Hidden Reentrant and Larkin-Ovchinnikov-Fulde-Ferrell Superconducting Phases in a Magnetic Field in a $(TMTSF)_2ClO_4$

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We solve a long-standing problem about a theoretical description of the upper critical magnetic field, parallel to conducting layers and perpendicular to conducting chains, in a $(TMTSF)_2ClO_4$ superconductor. In particular, we explain why the experimental upper critical field, $H^{b'}_{c2} \simeq 6$ T, is higher than both the quasiclassical upper critical field and the Clogston paramagnetic limit. We show that this property is due to the coexistence of the hidden reentrant and Larkin-Ovchinnikov-Fulde-Ferrell phases in a magnetic field in the form of three plane waves with nonzero momenta of the Cooper pairs. Our results are in good qualitative and quantitative agreement with the recent experimental measurements of $H^{b'}_{c2}$ and support a singlet d-wave-like scenario of superconductivity in $(TMTSF)_2ClO_4$.

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The physical properties of quasi-one-dimensional (Q1D) organic conductors $(TMTSF)_2X$ ($X = PF_6$, ClO_4, ReO_4, etc.) have been intensively studied [1,2] since the discovery of superconductivity in $(TMTSF)_2PF_6$ [3]. Early experiments [4,5] clearly showed that superconducting phases in these compounds were unconventional and that the corresponding order parameters changed their signs on Q1D Fermi surfaces (FS). In particular, it was shown that the Hebel-Slichter peak was absent in the NMR experiment [4] and superconductivity was destroyed by nonmagnetic impurities [5]. These results have been recently confirmed in a number of publications (see, for example, Refs. [6,7]). The first Knight shift measurements [7,8], performed in a $(TMTSF)_2PF_6$ conductor in a magnetic field $H = 1.43$ T, showed that the Knight shift was unchanged in the superconducting phase and were interpreted as evidence for triplet superconductivity. On the other hand, more recent Knight shift data [9], performed in a $(TMTSF)_2ClO_4$ conductor, clearly demonstrate the Knight shift change through the superconducting transition in a magnetic field $H = 0.957$ T. They are interpreted [9] in terms of singlet pairing in superconductor $(TMTSF)_2ClO_4$ at least at relatively weak magnetic fields.

Another source of information about a spin part of the superconducting order parameter was provided by the fact that the experimental upper critical magnetic field along conducting chains, H^a_{c2} [10], was clearly paramagnetically limited [11]. This has been recently confirmed in Refs. [12–14]. In addition, a new superconducting phase has been discovered in $(TMTSF)_2ClO_4$ [12,13] for a magnetic field, parallel to conducting chains. The suggested hypothesis [12,13] that it can be the Larkin-Ovchinnikov-Fulde-Ferrell (LOFF) phase [15,16] has recently been theoretically supported [17]. Note that the above-mentioned experimental and theoretical works are in favor of a singlet d-wave-like scenario of superconductivity in a $(TMTST)_2ClO_4$ [14,17–19].

In this situation, where support for a singlet d-wave-like scenario of superconducting pairing in a $(TMTST)_2ClO_4$ conductor is growing, it is important to theoretically reinvestigate the upper critical field, parallel to conducting layers and perpendicular to conducting chains, $H^{b'}_{c2}$. For many years, large experimental values of $H^{b'}_{c2}$ [10,12,13,20–23], which exceeds both the quasiclassical upper critical field $H^{b'}_{c2}(0)$ [24] and the Clogston paramagnetic limit H_p [25], have been considered as one of the main arguments in favor of triplet superconductivity. Although the exceeding of the values of $H^{b'}_{c2}(0)$ and H_p was predicted for $H^{b'}_{c2}$ in both singlet and triplet cases [26–30], it was also shown that, for realistic band parameters of $(TMTST)_2X$ conductors, it can happen only in a triplet case [11,20,26–30].

