Ground state of $(TMTSF)_2ClO_4$ in high magnetic fields: The creation of Su-Schrieffer-Heeger solitons

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It is shown that the problem of spin-density-wave formation in a quasi-one-dimensional organic conductor $(TMTSF)_2ClO_4$ in a magnetic field is equivalent to Brazovskii-Dzyaloshinskii-Kirova variant of an exactly solvable one-dimensional problem of Su-Schrieffer-Heeger (SSH) solitons. We demonstrate that the ground state in $(TMTSF)_2ClO_4$ in high magnetic fields corresponds to SSH solitonic superstructure. We calculate the magnetic moment and energy gap in an electron spectrum and discuss recent experimental data in terms of the creation of SSH solitonic superstructure. [S0163-1829(97)03203-7]

Quasi-one-dimensional (Q1D) organic conductors $(TMTSF)_2X$ ($X=ClO_4$, PF_6, etc.) demonstrate a complicated phase diagram in a magnetic field. The primary distinctive feature of this diagram is a cascade of phase transitions between different field-induced spin-density-wave (FISDW) subphases.[1] According to the so-called ''standard model,''[2–4] the explanation of a metal FISDW phase transition is based on the effective ''one-dimensionalization'' of an electron spectrum in a magnetic field.[2] This effect leads to the instability in the ''Peierls-channel,'' which results in the creation of FISDW subphases.[3,4] Most of the properties of FISDW subphases in $(TMTSF)_2PF_6$ can be described within the standard model.[2–4]

The organic conductor $(TMTSF)_2ClO_4$ exhibits some experimental features that cannot be understood in terms of the standard model. Unlike $(TMTSF)_2PF_6$, it demonstrates ''rapid magnetic oscillations'' (RMO) of specific heat[5] and magnetic moment[6,7] in the FISDW state at magnetic fields $15 \leqslant H \leqslant 27$ T . In addition, it was recently shown[7] that at $H \leqslant 27$ T there exist two SDW phases that demonstrate different properties. At higher fields, $H \geqslant 27$ T, experimental data are still controversial.[8–10] In our opinion, experimental data in steady magnetic fields[8] are in favor of the appearance of a semiconducting FISDW phase at $H \geqslant 27$ T that possesses anomalously strong oscillations of resistivity. On the contrary, similar measurements in pulsed magnetic fields[9,10] should be interpreted as resistivity oscillations in a semimetallic phase.

At the moment it is clear that the above-mentioned special features of the phase diagram in $(TMTSF)_2ClO_4$ come from the existence of an anion-ordering (AO) gap in its electron spectrum.[11] Most recent attempts to calculate the phase boundary of the metal-SDW transition in a magnetic field[12,13] utilize an effect of magnetic breakdown (MB) across the AO gap[14] that is believed to occur at $H > H_0 \sim 10$ T.[15] Unfortunately, no attempt to find a ground state of FISDW phase under the condition of MB has yet been made to our knowledge.

In this paper we reveal an analogy between the problem of FISDW formation in a quasi-one-dimensional (Q1D) $(TMTSF)_2ClO_4$ conductor in high magnetic fields and Brazovskii-Dzyaloshinskii-Kirova variant[16] of an exactly solvable 1D problem of Su-Schrieffer-Heeger (SSH) solitons.[17] We show that the production of solitons is energetically favorable at $H \gg H_0$. This allows us to conclude that the ground state of $(TMTSF)_2ClO_4$ in high fields corresponds to the appearance of SSH solitonic superstructure. The mean distance between solitons is found to be periodic in $1/H$. This leads to a periodic dependence of the FISDW energy gap and the magnetic moment on inverse magnetic field in accordance with the observation of RMO in Refs. 5–7.

In the presence of the AO gap, $\Box(y) = \Box \cos(\pi y / b^*)$, Q1D electron spectrum of $(TMTSF)_2ClO_4$ corresponds to four open sheets of the Fermi surface (FS) (see Fig. 1):

$$\epsilon_k^{\pm}(\mathbf{p}) = \pm v_F (p_x \mp p_F) + (-1)^k \sqrt{[2 t_b \cos(p_y b^*)]^2 + \Box^2} + 2 t_c \cos(p_z c^*), \tag{1}$$

where the first term represents free-electron motion along the chains; v_F and p_F are the Fermi velocity and Fermi momentum; $t_b \simeq 250$ K and $t_c \simeq 5-7$ K are the overlapping integrals of electron wave functions in perpendicular directions, **b** and **c**, correspondingly; $\Box \sim 50$ K $\ll 2 t_b$; and $k=1,2$.

