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Triplet electron pairing and anisotropic spin susceptibility in organic superconductors $(TMTSF)_2X$

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We argue that the $(TMTSF)_2PF_6$ compound under pressure is likely a triplet superconductor with a vector order parameter $\mathbf{d}(\mathbf{k}) \equiv (d_a(\mathbf{k}) \neq 0,\ d_c(\mathbf{k}) = ?,\ d_{b'}(\mathbf{k}) = 0)$; $|d_a(\mathbf{k})| > |d_c(\mathbf{k})|$. It corresponds to an anisotropic spin susceptibility at $T=0$: $\chi_{b'} = \chi_0$, $\chi_a \ll \chi_0$, where χ_0 is its value in a metallic phase. (The spin-quantization axis, $\mathbf{z}$, is parallel to a so-called $\mathbf{b}'$ axis.) We show that the suggested order parameter explains why the upper critical field along the $\mathbf{b}'$ axis exceeds all paramagnetic limiting fields, including that for a nonuniform superconducting state, whereas the upper critical field along the $\mathbf{a}$ axis ($\mathbf{a} \perp \mathbf{b}'$) is limited by the Pauli paramagnetic effects [I. J. Lee, M. J. Naughton, G. M. Danner, and P. M. Chaikin, Phys. Rev. Lett. **78**, 3555 (1997)]. The triplet order parameter is in agreement with the recent Knight shift measurements by I. J. Lee *et al.* as well as with the early results on a destruction of superconductivity by nonmagnetic impurities and on the absence of the Hebel-Slichter peak in the NMR relaxation rate.

Quasi-one-dimensional (Q1D) organic compounds $(TMTSF)_2X$ ($X = PF_6$, ClO_4, etc.) have been intensively investigated since the discovery of superconductivity[1,2] in the first organic superconductor $(TMTSF)_2PF_6$. From the beginning, it was clear that their properties were unusual. It was found[3–8] that superconductivity in $(TMTSF)_2X$ ($X = PF_6$, ClO_4) is destroyed by nonmagnetic impurities. This was interpreted in terms of a possible triplet pairing of electrons.[9] Another unusual feature, the absence of the Hebel-Slichter peak in the $1/T_1$ NMR data in $(TMTSF)_2X$ ($X = PF_6$, ClO_4),[10–12] was prescribed[13] to the existence of zeros of a superconducting order parameter on the Q1D Fermi surfaces (FS). As was stressed,[13] the early experiments[3–8,10,11] provided information only about an orbital part of the order parameter and could not distinguish between some triplet and singlet pairings.[2,13]

To reveal triplet superconductivity, experimental tests which probe a spin part of an order parameter are essential. Among them, are a surviving of triplet superconductivity in the Q1D case[14–17] at magnetic fields higher than both the upper orbital critical field and the Clogston paramagnetic limit,[18] observation of spin-wave excitations,[15] the Knight shift measurements,[12] and some others. Nowadays, interest in a possible triplet pairing has been renewed due to remarkable measurements of the upper critical fields (which are sensitive to a spin part of the order parameter) in $(TMTSF)_2ClO_4$ and in $(TMTSF)_2PF_6$ at $P \simeq 6$ kbar by Naughton, Lee, Chaikin, and Danner[19–21] and due to the theoretical analysis[16] of these experiments. The experimental fields along the $\mathbf{b}'$ axis (which are three times bigger[20,21] than the Clogston paramagnetic limit) were shown[16] to be even bigger than the paramagnetic limit[16,22] for the Larkin-Ovshinnikov-Fulde-Ferrell (LOFF) phase.[23] Therefore, measurements[19–21] were interpreted[16,19–21] in terms of triplet superconductivity. Recently, Lee *et al.*[12] have found no change of the Knight shift for $\mathbf{H} \parallel \mathbf{b}'$ in a superconducting phase of $(TMTSF)_2PF_6$ at $P \simeq 6$ kbar. This is consistent with the results[16,19–21] and strongly supports the triplet scenario[9,16,19–21] of superconductivity.

