Order parameter of quasi-one-dimensional superconductors:

symmetry features in quasiparticle density of states and spin susceptibility

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Recent experiments indicate that the Bechgaard Salts (TMTSF)$_2$ClO$_4$ and (TMTSF)$_2$PF$_6$ may be unconventional triplet superconductors. The quasiparticle density of states and the uniform spin susceptibility tensor are computed at low temperatures for order parameter symmetries, as an attempt to narrow the number of possibilities based on current experimental evidence.

In early theoretical investigations of quasi-one-dimensional superconductors (of the Bechgaard family (TMTSF)$_2$ClO$_4$ and (TMTSF)$_2$PF$_6$) in the presence of an external magnetic field, the possibility of triplet behavior in these systems was suggested [4]. The present experimental evidence indicates that these quasi-one-dimensional superconductors are indeed unconventional [5]. For instance, Lee et al. [6] measured the magnetic field versus temperature phase diagram for (TMTSF)$_2$PF$_6$ under pressure of 6 kbar. They found that the upper critical fields along the usual a, b', and c direction are highly anisotropic. Furthermore, the Pauli paramagnetic limit is exceeded by a factor of 3 or 4. In addition, Lee et al. [7] found that there is no Knight shift in (TMTSF)$_2$PF$_6$ for fields $\mathbf{H} \parallel b'$ at pressures $P \approx 6$ kbar. This suggests the existence of a triplet superconducting phase in this system. The temperature versus magnetic field phase diagram of (TMTSF)$_2$ClO$_4$ at ambient pressure was also measured by Lee et al. [8], for $\mathbf{H} \parallel b$. These measurements seem to indicate that the Pauli paramagnetic limit is exceeded in this compound, which suggests triplet superconductivity. Furthermore, Belin and Belnia [9] reported measurements of the thermal conductivity in the superconducting state of (TMTSF)$_2$ClO$_4$, indicating their data is inconsistent with the existence of gap nodes at the Fermi surface. These experimental results combined suggest the existence of gap nodes at the Fermi surface.

In the limit of weak interactions and low densities these quasi-one-dimensional systems exhibit a well defined Fermi surface which is open, being formed of two separate sheets which intersect the Brillouin zone boundaries. We work with the Hamiltonian $H = H_{kin} + H_{int}$, where the kinetic energy and the chemical potential contribution are $H_{kin} = \sum_{k,\alpha}(\epsilon_k - \mu)\psi_{k,\alpha}^\dagger\psi_{k,\alpha}$, and the interaction is

$$H_{int} = \frac{1}{2}\sum_{kk'qq'\alpha\gamma\delta} V_{\alpha\beta\gamma\delta}(k, k') b_{\alpha\beta}^\dagger(k, q) b_{\gamma\delta}(k', q)$$

with $b_{\alpha\beta}^\dagger(k, q) = \psi_{k+q/2,\alpha}^\dagger \psi_{k+q/2,\beta}^\dagger$, where $\alpha, \beta, \gamma$ and $\delta$ are spin indices and $k, k'$ and $q$ represent linear momenta. In the case of weak spin-orbit coupling and triplet pairing, the model interaction tensor can be chosen to be

$$V_{\alpha\beta\gamma\delta}(k, k') = \Gamma_{\alpha\beta\gamma\delta} V_{T}(k, k') \phi_{T}(k) \phi_{T}^\dagger(k'),$$

where $\Gamma_{\alpha\beta\gamma\delta} = v_{\alpha\beta} \cdot v_{\gamma\delta}/2$ with $v_{\alpha\beta} = (i\sigma\gamma_5)_{\alpha\beta}$. In addition, the interaction $V_T$ corresponds to the irreducible representation $\Gamma$ with basis function $\phi_{T}(k)$ representative of the orthorhombic group. In the case of strong spin-orbit coupling the interaction

$$V_{\alpha\beta\gamma\delta}(k, k') = V_{T}(k, k') [\Phi_{T}(k) \cdot v_{\alpha\beta}] [\Phi_{T}^\dagger(k') \cdot v_{\gamma\delta}/2$$

where the interaction $V_T$ corresponds to the irreducible representation $\Gamma$ with basis function vector $\Phi_{T}(k)$ representative of the orthorhombic group.

In this paper we are concerned with the symmetry of the order parameter of a triplet quasi-one-dimensional superconductor at zero magnetic field. Second, we calculate the quasiparticle density of states at zero temperature and the uniform spin susceptibility tensor at low temperatures for various candidate symmetries of the order parameter consistent with our group theoretical analysis. Third, we make connections to scanning tunneling microscopy (STM) of quasiparticle density of states and Knight shift measurements of the spin susceptibility tensor.

