Pairing symmetry and properties of iron-based high-temperature superconductors

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Abstract – Pairing symmetry is important to indentify the pairing mechanism. The analysis becomes particularly timely and important for the newly discovered iron-based multi-orbital superconductors. From the group theory point of view we classified all pairing matrices (in the orbital space) that carry irreducible representations of the system. The quasiparticle gap falls into three categories: full, nodal and gapless. The nodal-gap states show conventional Volovik effect even for on-site pairing. The gapless states are odd in the orbital space, have a negative superfluid density and are therefore unstable. In connection to experiments we proposed possible pairing states and implications for the pairing mechanism.

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Introduction. – The newly discovered family of iron-based FeAs high-temperature superconductors are raising great interests in the community [1]. The superconductor consists of layers of FeAs which are believed to be the conducting planes. Substitution of O by F, Fe by Co [2] or introducing O vacancies [3] dope electrons into the system, and contrarily substitution of rare-earth elements by alkaline-earth elements or alkaline-earth elements by alkaline elements can realize hole doping [4].

Experiments concerning superconductivity have been done, but the gap structure is still under debate. For single-layer FeAs compounds, the specific capacity measurement [5], penetration depth measurement [6] and nuclear magnetic resonance (NMR) [7] are consistent with nodal gap, while angle-resolved photoemission spectroscopy (ARPES) indicates nodeless gap [8]. Both nodal and nodeless pairing are suggested by different groups with tunnelling experiments [9]. For double-layer FeAs compounds, ARPES [10], specific heat [11] and tunnelling experiments [12] indicate nodeless pairing. On the theoretical side, local-density-approximation (LDA) calculations show that the states near the Fermi level are largely contributed by the five d-orbitals [13]. Moreover, some authors demonstrated that it is sufficient to keep only a few of them, for example the $d_{xz}$ and $d_{yz}$ orbitals, to reproduce qualitatively the LDA Fermi-surface topology [14]. As for the driving force for superconducting pairing, both electronic [15] and phonon-mediated mechanisms [16] are proposed, and various pairing symmetries are predicted.

Since the pairing symmetry is related to the pairing mechanism, a classification of all possible pairing symmetries [17] is important. This is more so given the fact that the pairing function becomes an orbitalwise matrix function in the multiorbital case, which we elaborate in this paper. The main results are as follows. 1) From a two-orbital ($d_{xz} + d_{yz}$) model we classify all possible on-site and bondwise pairing basis matrices. In addition to the momentum dependence, the matrices themselves carry nontrivial symmetries, so that even on-site pairing may lead to a nodal or gapless pairing. 2) In the gapless case the density of states (DOS) at the Fermi level is enhanced by the SC order. 3) Only the nodal-gap cases show the Volovik effect in an applied magnetic field. 4) Most surprisingly, the gapless SC state may have a negative superfluid density, and is therefore unstable against phase twisting. 5) In connection to available experiments we propose possible pairing bases and further experiments to reduce the candidate list.

Symmetry analysis. – In the FeAs layer there are two Fe ions per unit cell because of the As ions. In order to
simplify the analysis, we adopt a two-band model, i.e. we keep the two atomically degenerate \( d_{xz} \) and \( d_{yz} \) orbitals of the Fe ion, which are important for superconductivity, and neglect \( d_{xy} \) and \( d_{x^2-y^2} \) orbitals of the Fe ions and all orbitals of the As ions for a moment. Thus we will focus on the Fe lattice, for which there is only one Fe ion in each unit cell. The effect of As ions can be partially included in the effective hopping integrals for the \( d \)-orbitals. We define the \( x \)- and \( y \)-direction unitary vectors as connecting the nearest-neighbor Fe-atoms. The space group of our model is \( P4/mnm \), which has higher symmetry than the space group of the whole system \( P4/nmm \). The normal state of the model can be described, in the momentum space, by \( H_0 = \sum_{k, \sigma} \xi_k \phi_k \phi_{k \sigma} \), where \( \phi_k = (d_{xz}, d_{yz}, k_x, k_y) \), and \( \xi_k = \epsilon_{k 70} + \delta_{k73} + \gamma_71 \) in which \( \tau_{0,1,3} \) are unit and Pauli matrices defined in the orbital space. For point group operations under concern, the \( d_{xz} \) and \( d_{yz} \) orbital wave functions transform as \( x \) and \( y \), respectively. This dictates that \( \phi_k \tau_0 \phi_k \) transforms as \( A_{1g} \), \( \phi_k \tau_3 \phi_k \) as \( B_{2g} \), \( \phi_k \tau_2 \phi_k \) (which is actually absent in \( H_0 \) but is included here for later use) as \( A_{2g} \), and \( \phi_k \tau_1 \phi_k \) as \( B_{1g} \). Equivalently we claim that the \( \tau \)-matrices carry the above-mentioned irreducible representations, without further referring to the orbital wave functions. Finally to leave \( H_0 \) invariant, \( \epsilon_{k}, \delta_{k} \) and \( \gamma_{k} \) must transform as \( A_{1g}, B_{1g} \) and \( B_{2g} \), respectively. The concrete form of the dispersions in \( H_0 \) (see below) is irrelevant at this stage.

