TAO pairing: a fully gapped pairing scenario for the Iron-Based Superconductors

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Motivated by the fully gapped superconductivity in iron-based superconductors with uncompensated electron pockets, we propose a spin singlet, but orbital triplet analogue of the superfluid phase of $^3$He-B. We show that orbital triplets with a nominal d-wave symmetry at the iron sites can transform as s-wave pairs under rotations about the selenium sites. Linear combinations of such $d_{xy}$ and $d_{x^2-y^2}$ triplets form a fully gapped, topological superconductor. Raman-active excitations are predicted to descend below the superconducting transition temperature.

The discovery of superconductivity with $T_c = 26$K in LaFeAsO by Hosono et al. [1] has opened a new field of iron (Fe)-based multi-band high temperature superconductors (SCs). STM, ARPES and bulk experiments show a majority of these systems are fully gapped [2, 14, 15], while optical measurements also show a significant Coulomb interaction [15]. Most electronically mediated superconductors, avoid the on-site Coulomb interactions by building nodes into the pair wave, so that the on-site pairing $\langle \psi_{\uparrow \uparrow} \psi_{\uparrow \uparrow} \rangle_{Fe} = \sum_{\nu} \frac{\Delta_{\nu}}{2\nu} = 0$. In the organic, heavy fermion, cuprate and ruthenate superconductors, this “Coulomb orthogonalization” is guaranteed by a d- or p-wave symmetry of the order parameter [14, 18].

Superconductivity in the Fe-based systems is widely believed to derive from a gap function with symmetry $\pm s$ [2, 10], taking the form $\Delta(0) = \Delta_0 + 2\Delta_1 \cos k_x \cos k_y$, containing a nodal between the electron and hole pockets. This is supported by c-axis Josephson tunneling experiments [20], QPI measurements on Fe(Se,Te) [1], and the absence of the Wohlleben effect [7]. In this scenario, a phase cancellation between the electron and hole pockets eliminates on-site pairing, minimizing the Coulomb interaction. However, when the hole pockets are absent, such as in $A_2Fe_2Se_2$ [21] and single layer FeSe [22], the node in the $s^\pm$ order parameter is expected to intersect the the electron pockets to achieve Coulomb orthogonalization. These nodes have not been observed. One possibility is that the symmetry of the order parameter changes in the electron pocket materials, for example by the development of d-wave pairing, with nodes between the pockets [24]; yet this scenario appears to be inconsistent with the observation of fully gapped electron pockets around the Z-point in $Tl_{0.65}K_{0.35}Fe_{4.78}Se_2$ [12].

These observations motivate the search for an alternative description of the pairing symmetry to account for the Coulomb orthogonalization in single electron-pocket materials without a change in condensate symmetry. Here, we are inspired by superfluid $^3$He-B, where the Fermi surface is fully gapped, yet the pair wavefunction contains hidden nodes. In two-dimensional $^3$He-B, the gap function takes the form $\langle c_{k\sigma} c_{-k\sigma'} \rangle = (k_x \sigma_x + k_y \sigma_y) \sigma_2$; although the odd-parity $k_x$ and $k_y$ triplet components contain nodes, their anti-commuting spin structure causes them to add in quadrature to create a fully gapped condensate where $\Delta(0) \propto (k_x^2 + k_y^2)^{1/2}$. While spin-triplets are ruled out in iron-based superconductors by Knight-shift measurements [4, 5], here we show that the multi-orbital nature of the iron-based superconductors opens up an analogous class of non-trivial orbital triplet states.

The staggered tetrahedral structure of the iron based superconductors results in an enlarged unit cell containing two rotated Fe tetrahedra. Electrons on iron sites in “right” or “left” pointing tetrahedra (Fig. 1b) are labelled by a “tetrahedral” band index $\tau = \pm 1$ and the Pauli matrices linking these states will be denoted by $\tau = (\tau_1, \tau_2, \tau_3)$. Likewise, we associate an orbital index $\alpha = \pm 1$ with the degenerate $zx$ ($\alpha = +1$) and $zy$ ($\alpha = -1$) iron d-orbitals, denoting the Pauli matrices that link these states by $\tilde{\alpha} = (\alpha_1, \alpha_2, \alpha_3)$ (Fig. 1).

