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A phenomenological model of the superconducting state of the Bechgaard salts

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Abstract
We present a group theoretical analysis of the superconducting state of the Bechgaard salts, e.g., $(TMTSF)_2PF_6$ and $(TMTSF)_2ClO_4$ (TMTSF: tetramethyltetraselenafulvalene). We show that there are eight symmetry distinct superconducting states. Of these, only the (fully gapped, even frequency, odd parity, triplet) 'polar state' is consistent with the full range of the experiments on the Bechgaard salts. The gap function of the polar state is $\mathbf{d}(\mathbf{k}) \propto (\psi_{u\mathbf{k}}, 0, 0)$, where $\psi_{u\mathbf{k}}$ may be any odd parity function that is translationally invariant. This analysis also predicts that a phase transition, between two superconducting phases, occurs in weak magnetic fields.

1. Introduction

One of the central challenges facing theoretical physics is the development of a full microscopic understanding of unconventional superconductivity. An important first step towards this daunting task is the identification of the correct phenomenological description of the relevant materials. Indeed, our current understanding of the cuprate [1], heavy fermion [2], ruthenate [3], colbaltate [4], and quasi-two-dimensional organic [5] superconductors depends on phenomenological descriptions because in each of these cases there is no widely agreed upon microscopic description of the superconductivity. However, despite long standing evidence [6, 7] of unconventional superconductivity in the Bechgaard salts and theoretical proposals of triplet states [8], the correct phenomenological description of the superconducting state has not, until now, been identified.

Below we present a group theoretical classification of *all* of the possible superconducting states in the Bechgaard salts that respect translational symmetry. This shows that there are only eight symmetry distinct states. By considering the properties of these states we show that only one of them is consistent with the full range of thermodynamic measurements [9–17] that have been performed on both $(TMTSF)_2PF_6$ and $(TMTSF)_2ClO_4$. This state is somewhat analogous to the polar state, first discussed in the context of superfluid ^{3}He.

2. Symmetry analysis

The Bechgaard salts form triclinic crystals whose symmetry is represented by the C_i point group. C_i contains only two elements, the identity and inversion. Thus the point group only differentiates between even and odd parity states (which we henceforth refer to as s-wave and p-wave states respectively). Note that symmetry does not distinguish, say 'd-wave' states from s-wave states or 'f-wave' states from p-wave states as the crystal has neither rotational nor mirror symmetries. (Hence the terms 'd-wave' and 'f-wave' are rather meaningless in the context of the Bechgaard salts.) All the superconducting states unambiguously identified in bulk materials thus far, are even under frequency reversal. However, this is not required *a priori* in the superconducting state and so we must distinguish between even frequency or odd frequency pairing [18]. As the wavefunction of a fermionic system must be antisymmetric under the exchange of *all* labels, the allowed states are then: even frequency, s-wave singlet; odd frequency, p-wave, singlet; even frequency, p-wave triplet; and odd frequency, s-wave triplet.

The gap function of the singlet phases may be written as $\Delta(\mathbf{k}) = \eta\psi_{\mathbf{k}}$, where $\psi_{\mathbf{k}}$ may be any function with the appropriate parity that satisfies translation invariance, and η is the complex Ginzburg–Landau (GL) order parameter. Thus there are only two symmetry distinct singlet states: the conventional s-wave, even frequency singlet and a p-wave, odd frequency singlet.

To describe triplet superconductivity one must introduce a complex vector gap function, $\mathbf{d}(\mathbf{k})$ [19]. The interpretation of $\mathbf{d}(\mathbf{k})$ is straightforward (at least for unitary vectors[1]) as

[1] Unitary order parameters are defined as those for which $\mathbf{d}(\mathbf{k}) \times \mathbf{d}^*(\mathbf{k}) = 0$ [19].

