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Magnetic oscillations and field-induced spin-density waves in (TMTSF)₂ClO₄

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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 random-phase approximation for the SDW susceptibility. We find that the value of the anion potential fitting experiments in relaxed (TMTSF)₂ClO₄ is large, of the order of interchain hopping. In particular we predict the magnetic-field-induced transition of the first order between interband SDW₀ and intraband SDW_± phases, we reproduce the rapid oscillations with a period of 260 T and the overall profile of the (TMTSF)₂ClO₄ phase diagram.

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I. INTRODUCTION

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.¹ One of most spectacular phases of this kind are field-induced spin-density wave (FISDW), found in Bechgaard salts² and in some other low-dimensional compounds.³ The phenomenon of the FISDW is well understood in the Bechgaard salt (TMTSF)₂PF₆ where the cascade of SDW phases with quantized wave vector is induced by orbital effects of magnetic field to the quasi-one-dimensional (Q1D) orbits of band electrons. Theory based on the mechanism of quantized nesting⁴ reproduces satisfactorily main experimental data for this salt.

In this paper we concentrate on (TMTSF)₂ClO₄, a Bechgaard salt which after a slow cooling^{5,6} enters into a qualitatively different type of FISDW phase at low temperatures, with a phase diagram that is still, after more than 10 years of intensive studies,^{1,2} a matter of both experimental and theoretic controversies. In particular for magnetic field $B > 8$ T 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.^{2,7,8} This phase is at 8 T separated by a line of first-order transition from a cascade of FISDW phases which very much resembles that of the standard model. Another characteristic phenomenon, the rapid oscillations (RO) in $1/B$ with a frequency of 260 T, is visible in transport properties in both metallic and FISDW state.^{2,8,9} Similar RO are seen also in thermodynamic quantities such as torque, magnetization, sound velocity, and specific heat, but only in the ordered phase.^{1,2} The highest value of T_c in the $T_c(B)$ dependence is 5.5 K, instead of 12 K as expected from analogy with the (TMTSF)₂PF₆ salt.

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 ClO₄ anions.^{2,10} 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 interchain distance b . The magnitude of the dimerizing potential can be tuned to some extent by varying the cooling rate.^{5,6} 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.^{5,6,11} The anion ordering in slowly relaxed samples is at about 24 K, and coincides with the onset of rapid oscillations in the magnetoresistance.¹² The RO in (TMTSF)₂ClO₄ have been theoretically explained in two limiting cases. The limit of strong anion potential $V \gg t_b$, t_b being the interchain hopping integral, was calculated by Brazovskii and Yakovenko,¹⁰ while the opposite limit $V \ll t_b$ was solved by Lebed and Bak.¹³ In this paper we solve *exactly*, i.e., for any V and B , the one-particle problem, which determines the RO phenomenon.

The dimerized band has two pairs of Fermi sheets in the new Brillouin zone, as shown in Fig. 1. Already simple geometric arguments⁷ suggest three possible nesting wave vectors favoring various SDW phases. The interband nesting \mathbf{Q}_0 leads to SDW₀ that is the two-band version of the standard FISDW phase. Other two nesting vectors \mathbf{Q}_+ and \mathbf{Q}_- relate Fermi sheets within the same band. They give SDW₊ for bond nesting and SDW₋ for antibond nesting. However the interplay between SDW₀ and SDW_± 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^{14,15} in the calculation of the critical susceptibilities. This matrix aspect of the problem was ignored in all former theoretical approaches.^{16–19,21} We formulate the response matrix in the space of two order parameters Δ_h (homogeneous) and Δ_a (alternating) determining the magnetic pattern,

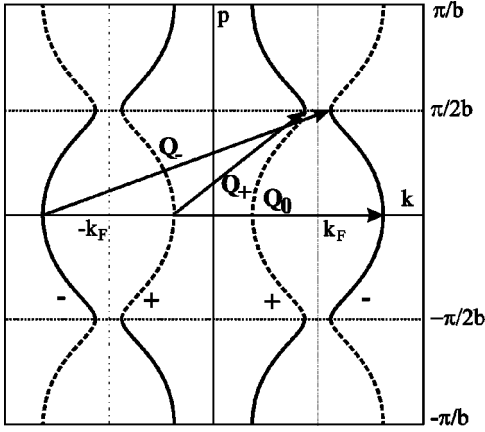


FIG. 1. Two Fermi surfaces of $(\text{TMTSF})_2\text{ClO}_4$ (wrapping is highly exaggerated). Wave vectors \mathbf{Q}_0 , \mathbf{Q}_+ , and \mathbf{Q}_- correspond, respectively, to SDW_0 , SDW_+ , and SDW_- .

