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# Hidden reentrant superconducting phase in a magnetic field in $(TMTSF)_2ClO_4$

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We suggest explanation of the high upper critical magnetic field, perpendicular to conducting chains and parallel to conducting layers, $H^{b'}_{c2} \simeq 6$ T, experimentally observed in the organic superconductor $(TMTSF)_2ClO_4$. In particular, we show that $H^{b'}_{c2}$ can be higher than both the quasiclassical upper critical field and Clogston–Chandrasekhar paramagnetic limit in a singlet quasi-one-dimensional superconductor. We predict the coexistence of the hidden Reentrant and Larkin–Ovchinnikov–Fulde–Ferrell phases in a magnetic field. Our results are compared to the recent experimental data and shown to be in a good agreement with the experiments.

Since a discovery of superconductivity in $(TMTSF)_2PF_6$ material [1], superconducting properties of quasi-one-dimensional (Q1D) organic conductors $(TMTSF)_2X$ ($X = PF_6$, $ClO_4$, etc.) have been intensively studied both experimentally and theoretically [2, 3]. Early experiments [4, 5] clear demonstrated that superconductivity in these compounds was unconventional. Indeed, it was found that superconductivity was destroyed by non-magnetic impurities [4] and the Hebel–Slichter peak was absent in the NMR-experiments [5]. These experimental results have been recently unequivocally confirmed (see, for example, Refs. [6, 7]). Although the experimental results [4–7] provided strong arguments that the superconducting order parameter changed its sign on Q1D Fermi surface (FS), they did not contain information about a spin part of the order parameter. In particular, they did not distinguished between singlet and triplet superconducting pairings.

At the moment, the problem about a spin part of the superconducting order parameter in $(TMTSF)_2X$ conductors is still controversial. The first Knight shift measurements [7, 8], performed in $(TMTSF)_2PF_6$ material, were interpreted as evidence of triplet superconductivity [7, 8]. On the other hand, the more recent NMR-experiments [9], performed in $(TMTSF)_2ClO_4$ conductor, clear demonstrate the Knight shift change through the superconducting transition and, thus, are interpreted [9] in terms of singlet pairing. Another argument in favor of a singlet superconducting pairing was theoretical analysis [10] of experiment [11], demonstrating that the upper critical magnetic field along conducting chains, $H^a_{c2}$, is paramagnetically limited (see also Refs. [12–14]). Moreover, for a magnetic field, parallel to conducting chains, the Larkin–Ovchinnikov–Fulde–Ferrell (LOFF) phase has been recently experimentally discovered in $(TMTSF)_2ClO_4$ [12, 13] and theoretically described [15].

Another piece of information about a spin part of the superconducting order parameter is the fact that the upper critical fields, perpendicular to conducting chains and parallel to conducting layers, $H^{b'}_{c2}$, are anomalously large in $(TMTSF)_2X$ materials. They are known [10–13, 16–19] to exceed both the quasi-classical upper critical field, $H^{b'}_{c2}(0)$ [20], and Clogston–Chandrasekhar paramagnetic limit, $H_p$ [21]. For many years, this has been considered as one of the major arguments in favor of triplet superconductivity. Note that anomalously high values of $H^{b'}_{c2}$ were predicted for both singlet and triplet cases [22]. Nevertheless, it was shown that, for realistic band parameters of $(TMTST)_2X$ superconductors, the exceeding of values of $H^{b'}_{c2}(0)$ and $H_p$ happens only in a triplet case [22–26]. In this situation, where there is a growing support for a singlet scenario of superconductivity in $(TMTST)_2ClO_4$ material, it is important theoretically to reinvestigate the upper critical field $H^{b'}_{c2}$.