FIG. 1. (a) Tetrahedral index $\tau = \pm 1$ denotes Fe in right or left pointing tetrahedra, (b) orbital index $\alpha = \pm 1$ denotes zx and zy orbitals. The electron operators now carry three discrete quantum numbers, so we replace the conventional electron operator $c_{k\sigma} \rightarrow c_{k\nu}$, where $\nu \equiv (\tau, \alpha, \sigma)$ denotes the triplet of site, orbital and spin indices. Since we are treating orbital triplets, we adopt a generalized Balian-Werthamer spinor to describe the electron fields

$$\Psi_k \equiv \begin{pmatrix} \psi_{k\nu} \\ -i(\lambda_2)_{\nu \nu'} \psi^\dagger_{-k\nu'} \end{pmatrix}$$

where $\lambda_2 = \tau_2 \alpha_2 \sigma_2$ flips the tetrahedral, orbital and spin quantum numbers so that so that the electron and
hole fields transform the same way under isospin rotations. In this notation an extended \( s^\pm \) pair, written \( \Delta^{s\pm}(k) = \cos(k_x)\cos(k_y)\tau_0\alpha_2 \mathbf{1} + \mathbf{i}\lambda_2 \), is an orbital and site triplet, the orbital analog of equal spin triplet pairing, for once multiplied by \( \lambda_2 \), it is diagonal in site and orbital indices. We now consider a more general class of orbital triplet pairing.

The 2D Fe As(Se) layer exhibits \( \text{C}_{4v} \) symmetry about the As(Se) atoms, and \( \text{D}_{2d} \) symmetry around the Fe atoms, with an inversion center on the Fe-Fe bond. We choose to use the \( \text{C}_{4v} \) symmetry as it naturally accounts for the two Fe atoms per unit cell. There is no 90° rotation symmetry about an iron site - instead, the unit cell is invariant under a combined 90° rotation and a translation between the two iron sites. This means that the site and orbital indices transform non-trivially under symmetry operations of the \( \text{C}_{4v} \) group. For example, under a 90° rotation, \( |zx\rangle \rightarrow |zy\rangle, |zy\rangle \rightarrow |−zx\rangle \), so that the orbital matrix \( \alpha_1 \equiv |zx|\langle zy| + |zy|\langle zx| \) changes sign, \( \alpha_1 \rightarrow −\alpha_1 \). This means that the global symmetry of the superconducting (SC) state need not correspond to the momentum-space symmetry of the pairs. For example, the orbital triplet \((k_x^2 − k_y^2)\alpha_1\) involving the product of a gap of nominal d-wave symmetry and an orbital field \( \alpha_1 \) forms an s-wave pair, because both \( k_x^2 − k_y^2 \) and \( \alpha_1 \) change sign under a 90° rotation. \( \text{[24, 25]} \). We shall argue that the strong Coulomb forces at the iron sites favor states with local orbital components of the pair wavefunction so that the winding number

\[
\oint_{\Gamma} \left( \hat{\mathbf{d}} \times \nabla_{\mathbf{k}} \hat{\mathbf{d}}(\mathbf{k}) \right) \cdot d\mathbf{k} = 2\pi \nu,
\]

evaluated around a path that encloses the \( \Gamma \) point, is a topological invariant. The “chirality” \( \nu = -\text{sgn}(\Delta_1\Delta_2) \) of the order parameter is selected by the underlying Hamiltonian without breaking any symmetry.