$$m_z(x, R_\perp) = (\Delta_h \pm \Delta_a) \cos[(2k_F + k)x + pnd]. \quad (1)$$

Here $d \equiv 2b$ and the upper and lower sign stay for even ($R_\perp = nd$) and odd ($R_\perp = nd + d/2$) chains, respectively. As it is shown in Refs. 14,15, three types of SDW modulations with wave vectors shown in Fig. 1 are candidates for ordering at the phase transition from the metallic state. SDW_0 is stabilized for low values of V (providing the imperfect nesting parameter t'_b allows for SDW stabilization), while SDW_+ or SDW_- get stable for $V/t_b > 1.6$ irrespective of the value of t'_b . The slowly relaxed $(\text{TMTSF})_2\text{ClO}_4$ 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 below, V/t_b fitting the experiments is close to unity, which is also in agreement with recent detailed x-ray data.²⁰ Still, Haddad *et al.*²¹ recently put forward some arguments in favor of the small V . In order to construct the phase diagram with dominant SDW_\pm already in the range of small V these authors put larger coupling constant for SDW_\pm than for SDW_0 . This assertion is not plausible because the renormalization group for the quasi-one-dimensional interacting fermions suggests that one obtains difference between intraband and interband effective coupling constants only if V is of the order or larger than t_b .^{22,23}

In the range $V/t_b \geq 1$ it is not allowed to use the quasi-classical approximation of Gor'kov and Lebed,¹⁷ which consists in making Peierls substitution $p \rightarrow p - eA$ in each subband separately and including the anions' effects only via magnetic breakdown (MB) junctions near the zone boundary. While this approximation is sufficient for $V/t_b \leq 1$, here one has to solve the whole quantum-mechanical problem instead.

It was pointed out several times^{8,12,17} that a mechanism of coherent interband tunneling, very similar to Stark overgap quantum interference (QI) in magnesium,²⁴ is essential for high-field physics in $(\text{TMTSF})_2\text{ClO}_4$. 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 $1/B$ can be seen already at the level of one-particle spectrum. This is the topic

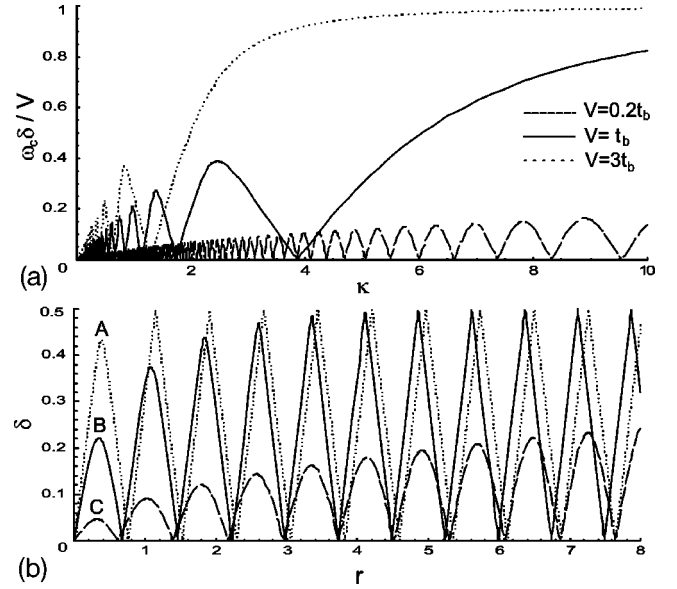


FIG. 2. (a) Energy ratio $\omega_c \delta / V$ as a function of the magnetic breakdown parameter κ for several values of V/t_b . (b) Dependence of δ on r for $\theta = 10^\circ$ (A), 45° (B), and 80° (C).

of the following section. In Sec. III we include interactions via the matrix random-phase approximation (RPA) for the *two-component* SDW order parameter and construct the phase diagram. The last section contains conclusions.

