Magnetic oscillations and field induced spin density waves in (TMTSF)$_2$ClO$_4$

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We report an analysis of the effects of magnetic field on a quasi-one-dimensional band of interacting electrons with a transverse dimerizing potential. One-particle problem in bond-antibond representation is solved exactly. The resulting propagator is used to calculate the spin-density-wave (SDW) response of the interacting system within the matrix RPA for the SDW susceptibility. We predict the magnetic field induced transition of the first order between interband SDW$_0$ and intra-band SDW$\pm$ phases. We reproduce the rapid oscillations with a period of 260 Tesla and the overall profile of the TMTSF$_2$ClO$_4$ phase diagram.

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Investigations of quasi-one-dimensional electronic systems at high magnetic fields and at low temperatures continue to give an important insight into the one-particle properties and interaction-induced phases such as spin- and charge-density-wave, superconductivity, and Mott localization. One of most spectacular phases of this kind are field-induced spin density wave (FISDW), found in Bechgaard salts and in some other low-dimensional compounds. The phenomenon of the FISDW is well understood in the Bechgaard salt (TMTSF)$_2$PF$_6$ where the cascade of SDW phases with quantized wave-vector is induced by orbital effects of magnetic field to the quasi-one-dimensional orbits of band electrons. Theory based on the mechanism of quantized nesting reproduces satisfactorily main experimental data for this salt.

In this letter we concentrate on (TMTSF)$_2$ClO$_4$, a Bechgaard salt which after a slow cooling enters into a qualitatively different type of FISDW phase at low temperatures, with a phase diagram that is still, after more than ten years of intensive studies, a matter of both experimental and theoretic controversies. In particular for magnetic field B>8T the nature of the ordering in the relaxed material is not a simple FISDW with some low integer quantum number \( N \), but a qualitatively different state containing several puzzling subphases. This phase is at \( 8T \) separated by a line of first order transition from a cascade of FISDW phases which very much resembles to that of the standard model. Another characteristic phenomenon, the rapid oscillations (RO) in \( 1/B \) with a frequency of 260 Tesla are visible in transport properties in both metallic and FISDW states. Similar RO are seen also in thermodynamic quantities like torque, magnetization, sound velocity and specific heat, but only in the ordered phase.

The incompatibility of above facts with the quantum nesting model (QNM) for a single quasi-1D band is believed to stem from the particular ordering of ClO$_4$ anions. This ordering introduces the new modulation with the wave vector \((0, \pi/b, 0)\), i.e. a dimerization in the low-conducting direction with the inter-chain distance \( b \). The magnitude of the dimerizing potential can be tuned to some extent by varying the cooling rate. Thus, anions presumably remain disordered in the rapidly quenched samples. Then there is no dimerization gap in the band, and the system shows properties of a single quasi-1D imperfectly nested band with a SDW order appearing already in the zero magnetic field. The anion ordering in slowly relaxed samples is at about 24K, and coincides with the onset of rapid oscillations in the magnetoresistance.

The dimerized band has two pairs of Fermi sheets in the new Brillouin zone. Already simple geometric arguments suggest three possible nesting wave vectors favoring various SDW phases. First, interband nesting, leads to SDW$_0$ that is the two-band version of the standard FISDW phase. Other two nesting vectors relate Fermi sheets within the same band. They give SDW$_+ \pm$ for antibond nesting and SDW$_-$ for bond nesting. However the interplay between SDW$_0$ and SDW$_\pm$ is not only a geometric question of the choice of the nesting vector. Due to a finite anion potential \( V \) in the kinetic part of the Hamiltonian an off-diagonal term appears in the SDW response, making necessary an appropriate matrix approach in the calculation of the critical susceptibilities. The response matrix is formulated in the space of two order parameters, \( \Delta_h \) (“homogeneous”) and \( \Delta_a \) (“alternating”), determining the magnetic pattern

\[
m_L(x, R_\perp) = (\Delta_h \pm \Delta_a) \cos [(2k_F + k)x + pnd] .
\]

Here \( d = 2b \) and the upper and lower sign stay for even \( (R_\perp = nd) \) and odd \( (R_\perp = nd + 1/2) \) chains respectively. As it is shown in Refs. SDW$_0$ \( [\Delta_h \neq 0, \Delta_a = 0] \) is stabilized for low values of \( V \) (providing the imperfect
nestic parameter $t'_0$ allows for SDW stabilization), while SDW$_\pm$ with $\Delta_h \geq \Delta_a \neq 0$ gets stable for $V/t_b > 1.6$ irrespectively to the value of $t'_0$. Here $t_b$ is the interchain hopping integral. The slowly relaxed (TMTSF)$_2$ClO$_4$ samples are expected to lie in the range of intermediate values of $V$ in which there is no SDW ordering at $B = 0$ down to $T = 0$.