We study quasi-one-dimensional systems with a single band, in an orthorhobic lattice, and allow for singlet or triplet pairing. We consider the following dispersion

$$\epsilon_k = -t_x \cos(k_x a) - t_y \cos(k_y b) - t_z \cos(k_z c),$$

where $|t_x| \gg |t_y| \gg |t_z|$. In the limit of weak interactions and low densities these quasi-one-dimensional systems exhibit a well defined Fermi surface which is open, being formed of two separate sheets which intersect the Brillouin zone boundaries. We work with the Hamiltonian $H = H_{kin} + H_{int}$, where the kinetic energy and the chemical potential contribution are $H_{kin} = \sum_{k,\alpha}(\epsilon_k - \mu)\psi_{k,\alpha}^\dagger\psi_{k,\alpha}$, and the interaction is

$$H_{int} = \frac{1}{2}\sum_{kk'qq'\alpha\gamma\delta} V_{\alpha\beta\gamma\delta}(k, k') b_{\alpha\beta}^\dagger(k, q) b_{\gamma\delta}(k', q)$$

with $b_{\alpha\beta}^\dagger(k, q) = \psi_{k+q/2,\alpha}^\dagger \psi_{k+q/2,\beta}^\dagger$, where $\alpha, \beta, \gamma$ and $\delta$ are spin indices and $k, k'$ and $q$ represent linear momenta. In the case of weak spin-orbit coupling and triplet pairing, the model interaction tensor can be chosen to be

$$V_{\alpha\beta\gamma\delta}(k, k') = \Gamma_{\alpha\beta\gamma\delta} V_{T}(k, k') \phi_{T}(k) \phi_{T}^\dagger(k'),$$

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$$V_{\alpha\beta\gamma\delta}(k, k') = V_{T}(k, k') [\Phi_{T}(k) \cdot v_{\alpha\beta}] [\Phi_{T}^\dagger(k') \cdot v_{\gamma\delta}/2$$

where the interaction $V_T$ corresponds to the irreducible representation $\Gamma$ with basis function vector $\Phi_{T}(k)$ representative of the orthorhombic group.
In both weak and strong spin-orbit coupling, we can use either the equation of motion method [11] or the functional integration method [12,13] in the zero center of mass momentum pairing approximation (which corresponds to the BCS limit in weak coupling) to obtain the anomalous Green’s function

\[ F_{\alpha\beta}(k, i\omega_n) = \frac{\Delta_{\alpha\beta}(k)}{\omega_n^2 + E_k^2}, \]

and the single particle Green’s function

\[ G_{\alpha\beta}(k, i\omega_n) = -\frac{i\omega_n + \xi_k}{\omega_n^2 + E_k^2} \delta_{\alpha\beta}, \]

where \( \xi_k = \epsilon_k - \mu, \mu \) is the chemical potential, \( E_k = \sqrt{\epsilon_k^2 + \Delta_k^2} \) is the quasiparticle excitation energy, and \( \Delta_k^2 \equiv Tr \left[ \hat{\Delta}(k)^2 \right] / 2 \). The matrix \( \hat{\Delta}(k) \) has matrix elements \( \Delta_{\alpha\beta}(k) \). The expressions for the single particle (Eq. [3]) and for the anomalous (Eq. [3]) Green’s functions are valid only in the unitary case where \( \hat{\Delta}(k)^2 \) is diagonal. We will not discuss here the non-unitary case. Using the single particle and anomalous Green’s functions defined above and standard many body methods [11,13] we obtain the familiar forms

\[ \Delta_{\alpha\beta}(k) = -\sum_{k'} V_{\alpha\gamma\delta}(k,k') \frac{\Delta_{\gamma\delta}(k')}{2E_{k'}} \tanh \left( \frac{E_{k'}}{2T} \right), \]

\[ N \equiv \sum_k n_k = \sum_k \left[ n_{qp}(k) + n_{qh}(k) \right], \]

where \( n_{qp}(k) = (1 + \xi_k/E_k)f(E_k) \) and \( n_{qh}(k) = (1 - \xi_k/E_k) (1 - f(E_k)) \), for the order parameter and number equations, respectively.

