Spin fluctuations and pairing symmetry in $A_xFe_{2-y}Se_2$: dual effect of the itinerant and the localized nature of electrons

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Abstract

We investigate the spin fluctuations and the pairing symmetry in $A_xFe_{2-y}Se_2$ by the fluctuation exchange approximation. Besides the on-site interactions, the next-nearest-neighbor antiferromagnetic coupling $J_2$ is also included. We find that both the itinerant and the localized natures of electrons are important to describe recent experimental results on the spin fluctuations and the pairing symmetry. In particular, a small $J_2$ coupling can change the pairing gap from the d-wave symmetry to the extended s-wave symmetry. We have also studied the real-space structures of the gap functions for different orbits in order to gain more insight into the nature of the pairing mechanism.

(Some figures may appear in colour only in the online journal)

1. Introduction

The discovery of a new family of iron-based superconductors (FeSCs) $A_xFe_{2-y}Se_2$ ($A = K, Rb, Cs$) with $T_c$ more than 30 K has attracted much attention [1–4]. Experiments on these materials show some surprising results which apparently differentiate them from other FeSC families (iron chalcogenide and iron pnictide). First, angle-resolved photoemission (ARPES) studies indicate that there are only electron-like Fermi surfaces (FSs) and no hole-like FSs in $A_xFe_{2-y}Se_2$ [5–7], while other FeSC families have both hole- and electron-like FSs simultaneously [8]. Second, they are the first FeSCs that approximate antiferromagnetic (AFM) insulators accompanied by Fe-vacancy order [4]. In addition, the AFM order has a novel $\sqrt{5} \times \sqrt{5}$ block-type structure with an unprecedented high transition temperature of $T_N = 559$ K and a large magnetic moment of $3.31 \mu_B$/Fe [9].

In iron pnictides, it is shown that the nesting between the hole and electron Fermi pockets gives rise to inter-band spin fluctuation with wavevector $(0, \pi)$ based on the weak coupling picture. This inter-band spin fluctuation leads to the dominant $s_{\pm}$-wave pairing state, in which the gap function changes sign between the hole and electron pockets [10–13]. Obviously, the absence of hole Fermi pockets in $A_xFe_{2-y}Se_2$ violates the nesting condition of the $s_{\pm}$ pairing symmetry, so that a reconsideration of the itinerant spin-fluctuation mechanism in these new systems is required. Based on the multi-orbital Hubbard model and functional-renormalization-group calculation, Wang et al [14] find that the $d_{x^2−y^2}$-wave instability is the leading pairing channel, with a subleading extended s-wave component. A similar conclusion has also been obtained by Maier et al [15] by performing random-phase-approximation (RPA) calculations. On the other hand, based on the local-antiferromagnetic-exchange interactions, Seo et al [16] predict an s-wave pairing symmetry in this system that can account for the ARPES experimental results [5–7]. Khodas et al [17] suggest an $s_{\pm}$ pairing symmetry, in which the gap changes sign between the hybridized pockets, by including the inter-pocket pairing.

Experimentally, ARPES measurements have reported a nodeless superconducting (SC) gap in the large Fermi pockets around the zone corner in these materials [5–7, 18, 19]. In particular, recent measurements further find an isotropic SC gap distribution in the small electron Fermi pocket around the Z point [20, 21], which favors the s-wave pairing symmetry. The experimental evidence from nuclear magnetic resonance (NMR) experiments suggests an $s_{\pm}$-wave pairing symmetry [22, 23], which is consistent with the isotropic gaps reported by ARPES. A similar conclusion is also obtained from muon spin spectroscopy experiments [24] and specific heat measurements [25]. Thus, the theoretical prediction of the $d_{x^2−y^2}$-wave pairing state is questioned by these experiments.
On the other hand, although inelastic neutron scattering (INS) measurements [26–28] report a spin resonance in the superconducting state at \( Q \approx (\pi, \pi/2) \) which is very close to the value predicted by Maier et al. [15] with a d-wave pairing symmetry, another spin excitation near \( Q = (0, \pi) \) was also observed in the superconducting state recently [27]. In the meantime, neutron scattering experiments on semiconducting \( \text{K}_2\text{Fe}_{2−y}\text{Se}_2 \) compounds, which neighbor the superconducting ones, have also revealed strong \((0, \pi)\) AFM long range order [29]. However, due to the absence of hole Fermi pockets in \( \text{A}_x\text{Fe}_{2−y}\text{Se}_2 \), the nesting condition between the hole and electron Fermi pockets is violated, so this \((0, \pi)\) spin fluctuation is hard to account for based on the weak coupling itinerant-electron picture.

