Possible pairing symmetry in the FeSe-based superconductors determined by quasiparticle interference

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We study the momentum-integrated quasiparticle interference (QPI) in the FeSe-based superconductors. This method was recently proposed theoretically and has been applied to determine the pairing symmetry in these materials experimentally. Our findings suggest that, if the incipient bands and the superconducting (SC) pairing on them are taken into consideration, then the experimentally measured bound states and momentum-integrated QPI can be well fitted, even if the SC order parameter does not change sign on the Fermi surfaces. Therefore, we offer an alternative explanation to the experimental data, calling for more careful identification of the pairing symmetry that is important for the pairing mechanism.

The superconducting (SC) mechanism and pairing symmetry in the FeSe-based superconductors, e.g. A2Fe2−ySe2 (A=Rb, Cs, K) [11,12], Li1−xFe2+yOHFe1−ySe [13], Li2(NH2)2(NH3)1−yFe2Se2 [14,15], as well as monolayer FeSe grown on SrTiO3 [16], remain hotly debated ever since their discovery. The hole bands sink below the Fermi level and become incipient in these materials while there are only electron-like Fermi surfaces, contrary to the electron- and hole-like ones in the usual iron pnictides [11,12,17,18,20,22]. However the transition temperature in these materials is the highest among all the iron pnictides, the reason of which is still unclear.

To resolve the SC mechanism, various pairing symmetries have been proposed, including the nodeless d-wave [19,20], sign-preserving s-wave [21,22], hidden s±-wave [23,24,25] and in&out s±-wave [26,27]. Among them, the nodeless d- and in&out s±-wave symmetries show apparent sign reversal of the SC order parameter (Δk) on the Fermi surfaces, while the sign-preserving s- and hidden s±-wave symmetries exhibit no such sign reversal. However for the hidden s±-wave symmetry, there is a hidden sign change of Δk between the incipient bands and the electron bands which cross the Fermi level.

Numerous experiments have been performed to distinguish the pairing symmetries. The SC gap magnitude measured by angle-resolved photoemission spectroscopy (ARPES) [11,15,18,20,22], the density of states (DOS) measured by scanning tunneling microscopy (STM) [10,30,40], as well as the temperature dependence of the London penetration depth [11], all suggest a nodeless SC gap, thus the nodeless d-wave symmetry seems to be ruled out since it would be nodal in the realistic Brillouin zone (BZ) where the Fermi surface warps along z [34]. Inelastic neutron scattering (INS) has observed a spin resonance, which is interpreted as a sign-reversing Δk on the Fermi surfaces [22,23]. The in-gap bound states induced by nonmagnetic impurities, which are usually believed to indicate a sign-changing Δk on the Fermi surfaces, have been observed in Ref. [40], but not in Refs. [37] and [39], therefore the former claimed that Δk must change sign on the Fermi surfaces while the latter reached the opposite conclusion.

Furthermore, by measuring the quasiparticle interference (QPI) in the presence of magnetic vortices, Refs. [37] and [39] claimed a sign-preserving s-wave symmetry. However recently, Refs. [50] and [51] pointed out that the above conclusion may be model dependent and unreliable. Instead Hirschfeld, Altenfeld, Eremin, and Mazin proposed a so called HAEM method to process the QPI data and this method has been applied to bulk FeSe [52] and Li1−xFe2+yOHFe1−ySe [40]. Based on this method, Ref. [40] implied a sign-reversing Δk on the Fermi surfaces.

In this work, we show that, when the incipient bands are present, nonmagnetic impurity-induced in-gap bound states can appear even if Δk does not change sign on the Fermi surfaces. In addition, the quantity based on the HAEM method shows similar behavior between the hidden s±- and in&out s±-wave symmetries. Therefore, we offered an alternative explanation to the pairing symmetry drawn from the QPI measurement in Ref. [40].

