Magnetic oscillations and field-induced spin-density waves in $(TMTSF)_2ClO_4$

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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 random-phase approximation for the SDW susceptibility. We find that the value of the anion potential fitting experiments in relaxed $(TMTSF)_2ClO_4$ is large, of the order of interchain hopping. In particular we predict the magnetic-field-induced transition of the first order between interband SDW_0 and intraband $SDW_\pm$ phases, we reproduce the rapid oscillations with a period of 260 T and the overall profile of the $(TMTSF)_2ClO_4$ 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 salts[2] and in some other low-dimensional compounds.[3] The phenomenon of the FISDW is well understood in the Bechgaard salt $(TMTSF)_2PF_6$ 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 nesting[4] reproduces satisfactorily main experimental data for this salt.

In this paper we concentrate on $(TMTSF)_2ClO_4$, 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 $(TMTSF)_2PF_6$ 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 ClO_4 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 $(TMTSF)_2ClO_4$ 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 arguments[7] suggest three possible nesting wave vectors favoring various SDW phases. The interband nesting $\mathbf{Q}_0$ leads to SDW_0 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 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[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,

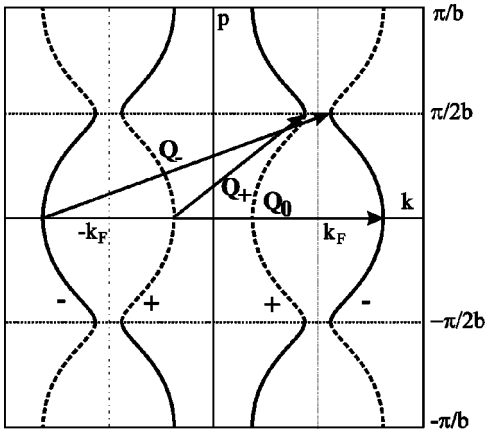

FIG. 1. Two Fermi surfaces of $(TMTSF)_2ClO_4$ (wrapping is highly exaggerated). Wave vectors $\mathbf{Q}_0$, $\mathbf{Q}_+$, and $\mathbf{Q}_-$ correspond, respectively, to SDW_0, SDW_+, and SDW_-.

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

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 $(TMTSF)_2ClO_4$ 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 $V/t_b\gtrsim 1$ it is not allowed to use the quasiclassical 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 $V/t_b\ll 1$, 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 $(TMTSF)_2ClO_4$. 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

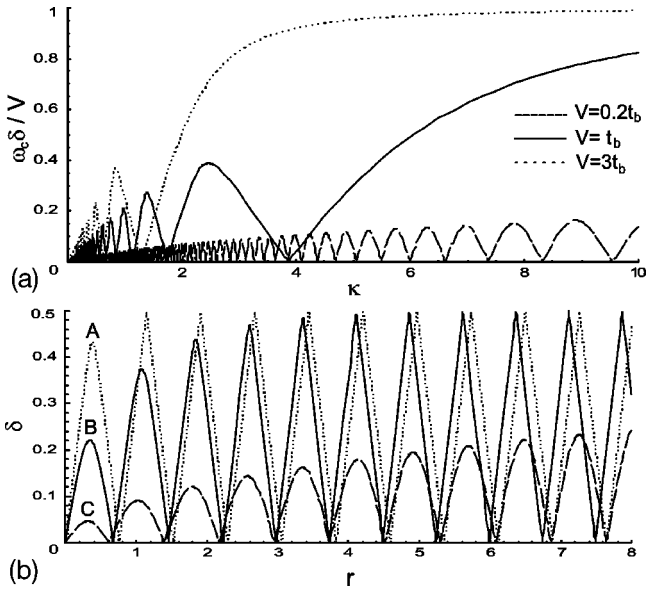

FIG. 2. (a) Energy ratio $\omega_c\delta/V$ as a function of the magnetic breakdown parameter κ for several values of V/t_b. (b) Dependence of δ on r for $\theta=10°(A)$, $45°(B)$, and $80°(C)$.