The goals of our paper are as follows: (1) To calculate the paramagnetic limited field along the $\mathbf{b}'$ axis, $H_p^{b'}$, for the LOFF phase in a Q1D superconductor, taking account of both the paramagnetic[16] and orbital destructive effects against superconductivity [we show that the calculated value of $H_p^{b'}$ is 4–5 times less than the experimental fields[20,21] in $(TMTSF)_2PF_6$]. (2) To demonstrate that the value of $H^{b'}$ becomes consistent with Refs. 20 and 21 if we switch off the paramagnetic effects (these indicate that an electron-spin susceptibility along the $\mathbf{b}'$ axis, $\chi_{b'}$, at $T=0$ is equal to its value in a metallic state, χ_0, which is a distinct feature of triplet superconductivity[24,27]). (3) To stress that the experimental critical fields[20,21] along the conducting chains (i.e., along the $\mathbf{a}$ axis), H_p^a, are strongly paramagnetically limited and thus the corresponding electron-spin susceptibility $\chi_a \ll \chi_0$ at $T=0$. (4) To show that the above described properties are naturally explained within the framework of a triplet superconductivity scenario with the following vector order parameter frozen into the crystalline lattice (i.e., the case of strong spin-orbit coupling[27]):

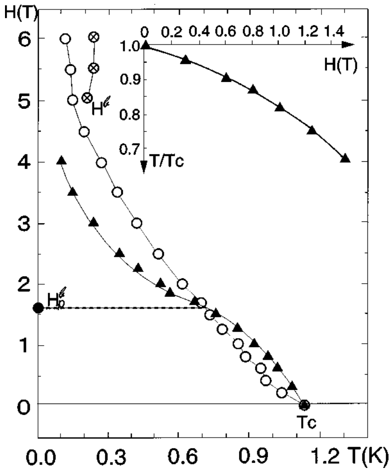

FIG. 1. Circles stand for the critical magnetic fields along the $\mathbf{b}'$ axis: open circles show an experimental curve (Refs. 20 and 21), a full circle corresponds to the calculated paramagnetically limited value of H_p^b at $T=0$ in a singlet superconductor, whereas crossed circles show the calculated nonparamagnetically limited critical fields $H^b(T)$ for a triplet order parameter (1). Triangles stand for the experimental critical fields (Refs. 20 and 21) along the $\mathbf{a}$ axis, $H_p^a(T)$, $(\mathbf{a}\perp\mathbf{b}')$. In the inset, the experimental values (Refs. 20 and 21) of $H_p^a(T)$ are shown in comparison with the calculated paramagnetically limited field (full line) for a triplet order parameter (1) (see the text).

$$\mathbf{d}(\mathbf{k})=(d_a(\mathbf{k})\neq 0,\quad d_c(\mathbf{k})=?,\quad d_{b'}(\mathbf{k})=0);$$
$$|d_a(\mathbf{k})|>|d_c(\mathbf{k})|, \tag{1}$$

corresponding to the BCS-pair's wave function

$$\Psi(\mathbf{k})=[-d_a(\mathbf{k})+id_c(\mathbf{k})]|\uparrow\uparrow\rangle+[d_a(\mathbf{k})+id_c(\mathbf{k})]|\downarrow\downarrow\rangle \tag{2}$$

and to the anisotropic spin susceptibility at $T=0$:

$$\chi_{b'}=\chi_0,\quad \chi_a\ll\chi_0, \tag{3}$$

where $|\uparrow\rangle$ ($|\downarrow\rangle$) stands for a spin-up (spin-down) electron with respect to the quantization axis $\mathbf{z}\|\mathbf{b}'$ $[\mathbf{a}(\mathbf{x})\perp\mathbf{b}'(\mathbf{z})\perp\mathbf{c}^*(\mathbf{y})]$ and the momentum $\mathbf{k}$ defines the position on the FS. [We stress that $\mathbf{b}'$ is the easy axis for a spin direction in a spin-density-wave (SDW) phase of $(TMTSF)_2PF_6$. Thus, one may expect that the order parameter (1) is the most stable since it corresponds to the BCS pairs (2) only with $S_{b'}\equiv S_z=\pm 1$.] At the end of the paper, we discuss some consequences of a group theory classification of the possible triplet phases, including the most probable orbital part of the order parameter and a possibility to break the time reversal symmetry.

The Q1D electron spectrum corresponds to two open sheets of the FS:[1,2]

$$\epsilon^{\pm}(\mathbf{p})=\pm v_F(p_a\mp p_F)-2t_b\cos(p_b b')-2t_c\cos(p_c c^*), \tag{4}$$

where + (−) stands for the right (left) sheet of the FS; $v_F=t_a a/\sqrt{2}$ and p_F are the Fermi velocity and Fermi momentum, respectively; $t_a\simeq 1600$ K, $t_b\simeq 200$ K, and $t_c\simeq 5$ K; $(\hbar=1)$.