Next we consider the allowed symmetries of the order parameter \( \Delta_{\alpha\beta} \) for an orthorhombic crystal [14], with a conventional symmetry normal state. Here, the relevant crystallographic point group is \( D_{2h} \), which has only one dimensional representations [13]. This implies that the order parameter matrix in the triplet channel,

\[ \hat{\Delta}(k) = i (\Delta_{\alpha\beta}(k)d(k) \cdot \sigma) \sigma_y, \]

must transform according to the one dimensional representations of the orthorhombic point group \( D_{2h} \), under the assumption that the order parameter does not break the crystal translational symmetry, i.e., the order parameter is invariant under all primitive lattice translations. Under the transformation \( k \to -k \) the three-dimensional vector \( d(k) \) is antisymmetric (odd), while the function \( \Delta_{\alpha\beta}(k) \) is symmetric (even).

Here, we will be interested in triplet states which do not break time reversal symmetry, and our analysis will be confined to zero magnetic field only. In Tables [II] and [III] we summarize the group theoretical analysis for \( \hat{\Delta}(k) \), in the weak and strong spin-orbit coupling cases respectively. The tables include the state nomenclature, the vector \( d(k) \), and the type of zeros of the quasiparticle excitation spectrum \( E_k \), when \( \tilde{\mu} = \mu - \min|\epsilon_k| \) is positive. In Table [II] the vector \( (0,0,1) \) is indicated up to an arbitrary rotation in spin space. In Table [III], the numerical coefficients \( A \) and \( C \) are determined through Eqs. [4] and [5]. It is crucial to emphasize that the basis functions \( X(k), Y(k) \) and \( Z(k) \) transform like \( k_x, k_y \) and \( k_z \) under the crystallographic point group operations. However, these functions cannot be chosen to be equal to \( k_x, k_y \) and \( k_z \) as done in the work by Lebed, Machida, and Ozaki [16] (LMO), since the Fermi surface \( \epsilon(k) = \mu \) intersects the Brillouin zone along the \( y \) and \( z \) directions. Thus, it is necessary to take into account the periodicity of the order parameter matrix \( \hat{\Delta}(k) \) and of the order parameter vector \( d(k) \) in reciprocal (momentum) space. As a result, the minimal basis set must be periodic and may be chosen to be \( X(k) = \sin(k_x), Y(k) = \sin(k_y) \) and \( Z(k) = \sin(k_z) \).

For weak spin-orbit coupling (Table [II]) the only candidate for weak attractive interaction \( (\tilde{\mu} > 0) \) is the state \( ^3B_{3u}(a) \), where the quasiparticle excitation spectrum \( E_k \) has no zeros and is fully gapped. For strong spin-orbit coupling (Table [III]) there are three candidates for weaker attractive interaction \( (\tilde{\mu} > 0) \), i.e., the states \( A_{1u}, B_{1u}, \) and \( B_{2u} \), where the quasiparticle excitation spectrum \( E_k \) may have no zeros and may be fully gapped. When \( \tilde{\mu} > 0 \), the state \( A_{1u} \) is fully gapped only for \( A \neq 0 \) and for any value of \( B \) and \( C \); the state \( B_{1u} \) is fully gapped only for \( B \neq 0 \) and for any value of \( A \) and \( C \); and the state \( B_{2u} \) is fully gapped only for \( C \neq 0 \) and for any value of \( A \) and \( B \). Note that in the case of strong attractive interactions where \( \tilde{\mu} < 0 \), the excitation spectrum \( E_k \) is fully gapped [13] for all states in both tables.

### Table I. Weak spin-orbit coupling.

| State | \( d(k) \) | \( E_k = 0 (\tilde{\mu} > 0) \) |
|-------|------------|-------------------------------|
| \( ^3A_{1u}(a) \) | \( (0,0,1)XYZ \) | lines |
| \( ^3B_{1u}(a) \) | \( (0,0,1)Z \) | lines |
| \( ^3B_{2u}(a) \) | \( (0,0,1)Y \) | lines |
| \( ^3B_{3u}(a) \) | \( (0,0,1)X \) | none |

### Table II. Strong spin-orbit coupling.

| State | \( d(k) \) | \( E_k = 0 (\tilde{\mu} > 0) \) |
|-------|------------|-------------------------------|
| \( A_{1u} \) | \( (AX, BY, CZ) \) | none, points or lines |
| \( B_{1u} \) | \( (AY, BX, CXYZ) \) | none or lines |
| \( B_{2u} \) | \( (AZ, BXYZ, CX) \) | none or lines |
| \( B_{3u} \) | \( (AXYZ, BZ, CY) \) | points or lines |
Next we turn our attention to the calculation of the quasiparticle density of states (QDOS) and uniform spin susceptibility at low temperatures for two potential triplet candidate states, which are fully gapped: a) the weak spin-orbit coupling state $^3B_{3u}(a)$; b) the strong spin-orbit coupling states $A_{1u}$.