We now discuss the pairing symmetry. The system is invariant under spin-\( SU(2) \) transformation and one can classify the pairing states into spin-singlet and spin-triplet cases, which we assign in the last step according to the local antisymmetry of the pairing function with respect to the combined exchange of spin, orbital and spatial position. We therefore concentrate first on the symmetry of pairing as a function of momentum and a matrix in the orbital space. Since the pairing operator \( \phi_\alpha(k) \phi_\beta(-k) \phi_\gamma \phi_\delta \) (where \( \phi_{\alpha, \beta} = d_{xz, yz} \) and \( i = 0, 1, 2, 3 \) transforms identically as \( \phi_\alpha(k) \tau_3 \phi_\beta \phi_\gamma \phi_\delta(k) \), we immediately see that the \( \tau \)-matrices in the pairing matrix transform exactly as they do in \( H_0 \). For on-site pairing, \( \tau_{0,1,2,3} \) carries the irreducible representations. Since \( \tau_{0,1,3} \) are even in orbital space and transform as \( A_{1g}, B_{2g} \) and \( B_{1g} \) under the point group, the spin channel must be a singlet. On the other hand, \( \tau_2 \) is odd in orbital space and transform as \( A_{2g} \), then the spin channel must be a triplet.

The extension to pairing on bonds is almost straightforward. We may multiply the above-mentioned \( \tau \)-matrices by trigonometric basis functions to form the pairing matrices corresponding to pairing on bonds in the real space. For the system under concern, the basis functions for nearest-neighbor bonds are \( \cos k_x + \cos k_y \) \( (A_{1g}) \), \( \cos k_x - \cos k_y \) \( (B_{1g}) \), \( \sin k_x, \sin k_y \) \( (B_{2g}) \) (carrying the two-dimensional \( E_g \) representation). The symmetry of the product is easily seen from that fact that the \( \tau \)-matrices only carry one-dimensional representations. For example, \( B_{1g} \cdot \tau_1 \sim B_{1g} \cdot B_{2g} \sim A_{2g} \). Since the orbital parity is even, the spin channel must be a singlet. In Table 1 we list all possible pairing basis matrices for on-site pairing and nearest-neighbor-bond pairing (extension to longer-range pairing is trivial), together with the irreducible representations, they carry the spin symmetry, orbital parity, and the behavior of the corresponding quasiparticle excitation gap in the momentum space (which will be discussed later). We notice that even on-site pairing can carry a non-trivial representation, indicating the unique role of the two (atomically) degenerate \( d \)-wave orbitals.

| No. | IR | Basis | Spin | Orbital parity | Gap |
|-----|----|-------|------|----------------|-----|
| 1   | \( A_{1g} \) | \( \tau_0 \) | S     | E              | Full |
| 2   | \( A_{1g} \) | \( (\cos k_x + \cos k_y)\tau_0 \) | S     | E              | Nodal |
| 3   | \( A_{1g} \) | \( (\cos k_x - \cos k_y)\tau_3 \) | S     | E              | Nodal |
| 4   | \( A_{2g} \) | \( (\cos k_x - \cos k_y)\tau_1 \) | S     | E              | Nodal |
| 5   | \( B_{1g} \) | \( \tau_3 \) | S     | E              | Nodal |
| 6   | \( B_{1g} \) | \( (\cos k_x - \cos k_y)\tau_0 \) | S     | E              | Nodal |
| 7   | \( B_{1g} \) | \( (\cos k_x + \cos k_y)\tau_3 \) | S     | E              | Nodal |
| 8   | \( B_{2g} \) | \( \tau_1 \) | S     | E              | Nodal |
| 9   | \( B_{2g} \) | \( (\cos k_x + \cos k_y)\tau_1 \) | S     | E              | Nodal |
| 10  | \( E_g \) | \[ \sin k_x i\tau_2 \] | S     | O              | Gapless |
| 10’ | \( E'_g \) | \[ (\sin k_x + i \sin k_y) i\tau_2 \] | S     | O              | Gapless |
| 11  | \( A_{2g} \) | \( i\tau_2 \) | T     | O              | Gapless |
| 12  | \( A_{2g} \) | \( (\cos k_x + \cos k_y)\tau_2 \) | T     | O              | Gapless |
| 13  | \( B_{2g} \) | \( (\cos k_x - \cos k_y)\tau_2 \) | T     | O              | Gapless |
| 14  | \( E_g \) | \[ \sin k_x \tau_0 \] | T     | E              | Nodal |
| 15  | \( E_g \) | \[ \sin k_x i\tau_0 \] | T     | E              | Nodal |
| 16  | \( E_g \) | \[ \sin k_x \tau_3 \] | T     | E              | Nodal |
| 16’ | \( E'_g \) | \[ (\sin k_x + i \sin k_y) \tau_1 \] | T     | E              | Nodal |
Quasiparticle excitation gap. – To illustrate the concrete gap structure in the various SC states, we need the band structure of iron-based superconductors. Here we adopt the tight-binding “minimal two-orbital model” introduced by ref. [18]. We point out that the Fermi surface topology given by this two-orbital model is inconsistent with LDA calculations, since it misplaces a hole Fermi pocket in the “large” Brillouin Zone (BZ) corresponding to one Fe per unit cell. However, this inconsistency disappears in the “small” BZ corresponding to a real unit cell. Since the purpose of this work is to simply discuss the pairing symmetry and the low-energy physical properties of various SC states in FeAs superconductors, this minimal two-orbital model exhibiting similar Fermi surfaces is acceptable.