In view of these experimental developments, we will study the spin fluctuations and the pairing symmetry in \( \text{A}_x\text{Fe}_{2−y}\text{Se}_2 \) with the fluctuation exchange (FLEX) approximation by including the next-nearest-neighbor (NNN) antiferromagnetic (AF) coupling \( J_2 \), a localized magnetic interaction, in the multi-orbital Hubbard model, which is treated within an itinerant-electron picture. In fact, INS experiments [30–33] have indicated that the NNN exchange coupling is AF and has a similar magnitude in many FeSCs, although the nearest-neighbor (NN) exchange coupling is quite different. This implies that the NNN exchange coupling may play an important role in the FeSCs. However, in FeSCs with both hole- and electron-like FSs the NNN exchange coupling has a similar effect to FS nesting on the spin excitations. However, we will show that both the itinerant and the localized natures of electrons are essential to describe properly the physical properties in \( \text{A}_x\text{Fe}_{2−y}\text{Se}_2 \), where the hole-like Fermi pocket disappears. In particular, a small \( J_2 \) can change the pairing gap from the d-wave symmetry to the extended s-wave symmetry, which is consistent with the nodeless gap observed in recent experiments. Considering the degeneracy of the two pairing states near the transition, we suggest a possible s + id-wave state in order to account for the spin resonance observed in INS experiments. We have also studied the real-space structures of the pairing gap at different orbits, from which we obtain the expressions for the favorable pairing gap functions.

2. Model and flex method

The model Hamiltonian consists of two parts,

\[
H = H_0 + H_{\text{int}}.
\]

The non-interacting part \( H_0 \) is described by a two-dimensional five-orbital tight-binding model in the unfolded (1-Fe) Brillouin zone (BZ) [14]. The tight-binding Hamiltonian reads

\[
H_0 = \sum_{k, \sigma} \sum_{\alpha, \beta = 1}^5 t_{k\sigma}^\alpha K_{\alpha\beta}(k)c_{\alpha\beta}^\dagger c_{k\beta\sigma},
\]

where \( c_{k\sigma}^\dagger \) (\( c_{k\sigma} \)) creates (annihilates) an electron with spin \( \sigma \) and momentum \( k \) in the orbital \( \alpha \). \( K_{\alpha\beta}(k) \) is obtained from \( H_{\alpha\beta}(k) \) in [14] through the following transformation:

\[
K(k) = G^\dagger H(k)G.
\]

with

\[
G = \begin{pmatrix}
1 & 0 & 0 & 0 & 0 \\
0 & 1 & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 \\
0 & \frac{1}{\sqrt{2}} & 1 & 0 & 0 \\
0 & 0 & 0 & 1 & 0 \\
0 & 0 & 0 & 0 & -1
\end{pmatrix}.
\]

This transformation changes the orbital symmetry from \( (d_{x^2−y^2}, d_{z^2}, d_{xy}, d_{xz}, d_{yz}) \) in this paper. Here, \( x, y \) refer to the nearest-neighbor Fe–Fe directions and \( X, Y \) refer to the diagonal directions. The details of \( H_{\alpha\beta}(k) \) can be found in [14], where the parameters of the model involve the nearest- and second-neighbor hopping among the Fe 3d orbits. The band structure and the associated FSs for 0.3 electron doping (six electrons per Fe for the parent compound) are shown in figure 1. We find that there are only electron FSs around \((0, \pm \pi)\) and \((\pm \pi, 0)\), which is consistent with the results of ARPES experiments [5–7]. Here, we do not consider the influence of Fe vacancies, because many experiments suggest that the antiferromagnetism with Fe vacancies and the superconductivity without Fe vacancies are in fact spatially separated [34–39].

The interactions between electrons are included in \( H_{\text{int}} \) as follows:

\[
H_{\text{int}} = U \sum_{i, \sigma} n_{i\sigma \uparrow} n_{i\sigma \downarrow} + \frac{1}{2} U' \sum_{i, \alpha \neq \beta \neq \sigma} n_{i\alpha \sigma} n_{i\beta \sigma'}
\]

\[
- J_{\text{H}} \sum_{i, \alpha \neq \beta} \left( S_{i\alpha} \cdot S_{i\beta} + \frac{1}{2} \sum_{\sigma \sigma'} n_{i\alpha \sigma} n_{i\beta \sigma} \right)
\]

\[
+ J_{2} \sum_{(0y), \alpha \beta} S_{i\alpha} \cdot S_{i\beta},
\]

where \( U \) (\( U' \)) is the intra-orbital (inter-orbital) Coulomb interaction, \( J_{\text{H}} \) the Hund coupling, and \( J_2 \) the NNN antiferromagnetic coupling. \( n_{i\alpha \sigma} = c_{i\alpha \sigma}^\dagger c_{i\alpha \sigma} \) and \( S_{i\alpha} = \frac{1}{2} \sum_{\sigma \sigma'} c_{i\alpha \sigma}^\dagger \sigma \sigma' c_{i\alpha \sigma} \) are the density and spin operators respectively, where the \( \tau \) are the Pauli matrices.