II. EXACT SOLUTION OF THE STARK INTERFEROMETER

We solved the one-particle problem of the Q1D band with anion potential V . Resulting electronic propagator with longitudinal momentum k has poles at

$$E_f = v_F [f(k - k_F) + GN] \pm v_F G \delta, \quad (2)$$

where f is left-right index, $G \equiv eBb/\hbar$ is the magnetic wave number, and N is integer number. The first term in Eq. (2) is the standard QNM dispersion and the last term is the splitting due to anions. Overgap resonances are present in $\delta(B)$ as will be discussed below (see Fig. 2). The expression for the spectrum (2) is common to perturbation calculations,¹⁶ to quasiclassical tunnelling analysis,¹⁷ and to our exact solution as well. What change from one approach to another are the dependence $\delta(B)$ and the result for electronic wave function. In order to obtain them exactly we start from the effective one-particle Hamiltonian for electronic operators $\Psi_f(x, p)$,

$$H_0 = iv_F \rho_3 \partial_x + \tau_3 \mathcal{T}(pb - Gx) + \bar{\mathcal{T}}(pb - Gx) - V\tau_1, \quad (3)$$

where ρ 's and τ 's are Pauli matrices in left-right and bond-antibond indices, respectively. The most general transverse dispersion was split into two parts,

$$\mathcal{T}(pb) \equiv 2 \sum_{j=1}^{\infty} t_j \cos[(2j-1)pb], \quad (4)$$

$$\tilde{T}(pb) \equiv 2 \sum_{j=1}^{\infty} t'_j \cos[2jpb],$$

corresponding to effective hoppings²⁵ between odd and even neighbors, respectively. We diagonalize H_0 by the unitary transform

$$\Psi_f = \begin{pmatrix} \alpha_f & \beta_f \\ -\beta_f^* & \alpha_f^* \end{pmatrix} e^{if\theta} \Phi_f, \quad (5)$$

with $|\alpha|^2 + |\beta|^2 = 1$, and functions α , β , and θ depending on x and p only through the combination $z = pb - Gx$. From the requirement that the effective Hamiltonian for field Φ be only $ifv_F\partial_x$ we get $\theta(z) = v_F^{-1} \int^z dz' \tilde{T}(z')$ and a system of differential equations for functions α and β ,

$$\begin{aligned} ifv_F\alpha'_f(z) &= -\mathcal{T}(z)\alpha_f(z) - V\beta_f^*(z), \\ ifv_F\beta'_f(z) &= -\mathcal{T}(z)\beta_f(z) + V\alpha_f^*(z). \end{aligned} \quad (6)$$

This ‘‘one dimensionalization’’ of the effective Hamiltonian is the two-component generalization of the standard phase transformation procedure for Q1D systems in magnetic field.⁴ 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 $\alpha(z) = A(z)\exp(-iz\delta)$; $\beta(z) = B(z)\exp(iz\delta)$. A and B are periodic with the period 2π , and closer inspection shows that the Floquet exponent δ for the system (6) is real for all values of parameters, at least after keeping in $\mathcal{T}(z)$ only the leading term $j=1$.

The Floquet exponent δ and the functions A and B are calculated using the Hill's theory and the fundamental matrix method.²⁶ In the present work we limit our calculations only to first harmonics in Eq. (4), parametrized with $t_1 = t_b$ and $t'_1 = t'_b$. Let us concentrate on the magnetic-field dependence of the Floquet exponent δ that splits the QNM spectrum as given by Eq. (2). Figure 2(a) shows the energy $\omega_c\delta$ (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 quasiclassical picture κ determines the probability of the overgap tunneling $P = \exp(-\pi/2\kappa)$.¹⁷ 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 δ is not universal in κ , but approximately in $r \equiv [(\gamma V)^2 + t_b^2]^{1/2} / \omega_c$, where the value of γ is 0.77. Figure 2(b) shows $\delta(r)$ for several ‘‘polar angles’’ defined by $\tan \theta \equiv t_b / \gamma V$. Oscillations of δ are approximately periodic in r with a period of 0.80. Choosing the parameters $t_b = 300$ K, $v_F = 2 \times 10^5$ m/s, and $b = 7.7 \times 10^{-10}$ m we fit RO at 260 T by putting $V \approx 0.8t_b$.