it points along the $S_z = 0$ projection, i.e., perpendicular to the spin, of the Cooper pairs. For triplet superconductors we must distinguish between weak and strong spin–orbit coupling (SOC). If the SOC is sufficiently weak we may rotate the spin and spatial degrees of freedom independently; therefore the symmetry group of the normal state is $\mathcal{G} = SO(3) \otimes G \otimes U(1) \otimes \mathcal{T}$, where $SO(3)$ is spin rotation symmetry, G is the point group of the crystal (C_i for the Bechgaard salts), $U(1)$ is gauge symmetry and $\mathcal{T}$ is time reversal symmetry. However, for strong SOC the independent rotation of the spin and spatial degrees of freedom is not a symmetry of the system. Therefore, the symmetry group becomes $\mathcal{G} = G^{(J)} \otimes U(1) \otimes \mathcal{T}$, where the group $G^{(J)}$ is identical to the usual point group of the crystal except that the operations of the point group simultaneously act on both the spin and the spatial degrees of freedom.

When SOC is strong we expect that $\mathbf{d}(\mathbf{k}) \propto (\hat{\mathbf{a}} X_{\mathbf{k}}, \hat{\mathbf{b}}' Y_{\mathbf{k}}, \hat{\mathbf{c}}^* Z_{\mathbf{k}})$, where $X_{\mathbf{k}}$, $Y_{\mathbf{k}}$, and $Z_{\mathbf{k}}$ are arbitrary functions which respect translational symmetry and transform like k_x, k_y, and k_z respectively under the symmetry operations of the point group [1, 2]. However, as C_i contains only the identity and inversion the only restriction on $X_{\mathbf{k}}$, $Y_{\mathbf{k}}$, and $Z_{\mathbf{k}}$ is that they have the appropriate parity. Thus, $\mathbf{d}(\mathbf{k}) = \eta(\hat{\mathbf{a}}\psi^a_{\mathbf{k}}, \hat{\mathbf{b}}'\psi^b_{\mathbf{k}}, \hat{\mathbf{c}}^*\psi^c_{\mathbf{k}})$, where $\psi^a_{\mathbf{k}}$, $\psi^b_{\mathbf{k}}$ and $\psi^c_{\mathbf{k}}$ may be any functions that have the required parity and satisfy translation symmetry, and the GL order parameter is a single complex number (η). Therefore the GL free energy is

$$F_s - F_n = \alpha|\eta|^2 + \beta|\eta|^4. \tag{1}$$

Clearly there is only one solution (up to the, broken, $U(1)$ gauge symmetry) and therefore there is only one symmetry distinct spin part of the wavefunction for triplet superconductivity when SOC is strong. These states are analogous to the BW phase, which is realized at ambient pressure in ^{3}He, but may be either an even frequency p-wave or an odd frequency s-wave state.

Because C_i only contains one-dimensional irreducible representations $\mathbf{d}(\mathbf{k}) = \vec{\boldsymbol{\eta}} \cdot \boldsymbol{\Psi}_{\mathbf{k}}$ for weak SOC, where $\vec{\boldsymbol{\eta}}$ is the complex vector GL order parameter and $\boldsymbol{\Psi}_{\mathbf{k}} = (\psi^a_{\mathbf{k}}, \psi^b_{\mathbf{k}}, \psi^c_{\mathbf{k}})$. The GL expression for the free energy is thus [1, 2]

$$F_s - F_n = \alpha|\vec{\boldsymbol{\eta}}|^2 + \beta_1|\vec{\boldsymbol{\eta}}|^4 + \beta_2\left|\vec{\boldsymbol{\eta}} \cdot \vec{\boldsymbol{\eta}}\right|^2. \tag{2}$$

The ground state, up to arbitrary rotations in spin-space, is $\vec{\boldsymbol{\eta}} \propto (1, i, 0)$ for $\beta_2 > 0$ and $\vec{\boldsymbol{\eta}} \propto (1, 0, 0)$ for $\beta_2 < 0$. We refer to these states as the β and polar phases respectively by analogy with ^{3}He [19]. The β phase corresponds to pairing in a single spin channel, while the polar phase corresponds to pairing in both equal spin pairing (ESP) channels. Note that a representation must be at least two-dimensional for the ABM phase (which is realized under pressure in ^{3}He [19] and probably in Sr_2RuO_4 [3]) to be possible and so this phase can be immediately ruled out of our consideration of the Bechgaard salts. Both the β and polar states may exist as either p-wave, even frequency, triplet states or as s-wave, odd frequency, triplet states. Thus there are four possible triplet states if SOC is weak.