Table. I classifies the even-parity order parameters, with nearest or next-nearest neighbor pairing in the Fe-based SCs according to the irreducible representations of the \( \text{C}_{4v} \) group. Within the s-wave \( A_1 \) and \( A_2 \) symmetry classes there are two one dimensional representations with local \( d_{xy} \) and local \( d_{x^2−y^2} \) symmetry, in which the product of an orbital field \( \alpha_{1,2} \) with a d-wave gap function creates an s-wave pair. Their identical symmetries under \( \text{C}_{4v} \) means that hopping terms in the Hamiltonian inter-convert the two types of pair, creating an internal Josephson coupling between them.

In this paper, we focus on the \( A_1 \) and \( A_2 \) states, which are invariant under inversion symmetry of a single layer. The \( A_1 \) and \( B_1^\ast \) representations, which involve staggered \( d_{xy} \) pairing are odd-parity within a single plane and can only form a tao state in bulk 3D systems by staggering the \( d_{xy} \) pairing between layers; in three dimensions, the \( A_1 \) and \( B_1^* \) states acquire an additional sin \( k_z \) dependence, restoring their even parity giving rise to the \( \Delta_{xyz} = (\text{staggered} \Delta_{xy}) \times \sin k_z \) form-factor. We note that the \( A_1^\ast \) state corresponds to the \( \eta \)-pairing proposed in [26].

Using this scheme, we are able to identify two fully gapped 2D tao states, with \( A_1 \) and \( A_2 \) symmetry (see

| Irreducible Rep. | Gap Fn. | Local Symmetry | Orbital Symmetry |
|------------------|--------|----------------|-----------------|
| \( A_1 \)       | \( c_x c_y \tau_0 \alpha_2 \) | \( s^z \)      | 11 + 22         |
| \( B_1 \)       | \( i\Delta_{x−y} \tau_0 \alpha_2 \) | \( d_{x−y} \)  | 11 + 22         |
| \( B_2 \)       | \( \Delta_{xy} \tau_0 \alpha_2 \) | \( d_{xy} \)   | 11 + 22         |
| \( A_1 \)       | \( i\Delta_{x−y} \tau_0 \alpha_3 \) | \( d_{x−y} \)  | 11 + 22         |
| \( A_2 \)       | \( i\Delta_{xy} \tau_0 \alpha_3 \) | \( d_{xy} \)   | 12 + 21         |
| \( A_1^\ast \)  | \( i\Delta_{xy} \tau_0 \alpha_1 \) | \( d_{xy} \)   | 11 + 22         |
| \( B_1^\ast \)  | \( \Delta_{xy} \tau_0 \alpha_1 \) | \( 3D \text{ staggered} d_{xy} \) | 11 + 22 |

TABLE I. Symmetry classification of the singlet superconducting order parameters in the Fe-based systems. The non-trivial s-wave representations \( A_1 \) and \( A_2 \) contain two independent entries involving a product of a d-wave gap function with an orbital field \( \alpha_{1,2} \) which also changes sign under 90° rotations. The \( A_1 \) and \( B_1^\ast \) representations involve staggered \( d_{xy} \) order. The \( \tau_1 \) operator which implements the staggered pairing is odd-parity under inversion, so that the “f-wave” combination \( \Delta_{xy} \tau_1 = (\Delta_{xy} \tau_1) \times \sin k_z \) has even parity. Since \( \alpha_2 \) and \( \tau_2 \) are odd under time-reversal, the time-reversal invariant combinations \( i\alpha_2 \) and \( i\tau_2 \) are used in the pair operator.
To illustrate these results, we now carry out a BCS treatment of the $A_1$ tao state\[^{[1]}\], with Hamiltonian

$$H = \sum_{k} \Psi_{k}^\dagger h(k)\Psi_{k} - \frac{g_{1}}{N_s} \sum_{k,k'} b_{k1}^\dagger b_{k'1} - \frac{g_{2}}{N_\gamma} \sum_{k,k'} b_{k2}^\dagger b_{k'2},$$