Singlet $(S=0)$ and triplet $(S=1)$ phases are characterized by the following wave functions of the BCS pair's:[27]

$$\psi_s(\mathbf{k},\mathbf{r})=(|\uparrow\downarrow\rangle-|\downarrow\uparrow\rangle)\psi(\mathbf{k},\mathbf{r}),\quad S=0; \tag{5}$$

$$\psi_t(\mathbf{k},\mathbf{r})=|\uparrow\uparrow\rangle[-d_x(\mathbf{k},\mathbf{r})+id_y(\mathbf{k},\mathbf{r})]+(|\uparrow\downarrow\rangle+|\downarrow\uparrow\rangle)d_z(\mathbf{k},\mathbf{r})$$
$$+|\downarrow\downarrow\rangle[d_x(\mathbf{k},\mathbf{r})+id_y(\mathbf{k},\mathbf{r})],\quad S=1. \tag{6}$$

[In Eqs. (5) and (6), S is the total spin of the BCS pair, $\mathbf{r}$ is its coordinate of a center of masses; $\psi_s(\mathbf{k},\mathbf{r})=\psi_s(-\mathbf{k},\mathbf{r})$, $\mathbf{d}(\mathbf{k},\mathbf{r})=-\mathbf{d}(-\mathbf{k},\mathbf{r})$.]

At $H\to 0$, $\psi(\mathbf{k},\mathbf{r})$ and $\mathbf{d}(\mathbf{k},\mathbf{r})$ do not depend on $\mathbf{r}$. The electron-spin susceptibility tensor, $\chi_{i,j}$, at $T=0$ for a singlet phase is $\chi_{i,j}=0$ whereas for a triplet phase is given by[27]

$$\chi_{i,j}=\chi_0\left\langle \delta_{i,j}-\frac{d_i^*(\mathbf{k})d_j(\mathbf{k})}{\mathbf{d}^*(\mathbf{k})\mathbf{d}(\mathbf{k})}\right\rangle_{\mathbf{k}}, \tag{7}$$

where $\delta_{i,j}=1$ if $i=j$ and $\delta_{i,j}=0$ if $i\neq j$; $\langle|\mathbf{d}(\mathbf{k})|^2\rangle_{\mathbf{k}}=1$, $\langle\cdots\rangle_{\mathbf{k}}$ means an averaging over the FS. [Here, we consider only unitary triplet phases[27] (i.e., $d_a(\mathbf{k})d_c^*(\mathbf{k})=d_a^*(\mathbf{k})d_c(\mathbf{k})$.]

At first we consider the case $\mathbf{H}\|\mathbf{b}'(\mathbf{z})$. In singlet phase (5), superconductivity is destroyed by paramagnetic effects in arbitrary directed magnetic fields. In a triplet phase (6), as follows from Eq. (7), the $d_{b'}(\mathbf{k})\equiv d_z(\mathbf{k})$ component is responsible for the deviation of the spin susceptibility $\chi_{b'}\equiv\chi_{zz}$ from χ_0. If $d_{b'}(\mathbf{k})\neq 0$ there exist two related phenomena: the paramagnetic destructive mechanism against superconductivity and a change of the Knight shift at $T<T_c(H)$. Let us calculate the upper critical field for $\mathbf{H}\|\mathbf{b}'$. By using a common approach[28] to the upper critical field of a clean superconductor[25] with open electron orbits and with one-component order parameter, it is possible to prove that Eq. (5) of Ref. 28,

$$\Delta(x)=\frac{g}{2}\int_{|x-x_1|>d}\frac{2\pi T dx_1}{v_F\sinh(2\pi T|x-x_1|/v_F)}$$
$$\times J_0\left[\frac{2\alpha\mu_B H(x-x_1)S_z}{v_F}\right]$$
$$\times J_0\left(2\lambda\sin\left[\frac{\omega_c(x-x_1)}{2v_F}\right]\sin\left[\frac{\omega_c(x+x_1)}{2v_F}\right]\right)$$
$$\times\cos\left[\frac{2\mu_B H(x-x_1)S_z}{v_F}\right]\Delta(x_1), \tag{8}$$