Table 1. Summary of the eight symmetry distinct states allowed for superconductors with C_i point groups. We describe whether these occur under weak or strong spin–orbit coupling (SOC) and define the vector order parameter for the triplet states. The basis functions $\psi^i_{u\mathbf{k}}$ ($\psi^i_{g\mathbf{k}}$) may be any odd (even) parity functions which satisfy translation symmetry.

State	SOC	$\mathbf{d}(\mathbf{k})$
s-singlet	Any	—
Polar s-triplet	Weak	$(1, 0, 0)$
β s-triplet	Weak	$(1, i, 0)$
BW s-triplet	Strong	$(\hat{\mathbf{a}}\psi^a_{g\mathbf{k}}, \hat{\mathbf{b}}'\psi^b_{g\mathbf{k}}, \hat{\mathbf{c}}^*\psi^c_{g\mathbf{k}})$
p-singlet	Any	—
polar p-triplet	**Weak**	$\mathbf{(1, 0, 0)}$
β p-triplet	Weak	$(1, i, 0)$
BW p-triplet	Strong	$(\hat{\mathbf{a}}\psi^a_{u\mathbf{k}}, \hat{\mathbf{b}}'\psi^b_{u\mathbf{k}}, \hat{\mathbf{c}}^*\psi^c_{u\mathbf{k}})$

3. Properties of the superconducting states

We list all eight symmetry distinct superconducting states for the group C_i, which represents the symmetry of crystals of the Bechgaard salts, in table 1. Our task is now to determine the properties of the of these states and to compare these properties with those found experimentally in $(TMTSF)_2PF_6$ and $(TMTSF)_2ClO_4$.

None of the four even frequency states are required by symmetry to have nodes in the order parameter. This extremely unusual property for an unconventional superconductor is *not*, as is often stated, because of the quasi-one-dimensionality of the Fermi surface, but is a direct consequence of the extremely low symmetry of the Bechgaard salts. Recall that the basis functions may be any function with the appropriate parity. Therefore, s-wave states have no symmetry required nodes and p-wave states are required to vanish only at the origin (Γ-point), which, by symmetry, the Fermi surface may not cross. In contrast, odd frequency pairing states are intrinsically gapless [18]. Specific heat measurements on $(TMTSF)_2PF_6$ [15] and thermal conductivity measurement on $(TMTSF)_2ClO_4$ [16] both show an exponentially activated behaviour, suggestive of a nodeless gap. As these experiments see a full gap they are inconsistent with odd frequency pairing. However, the NMR relaxation rate, $1/T_1$, has a power law temperature dependence [10, 11]. If this power law were assumed to arise from quasiparticles then it would be suggestive of nodes in the gap. Rostunov *et al* [20] have recently shown that, in a triplet superconductor, collective spin-wave excitations can also lead to a power law dependence of $1/T_1$. This theory may also resolve the puzzle of why the power law dependence of $1/T_1$ is seen even at temperatures very close to the critical temperature [10, 11], which is not expected from nodal quasiparticles. Thus these experiments may suggest a triplet pairing state.

An extremely small peak is seen in $1/T_1$, just below T_c [10, 9]. However, this peak more than an order of magnitude smaller than the Hebel–Slichter expected for an even frequency, s-wave, singlet order parameter [21]. This strongly suggests that the even frequency, s-wave, singlet order parameter is *not* realized in the Bechgaard salts. Further evidence for this conclusion comes from the observed strong suppression of the superconducting critical temperature by disorder [6, 7]. The only state for which this suppression of T_c by disorder is *not* expected is the even frequency, s-wave, singlet order parameter [22].