The goal of our Letter is to demonstrate that superconductivity in a $(TMTSF)_2ClO_4$ can exceed both critical magnetic fields $H^{b'}_{c2}(0) \simeq 3.5$ T and $H_p \simeq 2.7$ T and reach its experimental value, $H^{b'}_{c2} \simeq 6$ T [12,13,23], even in the case of a singlet d-wave-like superconducting pairing. Our first point is that the Pauli paramagnetic effects in all previous theories [11,20,26–30] were not treated completely correctly. Our second point is that the $3D \rightarrow 2D$ dimensional crossover [26] happens at magnetic fields $H^{b'} \simeq 5$–6 T, which are much lower than previously assumed. The latter statement is shown to result from theoretical analysis of both the Ginzburg-Landau (GL) slopes, $dH^{b'}_{c2}/dT|_{T=T_c}$ and $dH^{c}_{c2}/dT|_{T=T_c}$, measured in Refs. [12,13,23], and the so-called Lee-Naughton-Lebed oscillations [31,32]. In this Letter, we derive a novel gap equation, which accurately treats both the Pauli paramagnetic and the orbital destructive effects against superconductivity. By analyzing this equation, we show that it predicts the upper critical field, $H^{b'}_{c2} \simeq 6$ T, for real values of band parameters in a $(TMTSF)_2ClO_4$. The superconducting phase, which exists at such high magnetic fields, is shown to be very peculiar. It is characterized by an inhomogeneous order parameter in the form of the three LOFF-like waves, which appear due to both the 3D → 2D dimensional crossover and the Pauli paramagnetic effects. It is important that this phase is characterized by the Cooper pairs, localized on conducting layers, with the probability of the Cooper pair jumping from one layer to another being small. Therefore, it is not destroyed by the orbital effects in a parallel magnetic field. In the absence of the Pauli paramagnetic effects, such a phase would correspond to the reentrant superconductivity with $dT_c/dH > 0$; therefore, we call it the hidden reentrant superconducting phase.

Below we consider a tight-binding orthorhombic model of an anisotropic Q1D electron spectrum in a $(TMTSF)_2ClO_4$ conductor,

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

which can be simplified near two slightly corrugated sheets of Q1D FS as

$$\delta\epsilon^{\pm}(\mathbf{p}) = \pm v_x(p_y)[p_x \mp p_F(p_y)] - 2t_c \cos(p_z c^*). \tag{2}$$

[Here $t_a \gg t_b \gg t_c$ correspond to electron hoping integrals along $\mathbf{a}$, $\mathbf{b}'$, and $\mathbf{c}^*$ axes, respectively, and $+(-)$ stands for the right (left) sheet of the FS.]

In a magnetic field, parallel to conducting planes and perpendicular to conducting chains of a Q1D conductor,

$$\mathbf{H} = (0, H, 0), \qquad \mathbf{A} = (0, 0, -Hx), \tag{3}$$

we use the so-called Peierls substitution method, $p_x \mp p_F(p_y) \to -id/dx$, $p_z \to p_z - eA_z/c$. As a result, the effective Schrödinger equation for electron wave functions in a mixed representation, $\psi^{\pm}(x, p_y, p_z, \sigma)$, can be written as

$$\left[\mp i v_x(p_y)\frac{d}{dx} - 2t_c \cos\left(p_z c^* + \frac{\omega_c}{v_F}x\right) - \mu_B \sigma H\right] \times \psi^{\pm}_{\epsilon}(x, p_y, p_z, \sigma) = \delta\epsilon\, \psi^{\pm}_{\epsilon}(p_x, p_y, p_z, \sigma), \tag{4}$$

with the electron wave functions in a real space function being

$$\Psi^{\pm}_{\epsilon}(x, y, z, \sigma) = \exp[i p_F(p_y)x]\exp(i p_y y) \times \exp(i p_z z)\psi^{\pm}_{\epsilon}(x, p_y, p_z, \sigma), \tag{5}$$

where $\omega_c = e v_F H c^*/c$, μ_B is the Bohr magneton, and $\sigma = \pm 1$ stands for spin-up and -down, respectively.