We adopt a two-dimensional tight-binding model of the iron lattice, where each unit cell accommodates two inequivalent sublattices A and B [see Fig. 1(a)]. The coordinate of the sublattice A in the unit cell (i,j) is ri,j = (i,j) while that for the sublattice B...
is \( \mathbf{r}_i + \mathbf{d} \), with \( \mathbf{d} \) being \((0.5,0.5)\). Here we have taken \( \sqrt{2}a \) as the length unit, where \( a \) is the distance between the nearest-neighbor iron atoms. The Hamiltonian can be written as \( H = \sum_{\mathbf{k}} \psi^\dagger_{\mathbf{k}} A_{\mathbf{k}} \psi_{\mathbf{k}} \), where \( \psi_{\mathbf{k}} = (c^\dagger_{\mathbf{k}A1\uparrow}, c^\dagger_{\mathbf{k}B1\uparrow}, c^\dagger_{\mathbf{k}A2\uparrow}, c^\dagger_{\mathbf{k}B2\uparrow}, c_{-\mathbf{k}A1\downarrow}, c_{-\mathbf{k}B1\downarrow}, c_{-\mathbf{k}A2\downarrow}, c_{-\mathbf{k}B2\downarrow}) \) and

\[
A_{\mathbf{k}} = \begin{pmatrix}
M_{\mathbf{k}} & D_{\mathbf{k}} \\
D^T_{\mathbf{k}} & -M^T_{\mathbf{k}}
\end{pmatrix}, \\
M_{\mathbf{k}} =
\begin{pmatrix}
\epsilon_{A,\mathbf{k}} & \epsilon_{T1,\mathbf{k}} & \epsilon_{xy,\mathbf{k}} & 0 \\
\epsilon_{T1,\mathbf{k}} & \epsilon_{B,\mathbf{k}} & 0 & \epsilon_{xy,\mathbf{k}} \\
\epsilon_{xy,\mathbf{k}} & 0 & \epsilon_{A,\mathbf{k}} & \epsilon_{T2,\mathbf{k}} \\
0 & \epsilon_{xy,\mathbf{k}} & \epsilon_{T2,\mathbf{k}} & \epsilon_{B,\mathbf{k}}
\end{pmatrix}.
\]

Here \( c^\dagger_{\mathbf{k}A1\uparrow}/c^\dagger_{\mathbf{k}A2\uparrow} \) creates a spin up electron with momentum \( \mathbf{k} \) on the \( d_{xz}/d_{yz} \) orbital of the sublattice \( A \). \( \epsilon_{A,\mathbf{k}} = -2(t_x \cos k_x + t_y \cos k_y) - \mu \), \( \epsilon_{B,\mathbf{k}} = -2(t_x \cos k_x + t_y \cos k_y) \), \( \epsilon_{T1,\mathbf{k}} = -t_1[1+e^{-i(k_x+k_y)}]-t_2(e^{-i k_x}+e^{-i k_y}) \) and \( \epsilon_{T2,\mathbf{k}} = -t_2[1+e^{-i k_x}+e^{-i k_y}] \).

Throughout this work, the momentum \( \mathbf{k} \) is defined in the \( 2\text{Fe}/\text{cell} \) BZ and the energies are in units of 0.1 eV. In the following we set \( t_{1-5} = 1.0, 1.4, 0.4, -2, 0.04 \) and \( \mu = -1.8673 \) to fit the band structure measured by ARPES. Under this set of parameters, the average electron number is \( n \approx 2.12 \) (the system is about 12% electron doped). The band structure and Fermi surfaces in the normal state are plotted in Figs. 1(b) and 1(c). The top of the incident bands at \( \Gamma \) and the bottom of the electron bands at \( M \) are both located at about 80 meV below the Fermi level, while the Fermi momentum is \( k_F/\pi \approx 0.25 \), agreeing qualitatively with the ARPES measurements [11,12]. The band structure and the pairing function in the band basis can be obtained through a unitary transformation \( Q_{\mathbf{k}} \) as

\[
Q_{\mathbf{k}}^\dagger M_{\mathbf{k}} Q_{\mathbf{k}} = \begin{pmatrix}
E_{1\mathbf{k}} & 0 & 0 & 0 \\
0 & E_{2\mathbf{k}} & 0 & 0 \\
0 & 0 & E_{3\mathbf{k}} & 0 \\
0 & 0 & 0 & E_{4\mathbf{k}}
\end{pmatrix},
\]