Evidence for triplet pairing comes from the observation that the upper critical fields in the conducting planes of $(TMTSF)_2PF_6$ and $(TMTSF)_2ClO_4$ exceed the weak coupling Clogston–Chandrasekhar (or Pauli) limit by more than a factor of four [13, 14, 12]. The Pauli limit occurs when the Zeeman energy penalty for forming $S_z = 0$ Cooper pairs exceeds the condensation energy gained by entering the superconducting state and applies to singlet states and pairs in the $S_z = 0$ projection of a triplet state [23]. When SOC is weak the spin part of the order parameter is not 'pinned' to the lattice. Therefore, the superconductor may minimize its energy by aligning $\mathbf{d}(\mathbf{k}) \perp \mathbf{H}$ [23][2]. Thus, the triplet phases for weak SOC will always be ESP phases in the reference frame of the magnetic field and are therefore not Pauli limited.

In contrast the triplet phases for strong SOC are 'pinned' to the lattice as the symmetry group does not allow the independent rotation of the spin and spatial degrees of freedom. When a field exceeding the Pauli limit is applied to system, it will completely suppress the pairing in the $S_z = 0$ channel, i.e., $\mathbf{d}(\mathbf{k})$ goes to zero in the direction parallel to the field. However, ESP is not suppressed, i.e., $\mathbf{d}(\mathbf{k})$ remains finite perpendicular to the field. Both possible triplet states for strong SOC in the Bechgaard salts have finite components perpendicular to the conducting plane and thus we do not expect them to be Pauli limited. Hence, we do not expect any of the symmetry distinct triplet states to be Pauli limited. Therefore, while the large critical field is strong evidence against singlet pairing it does not differentiate among the six candidate triplet states. It is also worth noting that calculations suggest that the observed critical field is too large to be accounted for by FFLO singlet states which break translational symmetry [24]. We will therefore not further consider FFLO states.

If a superconducting order parameter breaks TRS then spontaneous supercurrents will flow around impurities and near grain boundaries [25]. The most sensitive probe of these tiny currents is the zero field muon spin relaxation (ZF-μSR) rate. Small fields consistent with broken TRS (BTRS) have been observed in UPt_3 [26], $U_{1-x}Th_xBe_{13}$ [27], $PrOs_4Sb_{12}$ [28], and Sr_2RuO_4 [29]. Importantly, as the currents due to a superconducting order parameter with BTRS are extremely small the magnetic fields they generate can be suppressed by very small longitudinal fields (50 G is sufficient in Sr_2RuO_4 [29]). Luke *et al* [17] measured the ZF-μSR rate in $(TMTSF)_2ClO_4$ and did not find any increase in the relaxation rate to within their experimental resolution (≈25 G). This indicates that the superconducting state of $(TMTSF)_2ClO_4$ does not break TRS. Both the even frequency and odd frequency pairing β phases are inconsistent with this experiment.

The spin susceptibility, $\chi_s(T)$, strongly distinguishes between different triplet states [19]. When an $S_z = 0$ pair forms it no longer contributes to the spin susceptibility and thus for a singlet superconductor or a triplet superconductor with $\mathbf{d}(\mathbf{k}) \parallel \mathbf{H}$, $\chi_s(T) \rightarrow 0$ as $T \rightarrow 0$. On the other hand ESP does not affect the susceptibility. Thus, $\chi_s(T)$ does not change upon passing though T_c for ESP-only states such as the ABM phase of ^{3}He [19]. For triplet states which contain both ESP and $S_z = 0$ pairs the decrease in $\chi_s(T)$ is proportional to the fraction of $S_z = 0$ pairs. As discussed above, the symmetry distinct states for weak SOC are all ESP states and thus we expect $\chi_s(T)/\chi_n = 1$ for all $T < T_c$, where χ_n is the spin susceptibility of the normal state. In contrast, in both the possible states for strong SOC contain pairing in all three S_z projections. Thus,

$$\frac{\chi_s^i(0)}{\chi_n} \rightarrow 1 - \frac{\langle|\psi_\mathbf{k}^i|^2\rangle_{FS}}{\langle|\mathbf{\Psi}_\mathbf{k}|^2\rangle_{FS}}, \tag{3}$$