As mentioned above, due to the absence of hole pockets in \( \text{A}_x\text{Fe}_{2−y}\text{Se}_2 \), the NNN antiferromagnetic coupling \( J_2 \) may play an essential role in determining the physical properties in these materials. Moreover, since the \( J_2 \) exchange coupling originates mostly from superexchange processes through Se, the effect of \( J_2 \) is underestimated in the itinerant-electron model based on the pure iron lattice with only the on-site interactions. On the other hand, INS experiments [30–33] have indicated that the NNN exchange couplings are AF and have similar values in many FeSCs. Hence, we consider the dual effect of both the on-site interactions and the \( J_2 \) exchange coupling here. A study based on similar considerations for the interactions in the two-orbital model for pnictide superconductors with both electron and hole FSs has been carried out using the strong coupling expansion [40].

We carry out the investigation using the FLEX approximation [13, 41–43], in which the Green’s function and
Figure 1. The energy band structure of the five-orbital model (a) and the FSs in the unfolded BZ for 0.3 electron doping (b). The colors indicate the majority orbital character (red: dₓ.z, green: dₓ.y, blue: dᵧ, yellow: d₂₋ₓ²/y², pink: d₁, orange: dₓ, and dᵧ with the same weight). The arrows with solid lines in (b) indicate the weak nesting vectors q = (π, π) and (−π, π); the arrows with dashed lines indicate the weak nesting vectors q = (π, 0,π) and (−0.6π, π).

The spin/charge fluctuations are determined self-consistently. Here, we consider the following spin fluctuations $\hat{\chi}^s(q, \omega_n)$ and charge fluctuations $\hat{\chi}^c(q, \omega_n)$:

$$
\hat{\chi}^s_{\alpha \beta}(q, \omega_n) = \int_0^1 d\tau e^{i\omega_n \tau} \langle T_\tau [S_\alpha(k, \tau) \cdot S_\beta(-k, 0)] \rangle, (6)
$$

$$
\hat{\chi}^c_{\alpha \beta}(q, \omega_n) = \int_0^1 d\tau e^{i\omega_n \tau} \langle T_\tau [\eta_\alpha(k, \tau) n_\beta(-k, 0)] \rangle, (7)
$$

where $S(k, \tau) = e^{\tau H} S(k) e^{-\tau H}$ with $S(k)$ the Fourier transformation of $S_i$. $T$ is the temperature, $\tau$ and $\omega_n$ are respectively the imaginary time and the Matsubara frequency. For the five-orbital model, the Green’s function $G$ and the self-energy $\Sigma$ are expressed in a $5 \times 5$ matrix form. As we use the spin and charge fluctuations expressed in equations (6) and (7), the susceptibility $\hat{\chi}^s$ and the effective interaction $\hat{V}$ also have a $5 \times 5$ matrix form. The Green’s function satisfies the Dyson equation $G(k)^{-1} = \tilde{G}^0(k)^{-1} - \tilde{\Sigma}(k)$, where the self-energy is given by $\Sigma_{\alpha \beta}(k) = \sum_q \sum_{\mu \nu} V_{\alpha \mu}(q) G_{\mu \nu}(k - q)$ and the bare Green’s function reads $\tilde{G}^0(k) = [\omega_n - k(k) + \mu]^{-1}$. Here, $k$ includes both momentum and frequency with $k = (k, \omega_n)$. The fluctuation exchange interaction is given by

$$
\hat{V}(q) = \frac{1}{2} \hat{U}_\alpha(q) [\hat{\Delta} - \hat{\chi}_c^0(q) \hat{U}_c(q)]^{-1} \hat{\chi}_c^0(q) \hat{U}_c(q) + \frac{1}{2} \hat{U}_c(q) [\hat{\Delta} + \hat{\chi}_c^0(q) \hat{U}_c(q)]^{-1} \hat{\chi}_c^0(q) \hat{U}_c(q) + \frac{1}{2} \hat{U}_\alpha(q) \hat{\chi}_c^0(q) \hat{U}_c(q) + \hat{U}_c(q) \hat{\chi}_c^0(q) \hat{U}_\alpha(q), (8)
$$

with the identity matrix $\hat{1}$ and the irreducible susceptibility $\chi_{\alpha \beta}(q) = -\frac{T}{N} \sum_\ell G_{\alpha \beta}(k + q) G_{\alpha \beta}(k)$. The interaction matrix for the spin (charge) fluctuation $\hat{U}_\alpha$ ($\hat{U}_c$) is given by the following: for $\alpha = \beta$, $U_{\alpha \beta}^c = U - 4 J_2 \cos k_x \cos k_y (U_{\alpha \beta}^c = U)$; for $\alpha \neq \beta$, $U_{\alpha \beta}^c = J_4 - 4 J_2 \cos k_x \cos k_y (U_{\alpha \beta}^c = 2 U - J_H)$.