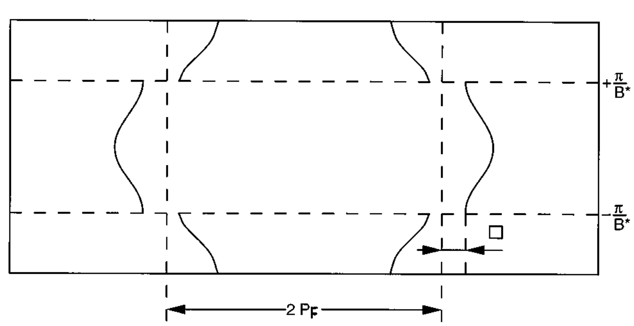

FIG. 1. Fermi surface of the Q1D conductor $(TMTSF)_2ClO_4$ below the AO phase transition.

In all previous papers the FISDW ground state was studied in relation to the formation of the most suitable *harmonic* SDW:[2–4]

$$\Delta'(x,y,z)=\Delta\exp[2ip_Fx+i(\pi/b^*)y+i(\pi/c^*)z]\exp(i\omega_c nx/v_F), \tag{2}$$

which provides the complete nesting of right and left sections of the FS in a magnetic field, where $\omega_c=eHv_Fb^*/c$, n is an integer.[3,4]

Below we choose a more common expression for SDW potential in an incommensurate case:

$$\Delta(x,y,z)=\Delta(x)\exp[2ip_Fx+i(\pi/b^*)y+i(\pi/c^*)z]\exp(i\omega_c nx/v_F), \tag{3}$$

where $\Delta(x)$ is supposed to be a smooth function of x on the scale $x_H\sim v_F/\omega_c$.

In a magnetic field $\mathbf{H}=(0,0,H)$ in the presence of both the AO gap, $\square(y)$, and the SDW gap ,$\Delta(x,y,z)$, the Schrodinger equations in the Landau gauge, $\mathbf{A}=(0,Hx,0)$, have the following form:

$$\epsilon\begin{pmatrix}\psi^+(p_y,x)\\ \psi^+\left(p_y+\dfrac{\pi}{b^*},x\right)\\ \psi^-(p_y,x)\\ \psi^-\left(p_y+\dfrac{\pi}{b^*},x\right)\end{pmatrix}=\hat{T}\begin{pmatrix}\psi^+(p_y,x)\\ \psi^+\left(p_y+\dfrac{\pi}{b^*},x\right)\\ \psi^-(p_y,x)\\ \psi^-\left(p_y+\dfrac{\pi}{b^*},x\right)\end{pmatrix} \tag{4}$$

where the matrix $\hat{T}$ is given by

$$\hat{T}=\begin{pmatrix}-iv_F\dfrac{d}{dx}+2t_b\cos\left(p_yb^*-\dfrac{\omega_cx}{v_F}\right),\square,0,\Delta^*(x)\\ \square,-iv_F\dfrac{d}{dx}-2t_b\cos\left(p_yb^*-\dfrac{\omega_cx}{v_F}\right),\Delta^*(x),0\\ 0,\Delta(x),+iv_F\dfrac{d}{dx}+2t_b\cos\left(p_yb^*-\dfrac{\omega_cx}{v_F}\right),\square\\ \Delta(x),0,\square,+iv_F\dfrac{d}{dx}-2t_b\cos\left(p_yb^*-\dfrac{\omega_cx}{v_F}\right)\end{pmatrix} \tag{5}$$

with e and c being the electron charge and the velocity of light. [Note that Eqs. (4) and (5) are written for the case $n=0$ in Eq. (3) since we consider the high-field limit, $\omega_c\gg t_b'$ (Refs. 2 and 12)].

At high magnetic fields, $H\gg H_0$, we can consider MB phenomenon in the framework of the perturbative approach (see Refs. 12, 18), when the solutions of Eqs. (4) and (5) in a metallic phase $[\Delta(x)=0]$ are symmetrical (S) and antisymmetrical (A) combinations of unperturbed wave functions:[12]

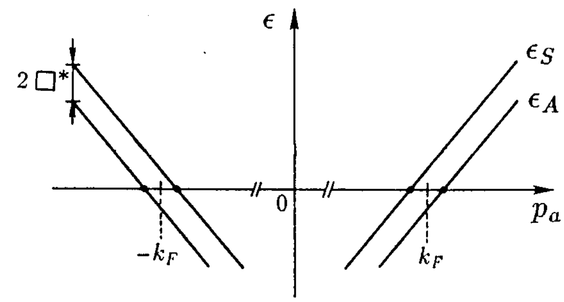

FIG. 2. Electron spectrum for $|p_x\mp p_F|\ll\omega_c/v_F$ corresponding to the magnetic breakdown phenomenon across the AO gap (μ stands for the chemical potential).