It is important that Eq. (4) can be analytically solved:

$$\psi^{\pm}_{\epsilon}(x, p_y, p_z, \sigma) = \frac{\exp[\pm i\delta\epsilon x/v_x(p_y)]}{\sqrt{2\pi v_x(p_y)}} \exp\left[\pm i\frac{\mu_B \sigma H x}{v_x(p_y)}\right] \times \exp\left[\pm i \frac{2t_c}{v_x(p_y)} \int_0^x \cos\left(p_z c^* + \frac{\omega_c}{v_F}u\right)du\right], \tag{6}$$

where wave functions (6) are normalized on $\delta(\epsilon_1 - \epsilon_2)$, $\delta\epsilon = \epsilon - \epsilon_F$. The corresponding finite temperature Green functions can be derived from Eq. (6) by means of the standard procedure [33]:

$$g^{\pm}_{i\omega_n}(x, x_1, p_y, p_z, \sigma) = -i\frac{\text{sgn}(\omega_n)}{v_x(p_y)}\exp\left[\mp\frac{\omega_n(x - x_1)}{v_x(p_y)}\right]\exp\left[\pm i\frac{\mu_B \sigma H(x - x_1)}{v_x(p_y)}\right] \times \exp\left[\pm i\frac{2t_c}{v_x(p_y)}\int_{x_1}^x \cos\left(p_z c^* + \frac{\omega_c}{v_F}u\right)du\right]. \tag{7}$$

[Note that, in contrast to previous works [11,20,26–30], Eqs. (6) and (7) take into account the dependence of electron velocity along conducting chains, $v_x(p_y)$, on a momentum component p_y. As shown below, it allows us to accurately describe the Pauli paramagnetic destructive effects against superconductivity.]

In this Letter, we consider a singlet d-wave-like scenario of superconductivity in a $(TMTSF)_2ClO_4$ conductor [14,17–19], which is consistent with all available experimental data. Therefore, we introduce the following superconducting order parameter:

$$\Delta(p_y, x) = \sqrt{2}\cos(p_y b')\Delta(x), \tag{8}$$

where the first term, $\sqrt{2}\cos(p_y b')$, is responsible for the existence of zeros on Q1D FS, whereas the second term describes both the orbital effects against superconductivity and possible LOFF-like phase formation. Below, we derive a so-called gap equation for the superconducting order parameter (8), using the Green functions (7). It is derived by means of the Gor'kov equations [33] for nonuniform superconductivity (see, for example, Refs. [34–36]. As a result of rather lengthy calculations, we obtain

$$\Delta(x) = g' \int \frac{dp_y}{v_x(p_y)} \int_{|x - x_1| > v_x(p_y)/\Omega} \frac{2\pi T dx_1}{v_x(p_y)\sinh[2\pi T|x - x_1|/v_x(p_y)]} J_0\left\{\frac{8t_c v_F}{\omega_c v_x(p_y)}\sin\left[\frac{\omega_c(x - x_1)}{2v_F}\right] \times \sin\left[\frac{\omega_c(x + x_1)}{2v_F}\right]\right\} 2\cos^2(p_y b')\cos\left[\frac{2\beta\mu_B H(x - x_1)}{v_x(p_y)}\right]\Delta(x_1), \tag{9}$$

where g' stands for electron coupling constant, Ω is a cutoff energy, parameter β takes into account possible deviation of the so-called electron g factor, $g = 2\beta$, from the value $g = 2$ [37]. We stress that Eq. (9) is different from the gap equations used so far, and, unlike Refs. [11,20,26–30], it describes accurately not only the orbital effects but also the Pauli paramagnetic ones. Note that Eq. (9) is based on a quantum-mechanical treatment of electron motion both parallel and perpendicular to conducting layers directions. It is the most general gap equation, which can be written for the Q1D conductor (2) in a magnetic field (3). It is possible to show that the major quantum parameter in Eq. (9) is $2t_c v_F/\omega_c v_x(p_y) \simeq 2t_c/\omega_c$. It is also possible to prove that in low magnetic fields, where $2t_c/\omega_c \gg 1$ and $(T_c - T)/T_c \ll 1$, Eq. (9) is reduced to the well-known GL equation [38].