The Dyson equation, the self-energy and the interaction matrix equation (8) form a closed set of equations and can be solved self-consistently. Since the equations must be solved at fixed electronic density $n$, they are subject to the additional constraint

$$
N(T) = \frac{T}{N} \sum_k e^{i\omega_0 \tau} \text{Tr} G(k, \mu(T)) = n, (9)
$$

where the chemical potential $\mu$ must be adjusted accordingly during the self-consistent calculation.

After obtaining the renormalized Green’s function $\hat{G}$, we can solve the ‘Elaisnberg’ equation

$$
\lambda \phi_{\alpha \beta}(k) = -\frac{T}{N} \sum_q \sum_{\mu \nu} \chi_{\alpha \beta}(q) G_{\alpha \beta}(k - q) \times G_{\mu \nu}(q - k) \phi_{\mu \nu}(k - q), (10)
$$

where the spin-singlet and spin-triplet pairing interactions $\hat{V}^s$ and $\hat{V}^t$ are given by

$$
\hat{V}^s(q) = \frac{1}{2} \hat{U}_\alpha(q) \hat{\chi}_c^0(q) \hat{U}_c(q) - \frac{1}{2} \hat{U}_c(q) \hat{\chi}_c^0(q) \hat{U}_\alpha(q) + \frac{1}{2} \hat{U}_\alpha(q) \hat{\chi}_c^0(q) \hat{U}_c(q) - \frac{1}{2} \hat{U}_c(q) \hat{\chi}_c^0(q) \hat{U}_\alpha(q), (11)
$$

$$
\hat{V}^t(q) = -\frac{1}{2} \hat{U}_\alpha(q) \hat{\chi}_c(q) \hat{U}_c(q) - \frac{1}{2} \hat{U}_c(q) \hat{\chi}_c(q) \hat{U}_\alpha(q) + \frac{1}{2} \hat{U}_\alpha(q) \hat{\chi}_c(q) \hat{U}_c(q) - \frac{1}{2} \hat{U}_c(q) \hat{\chi}_c(q) \hat{U}_\alpha(q). (12)
$$

The most favorable SC pairing symmetry corresponds to the eigenvector $\phi_{\alpha \beta}(k)$ with the largest eigenvalue $\lambda$.

Our numerical calculations are carried out on $64 \times 64$ k meshes with 4096 Matsubara frequencies. In the calculations, the intra-orbital Coulomb interaction $U = 1.2$ eV, the inter-orbital Coulomb interaction $U^t = 0.8$ eV and the relation $U = U^t + 2J_H$ are used. The electron density is set to be 6.3 electrons per Fe and the temperature is 0.008 eV.

3. Results and discussion

3.1. Spin fluctuation

As the spin fluctuations dominate over the charge fluctuations in the model described above, we only discuss the spin fluctuations and their contributions to the pairing interactions.