Taking the limit of strong magnetic field $\omega_c/t_b \gg 1$ and of weak anion potential $V/t_b \ll 1$ we can easily reproduce the 1D spectrum of Osada *et al.*,¹⁶ $E_k \rightarrow f v_F(k - k_f) \pm \omega_c\delta$ with $\delta \rightarrow (V/\omega_c) \mathcal{J}_0(4t_b/\omega_c)$, \mathcal{J}_0 being the Bessel function. On the other hand the spectrum of Gor'kov and Lebed¹⁷ is reproduced for weak anion potential, $V/t_b \ll 1$. The above fit,

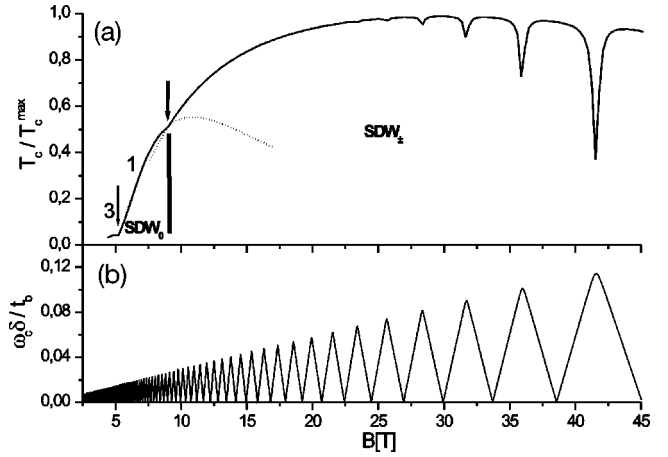


FIG. 3. (a) Phase diagram. (b) Energy ratio $\omega_c\delta/t_b$ on the same magnetic scale as the phase diagram.

as well as other insights^{20,28} however strongly suggest that V in $(\text{TMTSF})_2\text{ClO}_4$ is rather large, i.e., comparable to t_b .

The rapid oscillations in observable response functions are related to the oscillations of δ (Ref. 17), shown in Figs. 2 and 3(b). At 30 T the magnetic breakdown parameter has moderate value of $\kappa \sim 0.5$.

III. MANY-BODY EFFECTS

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. 14. The resulting relevant bare susceptibility is $\chi_1(\mathbf{q}; T) = \frac{1}{2} \{ \chi_{aa} + \chi_{hh} + [(\chi_{aa} - \chi_{hh})^2 + 4(\chi_{ha})^2]^{1/2} \}$, entering into the Stoner criterion

$$1 - U\chi_1(\mathbf{q}_c, T_c) = 0, \quad (7)$$

\mathbf{q}_c 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 $\chi_{aa}, \chi_{hh}, \chi_{ah}$ in the (a, h) basis (see Ref. 14). Their analytical expressions are

$$\begin{aligned} \chi_{hh} &= \sum_N \left[\left| I_{h0} \right|^2 P_0 + \frac{1}{2} I_{h+}^2 P_+ + \frac{1}{2} I_{h-}^2 P_- \right], \\ \chi_{aa} &= \sum_N \left[\left| I_{a0} \right|^2 P_0 + \frac{1}{2} I_{a+}^2 P_+ + \frac{1}{2} I_{a-}^2 P_- \right], \\ \chi_{ha} &= \sum_N \left[\text{Re}(I_{h0} I_{a0}^*) P_0 + \frac{1}{2} I_{h+} I_{a+} P_+ - \frac{1}{2} I_{h-} I_{a-} P_- \right], \end{aligned} \quad (8)$$

where P_0, P_{\pm} stand for $P(q_{\parallel} - NG, T)$ and $P[q_{\parallel} - G(N \pm 2\delta), T]$, respectively, $P(k, T)$ being the familiar 1D Lindhard function at the wave number $2k_F + k$. P_0 and P_{\pm} are the interband and the intraband susceptibilities of the N th split level of the spectrum (2). 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. The dependence on the transverse momentum is present in the amplitudes $I(q_\perp, N)$,

$$\begin{aligned}
 I_{h_0}(q_\perp, N) &= \sum_n (a_n b_{N-n} - \hat{b}_n \hat{a}_{N-n}) e^{i(n-N/2)q_\perp}, \\
 I_{h_+}(q_\perp, N) &= \sum_n (\hat{a}_n \hat{a}_{N-n} + b_n b_{N-n}) e^{i(n-N/2)q_\perp}, \\
 I_{h_-}(q_\perp, N) &= \sum_n (a_n a_{N-n} + \hat{b}_n \hat{b}_{N-n}) e^{i(n-N/2)q_\perp}, \\
 I_{a_0}(q_\perp, N) &= \sum_n (a_n \hat{a}_{N-n} - \hat{b}_n b_{N-n}) e^{i(n-N/2)q_\perp}, \\
 I_{a_+}(q_\perp, N) &= \sum_n (\hat{a}_n b_{N-n} + b_n \hat{a}_{N-n}) e^{i(n-N/2)q_\perp}, \\
 I_{a_-}(q_\perp, N) &= \sum_n (a_n \hat{b}_{N-n} + \hat{b}_n a_{N-n}) e^{i(n-N/2)q_\perp}. \quad (9)
 \end{aligned}$$

There are two important selection rules for these amplitudes, namely, for N even, $I_{h_0}(N) = I_{a_0}(N) = 0$ while for N odd, $I_{h_\pm}(N) = I_{a_\pm}(N) = 0$. 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 δ .