Let us estimate a value of the dimensionless quantum parameter $l_\perp(H) = 2t_c/\omega_c$ in Eq. (9), which, using classical language, represents a size of electron trajectory along the $\mathbf{z}$ axis in terms of interlayer distance [26]:

$$z(t, H) = l_\perp(H) c^* \cos(\omega_c t), \tag{10}$$

where t is time. It is easy to show that

$$l_\perp(H) = \frac{2\sqrt{2}}{\pi}\,\frac{\phi_0}{ac^* H}\,\frac{t_c}{t_a} \simeq \frac{2\times 10^3}{H(T)}\,\frac{t_c}{t_b}\,\frac{t_b}{t_a}, \tag{11}$$

where $H(T)$ is a magnetic field, measured in teslas. Here, according to Ref. [32], $t_a/t_b = 10$ and, according to Ref. [38], $t_c/t_b = (b^*/\sqrt{2}c^*)(H^c_{c2}/H^{b'}_{c2})_{\mathrm{GL}}$ with $(H^c_{c2}/H^{b'}_{c2})_{\mathrm{GL}}$ being a ratio of the GL slopes of the upper critical fields along the c^* and b' axes, correspondingly [39]. Note that the ratios $t_a/t_b = 10$ [31] and $(H^{b'}_{c2}/H^c_{c2})_{\mathrm{GL}} = 26$ [12,13] are very well measured in a $(\mathrm{TMTSF})_2\mathrm{ClO}_4$ conductor. If we take $H(T) = 6$ T, we obtain

$$l_\perp(H = 6\ \mathrm{T}) \simeq 0.48, \tag{12}$$

which means that a size of the electron classical trajectory along the $\mathbf{c}^*$ axis (10) is significantly less than the interlayer distance, c^*. In this case, which corresponds to the 3D $\to$ 2D dimensional crossover of electron motion in a magnetic field [26,40], it is possible to make sure directly from Eq. (9) that we can approximate the Bessel function as $J_0(z) \simeq 1 - z^2/4$.

Let us consider the above-mentioned approximation for the integral equation (9) at zero temperature, $T = 0$. It is possible to show that the solution for a superconducting gap, $\Delta(x)$, in this case can be written as

$$\Delta(x) = \exp(ikx)[1 + \alpha_1\cos(2\omega_c x/v_F) + \alpha_2\sin(2\omega_c x/v_F)], \tag{13}$$

where $|\alpha_1|, |\alpha_2| \ll 1$. Equation (9), which determines the upper critical field, in the same approximation and at $T = 0$ can be expressed as

$$\frac{1}{\tilde{g}} = \int_0^{2\pi/b'} \frac{dp_y b'}{2\pi} \int_{v_F/\Omega}^\infty \frac{dz}{z} 2\cos^2(p_y b')\cos\left(\frac{2\beta\mu_b H z}{v_F}\right) \times \frac{v_F}{v_x(p_y)}\left[1 - 2l_\perp^2(H)\sin^2\left(\frac{\omega_c z}{2v_F}\right)\right]\cos\left[\frac{v_x(p_y)}{v_F}kz\right], \tag{14}$$

where $\tilde{g}$ is the renormalized electron coupling constant, $x_1 - x = zv_x(p_y)/v_F$. [Note that we set $\alpha_1 = \alpha_2 = 0$ in Eq. (14), since we disregard all contributions of the order of $l_\perp^4(H) \ll l_\perp^2(H)$ to the upper critical field.]

Below, we simplify Eq. (14), taking into account that the electron velocity component along the conducting $\mathbf{x}$ axis is

$$v_x(p_y) = v_F[1 + \alpha\cos(p_y b')], \tag{15}$$

where $\alpha = \sqrt{2}t_b/t_a \simeq 0.14$ [20]. More specifically, Eq. (14) for $\alpha \ll 1$ can be written as follows:

$$\frac{1}{\tilde{g}} = \int_{v_F/\Omega}^\infty \frac{dz}{z}\cos\left(\frac{2\beta\mu_B H z}{v_F}\right)\cos(kz)[J_0(\alpha kz) - J_2(\alpha kz)]\left[1 - 2l_\perp^2(H)\sin^2\left(\frac{\omega_c z}{2v_F}\right)\right]. \tag{16}$$

It is important that Eq. (16) accurately takes into account the Pauli paramagnetic effects against superconductivity, unlike Refs. [11,20,26–30]. Note that, in the absence of the Pauli paramagnetic effects (i.e., at $\beta = 0$), Eq. (16) describes the reentrant superconducting phase [26] with $dT_c/dH > 0$. Therefore, we call the superconducting phase, described by Eqs. (16) and (18), the hidden reentrant superconductivity.