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of electrons in this paper. We first study the evolution of the spin fluctuations with the NNN antiferromagnetic coupling $J_2$. In figure 2, we present the physical spin susceptibilities $\chi(q, \omega = 0) = \sum_{\alpha \beta} \chi_{\alpha \beta}^s(q, \omega = 0)$ for four different values of $J_2$. For $J_2 = 0$, the absence of the hole pocket around the $\Gamma$ point in the unfolded BZ removes the dominant $q = (0, \pm \pi)$ and $(\pm \pi, 0)$ nestings in iron pnictide, so that the peaks of the spin fluctuations at $(0, \pm \pi)$ and $(\pm \pi, 0)$ disappear in $\alpha_x\text{Fe}_2\text{Se}_2$. Instead, there are weak intra-band nestings at $q = (\pm \pi, \pm 0.6\pi), (\pm 0.6\pi, \pm \pi)$ and $(\pm \pi, \pm \pi)$ in this system (see figure 1(b)), which lead to the broad peak around $(\pi, \pi)$, together with the four satellite peaks at $(\pi, \pm 0.6\pi)$ and $(\pm 0.6\pi, \pi)$ (figure 2(a)). We note that these results are not consistent with the experimental results from INS measurements [26, 27], in which the peak around $(\pi, \pi)$ is not observed. Thus, a simple Fermi surface nesting scenario within the itinerant-electron model is not sufficient to explain the neutron scattering experiments. When the $J_2$ interaction term is turned on, we find that the broad peak around $(\pi, \pi)$ decreases rapidly. Meanwhile, the intensity of the four satellite peaks increases and their positions move towards $(0, \pm \pi)$ and $(\pm \pi, 0)$, as shown in figures 2(b)–(d) for $J_2 = 0.01, 0.04$ and $0.05$ eV respectively. As the $J_2$ term favors the diagonal antiferromagnetic order in the square lattice of Fe, it competes with the spin fluctuations coming from the nesting of FSs with the nesting wavevector $(\pi, \pi)$ which are kinds of axial antiferromagnetic fluctuations. Thus, a sufficiently large $J_2$ can suppress the spin fluctuations from the intra-band nesting effect of the FSs. It is worth pointing out that we do not need a large value of $J_2$ compared to the band width (4 eV) to change the character of the spin fluctuations. We find that when $J_2 > 0.04$ eV, the peak at $(\pi, \pi)$ is completely suppressed, which is consistent with the results of INS experiments [26, 27]. Moreover, the $(\pi, 0)$ spin excitations observed in $\text{Rb}_0.82\text{Fe}_1.68\text{Se}_2$ [27] also emerge for $J_2 > 0.04$ eV. In addition, for $0.03$ eV $< J_2 < 0.05$ eV, the peaks around $(\pm \pi, \pm 0.6\pi)$ and $(\pm 0.6\pi, \pm \pi)$ move to about $(\pm \pi, \pm 0.5\pi)$ and $(\pm 0.5\pi, \pm \pi)$, which is also consistent with the INS results. If we further increase $J_2$ to $J_2 > 0.05$ eV, the peaks around $(\pm \pi, \pm 0.5\pi)$ and $(\pm 0.5\pi, \pm \pi)$, which come mainly from the FS nesting, disappear completely, and the four peaks of the spin fluctuations will move towards $(0, \pm \pi)$ and $(\pm \pi, 0)$, and their symmetric points, so that the consistency with INS experiments breaks down. These results imply that the dual effect of both the on-site interactions and the $J_2$ exchange coupling is appropriate and the $J_2$ exchange coupling has significant effects on the physical properties of $\alpha_x\text{Fe}_2\text{Se}_2$. Therefore, we will further study the pairing symmetries mediated by the spin fluctuations with the inclusion of the $J_2$ term.

3.2. Symmetry of the superconducting pairing

We can search for the most favorable pairing symmetry mediated by the spin fluctuations by solving the Eliashberg

![Figure 2. Physical spin susceptibilities for different NNN antiferromagnetic couplings $J_2$. (a) $J_2 = 0$, (b) $J_2 = 0.01$ eV, (c) $J_2 = 0.04$ eV and (d) $J_2 = 0.05$ eV.](image-url)
The s-wave pairing follows a monotonic increase with s-wave, so the d-wave pairing symmetry is dominant over the eigenvalue for the d-wave is larger than that for the s-wave and d-wave solutions of the Eliashberg equation at $U = 1.2$ eV, $V = 0.35$ eV, $J_{II} = 0.2$ eV and $T = 0.008$ eV.

Figure 3. The $J_2$ dependence of the eigenvalues for the extended s-wave and d-wave solutions of the Eliashberg equation.

The gap functions on the FSs for the s-wave and the d-wave symmetry corresponding to two representative values of $J_2 = 0$ and 0.05 eV are shown in figure 4. Here, the magnitudes of the gap have been normalized into the range from $-1$ to $+1$. In the case of $J_2 = 0$, the extended s-wave gap function has nodes (zero points of the gap) on the FS, yet the dominant pairing symmetry here is the d-wave. The gap of the d-wave symmetry for $J_2 = 0$ (figure 4(b)) has no node on the FS; this is consistent with the ARPES results of a nodeless SC gap in the large Fermi pockets around the zone corner [5–7, 18, 19]. However, it has line nodes along the diagonal direction in the Brillouin zone. Recent ARPES experiments further report an isotropic SC gap in the small electron Fermi pocket around the Z point [20, 21], so this rules out the d-wave symmetry. In addition, as argued by Mazin [44], this d-wave gap is rather fragile when the hybridization between the electron pockets in the folded (2-Fe) BZ is included. The only pairing symmetry that is stable and consistent with experiments is the extended s-wave gap for $J_2 \geq 0.05$ (figure 4(c)), which is also the dominant pairing symmetry in this case, as shown in figure 3. We note that the gap magnitude of this s-wave state has in fact a variation along the FS, though it has no node. To compare with the ARPES experiments, in figure 4(e), we present the variation of the gap along the FSs in the folded (2-Fe) BZ. We find that the gap on the inner FS is larger than that on the outer FS and it has a relatively small variation (about 25%) along the FS.