(4)

where $N_s$ is the number of sites in the lattice and

$$b_{k1}^\dagger = c_{k\nu}^\dagger \left[ \begin{array}{c} d_{xy}(k) i \gamma_{2} \gamma_{3} \varepsilon_{k} \varepsilon_{k'} \end{array} \right]_{\nu\nu'} c_{k'\nu'}^\dagger,$$

$$b_{k2}^\dagger = c_{k\nu}^\dagger \left[ \begin{array}{c} d_{x^2-y^2}(k) \gamma_{1} \gamma_{3} \varepsilon_{k} \varepsilon_{k'} \end{array} \right]_{\nu\nu'} c_{k'\nu'}^\dagger,$$

(5)

create the two components of a tao-pair with $A_1$ symmetry, where $d_{xy}(k)$ and $d_{x^2-y^2}(k)$ are the $d$-wave form factors. We note that a microscopic model is most likely to involve orbital and spin superexchange processes, giving rise to Kugel-Khomskii type $J_1-J_2$ interactions\[^{[28]}\]. For simplicity, we have omitted interaction terms that mix the two pairs, since as we shall see, this mixing is already provided by the hopping.

The kinetic part of the Hamiltonian

$$h(k) = \epsilon_1(k) \gamma_{3} + \epsilon_1(k) \gamma_{1} + \epsilon_3(k) \alpha_{1} + \epsilon_4(k) \gamma_{1} \gamma_{3},$$

(6)

is a tight-binding description of the hopping terms generated by direct hopping between the iron atoms, and virtual hopping via the out-of-plane As (Se) atoms. Note the absence of the Nambu matrix $\gamma_{3}$ in the second and third term of the hopping Hamiltonian: this is because the orbital and tetrahedral operators are invariant under the particle hole transformation, $\alpha \rightarrow -\lambda_{2}\alpha^{T}\lambda_{2} = \hat{\alpha}$, $\tau \rightarrow -\lambda_{2}\tau^{T}\lambda_{2} = \hat{\tau}$. Here $\epsilon_1(k) = 4t_1(c_x + c_y)$ and $\epsilon_0(k) = 4t_0(c_x c_y - \mu)$ are the orbitally-independent amplitudes for nearest and next nearest-neighbor hopping, where $\mu$ is the chemical potential and $c_l \equiv \cos kl$ ($l = x, y$). The terms $\epsilon_4(k) = 4t_4(c_x - c_y)$ and $\epsilon_3(k) = 4t_3 s_x s_y$ are the amplitudes for orbitally-dependent nearest and next-nearest neighbor hopping, where $s_l \equiv \sin kl$ ($l = x, y$). These terms can be regarded as orbital “Rashba” fields, which split the Fermi surface into separate electron and hole pockets with non-trivial band topology\[^{[29]}\]. The resulting normal state spectrum is given by $\epsilon_{\alpha\beta}(k) = E_{s}(k) + \alpha \sqrt{\epsilon_{3}(k)^{2} + \epsilon_{4}(k)^{2}}$, where $E_{s} = E_{0}(k) + sgn(s) \epsilon_{1}(k)$ and $s, \alpha = \pm 1$ are two sets of band indices. Since $\epsilon_{3}(k) = 4t_{3} d_{xy}(k)$ and $\epsilon_{4}(k) = 4t_{4} d_{x^2-y^2}(k)$ have d-wave symmetry, the product of the these terms generates matrix elements that inevitably hybridize the two locally d-wave components of an $A_1$ or $A_2$ tao pair.

Performing a mean-field decoupling of the interaction, we obtain

$$H = \sum_{k} \Psi_{k}^\dagger H(k)\Psi_{k} + N_{s} \left( \frac{\Delta_{2}^{2}}{g_{1}} + \frac{\Delta_{2}^{2}}{g_{2}} \right),$$

(7)

where

$$H(k) = h(k) + \Delta_{xy}(k) \gamma_{2} \gamma_{3} + \Delta_{x^2-y^2}(k) \gamma_{3} \gamma_{1}$$

