \( \Delta_{11}(\hat{x} \pm \hat{y}) = \pm \Delta_{22}(\hat{x} \mp \hat{y}) \), \( \Delta_{12}(\hat{x} \pm \hat{y}) = 0 \). Note that \( \Delta_{12}(\hat{x} \pm \hat{y}) = 0 \) for the \( d \)-wave state. The pairing strength with \( A_{2g} \) and \( B_{2g} \) symmetries [40–43] are found to be very small, and will not be discussed in the following context [44].

The mean field Hamiltonian of \( H_{\text{eff}} \) can be written in Nambu representation as:

\[ H_{\text{MF}} = \sum_k \psi_k^\dagger \left( \begin{array}{cc} \xi_{k} & V(k) \\ V^\dagger(k) & -\xi_{k} \end{array} \right) \psi_k \]

(11)

where \( \psi_k^\dagger = \left( \hat{c}_{k+1}, \hat{c}_{k-1}, \hat{c}_{-k+1}, \hat{c}_{-k-1} \right) \). Pair coupling non-diagonal component \( V(k) \) is a \( 2 \times 2 \) matrix in band picture, given by

\[ V_{a\beta}(k) = \sum_{nmnm'} A_{nmn'}(\tau) \Delta_{nmn'}^*(\tau) c_{k\tau}^\dagger u_{m'm}(k) u_{nm\beta}(k) \]

\[ = \sum_{nmnm'} S_{nmn'}(k) u_{m'm}(k) u_{nm\beta}(k) \]
where $S$ is the $2 \times 2$ matrix in orbital picture. The $S$ matrices can be written as $S(k) = S_0(k) \tau_0 + S_1(k) \tau_1 + S_2(k) \tau_2 + S_3(k) \tau_3$ with the Pauli matrix $\tau_i$. According to the symmetry analysis [40–43], $S_2 = 0$ for both $s_\pm$ states and $d$-wave states. According to Eqs. (3) and (9), the $S$ matrices for $s_\pm$ states are

$$S_0(k) = 4 \cos k_x + \cos k_y \left[ \left( t_2^2 - t_1 t_2 J \right) \Delta_{11}(\hat{x}) + \left( t_2^2 - t_1 t_2 J \right) \Delta_{22}(\hat{x}) \right]$$

$$+ 16 \cos k_x \cos k_y \left[ \left( t_2^2 - t_1 t_2 J \right) \Delta_{11}(\hat{x} + \hat{y}) - t_3 t_4 \left( t_2^2 - t_1 t_2 J \right) \Delta_{12}(\hat{x} + \hat{y}) \right]$$

$$S_1(k) = -16 \sin k_x \sin k_y \left[ \left( t_2^2 - t_1 t_2 J \right) \Delta_{11}(\hat{x}) - t_3 t_4 \left( t_2^2 - t_1 t_2 J \right) \Delta_{12}(\hat{x}) \right]$$

$$S_3(k) = 4 \cos k_x - \cos k_y \left[ \left( t_2^2 - t_1 t_2 J \right) \Delta_{11}(\hat{x}) - \left( t_2^2 - t_1 t_2 J \right) \Delta_{22}(\hat{x}) \right]$$

and the ones for $d$-wave states are

$$S_0(k) = 4 \cos k_x - \cos k_y \left[ \left( t_2^2 - t_1 t_2 J \right) \Delta_{11}(\hat{x}) + \left( t_2^2 - t_1 t_2 J \right) \Delta_{22}(\hat{x}) \right]$$

$$S_1(k) = 0$$

$$S_3(k) = 4 \cos k_x + \cos k_y \left[ \left( t_2^2 - t_1 t_2 J \right) \Delta_{11}(\hat{x}) - \left( t_2^2 - t_1 t_2 J \right) \Delta_{22}(\hat{x}) \right]$$

$$+ \frac{16 \cos k_x \cos k_y}{U - J} \left[ t_3^2 \Delta_{11}(\hat{x} + \hat{y}) + t_3 t_4 \Delta_{12}(\hat{x} + \hat{y}) \right]$$

$H_{MF}$ can be solved self-consistently, and the energy per site is $E = -\frac{1}{N} \sum_{k } E_{\pm}(k)$, with $E_{\pm}(k)$ the quasiparticle energy of the upper (+) and lower (−) bands, given by

$$E_{\pm}(k) = \sqrt{w_k^2 + V_{\pm}^2} \pm \sqrt{w_k^2 + V_{\pm}^2 \left[ (\xi_+ - \xi_-)^2 + (V_{++} + V_{--})^2 \right]}$$

where $w_k^2 = [\xi_+^2 + V_{++}^2 \pm (\xi_-^2 + V_{--}^2)]/2$, and the $k$-dependence is implied. In Fig. 2, the energies of the SC states are depicted as functions of $J/U$ for $t/U = 0.1$ and $t/U = 0.2$. At $t/U = 0.2$, the $s_\pm$ state is always energetically favorable. At $t/U = 0.1$, the ground state is $s_\pm$-wave if $J/U < 0.16$ and a $d$-wave if $J/U > 0.16$.

In Fig. 3, we plot the intra-band pairing amplitude $V_{++}(k)$ for the electron-band and $V_{--}(k)$ for the hole band. In the $s_\pm$ state, $V(k)$ is invariant under a $\pi/2$ rotation. However, unlike the conventional $s$-wave case, both $V_{++}(k)$ and $V_{--}(k)$ have a nodal line in the BZ, and the pairing amplitudes $V_{++}$ on $X$ and $Y$ pockets...
have the same sign, but are opposite to $V_{- -}$ on $\Gamma$, similar to the state proposed by Mazin et al. [19]. In the $d$-wave state which is stabilized by the strength and sign of the pair scattering between the two FS on $X$ and $Y$, the diagonal component $V_{\alpha \alpha}(k)$ changes a sign under a $\pi/2$ rotation, and has nodal lines along the diagonals in the BZ.