Although the nodeless s-wave gap is consistent with ARPES experiments, it cannot account for the spin resonance observed in neutron scattering [26, 27], because a sign change between the gaps connected by the wavevector at which the resonance occurs is essential to a nonzero spin response [45, 46]. However, we note that the s-wave and d-wave states are nearly degenerate around $J_2 = 0.045$ eV, as seen from figure 3. In this case, an s + id pairing state may be expected, which has been discussed in the early days of Fe pnictides [47]. It has been shown that the mixed state with s + id pairing symmetry does give rise to the spin resonance observed in INS experiments [48]. On the other hand, the gap magnitude for the s + id pairing function is $\Delta(k) = \sqrt{\Delta_s^2(k) + \Delta_d^2(k)}$, where $\Delta_s(k)$ and $\Delta_d(k)$ are the gaps for the s-wave and d-wave states, respectively. It is also nodeless, consistent with ARPES results [5–7, 18, 19]. Therefore, we consider that reasonable values of $J_2$ are about in the range from 0.04 to 0.05 eV in the model used here.

Because the spin fluctuation is dominant over the charge fluctuation, the pairing interaction in the spin-singlet channel is positive (repulsive) (see equation (11)) and has a maximum around the wavevectors $Q$ at which the spin fluctuation has a peak. For a repulsive pairing interaction, the most favorable SC gap must satisfy the condition $\Delta(k)\Delta(k + Q) < 0$ in order to get the largest eigenvalue $\lambda$ of the ‘Eliashberg’ equation, as can be seen from equation (10). In the case of $J_2 = 0$, the pairing interaction comes mainly from the spin fluctuations resulting from the nesting of the FSs. As demonstrated above in figures 2(a), two sets of spin fluctuations have been observed, with peaks around $q = (\pm \pi, \pm 0.6 \pi), (\pm 0.6 \pi, \pm \pi)$ and $(\pm \pi, \pm \pi)$ corresponding to the nesting wavevectors shown in figure 1(b). Therefore, the favorable gaps on FSs connected by these nesting vectors will change signs. This gives rise to two possibilities, namely the extended s-wave and the d-wave symmetry, as shown in figures 4(a) and (b). Energetically, the d-wave state will be more favored here because it opens a full gap around the Fermi pockets (figure 4(b)). Physically, the $J_3$ term will induce electron pairings along the NNN bonds, which gives rise to a pairing function of the form $\cos k_x \cos k_y$, i.e., the so-called $s_\pm$ pairing in the case of both electron and hole pockets. In the absence of hole pockets here, it exhibits itself as the s-wave gap function as shown in figure 4(c).

In order to quantify the detailed gap function and gain more understanding of the physical picture discussed above, we will consider the following Fourier transformation of the equation equation (10). We find that the eigenvalue $\lambda$ has the maximum values in the spin-singlet channel and it is nearly zero in the spin-triplet channel in the range of parameters we have used. The pairing functions corresponding to the two leading eigenvalues in the spin-singlet channel have the d-wave and extended s-wave symmetry. In figure 3, we show these eigenvalues as a function of $J_2$ for 0.3 electron doping corresponding to $K_{0.6}Fe_2Se_2$. For a small $J_2 (<0.045$ eV), the eigenvalue for the d-wave is larger than that for the s-wave, so the d-wave pairing symmetry is dominant over the s-wave pairing symmetry. However, the eigenvalue for the s-wave pairing follows a monotonic increase with $J_2$, and when $J_2 > 0.045$ eV the s-wave pairing surpasses the d-wave pairing and becomes the most favorable pairing state. We note that the threshold value for $J_2$ is much smaller than that needed in the local-antiferromagnetic-exchange model [16] and is comparable to the experimental value from INS measurements [33].

The gap functions on the FSs for the s-wave and the d-wave symmetry corresponding to two representative values of $J_2 = 0$ and 0.05 eV are shown in figure 4. Here, the magnitudes of the gap have been normalized into the range from $-1$ to $+1$. In the case of $J_2 = 0$, the extended s-wave gap function has nodes (zero points of the gap) on the FS, yet the dominant pairing symmetry here is the d-wave. The gap of the d-wave symmetry for $J_2 = 0$ (figure 4(b)) has no node on the FS; this is consistent with the ARPES results of a nodeless SC gap in the large Fermi pockets around the zone corner [5–7, 18, 19]. However, it has line nodes along the diagonal direction in the Brillouin zone. Recent ARPES experiments further report an isotropic SC gap in the small electron Fermi pocket around the Z point [20, 21], so this rules out the d-wave symmetry. In addition, as argued by Mazin [44], this d-wave gap is rather fragile when the hybridization between the electron pockets in the folded (2-Fe) BZ is included. The only pairing symmetry that is stable and consistent with experiments is the extended s-wave gap for $J_2 \geq 0.05$ (figure 4(c)), which is also the dominant pairing symmetry in this case, as shown in figure 3. We note that the gap magnitude of this s-wave state has in fact a variation along the FS, though it has no node. To compare with the ARPES experiments, in figure 4(e), we present the variation of the gap along the FSs in the folded (2-Fe) BZ. We find that the gap on the inner FS is larger than that on the outer FS and it has a relatively small variation (about 25%) along the FS.