and \( \Delta_{\mathbf{k}} = Q^\dagger_{\mathbf{k}} D_{\mathbf{k}} Q^\dagger_{-\mathbf{k}} = Q^\dagger_{\mathbf{k}} D_{\mathbf{k}} Q_{\mathbf{k}} \). Here \( E_{1\mathbf{k}}, E_{2\mathbf{k}} \) are the energies of the two incident bands and \( E_{3\mathbf{k}}, E_{4\mathbf{k}} \) are those of the two electron bands (\( E_{1\mathbf{k}} \leq E_{2\mathbf{k}} \leq E_{3\mathbf{k}} \leq E_{4\mathbf{k}} \)). The diagonal components in \( \Delta_{\mathbf{k}} \) represent the pairing function on each band while the off-diagonal ones signify the inter-band pairing, which we ignore for simplicity.

For the SC pairing, we consider two cases. The first one is the in\&out \( s_{\pm} \) pairing, where we set

\[
\Delta_{\mathbf{k}} = \begin{pmatrix}
\Delta_2 & 0 & 0 & 0 \\
0 & \Delta_2 & 0 & 0 \\
0 & 0 & \Delta_2 & 0 \\
0 & 0 & 0 & -\Delta_1
\end{pmatrix},
\]

with \( \Delta_1 = 0.14 \) and \( \Delta_2 = 0.08 \). It will lead to a sign-changing gap between the inner and outer electron pockets, as shown in Fig. 1(c). This pairing symmetry was suggested when the hybridization between the electron bands is strong enough [31,32]. Another one is the hidden \( s_{\pm} \) pairing, where we set

\[
\Delta_{\mathbf{k}} = \begin{pmatrix}
-\Delta_1 & 0 & 0 & 0 \\
0 & -\Delta_1 & 0 & 0 \\
0 & 0 & \Delta_2 & 0 \\
0 & 0 & 0 & \Delta_1
\end{pmatrix}.
\]

Contrary to the in\&out \( s_{\pm} \) pairing, the hidden \( s_{\pm} \) pairing will not lead to any sign change of the gap along the Fermi surfaces, as shown in Fig. 1(d). However, the sign of the order parameter on the incident bands is opposite to that on the electron bands. This pairing symmetry is predicted by the spin-fluctuation theory in the strong coupling limit [32,33]. In both cases, we have neglected the orbital selective renormalization effects [32,33] by assuming a k-independent \( \Delta_k \). A k-dependent \( \Delta_k \) may affect the momentum dependence of the QPI signal, but since we are focusing on the momentum-integrated QPI signal in the following, we believe this assumption is rea-
δρω(ω) − 8q/πρ(ω)

Fig. 2: (color online) (a) The LDOS at the impurity site in the SC state. The red dotted curve is for the in&out s± pairing (V1 = 6, V2 = −1), while the black solid one is for the hidden s± pairing (V1 = 10, V2 = 5). The inset shows the DOS in the clean system. (b) The difference of the FT-QPI δρω−(ω) for the in&out s± pairings, (c) is similar to (b), but is for the hidden s± pairing. (d) δρω−(ω) extracted from Figs. 3(b) and 3(c) of Ref. [40]. The black solid and red dotted curves are the original and filtered ones are both similar to (d), but are our calculated results for the δρω−(ω) by ignoring the incipient bands. The black and red curves are for the in&out s± and hidden s± pairings, respectively.

Fig. 3: (color online) (a) The filtered δρ−(ω). The black and red curves are our theoretical results for the in&out s± and hidden s± pairings, respectively, while the green and blue ones are the experimental data extracted from Figs. 3(c) and S7(d) of Ref. [40]. (b) Theoretically calculated δρ−(ω) for the two pairing symmetries exhibit identical DOS close to the Fermi level, with two pairs of SC coherence peaks located at ±Δ1 and Δ2.