(8)

is the Nambu Hamiltonian. $H(k)$ can be diagonalized, giving rise to four separate quasi-particle eigenvalues for the electron and hole pockets, given by

$$E_{k}^{\nu\alpha} = \left[ A_{s}(k) + \alpha \sqrt{A_{y}(k)^{2} - B_{s}(k)^{2}} \right]^{1/2},$$

(9)

where $s, \alpha = \pm 1$ and

$$A_{s} = \epsilon_{s}^{2} + \epsilon_{3}^{2} + \epsilon_{4}^{2} + \Delta_{xy}^{2} + \Delta_{x^2-y^2}^{2},$$

$$B_{s} = (A - 2\epsilon_{3}^{2} - 2\epsilon_{4}^{2})^{2} + 4(\Delta_{xy}\epsilon_{3} - \Delta_{x^2-y^2}\epsilon_{4})^{2},$$

(10)

and for clarity we have suppressed the explicit momentum labels $k$. From\[^{[9]}\] and\[^{[10]}\] we see that $(E_{k}^{+} E_{k}^{-})^{2} = B_{s}(k)^{2} > 0$ is positive definite, so that the excitation spectrum is fully gapped. However, the degree of anisotropy is strongly dependent on the chirality $\nu = -sgn(\Delta_{1} \Delta_{2})$ of the gap: for $\nu < 0$, (\Delta_{1}, \Delta_{2} in phase), the second term in $B_{s}$ vanishes at points on the Fermi surface, so the gap is highly anisotropic, almost closing with a tiny minimum value $\Delta_{min} \approx \min(\Delta_{1}, \Delta_{2})^{2}/W$, where $W$ is the bandwidth; for positive chirality $\nu > 0$ (\Delta_{1}, \Delta_{2} out-of-phase), all terms in $B_{s}^{2}$ remain positive, and the gap is weakly anisotropic, with a large minimum value $\Delta_{min} \approx \min(\Delta_{1}, \Delta_{2})$. The weakly anisotropic positive chirality state is thus kinetically favored. The winding number of the tao state is given by Eq.\[^{[1]}\] with $\nu = \pm 2$.

Figs.\[^{[2]}\&\[^{[3]}\]show the superconducting gaps for the two generic cases seen in the iron-based superconductors; two electron and hole pockets around $M$ and $\Gamma$, and also two electron pockets around $M$, showing the fully gapped structure in both cases. A mean field calculation shows that the on-site s-wave component induced by the $A_{1}$ tao state is less than 1/10-th of the d-wave components for both the hole and electron pockets and electron pocket only systems; thus minimizing the Hubbard interaction on the Fe sites compared to an s-wave state of similar magnitude.
To explore the internal Josephson coupling between the two condensates, we minimize the mean-field Free energy

\[
F = N_s \left[ \frac{\Delta_1^2}{g_1} + \frac{\Delta_2^2}{g_2} \right] - 2T \sum_{\mathbf{k}, n, \alpha} \ln \left[ 2 \cosh \left( \frac{E_{\mathbf{k}n}^{\alpha}}{2T} \right) \right]
\]  

(11)

with respect to \( \Delta_1 \) and \( \Delta_2 \), to obtain two coupled gap equations

\[
\begin{pmatrix}
\frac{1}{g_1} - \chi_{xy} \\
-\chi_J
\end{pmatrix}
\begin{pmatrix}
\Delta_1 \\
\Delta_2
\end{pmatrix}
= 0
\]

(12)

where, denoting \( f_{\mathbf{k}} = \int \frac{d^2 q}{(2\pi)^2} \),

\[
\chi_{xy} = \sum_{s\alpha} \int_{\mathbf{k}} \frac{\text{th} \left( \beta E_{\mathbf{k}}^{s\alpha} / 2 \right)}{2 E_{\mathbf{k}}^{s\alpha}} \left[ 1 + 2\alpha \frac{(4t_4 d_{x^2-y^2})^2}{\sqrt{A_s^2 - B_s^2}} \right] d_{xy}^2
\]