Although the nodeless s-wave gap is consistent with ARPES experiments, it cannot account for the spin resonance observed in neutron scattering [26, 27], because a sign change between the gaps connected by the wavevector at which the resonance occurs is essential to a nonzero spin response [45, 46]. However, we note that the s-wave and d-wave states are nearly degenerate around $J_2 = 0.045$ eV, as seen from figure 3. In this case, an s + id pairing state may be expected, which has been discussed in the early days of Fe pnictides [47]. It has been shown that the mixed state with s + id pairing symmetry does give rise to the spin resonance observed in INS experiments [48]. On the other hand, the gap magnitude for the s + id pairing function is $\Delta(k) = \sqrt{\Delta_s^2(k) + \Delta_d^2(k)}$, where $\Delta_s(k)$ and $\Delta_d(k)$ are the gaps for the s-wave and d-wave states, respectively. It is also nodeless, consistent with ARPES results [5–7, 18, 19]. Therefore, we consider that reasonable values of $J_2$ are about in the range from 0.04 to 0.05 eV in the model used here.

Because the spin fluctuation is dominant over the charge fluctuation, the pairing interaction in the spin-singlet channel is positive (repulsive) (see equation (11)) and has a maximum around the wavevectors $Q$ at which the spin fluctuation has a peak. For a repulsive pairing interaction, the most favorable SC gap must satisfy the condition $\Delta(k)\Delta(k + Q) < 0$ in order to get the largest eigenvalue $\lambda$ of the ‘Eliashberg’ equation, as can be seen from equation (10). In the case of $J_2 = 0$, the pairing interaction comes mainly from the spin fluctuations resulting from the nesting of the FSs. As demonstrated above in figures 2(a), two sets of spin fluctuations have been observed, with peaks around $q = (\pm \pi, \pm 0.6 \pi), (\pm 0.6 \pi, \pm \pi)$ and $(\pm \pi, \pm \pi)$ corresponding to the nesting wavevectors shown in figure 1(b). Therefore, the favorable gaps on FSs connected by these nesting vectors will change signs. This gives rise to two possibilities, namely the extended s-wave and the d-wave symmetry, as shown in figures 4(a) and (b). Energetically, the d-wave state will be more favored here because it opens a full gap around the Fermi pockets (figure 4(b)). Physically, the $J_3$ term will induce electron pairings along the NNN bonds, which gives rise to a pairing function of the form $\cos k_x \cos k_y$, i.e., the so-called $s_\pm$ pairing in the case of both electron and hole pockets. In the absence of hole pockets here, it exhibits itself as the s-wave gap function as shown in figure 4(c).

In order to quantify the detailed gap function and gain more understanding of the physical picture discussed above, we will consider the following Fourier transformation of the
Figure 4. Pairing functions on the FSs. Parts (a) and (b) are the s-wave and d-wave solutions for $J_2 = 0$, respectively; parts (c) and (d) are the s-wave and d-wave solutions for $J_2 = 0.05$ eV. Part (e) is the variation of the gap magnitude along the FSs for the s-wave pairing state shown in part (c). The inset in (e) is an illustration of the FSs in the folded BZ (dashed lines). The angle $\theta$ represents the position on the FS with respect to the $\Gamma$–$M$ direction. $\alpha$ labels the inner FS (blue), while $\beta$ labels the outer one (red).

The real-space structures of the gap functions $\Delta_{\alpha\beta}(R)$ in the $t_{2g}$ orbits up to the next nearest neighbors for

\[
\Delta_{\alpha\beta}(R) = \frac{1}{N} \sum_q e^{i q \cdot R} \Delta_{\alpha\beta}(q) \tag{13}
\]

at the lowest Matsubara frequency $\omega_n = \pi T$, where $R$ is the real-space vector and $\alpha$ and $\beta$ are the orbital indices. We find that the inter-orbital ($\alpha \neq \beta$) pairing magnitudes are negligible in comparison with the intra-orbital ($\alpha = \beta$) pairings, suggesting that the superconducting gaps come mainly from the intra-orbital pairings. In addition, the pairing magnitudes in the $e_g$ ($d_{z^2}$ and $d_{x^2-y^2}$) orbits are also much smaller than those in the $t_{2g}$ ($d_{xz}$, $d_{yz}$ and $d_{xy}$) orbits, because the contributions to the FSs are mainly from the $d_{xz}$, $d_{yz}$ and $d_{xy}$ orbits (see figure 1(b)). Thus, we will only consider the gap properties in the $t_{2g}$ orbits in the following.
Figure 5. Real-space structures of the pairing gaps at different orbits for the s-wave and the d-wave symmetry for \( J_2 = 0 \). The values are shown with the relative intensities of the pairing gap. \( d_{xz}, d_{yz} \) and \( d_{xy} \) indicate the \( t_{2g} \) orbits of Fe, which contribute mainly to the electronic states near the Fermi surface.

