New Type of Quantum Magnetic Resistance Oscillations in Quasi-One-Dimensional Conductors

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We suggest an explanation for the "rapid magnetic oscillations" (RMO) in the metallic phase of the organic superconductor $(TMTSF)_2ClO_4$. It is based on the possibility of quantum magnetic oscillations in the probability of electron-electron scattering and thus exceeds the bounds of the standard description of the magnetic oscillations in metals. The calculated RMO frequency is in agreement with experimental data. The estimated temperature and magnetic field dependences of RMO magnitude are unusual.

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The quasi-one-dimensional (Q1D) family of organic superconductors $(TMTSF)_2X$ (where $X = ClO_4$, PF_6, ReO_4, etc.) demonstrates unique properties both in a field-induced spin-density-wave (FISDW) state and a metallic one in a magnetic field perpendicular to the chains $\mathbf{H} \perp \mathbf{x}$ (for review, see [1]). Most features of the FISDW state, particularly, those of $(TMTSF)_2PF_6$, are well explained by the "standard model" [2,3]. This model utilizes the concept of "one dimensionalization" of the electron spectrum in a magnetic field [2]. Unusual properties in a metallic state, such as "magic angle" effects [4,5] and the unexpected observation of a torque in these metals with open Fermi surfaces (FS) [5], can also be attributed to the change of the effective dimensionality of the electron-electron ("*e-e*") interactions in strong enough magnetic fields $H_\perp \geq 5$ T [6–11].

The organic conductor $(TMTSF)_2ClO_4$ exhibits some new experimental features. One of the most interesting is the so-called "rapid magnetic oscillations" (RMO) of resistivity. In the $(TMTSF)_2ClO_4$ salt RMO exist both in the metallic state and FISDW one [12–16], whereas in the $(TMTSF)_2PF_6$ compound weak oscillations were found only in the SDW phase by Ulmet *et al.* [17,18].

The RMO phenomenon in a metallic phase was discovered by Chaikin *et al.* in 1983 [13] but its observation still has no theoretical explanation [19]. Unlike the $(TMTSF)_2PF_6$ salt, the $(TMTSF)_2ClO_4$ compound possesses an anion ordering (AO) of ClO_4^- ions at temperatures $T < T_{AO} \simeq 24$ K [1]. The AO creates a gap $\square$ in the electron spectrum, which, in spite of the gap, corresponds to the open sheets of the FS (see Fig. 1). It has become clear that the RMO are strongly dependent on the AO [14–16].

Yan *et al.* [12] first pointed out that the magnetic breakdown (MB) across the AO gap in $(TMTSF)_2ClO_4$ could provide oscillatory effects in the same way as the Stark interferometer [20]. This approach to RMO seems to be correct for resistivity perpendicular to the chains.

In this Letter we stress that the strong RMO of the resistivity along the chains $\delta\rho_{xx}/\rho_{xx} \geq 10^{-1}$ [12,13,15,16] in the metallic phase of the $(TMTSF)_2ClO_4$ should represent a new type of magnetic oscillation. It is easy to make sure that the description of ρ_{xx} in the framework of the Stark interferometer [20] leads to negligible magnitudes of the oscillations in the Q1D case, $\delta\rho_{xx}/\rho_{xx} \sim \rho_{xx}/\rho_{yy} \simeq 10^{-3}$. As it follows from present calculations, the reason the theory [20] meets with failure, while describing Q1D materials, is the neglect of the oscillations in the electron relaxation time τ.

We show that the effect of one dimensionalization of an electron spectrum under the condition of MB across the AO gap results in strong oscillatory behavior of the matrix elements of *e-e* scattering due to the quantum interference phenomena. We suggest that the magnetic oscillations of the probability of *e-e* scattering are responsible for strong RMO in the longitudinal component of resistivity, ρ_{xx}.