Indeed, as it will be shown, $V/t_b$ fitting the experiments is close to one. In this range it is not allowed to use the quasi-classical approximation of Gor’kov and Lebed [11], which consists in making Peierls substitution $p \rightarrow p - e A$ in each sub-band separately and including the anions’ effects only via magnetic breakdown (MB) junctions near the zone boundary. While this approximation is sufficient for $V/t_b \ll 1$, here one has to solve the whole quantum-mechanical problem instead.

It was pointed out several times (see [8] and references therein) that a mechanism of coherent inter-band tunnelling, very similar to Stark over-gap quantum interference (QI) in magnesium [4], is essential for high-field physics in (TMTSF)$_2$ClO$_4$. In particular, RO in metallic state can be explained only in terms of QI mechanism because no closed orbits exist. On the contrary, in the SDW state both closed orbits and Stark interference contribute to RO. Oscillating behavior periodic in $1/k$ is present in (TMTSF)$_2$ClO$_4$. In this range it is not allowed to use the quasi-classical approximation of Gor’kov and Lebed [11], and to our exact solution as well. What changes from one approach to another are the dependence $\delta (B)$ and the result for electronic wavefunction. In order to obtain them exactly we start from the effective one-particle Hamiltonian for electronic operators

$$\Psi_f(x, p)$$

$$H_0 = i v_F \rho_3 \partial_x + \tau_3 T(p b - G x) + \bar{T}(p b - G x) - V \tau_1,$$  

(3)

where $\rho$’s and $\tau$’s are Pauli matrices in left-right and bond-antibond indices respectively. The most general transverse dispersion was split into two parts

$$T(p b) \equiv 2 \sum_{j=1}^{\infty} t_j \cos[(2j - 1)p b], \bar{T}(p b) \equiv 2 \sum_{j=1}^{\infty} t'_j \cos[2jp b]$$

(4)

corresponding to effective hoppings between odd and even neighbors respectively. We diagonalize $H_0$ by the unitary transform

$$\Psi_f = \left( \frac{\alpha_f}{-\beta_f} \frac{\beta_f}{\alpha_f} \right) e^{i p_0} \Phi_f,$$

(5)

with $|\alpha|^2 + |\beta|^2 = 1$, and functions $\alpha$, $\beta$ and $\theta$ depending on $x$ and $p$ only through the combination $z = p b - G x$. From the requirement that the effective Hamiltonian for field $\Phi$ is only if $v_F \partial_x$ we get

$$\theta(z) = \frac{1}{v_F} \int dz \bar{T}(z)$$

(6)

and a system of differential equations for functions $\alpha$ and $\beta$

$$i f v_p \alpha_f' (z) = -\tau(z) \alpha_f (z) - V \bar{\beta}_f^* (z)$$

$$i f v_p \beta_f' (z) = -\tau(z) \beta_f (z) + V \alpha_f^* (z).$$

(7)

Note that $\theta(z + 2\pi) = \theta(z)$ and that $\alpha_+ (z) = \alpha_-^* (z)$ and $\beta_+ (z) = \beta_-^* (z)$, so that it suffices to follow e. g. solutions $\alpha_+ (z), \beta_+ (z)$ of the system (6). According to Floquet theory these solutions can be written in the form $\alpha (z) = A (z) \exp (-i z \delta); \beta (z) = B (z) \exp (i z \delta)$. $A$ and $B$ are periodic with the period $2 \pi$, and the closer inspection shows that the Floquet exponent $\delta$ for the system (6) is real for all values of parameters, at least after keeping in $\tau(z)$ only the leading term $t_1$.