\( J_2 = 0 \) and \( J_2 = 0.05 \) eV are plotted in figures 5 and 6, respectively. The general features are as follows. (i) The on-site pairing magnitude is always zero, simply because the strong on-site Coulomb repulsion prohibits on-site pairing. (ii) The magnitudes of the \( d_{xz} \) and \( d_{yz} \) orbits have only the \( C_2 \) symmetry due to the orbital character. In the case of \( J_2 = 0 \), the largest magnitude of the gap function appears at the \( d_{xy} \) orbit in the d-wave pairing channel, as can be seen from figure 5. It is nearly twice as much as those at the \( d_{xz} \) and \( d_{yz} \) orbits in the same channel, and is also larger than those in the extended s-wave pairing channel, so this pairing state is dominant. In this case, the pairing at the \( d_{xy} \) orbit occurs only along the NN bonds and the pairing amplitude changes sign between the \( x \) and \( y \) directions, so it gives rise to the usual d-wave gap function of the form \( \cos k_x \cos k_y \). The induction of the pairings along the NNN bonds due to the \( J_2 \) term is obvious from figure 6. However, the important feature is that only at the \( d_{xy} \) orbit in the d-wave channel is the NNN bond pairing not induced at all; all others are induced nearly equally and these pairings are larger than those along the NN bonds. Thus, the extended s-wave pairing is favored in the case of \( J_2 = 0.05 \) eV, which has a gap function of the form \( \cos k_x \cos k_y \) due to the pairings along the NNN bonds.

From the real-space distributions of the pairing bonds, we can obtain the expressions of the pairing gaps at different orbits. For \( J_2 = 0 \), the pairing state has the dominant d-wave symmetry and the pairing functions are

\[
\Delta_{xz}(k) = \Delta_0(0.63 \cos k_x - 1.26 \cos k_y) \\
+ 0.14 \cos k_x \cos k_y),
\]

(14)

\[
\Delta_{yz}(k) = \Delta_0(1.26 \cos k_x - 0.63 \cos k_y) \\
- 0.14 \cos k_x \cos k_y),
\]

(15)

\[
\Delta_{xy}(k) = \Delta_0(2.57 \cos k_x - 2.57 \cos k_y).
\]

(16)

For \( J_2 = 0.05 \) eV, the pairing state has the dominant extended s-wave symmetry and the pairing functions are

\[
\Delta_{xz}(k) = \Delta_0(0.06 \cos k_x + 1.15 \cos k_y) \\
+ 2.84 \cos k_x \cos k_y),
\]

(17)

\[
\Delta_{yz}(k) = \Delta_0(1.15 \cos k_x + 0.06 \cos k_y) \\
+ 2.84 \cos k_x \cos k_y),
\]

(18)

\[
\Delta_{xy}(k) = \Delta_0(-0.02 \cos k_x - 0.02 \cos k_y) \\
+ 2.90 \cos k_x \cos k_y).
\]

(19)

We note that the dominant gap function in this case is of the form \( \cos k_x \cos k_y \), which is the so-called \( s_\pm \) pairing state although there are only electron pockets. This mainly comes from the next-nearest-neighbor pairings induced by the \( J_2 \)
interaction term. These pairing functions may be helpful for the experimental analysis of the pairing states.

We note that the importance of the NNN coupling and a similar gap function have also been proposed recently in FeTeO$_{5.55}$Se$_{0.45}$ based on ARPES results and the strong coupling local pairing picture [49]. In FeTeO$_{5.55}$Se$_{0.45}$, apart from the NNN coupling, the next-next-nearest-neighbor coupling $J_3$ is also important, which leads to considerable next-next-nearest-neighbor pairing.

4. Summary

In summary, we have investigated the spin fluctuations and the pairing symmetry in $A_x$Fe$_2-y$Se$_2$, which has only electron Fermi pockets, by the fluctuation exchange approximation. Besides the on-site interactions, the next-nearest-neighbor antiferromagnetic coupling $J_2$ is also included. We find that both the itinerant and the localized natures of electrons are important to describe recent experimental results on the spin fluctuations and the pairing symmetry in these materials. In particular, a small $J_2$ can change the pairing gap from the d-wave symmetry to the extended s-wave symmetry, and in a certain range the two pairing states are nearly degenerate, so these materials can realize a mixed pairing state of d-wave and s-wave, which is consistent with recent ARPES and neutron scattering experiments. In addition, we have studied the real-space structures of the pairing gap at different orbits, from which we obtain the expressions for the favorable pairing gap functions.

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