The Q1D electron spectrum of the $(TMTSF)_2Cl_4$ salt in the absence of the AO gap takes the form

$$\epsilon^{\pm}(\mathbf{p}) = \pm v_F(p_x \mp p_F) + 2t_b \cos(p_y b) + 2t_c \cos(p_z c^*). \quad (1)$$

In Eq. (1) the first term represents free motion of electrons along the chains in the right (+) and left (−) sheets of the FS, with p_F and v_F the Fermi momentum and the Fermi velocity, correspondingly. The second and third terms correspond to the hopping of electrons in

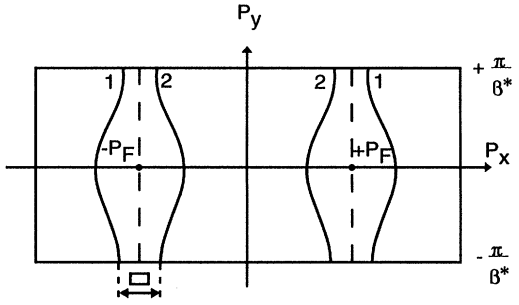

FIG. 1. Electron spectrum of the $(TMTSF)_2ClO_4$ salt in the presence of anion gap $\square$.

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the perpendicular directions **b** and **c*** ($t_b \simeq 200$ K, $t_c \simeq 5 - 10$ K [1]); b and c^* are the lattice parameters in the orthorhombic model of the $(TMTSF)_2X$ unit cell.

The AO introduces a periodic potential $\Box(y) = \Box\cos(\pi y/b)$, which leads to a doubling of the crystalline lattice. In the presence of the AO gap four components of electron wave function

$$(\psi^{\pm}(p_y), \psi^{\pm}(p_y + \pi/b)) = \exp[\pm i p_F x](\Psi^{\pm}(p_y), \Psi^{\pm}(p_y + \pi/b)) \quad (2)$$

obey the following Schrödinger equations:

$$[\pm v_F(p_x \mp p_F) + 2t_b\cos(p_y b)]\Psi^{\pm}(p_y) + \Box\Psi^{\pm}(p_y + \pi/b) = \epsilon\Psi^{\pm}(p_y), \quad (3)$$

$$[\pm v_F(p_x \mp p_F) - 2t_b\cos(p_y b)]\Psi^{\pm}(p_y + \pi/b) + \Box\Psi^{\pm}(p_y) = \epsilon\Psi^{\pm}(p_y + \pi/b). \quad (4)$$

The electron spectrum splits into four branches (see Fig. 1):

$$\epsilon_k^{\pm}(\mathbf{p}) = \pm v_F(p_x \mp p_F) + (-1)^k\sqrt{[2t_b\cos(p_y b)]^2 + \Box^2}\,. \quad (5)$$

[In Eqs. (3) and (4) the term which contains t_c is omitted; $k = 1, 2$.]

Under the magnetic field $\mathbf{H} = (0, 0, H)$, the Schrödinger equations in the gauge $\mathbf{A} = (0, Hx, 0)$ are given by the substitution $\mathbf{p} \to \mathbf{p} - e\mathbf{A}/c$ [21]

$$\left[\pm i v_F\frac{d}{dx} + 2t_b\cos\left(p_y b - \frac{\omega_c x}{v_F}\right)\right]\times \Psi^{\pm}(p_y) + \Box\Psi^{\pm}\left(p_y + \frac{\pi}{b}\right) = \epsilon\Psi^{\pm}(p_y), \quad (6)$$

$$\left[\pm i v_F\frac{d}{dx} - 2t_b\cos\left(p_y b - \frac{\omega_c x}{v_F}\right)\right]\times \Psi^{\pm}\left(p_y + \frac{\pi}{b}\right) + \Box\Psi^{\pm}(p_y) = \epsilon\Psi^{\pm}\left(p_y + \frac{\pi}{b}\right), \quad (7)$$

where $\omega_c = eHv_F b/c$ is the characteristic frequency of electron motion in a magnetic field; e and c are the electron charge and the velocity of light.

Note that the main contribution to the oscillatory part of e-e scattering does not contain Zeeman terms (see Fig. 2). That is why we omit electron spins in Eqs. (6) and (7) and everywhere below.