Once we find $A$, $B$ and $\delta$ the wave functions $\langle x, p | F_k \rangle$ of the states created by $\Psi^*(x, p)$ are known. The corresponding spectrum is one-dimensional, $E_k = v_F f(k - k_F)$. Projection of $|F_k\rangle$ to the plane wave $\langle k', p |$ is

$$\sum_N \left\{ \mu_k e^{i(N-\delta)p} \delta[k' - k + G(N-\delta)] \left( \frac{a_N}{b_N} \right) + v_k e^{i(N+\delta)p} \delta[k' - k + G(N+\delta)] \left( \frac{b_N}{a_N} \right) \right\}$$

(8)

Coefficients $a_N$, $b_N$, $\hat{a}_N$ and $\hat{b}_N$ are Fourier components of the products $A \exp (i \theta), B \exp (i \theta), A^* \exp (i \theta)$ and $B^* \exp (i \theta)$ respectively. Coefficients $\mu_k$ and $v_k$ are fixed by initial conditions. The expression (6) tells us how the plane wave $\exp (i k x + p R_1)$ is decomposed into discrete states $N$. Each state $N$ is split by $\delta$ in a way that components with the tilt $-\delta$ have the statistical weight $|\mu_k|^2 |a_N|^2 + |\hat{b}_N|^2$ and the ones with the tilt $+\delta$ have the weight $|v_k|^2 |b_N|^2 + |\hat{a}_N|^2$. Green function $\langle \Psi(x, p) \Psi^*(x', p) \rangle$ is easily constructed using transformation (8) and knowing that $\langle \Phi^\dagger \rangle = (i \omega_n - iv_F \theta^\dagger )^{-1} = G_{1D}$.

The Floquet exponent $\delta$ and the functions $A$ and $B$ are calculated using the Hill’s theory and the fundamental matrix method [15, 16]. In the present work we limit our calculations to first harmonics in Eq. (8). The expression (8) shows the energy $\omega_n$ (in units of $V$) as a function of
the magnetic breakdown parameter \( \kappa \equiv 2\omega_c t_b/V^2 \), where \( \omega_c = v_F G \) is the cyclotron frequency. In quasi-classical picture \( \kappa \) determines the probability of the over-gap tunnelling \( P = \exp(-\pi/2\kappa) \). One sees that the crossover from oscillating to saturating behavior does not coincide with the crossover from the weak \( (\kappa < 1) \) to the strong \( (\kappa > 1) \) MB. The position of the last zero of \( \delta \) is rather large, i.e. comparable to \( E \). The 1D spectrum of Osada et al. [15] however strongly suggest that \( \omega_c \) is rather large, i.e. comparable to \( t_b \).

We proceed with the solution of the interacting problem. Neglecting the absence of a presumably small Umklapp scattering, the effective coupling for SDW is the forward scattering amplitude \( g_2 \), here simply denoted by \( U \). We employ the matrix RPA formalism developed in Ref. [12]. The resulting relevant bare susceptibility is \( \chi_1(\mathbf{q}, T) = \frac{1}{2}\{\chi_{aa} + \chi_{hh} + [\chi_{aa} - \chi_{hh}]^2 + 4(\chi_{ha})^2]^{1/2}\} \), playing in the Stoner criterion

\[
1 - U\chi_1(\mathbf{q}, T_c) = 0, \tag{9}
\]

for several values of the magnetic field are given by \( \mathbf{q} \), being the wave vector at which \( \chi_1(\mathbf{q}) \) has the maximum. The ratio of two SDW order parameters from Eq. (1) is also a function of bare correlators \( \chi_{aa}, \chi_{hh}, \chi_{ab} \) in the \( (a, h) \) basis (see [13]). The bare correlators in the magnetic field are given by

\[
\begin{align*}
\chi_{hh} &= \sum_N \left| I_{0h0} \right|^2 P_0 + \frac{1}{2} I_{h+}^2 P_+ + \frac{1}{2} I_{h-}^2 P_- ,
\chi_{aa} &= \sum_N \left| I_{0a0} \right|^2 P_0 + \frac{1}{2} I_{a+}^2 P_+ + \frac{1}{2} I_{a-}^2 P_- ,
\chi_{ha} &= \sum_N \left| R(I_{0h0}^* I_{0a0}) \right|^2 P_0 + \frac{1}{2} I_{h+} I_{a+} P_+ - \frac{1}{2} I_{h-} I_{a-} P_- ,
\end{align*}
\tag{10}
\]