The main goals of the Letter are as follows. We derive novel gap equation for the case of $d$-wave nodeless superconducting pairing [27, 28]. It accurately takes into account not only the orbital effects but also the Pauli paramagnetic effects against superconductivity, unlike the previous gap equations of Refs. [10, 16, 22–26]. We evaluate band parameters of Q1D-electron spectrum of $(TMTSF)_2ClO_4$, using experiments on Lee–Naughton–Lebed oscillations [29, 30] and measured Ginzburg–Landau (GL) slopes of the upper critical fields [12, 13]. By solving the gap equation, we show that superconductivity in $(TMTSF)_2ClO_4$ can exceed both the quasi-classical upper critical field and Clogston–Chandrasekhar paramagnetic limit for realistic band parameters in a singlet case. Such superconducting phase, in the absence of the Pauli paramagnetic effects, has transition temperature increasing with increasing magnetic field (i.e., is the Reentrant superconductivity [22–26]). Therefore, we call high magnetic field superconducting phase in $(\mathrm{TMTSF})_2\mathrm{ClO}_4$ the hidden Reentrant superconductivity.

Below, we consider Q1D-electron spectrum of $(\mathrm{TMTSF})_2\mathrm{ClO}_4$ conductor in a tight-binding approximation,

$$\epsilon(\mathbf{p}) = -2t_a\cos(p_x a/2) - 2t_b\cos(p_y b') - 2t_c\cos(p_z c^*), \tag{1}$$

in a magnetic field, perpendicular to its conducting chains and parallel to its conducting layers,

$$\mathbf{H} = (0, H, 0), \quad \mathbf{A} = (0, 0, -Hx), \tag{2}$$

where $t_a \gg t_b \gg t_c$ correspond to electron hoping integrals along $\mathbf{a}$-, $\mathbf{b}'$-, and $\mathbf{c}^*$-axes, respectively. Electron spectrum (1) can be simplified near two slightly corrugated sheets of Q1D Fermi surface (FS) as

$$\delta\epsilon^{\pm}(\mathbf{p}) = \pm v_x(p_y)[p_x \mp p_{\mathrm{F}}(p_y)] - 2t_c\cos(p_z c^*), \tag{3}$$

where "+" ("−") stands for right (left) sheet of Q1D FS.

According to Ref. [22], electron spectrum (3) is "two-dimensionalized" in a magnetic field (2). More specifically, electrons are characterized by free unrestricted motion along $(\mathbf{a}, \mathbf{b}')$ conducting plane, whereas their motion along $\mathbf{z}$-axis is periodic and restricted [22]:

$$z(t, H) = l_\perp(H)\ c^*\cos(\omega_c t), \quad l_\perp(H) = 2t_c/\omega_c, \tag{4}$$

where $\omega_c = ev_{\mathrm{F}}Hc^*/c$, $v_{\mathrm{F}} = \langle v_x(p_y)\rangle$, $\langle ...\rangle$ stands for averaging procedure over momentum component $p_y$. Let us estimate a value of the dimensionless parameter $l_\perp(H)$, which represents a size of quasiclassical electron trajectory (4) in terms of interlayer distance, $c^*$. It is possible to show that

$$l_\perp(H) = \frac{2\sqrt{2}}{\pi}\frac{\phi_0}{ac^*H}\frac{t_c}{t_a} \simeq \frac{2\cdot 10^3}{H(T)}\frac{t_c}{t_b}\frac{t_b}{t_a}, \tag{5}$$

where $H(T)$ is a magnetic field measured in Teslas. Note that the ratios $t_a/t_b$ and $t_c/t_b$ can be determined in $(\mathrm{TMTSF})_2\mathrm{ClO}_4$ conductor with good accuracy. Indeed, according to Ref. [30], $t_a/t_b = 10$, whereas, according to Ref. [31], $t_c/t_b = (b^*/c^*)(H^c_{c2}/H^{b'}_{c2})_{\mathrm{GL}}$, where $(H^c_{c2}/H^{b'}_{c2})_{\mathrm{GL}}$ is a ratio of the GL-slopes of the upper critical fields along $c^*$- and $b'$-axes, correspondingly. If we take $(H^{b'}_{c2}/H^c_{c2})_{\mathrm{GL}} = 30$ and $H(T) = 6\,\mathrm{T}$, we obtain