We estimate the AO gap as $\Box \simeq 1.78T_{AO} \simeq 50$ K. Taking $\Box \simeq 50$ K, $v_F \simeq 2 \times 10^7$ cm/sec, $t_b \simeq 200$ K, and $b \simeq 7.7$ Å [1], we find for the breakdown field: $H_0 \simeq 15$ T [19]. From general theory [21] it follows that the perturbation approach to our problem is valid for experimental magnetic fields $H \geq H_0 \simeq 15$ T.

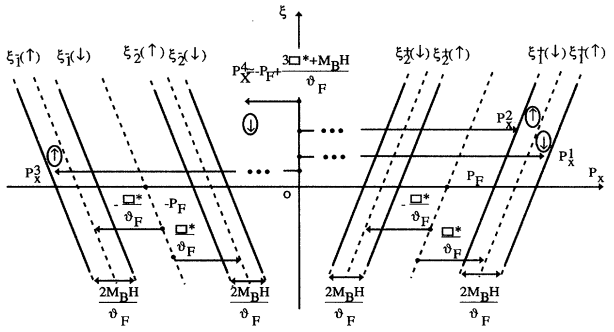

FIG. 2. One of the possible processes of e-e scattering for which electron Fermi functions depend on oscillatory parameter $\Box^*$; arrows stand for electron spins, μ_B is the Bohr magneton.

In the absence of AO gap the Schrödinger equations (6) and (7) have the solutions

$$\Phi^{\pm}(p_y, x) = \exp\left[\mp\frac{i\epsilon x}{v_F} \mp i\frac{\lambda}{2}\sin(p_y b - \omega_c x/v_F)\right], \quad (8)$$

$$\Phi^{\pm}(p_y + \pi/b, x) = \exp\left[\mp\frac{iex}{v_F} \pm i\frac{\lambda}{2}\sin(p_y b - \omega_c x/v_F)\right], \quad (9)$$

where we introduce parameter $\lambda = 4t_b/\omega_c$.

According to perturbation theory (see, for example, [22]) the zeroth-order wave functions have to be chosen in the form

$$\left(\Psi_n^{\pm}(p_y, x), \Psi_n^{\pm}\left(p_y + p_y + \frac{\pi}{b}, x\right)\right) = \left(C_{1,n}\Phi^{\pm}(p_y, x), C_{2,n}\Phi^{\pm}\left(p_y + \frac{\pi}{b}, x\right)\right). \quad (10)$$

[In Eq. (10) the number n enumerates the two solutions of Eqs. (6) and (7).]

After the solution of so-called "secular" equations [19,22] one can find the matrix $C_{m,n}$ and the first order corrections to the energy $\delta\epsilon_n$:

$$C_{m,n} = \frac{1}{\sqrt{2}}\begin{pmatrix} +1 & +1 \\ +1 & -1 \end{pmatrix}, \quad (11)$$

$$\delta\epsilon_n = (-1)^n\Box^* \equiv (-1)^n\Box J_0(\lambda) \simeq (-1)^n(\Box\sqrt{\omega_c/2\pi t_b}\,)\cos(4t_b/\omega_c), \quad (12)$$

where $J_0(\lambda)$ is a zeroth-order Bessel function $\lambda = 4t_b/\omega_c \gg 1$.

At this point, we present the wave functions for the case $\epsilon, \Box^* \ll \omega_c$:

$$(\Psi_1^{\pm}(p_y, x), \Psi_1^{\pm}(p_y + \pi/b, x)) = \frac{1}{\sqrt{2}}\exp[\mp i(\epsilon - \Box^*)x/v_F] \times \left(+\exp\left[\mp\frac{i\lambda}{2}\sin\left(p_y b - \frac{\omega_c x}{v_F}\right)\right], +\exp\left[\pm\frac{i\lambda}{2}\sin\left(p_y b - \frac{\omega_c x}{v_F}\right)\right]\right), \quad (13)$$

$$(\Psi_2^{\pm}(p_y,x),\Psi_2^{\pm}(p_y+\pi/b,x)) = \frac{1}{\sqrt{2}}\exp[\mp i(\epsilon+\Box^*)x/v_F] \times\left(+\exp\left[\mp\frac{i\lambda}{2}\sin\left(p_y b-\frac{\omega_c x}{v_F}\right)\right],-\exp\left[\pm\frac{i\lambda}{2}\sin\left(p_y b-\frac{\omega_c x}{v_F}\right)\right]\right). \quad (14)$$