The BdG Hamiltonian is given by $H = \sum_k \psi_k^\dagger H_k \psi_k$, where we recall that the momentum is defined in the “large” BZ, $\psi_k = (d_{xz,k}, d_{yz,k}, q_{xz,k}^d, q_{yz,k}^d)^T$ is the four-component spinor in the orbital and Nambu space, and $H_k$ is, in the form of a block-matrix, 

$$H_k = \begin{pmatrix} \xi_k & V \Delta_k^\dagger \\ V \Delta_k & -\xi_k \end{pmatrix}.$$ 

Here $\xi_k = \epsilon_k \tau_0 + \delta_k \tau_3 + \gamma_k \tau_1$ is a tight-binding dispersion defined in $H_0$, with $\epsilon_k = -(t_1 + t_2)(\cos k_x + \cos k_y) - 4t_3 \cos k_x \cos k_y - \mu$, $\delta_k = -(t_1 - t_2)(\cos k_x - \cos k_y)$, and $\gamma_k = -4t_4 \sin k_x \sin k_y$. Here $t_1 = -1$, $t_2 = 1.3$, $t_3 = t_4 = -0.85$, and $\mu = 1.45$, in unit of $|t_1|$. The gap amplitude $V = 0.4$ is chosen for illustration, and $\Delta_k$ is selected from the basis matrix functions listed in table 1. The Hamiltonian can be exactly diagonalized to obtain the quasiparticle excitations and other SC properties. Zero energy excitations are determined by $\det(H_k) = 0$. In the case of SC state No. 1 in table 1, $\det(H_k) = (\epsilon_k^2 - \delta_k^2 - \gamma_k^2 - V^2)^2 + (2V \epsilon_k)^2 = 0$ has no solution. This corresponds to the full-gapped case. For SC state No. 5, $\det(H_k) = (\epsilon_k^2 - \delta_k^2 - \gamma_k^2 - V^2)^2 + (2V \delta_k)^2 = 0$ is satisfied at four sets of nodal points in the BZ. Another interesting example is SC state No. 11, for which $\det(H_k) = (\epsilon_k^2 - \delta_k^2 - \gamma_k^2 + V^2)^2 - (2V \delta_k)^2 = 0$ holds along lines in the BZ. Such zero-lines form the Fermi surface (FS) (slightly different from the normal state FS) for the BdG quasiparticles. This is the gapless case.

Due to the two-orbital character of the SC state, the nodal points or the BdG FS may be located away from the normal state FS. We therefore need the quasiparticle excitation energy in the whole BZ to characterize the gap structure. Several typical results are shown in fig. 1 where the minimal quasiparticle excitation energy as a function of momentum in (a) the normal state, (b) the full-gap SC state No. 1, (c) the nodal-gap SC state No. 5 and (d) the gapless SC state No. 11 are presented. Here thick white lines (symbols) highlight the zero-excitation-energy contour (nodal points). By checking the excitation spectra of all possible cases in table 1, we summarize that: i) all $\tau_1, \tau_3$-bases are nodal, consistent with the fact that these $\tau$ matrices carry $B_{2y,1y}$ representations; ii) all $\tau_2$-bases (which carries odd orbital parity) are gapless (unless the pairing energy scale is of the order of the band width); iii) the $\tau_0$-bases leads to full or nodal gaps, depending on the momentum basis function.