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

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

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 $\Psi_f(x,p)$,

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

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,

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

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

corresponding to effective hoppings[25] between odd and even neighbors, respectively. We diagonalize H_0 by the unitary transform

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

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 $if v_F \partial_x$ we get $\theta(z)=v_F^{-1}\int^z dz \tilde{\mathcal{T}}(z)$ and a system of differential equations for functions α and β,

$$if v_F \alpha_f'(z) = -\mathcal{T}(z)\alpha_f(z) - V\beta_f^*(z),$$

$$if v_F \beta_f'(z) = -\mathcal{T}(z)\beta_f(z) + V\alpha_f^*(z). \tag{6}$$

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, $v_F=2\times10^5$ m/s, and $b=7.7\times10^{-10}$ 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] $E_k \to f v_F(k-k_f) \pm \omega_c\delta$ with $\delta \to (V/\omega_c)\mathcal{J}_0(4t_b/\omega_c)$, $\mathcal{J}_0$ being the Bessel function. On the other hand the spectrum of Gor'kov and Lebed[17] is reproduced for weak anion potential, $V/t_b \ll 1$. The above fit, as well as other insights[20,28] however strongly suggest that V in $(TMTSF)_2ClO_4$ is rather large, i.e., comparable to t_b.

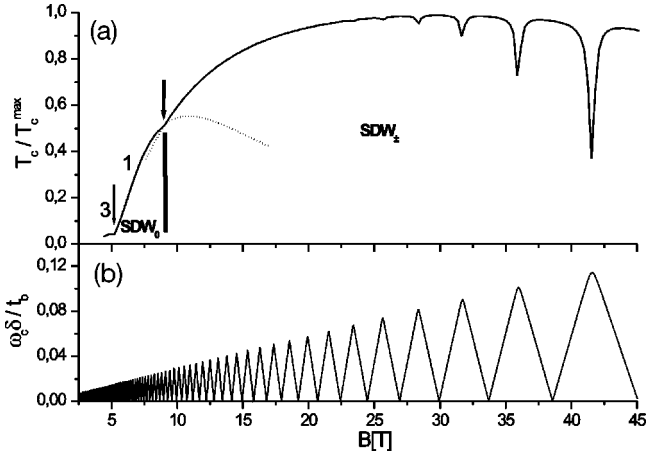

FIG. 3. (a) Phase diagram. (b) Energy ratio $\omega_c\delta/t_b$ on the same magnetic scale as the phase diagram.

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

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

$\mathbf{q}_c$ 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

$$\chi_{hh} = \sum_N \left[\left| I_{h0} \right|^2 P_0 + \frac{1}{2} I_{h+}^2 P_+ + \frac{1}{2} I_{h-}^2 P_- \right],$$

$$\chi_{aa} = \sum_N \left[\left| I_{a0} \right|^2 P_0 + \frac{1}{2} I_{a+}^2 P_+ + \frac{1}{2} I_{a-}^2 P_- \right],$$

$$\chi_{ha} = \sum_N \left[\mathrm{Re}(I_{h0} I_{a0}^*) P_0 + \frac{1}{2} I_{h+} I_{a+} P_+ - \frac{1}{2} I_{h-} I_{a-} P_- \right], \tag{8}$$

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 Nth 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 $I(q_\perp, N)$,

$$I_{h0}(q_\perp, N) = \sum_n (a_n b_{N-n} - \hat{b}_n \hat{a}_{N-n}) e^{i(n-N/2)q_\perp},$$

$$I_{h+}(q_\perp, N) = \sum_n (\hat{a}_n \hat{a}_{N-n} + b_n b_{N-n}) e^{i(n-N/2)q_\perp},$$

$$I_{h-}(q_\perp, N) = \sum_n (a_n a_{N-n} + \hat{b}_n \hat{b}_{N-n}) e^{i(n-N/2)q_\perp},$$

$$I_{a0}(q_\perp, N) = \sum_n (a_n \hat{a}_{N-n} - \hat{b}_n b_{N-n}) e^{i(n-N/2)q_\perp},$$

$$I_{a+}(q_\perp, N) = \sum_n (\hat{a}_n b_{N-n} + b_n \hat{a}_{N-n}) e^{i(n-N/2)q_\perp},$$

$$I_{a-}(q_\perp, N) = \sum_n (a_n \hat{b}_{N-n} + \hat{b}_n a_{N-n}) e^{i(n-N/2)q_\perp}. \quad (9)$$

There are two important selection rules for these amplitudes, namely, for N even, $I_{h0}(N) = I_{a0}(N) = 0$ 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, $V = 0.85 t_b$, $t_b' = 0.03 t_b$, 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 $TMTSF_2PF_6$, 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 calculated[29] the phase diagram for all values of V and concluded that $V \approx 0.85 t_b$ is indeed the only value fitting the phase diagram obtained in experiments. Namely, increasing V or t_b' by a few percent one reduces $T_c(SDW_0)$ 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 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 experiments[20] suggest that the anion ordering in $TMTSF_2ClO_4$ 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 $T_c(SDW_+)$ and $T_c(SDW_-)$ 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 $(TMTSF)_2ClO_4$. 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 $(TMTSF)_2ClO_4$ is large, of the order of interchain hopping t_b.

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