For a single nonmagnetic impurity located at the A sublattice of the unit cell R = (0, 0), the impurity Hamiltonian can be expressed as $H_{\text{imp}} = \sum_{\alpha,\beta=1}^2 \sum_{r,s=\pm}^4 V_{\alpha\beta}^{rs} c_{rA\alpha}^\dagger c_{sA\beta}$. Since it is a multiorbital system, the scattering may consist of both the intraribital ($V_{\alpha=\beta} = V_1$) and interorbital ($V_{\alpha\neq\beta} = V_2$) components. Following the standard T matrix procedure, we can obtain $\rho_{ij}(r_{ij}, \omega)$, which is the local density of states (LDOS) on the sublattice A/B of the unit cell $(i, j)$. After that, we follow the same procedure in Ref. [40] and select an area enclosed by the dashed square in Fig. 1(a). The location of the impurity is at the center of the square and is set to be the origin. Using this area (contains 257² atoms in our calculation), we perform the Fourier transformation to get the FT-QPI as

$$\rho(\mathbf{q}, \omega) = \sum_{\mathbf{r}} \rho(r, \omega) e^{i\mathbf{q}\cdot\mathbf{r}}.$$ 

The anti-symmetrized FT-QPI is calculated as

$$\delta\rho^- (\mathbf{q}, \omega) = \sum_{\mathbf{q} \in A} \text{Re} \{ \rho(\mathbf{q}, \omega) - \rho(-\mathbf{q}, -\omega) \},$$

where the area $A$ is defined as $0.13\pi/a \leq |\mathbf{q}| \leq 0.55\pi/a$, which is exactly the same area used in the experiment [40]. According to the HAEM theory [40, 50, 51], $\delta\rho^- (\omega)$ should change sign when $\Delta_2 \leq \omega \leq \Delta_1$ if the SC order parameter does not exhibit any sign reversal on the Fermi surfaces. Otherwise it will maintain the same sign when $\Delta_2 \leq \omega \leq \Delta_1$ if the SC order parameter changes sign on the Fermi surfaces.