$$l_\perp(H) \simeq 0.58, \tag{6}$$

which means that a size of electron quasiclassical trajectories along $\mathbf{c}^*$-axis (4) is significantly less than interlayer distance, $c^*$. In this case, electrons are almost two-dimensional and, therefore, such superconducting phase, in the absence of the Pauli paramagnetic effects, corresponds to the Reentrant superconductivity [22–26].

Let us represent electron wave functions in a real space in the following way:

$$\Psi^{\pm}_{\epsilon}(x, y, z, \sigma) = \exp[ip_{\mathrm{F}}(p_y)x]\exp(ip_y y)\exp(ip_z z)\ \psi^{\pm}_{\epsilon}(x, p_y, p_z, \sigma). \tag{7}$$

Then, in a magnetic field (2), the Hamiltonian for electron wave functions in a mixed representation, $\psi^{\pm}_{\epsilon}(x, p_y, p_z)$, can be obtained by means of the Peierls substitution method, $p_x \mp p_{\mathrm{F}}(p_y) \to -id/dx$, $p_z \to p_z - eA_z/c$. As a result, the Schrodinger like equation for wave functions $\psi^{\pm}(x, p_y, p_z, \sigma)$ can be written as

$$\left[\mp iv_x(p_y)\frac{d}{dx} - 2t_c\cos\left(p_z c^* + \frac{\omega_c}{v_{\mathrm{F}}}x\right) - \mu_{\mathrm{B}}\sigma H\right] \times \psi^{\pm}_{\epsilon}(x, p_y, p_z, \sigma) = \delta\epsilon\ \psi^{\pm}_{\epsilon}(p_x, y, p_z, \sigma), \tag{8}$$

where $\mu_{\mathrm{B}}$ is the Bohr magneton, $\sigma = \pm 1$ stands for spin up and down, respectively; $\delta\epsilon = \epsilon - \epsilon_{\mathrm{F}}$.

Note that Eq. (8) can be solved analytically,

$$\psi^{\pm}_{\epsilon}(x, p_y, p_z, \sigma) = \frac{\exp[\pm i\delta\epsilon x/v_x(p_y)]}{\sqrt{2\pi v_x(p_y)}}\exp\left[\pm i\frac{\mu_{\mathrm{B}}\sigma Hx}{v_x(p_y)}\right] \times \exp\left[\pm i\frac{2t_c}{v_x(p_y)}\int_0^x \cos\left(p_z c^* + \frac{\omega_c}{v_{\mathrm{F}}}u\right)du\right], \tag{9}$$

and the corresponding finite temperatures Green functions can be derived from Eq. (9), using the standard procedure [32]:

$$\begin{aligned} g^{\pm}_{i\omega_n}(x, x_1, p_y, p_z, \sigma) = \\ = -i\frac{\mathrm{sgn}(\omega_n)}{v_x(p_y)}\exp\left[\mp\frac{\omega_n(x - x_1)}{v_x(p_y)}\right] \times \\ \times \exp\left[\pm i\frac{\mu_{\mathrm{B}}\sigma H(x - x_1)}{v_x(p_y)}\right] \times \\ \times \exp\left[\pm i\frac{2t_c}{v_x(p_y)}\int_{x_1}^x \cos\left(p_z c^* + \frac{\omega_c}{v_{\mathrm{F}}}u\right)du\right]. \end{aligned} \tag{10}$$