is extended to a singlet phase $\psi_s(\mathbf{k},\mathbf{r})\equiv f(\mathbf{k})\Delta(x)$ as well as to the triplet phases $\mathbf{d}_1(\mathbf{k},\mathbf{r})\equiv(d_a=1,\ d_c=0,\ d_{b'}=0)f(\mathbf{k})\Delta(x)$ and $\mathbf{d}_2(\mathbf{k},\mathbf{r})\equiv(d_a=0,\ d_c=0,\ d_{b'}=1)f(\mathbf{k})\Delta(x)$. [Here, $\langle|f(\mathbf{k})|^2\rangle_{\mathbf{k}}=1$; g is an effective electron interaction constant, d is a cutoff distance; $\alpha=\sqrt{2}t_b/t_a$, $\omega_c=ev_F Hc^*/c$, $\lambda=4t_c/\omega_c$; μ_B is a Bohr magneton, e and c are the electron charge, and the velocity of light, correspondingly; $S_z=1$ for singlet and for $\mathbf{d}_2$-triplet phases whereas $S_z=0$ for $\mathbf{d}_1$-triplet phases. By solving Eq. (8) numerically for $S_z=1$, $\alpha=0.17$, $|dH^{b'}/dT|_{T_c}\simeq 2$ T/K, $v_F=10^7$ cm/sec, $t_c\simeq 3$ K, $T_c(0)=1.14$ K, $c^*=13.6$ Å (see Refs. 1, 2, 19–21, and 29), we found that the calculated value of the paramagnetic limited critical field, $H_p^{b'}\simeq 1.3$–1.4 T, is 4–5 times less than the experimental ones[20,21] (see Fig. 1). A similar analysis for the $\mathbf{d}_1$-triplet phase (which is not paramagnetically limited) shows that superconductivity survives at $H^{b'}\simeq 6$ T and $T\simeq 0.2$–0.25 K in qualitative agreement with experiments[20,21] (see Fig. 1). On the basis of the calculation of $H_p^{b'}$ and $H^{b'}$, we can conclude that $|d_{b'}(\mathbf{k})|\equiv|d_z(\mathbf{k})|\simeq 0$ in Eqs. (6), (7) and thus $\chi_{b'}\equiv\chi_{zz}\simeq\chi_0$. Note that the recent Knight shift measurements[12] are also in favor of $\chi_{b'}=\chi_0$ below $T_c(H)$.

If we consider the case $\mathbf{H}\|\mathbf{a}(\mathbf{x})$ then the $d_a\equiv d_x$ component of the order parameter (6) is responsible for the destructive paramagnetic effects against superconductivity and for the change of the Knight shift at $T<T_c(H)$ [see Eq. (7)]. Let us calculate the critical field for $\mathbf{H}\|\mathbf{a}$ in $\mathbf{d}_1(\mathbf{r})\equiv(d_a\neq 0,\ d_c\neq 0,\ d_{b'}=0)$ $\Delta(x)$ triplet phase (which is paramagnetically limited for such direction of a magnetic field). The corresponding linearized gap equation can be obtained from the common Eq. (5) of Ref. 28:

$$\Delta(x)=\frac{g}{2}\int_0^{2\pi}\frac{d\phi}{2\pi}\int_{|x-x_1|>\sqrt{2}d|\sin\phi|/\gamma}^{\infty}\Delta(x_1)\times\frac{\sqrt{2}\gamma\pi T dx_1}{v_F\sin\phi\,\sinh[\sqrt{2}\gamma\pi T|x-x_1|/v_F\sin\phi]}\times J_0\left(\frac{\sqrt{2}\lambda\gamma}{\sin\phi}\sin\left[\frac{\omega_c(x-x_1)}{2v_F}\right]\sin\left[\frac{\omega_c(x+x_1)}{2v_F}\right]\right)\times\cos\left[\frac{\sqrt{2}\gamma\mu_B H S_z(x-x_1)}{v_F\sin\phi}\right], \tag{9}$$

where $\gamma=t_a a/(2t_b b)$. Numerical solution of Eq. (9) [with the same values of parameters as Eq. (8)] shows that the best fitting of the data[20,21] at $H\leqslant 1.5$ T (see Fig. 1) corresponds to $S_z\simeq 0.9$ (i.e., $d_a\simeq 0.9$, $\chi_a\simeq 0.2\chi_0\ll\chi_0$) and $|dH^a/dT|_{T_c}\simeq 8$ T/K. The latter is in a good agreement with the experimental slopes[20,21] $|dH^{b'}/dT|_{T_c}\simeq 2$ T/K since the value of $t_b/t_a\simeq 8.5$ is known.[29] Note that the accuracy of our calculations does not allow us to distinguish between the triplet phases with $d_c=0$ and $|d_a|>|d_c|$.