(13)

\[
\chi_{x^2-y^2} = \sum_{s\alpha} \int_{\mathbf{k}} \frac{\text{th} \left( \beta E_{\mathbf{k}}^{s\alpha} / 2 \right)}{2 E_{\mathbf{k}}^{s\alpha}} \left[ 1 + 2\alpha \frac{(4t_3 d_{x^2-y^2})^2}{\sqrt{A_s^2 - B_s^2}} \right] d_{x^2-y^2}^2
\]

(14)

are the \( d_{xy} \) and \( d_{x^2-y^2} \) pair-susceptibilities, while

\[
\chi_J = \sum_{s\alpha} \int_{\mathbf{k}} \frac{\text{th} \left( \beta E_{\mathbf{k}}^{s\alpha} / 2 \right)}{2 E_{\mathbf{k}}^{s\alpha}} \left[ \frac{2t_3 t_4}{\sqrt{A_s^2 - B_s^2}} \right] (4d_{xy} d_{x^2-y^2})^2
\]

(15)

is the internal Josephson coupling between them. Since the \( E^{s\alpha}_{\mathbf{k}} \) have the lowest magnitude when \( \alpha = -1 \), \( \chi_J \) is negative, inducing a \( \pi \) coupling between the d-wave condensates, favoring selection of a positive chirality \( \nu > 0 \).

The mean-field transition temperature is determined by

\[
(1/g_1 - \chi_{xy})(1/g_2 - \chi_{x^2-y^2}) - \chi_J^2 = 0.
\]

In a conventional d-wave superconductor, pairing develops exclusively in the single most attractive d-wave channel, but in this system, the strong Josephson coupling between the two states will immediately lock the two gaps into a topological state with a single, enhanced transition temperature.

We end with a discussion of the experimental consequences of tao-pairing. There are a number of well established features of the experiments, such as the the absence of a Hebel-Slichter peak in NMR measurements \[1, 2\], and resonance peaks in the neutron scattering \[30, 31\] that are qualitatively consistent with the d-wave character of tao pairing. In multi-layer iron systems, tao pairing could occur in four different global symmetry representations, \( A_1 \), \( A_2 \) and the staggered \( A_1^* \) and \( B_1^* \) representations, driven by interlayer physics. The uniform \( A_1 \) (s-wave) symmetry driven by a strong in-plane Josephson coupling appears to be the best candidate: it allows us to understand the observed c-axis tunneling into s-wave superconductors \[20\], and possesses the most isotropic gap, a feature consistent with STM experiments \[10\].

The staggered d-wave \( A_1^* \) and \( B_1^* \) states may be relevant to systems with strong interplane coupling. Some experiments suggest a multi-gap character, including STM \[10, 11\], ARPES experiments \[32\], and the observation of a cross-over in the NMR relaxation rate from \( T_2^\prime \) from \( T_1 \) at high temperatures to \( T_2 \) \[4, 5\] behavior at low temperatures, features consistent with the \( A_1^* \) or \( B_1^* \) condensates, where the staggered d-wave gap \( \Delta_{xy} = \Delta_{s^x s_y s_z} \) will lead to out-of-plane point nodes.

An interesting feature of the theory is the internal chirality \( \nu \) of the pairs. Pairs of reversed chirality, generated by anti-phase fluctuations of the two gaps are expected to lead to a low-lying Raman-active Leggett mode \[33, 34\]. There may also be higher-energy excitations of Cooper pairs of a different symmetry, for e.g. from \( A_1 \) to \( B_1 \), which will couple to corresponding Raman modes. The emergence of these low-lying “failed d-wave” excitations below \( T_c \) is key property of the tao-condensate.

The chirality of the gap gives it a topological character. Symmetry analyses of the pairing Hamiltonian shows that it lies in the “CT” class of topological superconductors \[35\]. In two dimensions, these states are topologically trivial, but their three dimensional extension contains a topological integer \( (Z) \) invariant which allows for the possibility of non-trivial surface Andreev states.

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