In this Letter, we consider d-wave nodeless scenario of superconductivity in $(\mathrm{TMTSF})_2\mathrm{ClO}_4$ conductor [27, 28], which is mathematically equivalent to s-wave case. We derive the so-called gap equation for superconducting order parameter, $\Delta(x)$, using the known Green functions (10). Below, it is derived by means of the Gor'kov equations [32] for non-uniform superconductivity (see, for example, Ref. [33]). As a result of rather lengthly but straightforward calculations, we obtain:

$$\Delta(x) = U \int \frac{dp_y}{v_x(p_y)} \times \\ \times \int_{|x-x_1|>\frac{v_x(p_y)}{\Omega}} \frac{2\pi T dx_1}{v_x(p_y)\sinh(\frac{2\pi T|x-x_1|}{v_x(p_y)})} \times \\ \times J_0\left\{\frac{8t_c v_F}{\omega_c v_x(p_y)} \sin\left[\frac{\omega_c(x-x_1)}{2v_F}\right] \sin\left[\frac{\omega_c(x+x_1)}{2v_F}\right]\right\}\times \\ \times \cos\left[\frac{2\beta\mu_B H(x-x_1)}{v_x(p_y)}\right] \Delta(x_1), \tag{11}$$

where $U$ is effective electron coupling constant, $\Omega$ is a cutoff energy, parameter $\beta$ takes into account possible small deviations of superconducting pairing from a weak coupling scenario. It is important that Eq. (11) is different from the gap equations used so far. The main feature of Eq. (11) is that, unlike Refs. [22–26, 10, 16], it describes accurately not only the orbital effects against superconductivity, but also the Pauli paramagnetic ones.

We note that, in the absence of effects of a non-linearity of Q1D-electron spectrum (1), (3) along **a**-axis, the upper critical field along **b**-axis is divergent at $T \to 0$ [22–25]. These effects are taken into account in Eq. (11) of the Letter, since $v_x(p_y)$ depends on a position on the FS [see also Eq. (14)]. The effects of a non-linearity of Q1D electron spectrum along **a** axis were first considered in Ref. [16] to explain finite experimental values of the upper critical magnetic fields along **b** axis in $(TMTSF)_2X$ superconductors. Nevertheless, in Refs. [16, 26], these efects were calculated in the so-called nesting model (NM), where $p_F(p_y)$ depends on momentum component $p_y$, whereas velocity, $v_x = v_F$, does not depend on $p_y$. Our current analysis shows that the NM is approximately valid at low temperatures, whereas Eq. (11) is valid at any temperatures. We also pay attention that even at $T = 0$ Eqs. (13), (15), (17), obtained in the Letter, are different from the corresponding approximate equations of the NM [16, 26].

Note that it is possible to approximate the Bessel function in Eq. (11) as $J_0(z) \simeq 1 - z^2/4$ under condition (6) and, thus, below, we consider the above mentioned approximation for integral equation (11) [34]. In this case, it is possible to show that solution for a superconducting gap, $\Delta(x)$, can be written as

$$\Delta(x) = \exp(ikx)[1 + \alpha_1 \cos(2\omega_c x/v_F) + \alpha_2 \sin(2\omega_c x/v_F)], \tag{12}$$

where $|\alpha_1|, |\alpha_2| \ll 1$. Eq. (11), determining the upper critical field, in this case and at $T = 0$ can be expressed as

$$\frac{1}{g} = \left\langle \frac{v_F}{v_x(p_y)} \int_{\frac{v_F}{\Omega}}^{\infty} \frac{dz}{z} \cos\left(\frac{2\beta\mu_B H z}{v_F}\right) \times \\ \times \left[1 - 2l_\perp^2(H) \sin^2\left(\frac{\omega_c z}{2v_F}\right)\right] \cos\left[\frac{v_x(p_y)}{v_F} kz\right] \right\rangle_{p_y}, \tag{13}$$

where $g$ is renormalized electron coupling constant, $x_1 - x = z v_x(p_y)/v_F$.