where, $\langle\cdots\rangle_{FS}$ indicates the average over the Fermi surface, $i \in \{a, b, c\}$, and the superscript on the susceptibility indicates the orientation of the field [19]. If the averages over the three basis functions are the same, as they are in the BW phase of ^{3}He, $\chi_s(0)/\chi_n = 2/3$ for all field orientations. No decrease in $\chi(T)$ is detected below T_c in $(TMTSF)_2PF_6$ with the field aligned along either the $\mathbf{a}$ [10] or $\mathbf{b}'$ [11] axes. $\chi_s^a(0)/\chi_n = \chi_s^b(0)/\chi_n = 1$ if and only if $\psi_\mathbf{k}^a$ and $\psi_\mathbf{k}^b$ vanish everywhere on the Fermi surface. As we do not, in general, expect this to be the case the BW state is incompatible with the measured Knight shift.

We summarize the properties of the eight symmetry distinct superconducting phases in table 2. It can readily be seen that the only state consistent with all of the experiments is the 'polar' p-wave triplet state realized for weak SOC which is specified by $\mathbf{d}(\mathbf{k}) \propto (\psi_\mathbf{k}, 0, 0)$. It is worth stressing the very small number of assumptions that have been made to reach this conclusion: (i) translational symmetry is not violated by superconducting state of the Bechgaard salts; (ii) there is not an accidental vanishing of two independent basis functions if SOC is strong; and (iii) that the superconducting states of $(TMTSF)_2PF_6$ and $(TMTSF)_2ClO_4$ have the same symmetry, i.e., that the materials are related by 'chemical pressure'. (i) is supported by calculations of the critical field for the FFLO state in these materials [24]. (ii) can be tested experimentally: $\chi_s(T) \rightarrow 0$ as $T \rightarrow 0$ with $\mathbf{H} \parallel \mathbf{c}^*$ for the BW state with $\langle|\psi_\mathbf{k}^a|^2\rangle_{FS} = \langle|\psi_\mathbf{k}^b|^2\rangle_{FS} = 0$; whereas for the polar state $\chi_s(T) = \chi_n$ for $\mathbf{H} \parallel \mathbf{c}^*$. (iii) is easily tested experimentally; in particular measurements of the Knight shift in $(TMTSF)_2ClO_4$ and the ZF-μSR rate in $(TMTSF)_2PF_6$ would complete the set of measurements required to uniquely determine the superconducting state of each material individually.

The different states correspond to different broken symmetries therefore measurements of collective modes [19, 20] could also provide confirmation of our identification of the superconducting state.

For sufficiently small magnetic fields SOC will be strong therefore our analysis predicts that a phase transition from the BW state to the polar state occurs at extremely low fields. This phase transition is somewhat reminiscent of the Fredericks transitions that occurs in superfluid ^{3}He in a slab geometry [19]. As this phase transition corresponds to the

[2] A complex vector, e.g., the order parameter of the β phase, $(1, i, 0)$, is perpendicular to a field if $\mathbf{d}(\mathbf{k}) \cdot \mathbf{H} = 0$.

Table 2. Summary of the thermodynamic properties of the eight symmetry distinct states allowed for superconductors with C_i point groups and comparison with experiments on the Bechgaard salts. We see that only the polar p-wave triplet state is compatible with experiment. $\chi_s(0)/\chi_n$ is the ratio of the spin susceptibility in the limit $T \to 0$ to that in the normal state above T_c. The symbol '?' in the experimental sections indicates that an experiment has not been performed. See section 3 for discussion and caveats.