Volovik effect and superfluid density. – In this section, we discuss the physical properties of various SC states, especially the DOS, Volovik effect and superfluid density. We did the calculations for all SC states listed in table 1. Since we found that the qualitative behavior is the same for SC states belonging to the same category.
(full-gap, nodal-gap or gapless), we only present the results for representative states, namely, the full-gap SC state No. 1, the nodal-gap SC state No. 5 and gapless SC state No. 11. Figure 2(a) shows the DOS. The low energy U-shaped (V-shaped) DOS in the full-gap (nodal-gap) SC states are conventional. The gapless case is rather exotic. The low-energy DOS is not gapped but slightly piled up by SC order, due to the presence of BdG FS. Figure 2(b) shows the effect of a magnetic field on the DOS, which is calculated by a semi-classical method as follows [19]. The effect of vortices is simulated by averaging over the direction of the superflow momentum $q$ with $q \propto \sqrt{H}$. The change in the zero-energy DOS $\Delta N(0, q) = (N(0, q)) - N(0, 0)$ can be probed by the magnetic-field–dependent specific heat [5]. It is seen in fig. 2(b) that the Volovik effect is absent in the full-gap SC state, but is manifest as a linear rise of $\Delta N$ with $q$ in the nodal-gap SC state. Moreover, for the gapless state, the presence of magnetic field actually reduces the DOS (as seen for moderate $q$). Figure 2(c) shows the temperature dependence of superfluid density. The full-gap and nodal-gap SC states exhibit activated and linear drop, respectively. Contrarily, the superfluid density is negative at all temperatures in the gapless case. This arises from the fact that piling-up of low-energy DOS leads to an overwhelming paramagnetic contribution against the diamagnetic part. It indicates that the gapless state is unstable upon phase twisting, implying a tendency toward a possible Fulde-Ferrell-Larkin-Ovchinnikov state or magnetic ordering.

**Discussion.** – Basing upon our analysis presented above and recent experiments, we could discuss the possible pairing symmetry of iron-based superconductors. A sharp conclusion from our analysis is that a nodeless gap must be associated with the intra-orbital pairing in the $\tau_0$ channel (which maps to intra-band pairing in the band language), since nodes are unavoidable in the other channels. As stated in previous sections, there are still several inconsistencies among experimental results up to now. For double-layer FeAs compounds, the nodeless pairing is confirmed by ARPES, penetration length and specific-heat measurements, and further the so-called $s \pm$ pairing symmetry is suggested theoretically [20]. Consequently, the most favorable candidate is described by the pairing matrix $\Delta_k = \cos(k_x)\cos(k_y)\tau_0$ with $A_{1g}$ symmetry. This SC state is not listed in table 1, because next-nearest-neighbor pairing is involved. However, it can be included with trivial extension in table 1 as we have pointed out previously. For single-layer FeAs compounds, the situation is less clear, and both nodal and nodeless pairings are suggested by different groups. If there is no qualitative difference in electronic structure between the single-layer and double-layer compounds, the single-layer FeAs compound is expected to share the same pairing symmetry, namely $s \pm$ pairing. However, if the situation in single-layer compounds is different, further analysis is needed. On-site singlet pairing is unfavorable to Hund’s rule coupling, while bondwise triplet pairing is inconsistent with the antiferromagnetic exchange [21]. Given nodal pairing, we are left with the bondwise singlet pairing cases No. 3, 4, 6, 7, and 9 in table 1. Out of these cases we observe that only the $A_{2g}$ case No. 4 and the $B_{2g}$ case No. 9 have nodal points in the $x$- and $y$-directions, which are more relevant if the electron pockets are important. We propose that Raman scattering and phase-sensitive probes could further reduce the redundancy.

Some remarks are in order before closing. First, it should be emphasized that the symmetry classification is robust, while the assignment of a particular pairing symmetry has to be sharpened or even altered according to further systematic and intrinsic experimental results. Second, a general pairing matrix can be decomposed into a linear combination of the bases. In principle, bases belonging to different irreducible representations do not mix, but mixing in other cases cannot be ruled out on symmetry grounds alone. In particular, the two-dimensional representations, like the case No. 10 may be mixed into one of the chiral-symmetry–breaking states shown in No. 10'. Third, even if the effect of As ions neglected so far is reconsidered, the point group is still $D_{4h}$, so that the above symmetry classification still holds. Finally, several authors propose pairing including $d_{xy}$ or even all five $d$-orbitals [22]. These may be included in our analysis and result in more pairing bases. For example $xy$ can pair with $yz$, which transform as $x$, forming one component of the $E$ representations.

We became aware of a related work [23] after we finished the present paper.

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