The symmetrical combination (13) of unperturbed wave functions (8) and (9) and the antisymmetrical one (14) have different energies. In our case the difference in the energies between them, $\delta\epsilon = 2\Box^*$ [see Eq. (12)], rapidly oscillates with inverse magnetic field. As a conclusion, we show in Fig. 2 those parts of the electron spectrum which are responsible for *e-e* scattering in the significant for resistivity measurements quantum limit (QL), when $\omega_c \gg T$.

Let us consider the longitudinal resistivity ρ_{xx}, which is determined by the *e-e umklapp* scattering

$$p_x^1 + p_x^2 = p_x^3 + p_x^4 + 4p_F \quad (15)$$

in the commensurate $(TMTSF)_2X$ compounds [1].

The physical meaning of the RMO is now evident from Fig. 2. Indeed, under the condition of MB the distances in **p** space between electrons on the Fermi level depend on the magnetic field and are periodic in $1/H$. If, for example, two electrons with the wave functions (13) jump from the right side of the FS to the left one, due to the *umklapp* process (15), they have Fermi functions which depend on the oscillatory parameter $\Box^* \equiv \Box J_0(\lambda)$. From this particular example it is clear that the probability of *e-e* scattering should oscillate in a magnetic field.

To estimate the longitudinal resistivity ρ_{xx} we make use of the variational principle for Boltzmann equation for *e-e* scattering. According to this principle, one has to average the probability of the *umklapp* process (15) over the energies of scattering electrons on the scale $\epsilon \sim T$ by using Fermi functions [6,8,23]. The calculations of the amplitudes of *e-e* scattering in a magnetic field are convenient to perform using the diagram technique for Green functions [2]. Taking into account that in Q1D case $\rho_{xx} = 1/\sigma_{xx} \sim 1/\tau$, we obtain in QL

$$\frac{1}{\tau} = g^2 T\int_{-\infty}^{+\infty} dx\,\frac{2\pi T/v_F}{\sinh^2(2\pi Tx/v_F)}\left[\frac{2\pi T|x|/v_F}{\exp(4\pi T|x|/v_F)-1}+\frac{2\pi T|x|/v_F-1}{2}\right] \times\int_0^{2\pi}dk_1\int_0^{2\pi}dk_2\,J_0^2[(4t_c x/v_F)\sin(k_1)] \times\left\{J_0^2\left[2\lambda\sin\left(\frac{\omega_c x}{2v_F}\right)\sin(k_2)\right]\cos^4\left(\frac{\Box^* x}{v_F}\right)+J_0^2\left[2\lambda\cos\left(\frac{\omega_c x}{2v_F}\right)\cos(k_2)\right]\sin^4\left(\frac{\Box^* x}{v_F}\right)\right\}. \quad (16)$$

[Since $\lambda = 4t_b/\omega_c \simeq 10^2 \gg 1$ we have retained in Eq. (16) only the leading order terms in λ; g is the dimensionless constant of *umklapp e-e* scattering.]

Let us consider the final formula (16). We suggest that the oscillations of the renormalized AO gap

$$\Box^* = \Box J_0(\lambda) \simeq \Box\sqrt{\omega_c/2\pi t_b}\cos(4t_b c/eHv_F b) \quad (16')$$

are responsible for the experimentally observed strong RMO in the $(TMTSF)_2ClO_4$.