The QDOS for these different symmetries can be obtained from the single particle Green’s function as

$$\mathcal{N}(\omega) = -\frac{1}{\pi} Tr \sum_k \text{Im} G_{\alpha\beta}(k, i\omega_n = \omega + i\delta), \quad (8)$$

where $G_{\alpha\beta}(k, i\omega_n)$ is defined in Eq. (3). The QDOS, shown in Fig. 1, can be measured in STM experiments. Although these experiments have not yet been performed in (TMTSF)$_2$ClO$_4$ and (TMTSF)$_2$PF$_6$, our theoretical predictions can serve as qualitative guides for the extraction of gaps and symmetry dependent features when experimental results become available. In particular, STM measured gaps could be compared with gaps measured either thermodynamically (e.g., specific heat) or in transport experiments (e.g., thermal conductivity [3]).

![FIG. 1. Plot of QDOS versus frequency: the weak spin-orbit coupling state $^3B_{3u}(a)$ is shown in (a); the strong spin-orbit coupling state $A_{1u}$ is shown in (b) for $A = 0.01$, $B = \sqrt{2} - A^2$, $C = 0$; in (c) for $A = B = 1$, $C = 0$; in (d) for $A = \sqrt{2} - B^2$, $B = 0.01$, $C = 0$. The parameters used are $|t_6| = 1000K$, $|t_9| = 100K$ and $|t_s| = 5K$, $\Delta_{tr} = \Delta_0 = 3.0K$ and $\mu = -250K$.

We compare in Fig. 1 the QDOS for the states $^3B_{3u}(a)$ (weak spin-orbit coupling) and $A_{1u}$ (strong spin-orbit coupling) for various values of the constants $A$, $B$, and $C$. The symmetry dependent features of the QDOS are manifested through the magnitude of the order parameter vector $d(k)$. For the $^3B_{3u}(a)$ state $|d(k)| \propto |\sin(k_xa)|$, while for the $A_{1u}$ state $|d(k)| \propto \sqrt{A^2|\sin(k_xa)|^2 + B^2|\sin(k_yb)|^2 + C^2|\sin(k_zc)|^2}$. In Fig. 1 (b), (c) and (d), we study only the case corresponding to $C = 0$, where $A \gg B$, $A = B$, and $A \ll B$, respectively. Notice that Fig. 1(d) is nearly identical to Fig. 1(a) given that $|d(k)|$ is essentially the same in this case. Furthermore, notice that while the position of the peaks in Figs. 1 (a), (b), (c) and (d) are essentially the same in scaled units ($\omega_g/\Delta_0 \approx \pm 1.40$), the corresponding gap sizes in scaled units are respectively $\omega_g/\Delta_0 = 1.32; 0.01; 0.04; 1.32$. Gap sizes, peaks and the general shape of the QDOS should be in principle identifiable in an STM experiment. However, such experiments alone cannot uniquely determine the symmetry of the order parameter in triplet quasi-one-dimensional superconductors, since the QDOS depends only on $|d(k)|$.

Now, we turn our attention to the calculation of the spin susceptibility tensor, which explicitly depends on both the magnitude and direction of $d(k)$, and, thus, may help elucidate the symmetry of the order parameter in quasi-one-dimensional superconductors. The spin susceptibility tensor for a triplet superconductor is

$$\chi_{mn}(q_\mu) = -\mu_B^2 (P_{nm})_{\alpha\beta\gamma\delta} [A_{\alpha\beta\gamma\delta}(q_\mu) + S_{\alpha\beta\gamma\delta}(q_\mu)],$$

where we use the Einstein summation convention, and the four-vector $q_\mu = (q, i\nu)$. The tensor $(P_{nm})_{\alpha\beta\gamma\delta} = (\sigma_m)_{\alpha\beta}(\sigma_n)_{\gamma\delta}$, contains Pauli spin matrices, and the tensors

$$A_{\alpha\beta\gamma\delta}(q_\mu) = \frac{1}{\beta} \sum_{k,i,\omega} F^+_{\beta\gamma}(-k + q, -i\nu + i\omega)F_{\delta\beta}(k, i\omega),$$

$$S_{\alpha\beta\gamma\delta}(q_\mu) = \frac{1}{\beta} \sum_{k,i,\omega} G_{\delta\alpha}(k - q, -i\nu + i\omega)G_{\gamma\beta}(k, i\omega)\delta_{\delta\alpha}\delta_{\beta\gamma}$$

contain the Green’s functions described in Eqs. (3) and (4), and $\beta = 1/k_BT$. For $\omega \to 0$ and $q \to 0$,