According to Eq. (7) the maximum of $\chi_1(\mathbf{q})$ attains the value $1/U$ at $T = T_c$. Figure 3 shows the resulting phase diagram for a realistic choice of parameters, $V = 0.85t_b$, $t'_b = 0.03t_b$, and $T_c(V = t'_b = 0) = 13$ K. The resulting maximal critical temperature within the present field range is $T_c^{\max} \approx 1.1$ K. The most obvious characteristic of the obtained phase diagram is the first-order transition from SDW_0 to SDW_\pm at $B_c \approx 9$ T. Dependence $T_c(B)$ for $B < B_c$ is similar to the FISDW cascade in $\text{TMTSF}_2\text{PF}_6$, with the difference that here only odd phases appear because the even ones are suppressed by splitting. We expect that at lower temperatures the first-order transition from SDW_0 to SDW_\pm is driven by stabilization of soliton lattices with competing SDW_0 and SDW_\pm domains.²⁷ For $B > B_c$ the critical temperature increases towards the highest value T_c^{\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. We can also estimate the quantum Hall effect in the phase SDW_+ or SDW_- . The shift from the perfect nesting in these phases is $v_F \Delta k = \sqrt{V^2 + 2t_b^2} - (\sqrt{V^2 + 4t_b^2} + V)/2$.¹⁴ For $V \sim t_b$ this gives $v_F \Delta k \sim t_b/10$ and the quantum number of the Hall effect, $N_H \sim v_F \Delta k / \omega_c$, takes values between 3 and 1 for magnetic fields between 10 and 30 T. However, the precise values of N_H and whether N_H is integer or not are the questions beyond the present analysis.

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. We have calculated²⁹ the phase diagram for all values of V and concluded that $V \approx 0.85t_b$ is indeed the only value fitting the phase diagram obtained in experiments. Namely, increasing V or t'_b by a few percent one reduces $T_c(\text{SDW}_0)$ below $T_c(\text{SDW}_\pm)$ in the whole B domain. On the other hand by decreasing V by a few percents one gets a hump in $T_c(\text{SDW}_0)$ on the left of the transition $\text{SDW}_0 - \text{SDW}_\pm$.

The maximal value of the critical temperature in Fig. 3, $T_c^{\max} \approx 1.1$ K, is considerably smaller than the experimental value of 5.5 K. In this respect we note that T_c^{\max} is essentially model dependent quantity, i.e., the Hamiltonian (3) represents a *minimal* model for understanding the interplay between two SDW phases in the magnetic field. Namely, recent experiments²⁰ suggest that the anion ordering in $\text{TMTSF}_2\text{ClO}_4$ 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. 14. Similar conclusions were obtained also by numerical calculations,¹⁹ but without taking into account the two-component aspect of the order parameter (1). The critical temperature for the SDW_+ subphase can be calculated within Landau theory as in Ref. 15, 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. On this point our picture differs again from the one advanced in Ref. 21, where it was argued that SDW_+ and SDW_- *must* order simultaneously because otherwise T_c would disappear exponentially. As far as we see this kind of locking of the two critical temperatures is not possible. The splitting of the single T_c to $T_c(\text{SDW}_+)$ and $T_c(\text{SDW}_-)$ is a smooth function of the appropriate band parameters, the simplest one being the effective third-neighbor interchain hopping t_3 .¹⁵

IV. CONCLUSION

We 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 T oscillations in normal and SDW phases of $(\text{TMTSF})_2\text{ClO}_4$. Using matrix RPA for SDW susceptibility we reproduce the overall profile of the experimental phase diagram, containing the first-order transition from the (low-field) interband SDW_0 to the (high-field) intraband SDW_- (or SDW_+). The value of the anion potential V fitting experiments in relaxed $(\text{TMTSF})_2\text{ClO}_4$ is large, of the order of interchain hopping t_b .

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