Let us further simplify Eq. (16) by taking into account the fact that

$$\frac{1}{\tilde{g}} = \int_{v_F/\Omega}^\infty \frac{2\pi T_c dz}{v_F\sinh(2\pi T_c z/v_F)}, \tag{17}$$

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

$$\ln\left(\frac{H}{H^*}\right) = \int_0^\infty \frac{dz}{z}\cos\left(\frac{2\beta\mu_B H z}{v_F}\right)\left\{\cos(kz)[J_0(\alpha kz) - J_2(\alpha kz)]\left[1 - 2l_\perp^2(H)\sin^2\left(\frac{\omega_c z}{2v_F}\right)\right] - 1\right\}, \tag{18}$$

where $\mu_B H^* = \pi T_c/2\gamma$, γ is the Euler constant. Numerical analysis of Eq. (18) shows that the upper critical field along the $\mathbf{b}'$ axis, $H^{b'}_{c2}$, for $l_\perp(H) = 0.48$ and $\beta = 0.84$ has a maximum at $k = 0.88(2\beta\mu_B H/v_F)$ and is equal to

$$H^{b'}_{c2} \simeq 5.9\ \mathrm{T}. \tag{19}$$

[We pay attention to the fact that the obtained value of the upper critical field (19) well corresponds to the value of a magnetic field (12).] For the same values of the parameters $l_\perp(H)$ and β, numerical analysis of Eq. (9) gives the following values for factors α_1 and α_2 in Eq. (13):

$$\alpha_1 = -0.139, \qquad \alpha_2 = 0.021i. \tag{20}$$

Below we summarize the main results of the Letter. We have derived gap equations (9), (14), (16), and (18), which, unlike gap equations in the previous publications, take accurately into account not only the orbital effects but also the Pauli paramagnetic effects against superconductivity. We have analyzed the experimental data [12,13,32] and shown that, in contrast to the common belief, the quantum effects of electron motion in a magnetic field [26,41] are strong in relatively weak magnetic fields of the order of 5–6 T in a $(TMTSF)_2ClO_4$ conductor. By analyzing the above-mentioned gap equations, we have explained how superconductivity in a $(TMTSF)_2ClO_4$ can exceed both the quasiclassical upper critical field [24] and the Clogston paramagnetic limit [25] and how it can reach its experimental value, $H \simeq 6$ T [12,13]. We have shown that, due to the reentrant quantum effects [26,41], superconductivity survives in the form of the hidden reentrant superconducting phase, corresponding to three LOFF-like phases. Although we have not calculated in this Letter the phase diagram of the $(TMTSF)_2ClO_4$ superconductor in all ranges of temperatures and magnetic fields, we anticipate the existence of a phase transition between the BCS and LOFF phases at $H \simeq 2.5$ T, which can be experimentally studied.

In conclusion, we note that the above-considered hidden reentrant superconductivity is a rather general phenomenon. It is expected to exist in other $(TMTSF)_2X$ conductors and may exist in quasi-two-dimensional superconductors in a parallel magnetic field. Nevertheless, this phase in $(TMTSF)_2PF_6$ material, which is stable in a mixed superconducting-spin-density-wave state [21,22,42] in a magnetic field up to $H = 9$ T, possesses some peculiarities. Our preliminary analysis shows that, to describe the hidden reentrant superconducting phase in $(TMTSF)_2PF_6$, it is necessary to take into account some additional effects such as the singlet-triplet mixing phenomenon [43] or possible singlet-triplet phase transition (see, for example, [20,30]).

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