Let us examine the pairing strength at the FS around the Fermi pockets $Y$ and $\Gamma$. For a Fermi wavevector $k_F$ on the Fermi pocket centered at $C = (k_c^x, k_c^y)$, we define an angle $\theta = \arctan[(k_F^y - k_c^y)/(k_F^x - k_c^x)]$. The $\theta$-dependences of $V(k)$ on the electron pocket around $Y$ and the hole pocket around $\Gamma$ of the $s_{\pm}$ state are plotted in Fig. 4. For the $s_{\pm}$ state, $|V_{++}| \gg |V_{+-}|, |V_{- -}|$ on $Y$-pocket [see Fig. 4(a)]. This suggests that the SC pairing is mainly due to the electron pairing of the same orbital. At the pocket centered at $\Gamma$, $V_{+-}$ is negligibly small, so that the SC pairing is mainly due to the hole pairings. We emphasize that although there are nodal lines, $V_{++}$ on pocket $Y$ and $V_{- -}$ on pocket $\Gamma$ are always finite. There is a full gap on both Fermi pockets around $Y$ and $\Gamma$, consistent with recent ARPES and Andreev reflection spectroscopy results. Because of the above analyses, we have $E_-(k) \approx V_{++}(k)$ around $Y$ and $E_-(k) \approx V_{- -}(k)$ around $\Gamma$ as shown in Fig. 4(c) and (d). The results for the $d$-wave state are also shown in Fig. 5. The $V(k)$ on the electron pocket are similar with the $s_{\pm}$ result. However, the nodal line of $V_{--}(k)$ crosses the hole Fermi pocket and leads to a $d$-wave like quasiparticle spectrum. The quasiparticle energy at the nodal point is given by $E_k = V_{++}^2(k)/E_{++}(k)$. Since $V_{+-}(k) \neq 0$, but small, $E_k$ is non-zero but very small [not distinguishable from 0 in Fig. 5(d)].

To better understand the SC pairing and its symme-
Fig. 4 Angle dependence of pairing amplitude $V_{++}^{(k)}$ (red line), $V_{--}^{(k)}$ (blue line), and $V_{+-}^{(k)}$ (green line) along the Fermi pocket around $Y$ [panel (a)] and $\Gamma$ [panel (b)] in the $s_{\pm}$-state. (c) and (d): The quasiparticle gap on the electron Fermi pocket and hole Fermi pocket for $s_{\pm}$-states respectively.

Fig. 5 Angle dependence of pairing amplitude $V_{++}^{(k)}$ (red line), $V_{--}^{(k)}$ (blue line), and $V_{+-}^{(k)}$ (green line) along the Fermi pocket around $Y$ [panel (a)] and $\Gamma$ [panel (b)] in the $d$-state. (c) and (d): The quasiparticle gap on the electron Fermi pocket and hole Fermi pocket for $d$-states respectively.
From Eq. (16), we find that the intra-pocket pair scattering between hole and electron pockets are always repulsive.

$A_{nm}^{\pi,\pi}(q) = 4U(t_1^3 \cos q_x + t_2^3 \cos q_y) / (U^2 - J^2) + 4J \cos q_x \cos q_y$

$A_{21}(q) = A_{12}(q) = 4t_1^2 \cos q_x \cos q_y / (U + J) + 2t_1^2 \cos q_x \cos q_y / (U - J)$

but they are much less important than the orbital diagonal ones since substantial orbital hybridization only happens on the diagonal direction of the hole pocket and the pairing mainly formed between the same orbitals.

Finally, we discuss the effect to the superconductivity due to the simplification in the 2-orbital model, which results in the shift of a hole Fermi pocket from the $\Gamma$- to $M$-point. Let us consider the effect of this Fermi pocket to the pairings. From Eqs. (15) and (16), the pair scatterings between the hole pocket and the electron pockets are repulsive for both hole pocket centered at the $\Gamma$ or $M$ points. The pair scattering between a hole pocket at $\Gamma$ and a hole pocket at $M$ is very small because of the cancelation from the intra and inter-orbital scatterings, while the pair scattering between the two hole pockets both centered at the $\Gamma$ point is attractive and strong, which further favors the extended $s$-wave over the $d$-wave states. In brief, the qualitative physics obtained from our study of the 2-orbital model is expected to remain the same except that the parameter space for the extended $s$-wave state is expected to be enlarged when more accurate band structure is considered. To further ensure the qualitative conclusions of our theory, we have examined a 3-orbital model as in Ref. [29], in which there are two hole pockets around $\Gamma$ in the unfolded BZ, which is better in agreement with the LDA calculations. We have extended our analyses of Eq. (15) to that model and the pairing symmetries are found essentially the same as from the 2-orbital model.

In summary, we have examined superconductivity in iron pnictides using a 2-orbital Hubbard model at the large $U$ limit. An extended $s$-wave pairing is found most stable in a large parameter space, consistent with early theories starting with weak coupling (small $U$) and with ARPES [15] and tunneling experiments [16]. Contrary to some weak coupling theories, we find the pairing to be mainly from the pair scattering within the same Fermi pocket. Our analyses suggest some similarities between the superconductivity in iron pnictides and in the cuprates.

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