where \( P_0, P_\pm \) stand for \( P(\mathbf{q} \parallel -NG,T) \) and \( P(\mathbf{q} \parallel -G(N \pm 2\delta),T) \) respectively, \( P(k,T) \) being the familiar 1D Lindhard function at the wave number \( 2k_F + k \). The above fit, as well as other insights [21] however strongly suggest that \( V \) in \( \text{TFM} \) is rather large, i.e. comparable to \( t_b \).

FIG. 1: (a) Energy ratio \( \omega_c \delta/V \) as function of the magnetic breakdown parameter \( \kappa \) for several values of \( V/t_b \). (b) Dependence of \( \delta \) on \( r \) for \( \theta = 10^\circ \) (A), \( 45^\circ \) (B), and \( 80^\circ \) (C).
The $N$-th split level in the decomposition $I(q,N)$.

\[ I_{b0}(q,N) = \sum_n (a_n b_{N-n} - \hat{b}_n \hat{a}_{N-n}) e^{i(n-N/2)q} \]
\[ I_{b+}(q,N) = \sum_n (\hat{a}_n \hat{a}_{N-n} + b_n b_{N-n}) e^{i(n-N/2)q} \]
\[ I_{b-}(q,N) = \sum_n (a_n \hat{a}_{N-n} - \hat{b}_n b_{N-n}) e^{i(n-N/2)q} \]
\[ I_{a0}(q,N) = \sum_n (a_n \hat{a}_{N-n} - \hat{b}_n b_{N-n}) e^{i(n-N/2)q} \]
\[ I_{a+}(q,N) = \sum_n (\hat{a}_n b_{N-n} + b_n \hat{a}_{N-n}) e^{i(n-N/2)q} \]
\[ I_{a-}(q,N) = \sum_n (\hat{a}_n b_{N-n} + b_n \hat{a}_{N-n}) e^{i(n-N/2)q} \]

There are two important selection rules for these amplitudes, namely for $N$ even, $I_{b0}(N) = I_{a0}(N) = 0$ while for $N$ odd, $I_{b\pm}(N) = I_{a\pm}(N) = 0$. Thus the interband processes contribute only to FISDW phases with odd $N$ while the intraband processes contribute only to phases with even $N$. Consequently only phases with even $N$ "see" the splitting by $\delta$.

The $q$-dependence of the susceptibility $\chi_1(q)$ for the particular choice of parameters, $\omega_c = 0.1 t_b, V = 0.85 t_b, t'_b/t_b = 0.03, T/t_b = 0.001$, is shown in Fig. 2. The overall envelope assumes the shape present already in the absence of magnetic field $B$. It is now superimposed by a well known characteristic of FISDW susceptibilities, logarithmic peaks corresponding to single one-dimensional bubbles $P(k)$, weighted by $p$-dependent amplitudes as defined by Eqs. (10,11). Qualitatively new feature regarding these peaks is the splitting of peaks with even $N$ by $\pm \delta$ around the positions at $k = N\pi/2B$.

According to Eq. (11) at $T = T_c$ the highest of peaks in Fig. 2 attains the value $1/U$. Fig. 3 shows the resulting phase diagram for a realistic choice of parameters, $V = 0.85 t_b, t'_b/t_b = 0.03, t_b$ and $T_c(V = t'_b = 0) = 13K$. The resulting maximal critical temperature within the present field range is $T_c^{\text{max}} \approx 1.1K$. The most obvious characteristic of the obtained phase diagram is the first order transition from SDW$_0$ to SDW$_\pm$ at $B_c \approx 9$ Tesla. Dependence $T_c(B)$ for $B < B_c$ is similar to the FISDW cascade.
in TMTSF$_2$PF$_6$, with the difference that here only odd phases appear because the even ones are suppressed by splitting. For $B > B_c$ the critical temperature increases towards the highest value $T_c^{\text{max}}$. As the magnetic field further increases the critical temperature $T_c(B)$ starts to oscillate, with the sharp dips corresponding to commensurability condition $2G\delta = G$ between the Floquet wave number and the magnetic wave number.