State	(Symmetry required) nodes	BTRS	Pauli limited $\mathbf{H} \parallel \mathbf{a}$	Pauli limited $\mathbf{H} \parallel \mathbf{b}'$	$\frac{\chi_s(0)}{\chi_n}$ $\mathbf{H} \parallel \mathbf{a}$	$\frac{\chi_s(0)}{\chi_n}$ $\mathbf{H} \parallel \mathbf{b}'$	Disorder suppresses T_c	Hebel–Slichter peak
s-singlet	**No**	**No**	Yes	Yes	0	0	No	Yes
s-polar	Gapless	**No**	**No**	**No**	**1**	**1**	**Yes**	**No**
s-β	Gapless	Yes	**No**	**No**	**1**	**1**	**Yes**	**No**
s-BW	Gapless	**No**	**No**	**No**	$\sim 2/3$	$\sim 2/3$	**Yes**	**No**
p-singlet	Gapless	**No**	Yes	Yes	0	0	**Yes**	**No**
p-polar	**No**	**No**	**No**	**No**	**1**	**1**	**Yes**	**No**
p-β	**No**	Yes	**No**	**No**	**1**	**1**	**Yes**	**No**
p-BW	**No**	**No**	**No**	**No**	$\sim 2/3$	$\sim 2/3$	**Yes**	**No**
$X = PF_6$	No [15]	?	No [13]	No [13]	1 [10]	1 [11]	Yes [6]	No [10]
$X = ClO_4$	No [16]	No [17]	?	No [12]	?	?	Yes [7]	No [9]

vanishing of superconductivity in the $S_z = 0$ channel, and, by definition, occurs in a finite magnetic field, we expect that the phase transition will be first order.

4. Previous theoretical work

Early theoretical work only focused on whether the superconductivity was singlet or triplet and did not propose a specific triplet state [8]. The state discussed by Lebed and co-workers [24] assumes strong SOC and is therefore a special case of the BW phase. Lebed *et al*'s state has $\langle|\psi^b_{\mathbf{k}}|^2\rangle_{FS} = 0$, but $\langle|\psi^a_{\mathbf{k}}|^2\rangle_{FS} \neq 0$. Equation (3) shows that this theory will predict a large decrease in the spin susceptibility below T_c when $\mathbf{H} \parallel \mathbf{a}$. This prediction is clearly contradicted by experiment [10] and therefore this theory can be ruled out. Duncan *et al* [30] have discussed the symmetry distinct triplet states in an orthorhombic (D_{2h}) crystal. As the Bechgaard salts are triclinic and the angles involved are rather large this is *not* a good approximation. Nevertheless the state they propose (a 'p_x' state, $\mathbf{d}(\mathbf{k}) \propto (k_x, 0, 0)$ and weak SOC) is a special case of the polar state we have shown to be the actual superconducting state. However, the 'p_x' state has an accidental node in the plane $k_x = 0$. As this plane does not cut the Fermi surface the physical properties of the 'p_x' state are rather similar to polar state. However, we stress that the node in the 'p_x' state is *not* required by symmetry and will raise the energy of the state, therefore this node is unphysical. Shimahara [31] proposed that a singlet state is found at low field and a triplet state is found in large fields. As inversion is a symmetry of the crystal, such a change must be accompanied by a phase transition. This has not been observed and so this theory does not seem compatible with experiment.

5. Conclusions

We have shown that their are eight symmetry distinct superconducting states in monoclinic crystals with the C_i point group. Of these only the p-wave, even frequency, polar state $[\mathbf{d}(\mathbf{k}) \propto (\psi_{u\mathbf{k}}, 0, 0)]$ is consistent with the full range of experiments on both $(TMTSF)_2ClO_4$ and $(TMTSF)_2PF_6$. There is not yet sufficient experimental evidence to determine the superconducting state of either these materials individually, but the chemical pressure hypothesis suggests that the polar state is also realized in these materials. This analysis also led to the prediction of a magnetic field induced phase transition from the BW phase in low fields to the polar state in moderate fields. Finally, we note that the same symmetry analysis applies to the Fabre salts.

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Note added in proof. Since this manuscript was placed on the arXiv experimental results have been reported [32] that are entirely consistent with our prediction of the superconductor to superconductor phase transition. It appears that the critical field for this transition is about 10 kOe. Further, from the data reported in [32] and equation (3) it is straightforward to show that in the low field BW state $\langle|\psi^a_{\mathbf{k}}|^2\rangle_{FS} \sim 0.4\langle|\mathbf{\Psi_k}|^2\rangle_{FS}$ and $\langle|\psi^b_{\mathbf{k}}|^2\rangle_{FS} \sim 0.6\langle|\mathbf{\Psi_k}|^2\rangle_{FS}$ which implies that $\langle|\psi^c_{\mathbf{k}}|^2\rangle_{FS} \sim 0$.

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