$$\chi_{mn}(0,0) = \sum_k \left[\chi_{mn,1}(k) + \chi_{mn,2}(k)\right],$$

where the $k$-dependent tensors have the forms

$$\chi_{mn,1}(k) = \chi_{||}(k) Re \, d^*_n(m)\hat{d}_n(k),$$

$$\chi_{mn,2}(k) = \chi_{\perp}(k) \left(\delta_{mn} - Re \, d^*_n(m)\hat{d}_n(k)\right),$$

with $\hat{d}_n(k) = d_n(k)/|d_n(k)|$. The parallel component is

$$\chi_{||}(k) = -2\mu_B \frac{\partial f(E_k)}{\partial E_k},$$

while the perpendicular component is

$$\chi_{\perp}(k) = 2\mu_B^2 \frac{d}{d\xi_k} \frac{\xi_k}{2E_k} \left(1 - 2f(E_k)\right).$$

This result is more general than the expression quoted in LMO [3] in a couple of ways. First, the expression derived in Eqs. (5), (6), and (7) includes particle-hole symmetry effects. Second they are valid at finite $T$. They do not include, however, Fermi liquid corrections.
In Fig. 2, we show the theoretical uniform $\chi_{mn}$ only for the $^3B_{3u}$ and $A_{1u}$ states [13], where triangles correspond to $\chi_{11}$ circles to $\chi_{22}$, and squares to $\chi_{33}$. It is known experimentally (Knight shift) [14] that the spin susceptibility of (TMTSF)$_2$PF$_6$ for $H \parallel b'$ is very close to $\chi_N$. Experiments for magnetic field along other directions have not yet been performed. Thus, for definiteness, we choose the unit vectors $\hat{3}$ ($m = 3$), $\hat{2}$ ($m = 2$) and $\hat{1}$ ($m = 1$) to point along the $b'$, $a$ and $c$ direction, respectively.

For the orthorhombic symmetry $\chi_{mn}$ is diagonal, and is calculated here under the assumption of constant $d(k)$, i.e., the direction of $d(k)$ is not changed upon application of a small magnetic field. In the case of state $A_{1u}$ (strong spin-orbit coupling) a small magnetic field cannot rotate $d(k)$ which is pinned to a particular lattice direction. Here, $\chi_{mm}$ is still diagonal, however the diagonal components are not equal in general. Thus, the experimentally measured $\chi^e_{mm}$ and the theoretically calculated $\chi^{th}_{mm}$ (at constant $d(k)$) should agree for small enough magnetic fields. However, in the case of state $^3B_{3u}(a)$ (weak spin-orbit coupling) a small magnetic field can always rotate $d(k)$ to be perpendicular to $H$, and thus minimize the magnetic free-energy $F_{mag} = -H_m \chi_{mn} H_n/2$. In this case, $\chi^e_{mn} \approx \chi_N \delta_{mn}$, where $\chi_N$ is the normal state value, for any direction $H$. Thus, $\chi^e_{mn}$ and $\chi^{th}_{mn}$ (shown in Fig. 2) are different.

![Fig. 2](image-url)

**FIG. 2.** Plot of the theoretical uniform spin susceptibility tensor components $\chi_{11}$ (triangles); $\chi_{22}$ (circles); $\chi_{33}$ (squares) at low temperatures. The weak spin-orbit coupling state $^3B_{3u}$ is shown in (a); strong spin-orbit coupling state $A_{1u}$ is shown in (b) for $A = 0.01$, $B = \sqrt{2 - A^2}$, $C = 0$; in (c) for $A = B = 1$, $C = 0$; in (d) for $A = \sqrt{2 - B^2}$, $B = 0.01$, $C = 0$. The parameters used are $|t_x| = 1000K$, $|t_y| = 100K$ and $|t_z| = 5K$, $\Delta_{tr} = \Delta_0 = 3.0K$ and $\mu = -250K$.

In conclusion, we have studied order parameter symmetry features in the quasiparticle density of states and spin susceptibility tensor of orthorhombic quasi-one-dimensional superconductors. We studied both the weak and strong spin-orbit coupling cases from a group theoretical point of view at zero magnetic field. Based on experimental evidence, we would like to suggest that the weak spin-orbit coupling state $^3B_{3u}(a)$ is the best candidate for the order parameter symmetry for these systems since this state is: (1) fully gapped and consistent with thermal conductivity measurements [3]; (2) characterized by weak spin-orbit coupling and consistent with weak spin-orbit coupling fits of $T_c(H)$ for (TMTSF)$_2$PF$_6$ [15] at low magnetic fields; (3) consistent with no observable Knight shift when $H \parallel b'$, and predicted to have no observable Knight shift for any direction of $H$.

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