Note that the present choice of anion potential $V = 0.85t_b$ places us in the intermediate regime on scale $V$ where the response of both SDW$_0$ and SDW$_\pm$ is suppressed for $B = 0$. Hence, being of comparable magnitude the two instabilities compete once they are restored by magnetic field. Both phases in fig.3 are sensitive to $V$, general trend being that by increasing $V$ one reduces $T_c(SDW_0)$ and increases $T_c(SDW_\pm)$. The parameter of imperfect nesting in the standard model, $t'_b$, here affects only SDW$_0$, while for SDW$_\pm$ $t'_b$ plays a role of an effective nearest neighbor hopping. We remind that the effective parameter of imperfect nesting for SDW$_\pm$ is a function of $t_b/V$, as pointed out in Ref. [12]. $t'_b$ acts on SDW$_0$ in a standard way [12], i.e. it fixes the width of the FISDW cascade $\Delta \omega_c \sim t'_b$ so that by increasing $t'_b$ one reduces $T_c(SDW_0)$ at fixed $B$.

The result of the subtle interplay between two scales $V$ and $t'_b$ is that the realistic profile of the phase diagram is possible only within a rather restricted range of the $(V, t'_b)$ space. By increasing $V$ or $t'_b$ by a few percent one reduces $T_c(SDW_0)$ below $T_c(SDW_\pm)$ in the whole $B$ domain. On the other hand by decreasing $V$ by a few percents one gets a hump in $T_c(SDW_0)$ on the left of the transition SDW$_0$ – SDW$_\pm$.

The rapid oscillations in observable response functions are related to the oscillations of $\delta$ [16], shown in Fig.3(b). Generally, we expect RO to be visible if two conditions are fulfilled. First, in order to have an overgap interference one needs a moderate MB parameter, $\kappa \sim 1$. Second, one has to be in the oscillating regime on the scale $r$, which is equivalent to $\kappa < \rho(V/t_b) \equiv 2(V/t_b)^{-2} \sqrt{(\gamma V/t_b)^2 + 1}$. Fig.3(b) shows how the energy tilt $\omega_c \delta$ varies in various parts of the phase diagram. At 30 Tesla we have $\kappa \sim 0.5$ and $\rho(0.85) \approx 3.3$, so that both conditions are fulfilled. The effect is expected to be even stronger for higher fields because $\kappa$ then increases.

The maximal value of the critical temperature in Fig.3, $T_c^{\text{max}} \approx 1.1K$, is considerably smaller than the experimental value of 5.5K. In this respect we note that $T_c^{\text{max}}$ is essentially model dependent quantity, i.e. that the Hamiltonian [6] represents a minimal model for understanding the interplay between two SDW phases in the magnetic field. Namely, recent experiments [12] suggest that the anion ordering in TMTSF$_2$ClO$_4$ induces
also, beside a strong dimerizing potential $V$, rather large changes in other band parameters.

The present treatment also does not include the quantitative analysis of the splitting of degeneracy of two intraband phases, $SDW_+$ and $SDW_-$. Physically the degeneracy is lifted because the realistic tight-binding dispersion along the chain is not strictly linear. Consequently the dominant instability will be that of $SDW_-$, as discussed in Ref. [12]. Similar conclusions were obtained also by numerical calculations [22], but without taking into account the two component aspect of the order parameter $\Omega$. The second critical temperature can be calculated within Landau theory as in Ref. [13], and by taking the nonlinearity of the band dispersion into account. The subphases of the high-field phase correspond to $SDW_+$ phases within $SDW_-$, each one nesting its own pair of Fermi sheets. Such scenario is impossible for $SDW_0$ since it proceeds through nesting of all four sheets at the single critical temperature.

In conclusion, we have solved exactly the one-particle problem of dimerized Q1D band of electrons in magnetic field. Observables contain characteristic periodicity in $1/B$, consistent with 260 Tesla oscillations in normal and $SDW$ phases of $(TMTSF)_2\text{ClO}_4$. Using matrix RPA for $SDW$ susceptibility we reproduce the first-order transition between two types of FISDW ordering, as well as the overall profile of the experimental phase diagram.

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