Summarizing, our analysis of the experimental critical fields[20,21] measured in $(TMTSF)_2PF_6$ at $P\simeq 6$ kbar has shown that paramagnetic destructive effects against superconductivity do not affect $H^{b'}$ whereas H^a is paramagnetically limited at $H\leqslant 1.5$ T. These are naturally explained within a triplet scenario of superconductivity[9,16,19–21] with the triplet order parameter (1). We suggest to measure the Knight shift along the **a** axis at $H\leqslant 1.5$ T and $T<T_c(H)$ to prove the order parameter (1). Note that temperature dependence of the critical field along the **a** axis, $H^a(T)$, changes drastically[20,21] at $H\geqslant 1.5$ T. We speculate that at $H\geqslant 1.5$ T there may appear a triplet phase with $\mathbf{d}(\mathbf{k})\perp\mathbf{H}$, which minimizes the magnetic contribution to the free energy.[30] Nevertheless, we cannot completely exclude another possibility—the appearance of the LOFF state at $H\geqslant 1.5$ T for $\mathbf{H}\|\mathbf{a}$. Note that our theoretical analysis of the critical fields is based on the Fermi-liquid picture[29] proved at $P\simeq 6$ kbar in $(TMTSF)_2PF_6$. At higher pressures, $P\simeq 9.8$ kbar, the behavior of $(TMTSF)_2PF_6$ may deviate from the Fermi liquid one.[31]

TABLE I. Triplet order parameter $\hat{\Delta}(k)$ for D_{2h} and C_i groups.

Group	Representation	Order parameter $\hat{\Delta}(k)$
D_{2h}	A_{1u}	$Ak_x\hat{\tau}_x+Bk_y\hat{\tau}_y+Ck_z\hat{\tau}_z$
	B_{1u}	$Ak_x\hat{\tau}_y+Bk_y\hat{\tau}_x$
	B_{2u}	$Ak_x\hat{\tau}_z+Bk_z\hat{\tau}_x$
	B_{3u}	$Ak_y\hat{\tau}_z+Bk_z\hat{\tau}_y$
C_i	A_u	$Ak_x\hat{\tau}_x+Bk_y\hat{\tau}_y+Ck_z\hat{\tau}_z+Dk_y\hat{\tau}_x$ $+Fk_x\hat{\tau}_z+Gk_z\hat{\tau}_x+Hk_y\hat{\tau}_z+Ik_z\hat{\tau}_y$

At the end of the paper, we would like to make a few comments based on symmetry arguments. We classify the possible triplet phases in the case of strong spin-orbit coupling for orthorhombic (D_{2h}), and triclinic (C_i) point group symmetries (see Table I), where the matrix order parameter $\hat{\Delta}(\mathbf{k})=d_i(\mathbf{k})\hat{\tau}_i$, ($\hat{\tau}_i=i\hat{\sigma}_i\hat{\sigma}_y$; $\hat{\sigma}_i$ are the Pauli matrices). As is seen from Table I, there are no degenerated orbital states, thus a time reversal symmetry is broken only if a nonunitary triplet phase appears.[27] In our particular case, this happens when $d_a(\mathbf{k})d_c^*(\mathbf{k})\neq d_a^*(\mathbf{k})d_c(\mathbf{k})$. Using the expression for a gap in a quasiparticle spectrum,[27] $\delta(k)=|\mathbf{d}(\mathbf{k})|$ (the unitary case), it is possible to make sure that there are no generic phases with the lines of zeros on the FS in accordance with a common theorem.[32] This is in agreement with the experimental data[26,33] which seem to be in favor of fully gapped FS and against the existence of isolated zeros on the FS.[32] Therefore, we speculate that the orbital part of the order parameter is likely $d_a(\mathbf{k})\sim d_c(\mathbf{k})\sim\mathrm{sgn}(k_a)$, which corresponds to a fully gapped Q1D sheets of the FS. From Table I, it is possible to conclude that, for a triclinic space group of $(TMTSF)_2PF_6$, the most generic case is $d_a\neq 0$, $d_c\neq 0$, and $d_{b'}\neq 0$. However, it is known[1,2,34] that the spin-dependent interactions in a SDW phase of $(TMTSF)_2PF_6$ (which has a common boundary with the superconducting phase) result in an alignment of spins along the $\mathbf{b}'$ axis. Therefore, it is natural to expect the form (1) for the superconducting order parameter corresponding to the absence of the BCS pairs with $S_{b'}=0$ [see Eq. (2)].

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