Because of the oscillations of $\Box^*$ (16′) expression (16) oscillates in $1/H$ with the following period:

$$\delta(1/H) = \pi e b v_F/4t_b c\,. \quad (17)$$

The experimental frequency of the RMO, $H^2/\delta H \simeq$ 250 T, corresponds to the value $t_b \simeq 200$ K in Eq. (17), which is in qualitative agreement with independent estimations [1].

Note that even in the absence of the AO gap [$\Box^* = 0$ in (16)] an integration of the Bessel function $J_0^2[2\lambda\sin(\omega_c x/2v_F)\sin(k_2)]$ over the variables x and k_2 still gives a small oscillatory contribution to Eq. (16), as was shown for resistivity and magnetic moment by Yamaji [6] and Yakovenko [8], correspondingly. The magnitude of those oscillations, $\delta(1/\tau_0)$, is much smaller than the magnitude of the oscillations suggested in this Letter, $\delta(1/\tau_0)/\delta(1/\tau) \sim \lambda^{-1} \sim 10^{-2}$. The period of Yamaji-Yakovenko's (YY) oscillations in $1/H$ differs from the period calculated by us [see (17)], since they have only considered the case when the AO gap is absent. From a mathematical point of view, the oscillations (17) are related to the renormalized AO gap $\Box^*$ and correspond to the Bessel function (16′), which is distinct from the Bessel function that gives contribution to YY oscillations.

In our opinion, weak YY oscillations cannot be considered as a primary effect in the $(TMTSF)_2ClO_4$ salt that has the AO gap in an electron spectrum. Although they might be important in $(TMTSF)_2PF_6$ and $(TMTSF)_2AsF_6$ salts, there is no evidence of oscillations in the metallic phase of these two compounds yet.

We would also like to point out that existing theories of thermodynamical properties, such as the temperature of the phase transition between the metallic phase and the SDW state in $(TMTSF)_2ClO_4$ [19], predict the oscillations with the same frequency as we calculated for the oscillations of resistivity (17). Moreover, recently we have shown [24] that experimentally observed oscillations of magnetization in $(TMTSF)_2ClO_4$ [12] could also be attributed to the oscillations of the renormalized AO gap,

$\square^*$ (16′). All of the above mentioned features of RMO in the $(TMTSF)_2ClO_4$ are in qualitative agreement with experimental data.

The temperature dependence of the magnitude of the oscillatory part in (16) is unusual. It has a peculiarity at crossover line

$$T^*(H) \simeq (\square/\pi)\sqrt{\omega_c(H)/2\pi t_b} \simeq (5\ \mathrm{K})(\sqrt{H/H_1}), \quad H_1 \simeq 50\ \mathrm{T}. \tag{18}$$

Under the crossover value $T \leq T^*(H)$ the relative magnitude of RMO is of the order of unity, $\delta\rho_{xx}/\rho_{xx} \sim 1$, but peaks of the resistivity are very narrow. Above $T^*(H)$ the oscillations become more harmonic.

It is possible to obtain an analytical expression for the relative magnitudes of RMO at temperatures $T \geq T^*, t_c/\pi$:

$$\frac{\delta\rho_{xx}}{\rho_{xx}} = \frac{\delta(1/\tau)}{1/\tau} = \alpha_1 \frac{\square^2\omega_c}{\pi^3 T^2 t_b}\cos(8t_b/\omega_c) + \alpha_2 \frac{\square^4\omega_c^2}{\pi^6 T^4 t_b^2}\cos(16t_b/\omega_c), \tag{19}$$

where the numbers $\alpha_1, \alpha_2 \sim 1$.

We mention a point of experimental significance regarding metals with open FS. Unlike Stark oscillations that do not depend on temperature *per se* [20] the oscillations suggested in this Letter are strongly temperature dependent, disappearing when $T \geq \omega_c(H)$. The calculated temperature dependence [19] is in agreement with recent data by Uji and Brooks [16].

In conclusion, we point out that another organic conductor, $(TMTSF)_2NO_3$, is believed to have closed electron orbits [1]. Therefore, RMO phenomenon in this material could be SdH-like, as was recently proposed by Maki [25].

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