Let us simplify Eq. (13), taking into account that electron velocity component along conducting **x**-axis is

$$v_x(p_y) = v_F[1 + \alpha\cos(p_y b')], \tag{14}$$

where $\alpha = \sqrt{2} t_b/t_a \simeq 0.14$ [16]. In this case, Eq. (13) for $\alpha \ll 1$ can be rewritten in the following way:

$$\frac{1}{g} = \int_{\frac{v_F}{\Omega}}^{\infty} \frac{dz}{z} \cos\left(\frac{2\beta\mu_B H z}{v_F}\right) \cos(kz) J_0(\alpha k z) \times \\ \times \left[1 - 2l_\perp^2(H)\sin^2\left(\frac{\omega_c z}{2v_F}\right)\right]. \tag{15}$$

We stress that Eq. (15) accurately takes in to account not only the orbital effects but also the Pauli paramagnetic effects against superconductivity, unlike Refs. [10, 16, 22–26]. In the absence of the Pauli paramagnetic effects (i.e., at $\beta = 0$), Eq. (15) describes the Reentrant superconducting phase [22] with $dT_c/dH > 0$. Therefore, we call superconducting phase, described by Eqs. (15), (17), the hidden Reentrant superconductivity.

Here, we transform Eq. (15), using that

$$\frac{1}{g} = \int_{\frac{v_F}{\Omega}}^{\infty} \frac{2\pi T_c dz}{v_F \sinh(2\pi T_c z/v_F)}, \tag{16}$$

where $T_c$ is superconducting transition temperature at $H = 0$. As a result, we obtain:

$$\ln\frac{H}{H^*} = \int_0^{\infty} \frac{dz}{z} \cos\left(\frac{2\beta\mu_B H z}{v_F}\right) \left\{\cos(kz) J_0(\alpha k z) \times \\ \times \left[1 - 2l_\perp^2(H)\sin^2\left(\frac{\omega_c z}{2v_F}\right)\right] - 1\right\}. \tag{17}$$

(Here, $\mu_B H^* = \pi T_c/2\gamma$, $\gamma$ is the Euler constant.) Numerical analysis of Eq. (17) shows that the upper critical field along $b'$-axis, $H_{c2}^{b'}$, for $l_\perp(H) = 0.58$ and $\beta = 0.72$ has a maximum at $k = 0.88(2\mu_B H/v_F)$ and is equal to

$$H_{c2}^{b'} \simeq 5 \text{ T}. \tag{18}$$

For the same values of the parameters $l_\perp(H)$ and $\beta$, numerical analysis of Eq. (11) gives the following results for factors $\alpha_1$ and $\alpha_2$ in Eq. (12):

$$\alpha_1 = -0.099, \quad \alpha_2 = -0.018\, i. \tag{19}$$

Let us summarize the main results of the Letter. We have derived gap equations for a singlet $d$-wave nodeless superconducting order parameter, which take accurately into account both the orbital and Pauli paramagnetic effects against superconductivity. By analyzing the above mentioned equations, we have explained how superconductivity in $(TMTSF)_2ClO_4$ can exceed both the quasiclassical upper critical field and Clogston paramagnetic limit and how it can reach its experimental value, $H \simeq 5{-}6$ T [12, 13, 19].

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34. Note that the corresponding gap equation for a magnetic field, applied along **a**-axis [15], contains extra factor $v_x(p_y)/|v_y(p_y)| \gg 1$ in an argument of the Bessel function. Therefore, the Bessel function argument in Eq. (13) of Ref. [15] is large in experimental magnetic fields, $H \simeq 5$ T, which results in qualitatively different behavior of the upper critical magnetic field along **a**-axis. From physical point of view, large value of argument in the Bessel function in Eq. (13) of Ref. [15] corresponds to large sizes of the quasi-classical electron orbits in a magnetic field. In this case, electrons can be considered as three-dimensional ones, in contrast to our case, and, thus, the Reentrant superconductivity effects are absent at $\mathbf{H} \parallel \mathbf{a}$ [15].