In the following, we show our calculated results and compare them with the experiment. In Fig. 2(a), we plot the LDOS at the impurity site for the two pairing symmetries. As can be seen from the inset, in the clean system, the two pairing symmetries exhibit identical DOS close to the Fermi level, with two pairs of SC coherence peaks located at ±Δ1 and ±Δ2. At the impurity site, with appropriate scattering potential [(V1, V2) = (6, −1)/(10, 5) for the in&out/hidden s± pairing], clear in-gap bound states show up, which are located at $\omega = ±0.04$ and ±0.055 for the in&out s± and hidden s± pairings, respectively. Furthermore, the intensity of the bound states at positive ω is much larger than that at negative ω. The two-gap DOS in the clean system, as well as the location and the asymmetrical height of the impurity bound states, are all qualitatively consistent between our theoretical results and the experimental measurements (see Fig. 1 in Ref. [40]). In Figs. 2(b) and 2(c), we plot the difference of the FT-QPI $\delta\rho^- (\mathbf{q}, \omega) = \text{Re} \{ \rho(\mathbf{q}, \omega = 0.085) - \rho(\mathbf{q}, \omega = 0.085) \}$ for the in&out s± and hidden s± pairings, respectively. The two pairing symmetries show no qualitative difference and both agree with the experiment (see Fig. 3(a) in Ref. [40].
We have verified that the DOS in the clean system calculation from the incipient bands can be completely removed. The order parameter, contributes significantly to $\delta \rho$ for the two pairings are both qualitatively consistent with experimental data in Fig. 3(a). Our theoretical results $\delta \rho$ the results are shown in Fig. 2(f). We then rescale the $\omega$ the same filtering scheme from $|\omega|$ 3.55 meV and the filtered $\delta \rho^{-} (\omega)$ is shown as the red curve. In Ref. [40], they considered only the two electron bands and neglected the incipient bands. They claimed that, if the SC order parameter changes sign between the electron pockets (i.e., the $s^{\pm}$ pairing state defined in their paper), then $\delta \rho^{-} (\omega)$ will not change sign between $\Delta_1$ and $\Delta_2$, while it will change sign if the pairing state is $s^{\mp}$ (the SC order parameter does not change sign between the electron pockets). Since the experimental data show no sign change of $\delta \rho^{-} (\omega)$ between $\Delta_1$ and $\Delta_2$, therefore they concluded that there should exist a sign reversal of the SC order parameter on the electron pockets. The results of $\delta \rho^{-} (\omega)$ from our calculation are plotted in Figs. 2(e) and 2(f). The black solid curve in Fig. 2(e) shows $\delta \rho^{-} (\omega)$ for the in&out $s^{\pm}$ pairing and we can see that there is a sharp peak at $\omega = 0.04$, which is due to the impurity bound state. To eliminate the effect of the bound state, we use a parabolic function $\delta \rho^{-} (\omega) = A\omega^2 + B\omega$ to substitute the original one from $\omega = 0.03$ to 0.06, as has been done in the experiment, and show the filtered $\delta \rho^{-} (\omega)$ as the red dotted curve. Similarly, for the hidden $s^{\pm}$ pairing, since the impurity bound state is located at $|\omega| = 0.055$, therefore we employ the same filtering scheme from $\omega = 0.045$ to 0.075, and the results are shown in Fig. 2(f). We then rescale the filtered $\delta \rho^{-} (\omega)$ from our calculation and plot it with the experimental data in Fig. 2(a). Our theoretical results for the two pairings are both qualitatively consistent with the experimental data, that is, $\delta \rho^{-} (\omega)$ exhibits no sign change between $\Delta_1$ and $\Delta_2$. In the hidden $s^{\pm}$ pairing, the off-shell scattering process denoted by the green arrow in Fig. 1(b), which connects states with sign-reversed order parameter, contributes significantly to $\delta \rho^{-} (\omega)$ and makes $\delta \rho^{-} (\omega)$ in this case similar to that in the in&out $s^{\pm}$ pairing case. Therefore, the experimental data does not exclusively imply a sign-changing order parameter on the electron Fermi surfaces. A detailed derivation of the HAEM theory in the presence of incipient bands can be found in Ref. [55].

In summary, we have investigated the momentum-integrated QPI in the FeSe-based superconductors, by taking the incipient bands into consideration. We found that, if there is SC pairing on the incipient bands, then special caution has to be taken when interpreting the pairing symmetry from the experimental data. For example, naively people may expect that the in-gap bound states induced by nonmagnetic impurities should suggest a sign-reversing order parameter on the Fermi surfaces, while our theoretical calculation indicates that this is not the case. In addition, the HAEM theory proposed in Refs. 50 and 51, which has been used to process the experimental data in Ref. [40], may be unable to determine the pairing symmetry in these materials. In this work, the strength of the scattering potential ($V_1, V_2$) is chosen so as to fit the location and asymmetrical height of the bound states observed in experiment. We have also verified that, for other scattering potentials, for example, if we set the interorbital scattering potential $V_2$ to be zero, the main conclusions still hold. The comparison between our results and the experimental data implies that, the QPI measurement cannot distinguish the hybridization-induced in&out $s^{\pm}$ pairing from the strong-coupling-spin-fluctuation-induced hidden $s^{\pm}$ pairing. Finally we would like to comment on the spin resonance observed in INS. For example, in Refs. [42] and [43], the energy of the spin resonance is at 21 meV. However in Li$_{1-x}$Fe$_x$OFe$_{1-x}$Se, $2\Delta_2 \approx 17$ meV, as determined by the STM data in Ref. [40]. Therefore the spin resonance energy is actually above $2\Delta_2$ and this can happen even if the SC order parameter preserves its sign on the Fermi surfaces. Therefore, the sign of the SC order parameter in the FeSe-based superconductors is far from settled.

This work is supported by the Natural Science Foundation from Jiangsu Province of China (Grant No. BK20160094, Y.G.), the Start-up Foundation from South China Normal University (T.Z.) and NSFC (Grant No. 11574134, Q.H.W.).
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