$$\begin{pmatrix}\psi_S^{0\pm}(p_y,x)\\ \psi_S^{0\pm}\left(p_y+\dfrac{\pi}{b^*},x\right)\end{pmatrix}=\exp\left[\pm i\frac{(\epsilon-\square^*)x}{v_F}\right]\times\begin{pmatrix}+\exp\left[\pm\dfrac{i\lambda}{2}\sin\left(p_yb^*-\dfrac{\omega_cx}{v_F}\right)\right]\\ +\exp\left[\mp\dfrac{i\lambda}{2}\sin\left(p_yb^*-\dfrac{\omega_cx}{v_F}\right)\right]\end{pmatrix}, \tag{6}$$

$$\begin{pmatrix}\psi_A^{0\pm}(p_y,x)\\ \psi_A^{0\pm}\left(p_y+\dfrac{\pi}{b^*},x\right)\end{pmatrix}=\exp\left[\pm i\frac{(\epsilon+\square^*)x}{v_F}\right]\times\begin{pmatrix}+\exp\left[\pm\dfrac{i\lambda}{2}\sin\left(p_yb^*-\dfrac{\omega_cx}{v_F}\right)\right]\\ -\exp\left[\mp\dfrac{i\lambda}{2}\sin\left(p_yb^*-\dfrac{\omega_cx}{v_F}\right)\right]\end{pmatrix} \tag{7}$$

$$\square^*=\square J_0(\lambda)\simeq\square\sqrt{\omega_c/2\pi t_b}\cos(4t_bc/eHv_Fb^*), \tag{8}$$

where $|\epsilon|,|\square^*|\ll\omega_c$, $J_0(x)$ is the zeroth-order Bessel function, $\lambda=4t_b/\omega_c$.

The calculated energy spectrum of electrons with wave functions (6) and (7) consists of four one-dimensional branches:

$$\epsilon_{S,A}^+(\mathbf{p})=+v_F(p_x-p_F)\pm\square^*,$$
$$\epsilon_{S,A}^-(\mathbf{p})=-v_F(p_x+p_F)\pm\square^* \tag{9}$$

(see Fig. 2). (We have omitted the dependence of energy on p_z since an ideal nesting of the right and the left parts of the FS along the p_z direction is supposed.)

Note that energy splitting between S and A combinations of unperturbed wave functions, $2|\square^*|$, rapidly oscillates in inverse magnetic field with frequency:

$$\Delta(1/H)=4t_b c/ev_F b^*. \tag{10}$$

From Eq. (9) and Fig. 2, it is evident that the wave vector $Q_S=2p_F-2\square^*/v_F$ [which corresponds to the interaction of S electrons with potential (3)] differs from the wave vector $Q_A=2p_F+2\square^*/v_F$ (which corresponds to the interaction of A electrons with the same potential). Therefore, in the presence of the AO gap we have to consider the possibility of an opening of two SDW gaps on the Fermi level. That is why we have introduced the function $\Delta(x)$ in Eq. (3).

It is convenient to represent electron wave functions in the presence of the SDW potential (3) in the form

$$\begin{pmatrix} \psi_S^{\pm}(p_y,x) \\ \psi_S^{\pm}\left(p_y+\dfrac{\pi}{b^*},x\right) \end{pmatrix} = f_S^{\pm}(x)\left\{+\exp\left[\pm i\frac{\lambda}{2}\sin\left(p_y b^*-\frac{\omega_c x}{v_F}\right)\right]\right.$$
$$\left. = r\left\{+\exp\left[\mp i\frac{\lambda}{2}\sin\left(p_y b^*-\frac{\omega_c x}{v_F}\right)\right]\right\}\right\}, \tag{11}$$

$$\begin{pmatrix} \psi_A^{\pm}(p_y,x) \\ \psi_A^{\pm}\left(p_y+\dfrac{\pi}{b^*},x\right) \end{pmatrix} = \pm f_A^{\pm}(x)$$
$$\times\begin{pmatrix} +\exp\left[\pm i\dfrac{\lambda}{2}\sin\left(p_y b^*-\dfrac{\omega_c x}{v_F}\right)\right] \\ -\exp\left[\mp i\dfrac{\lambda}{2}\sin\left(p_y b^*-\dfrac{\omega_c x}{v_F}\right)\right] \end{pmatrix}. \tag{12}$$

Let us substitute Eqs. (11) and (12) into Eq. (4). Taking into account that $|\epsilon,\square^*|\ll\omega_c$, we can average quickly oscillating exponential functions over x on the scale $x\sim\min|v_F/\epsilon,v_F/\square^*|\gg v_F/\omega_c$. As a result of this procedure, we get the following equations:

$$\begin{pmatrix} \epsilon_S-\square^*+iv_F\dfrac{d}{dx}, -\Delta^*(x) \\ -\Delta(x),\epsilon_S-\square^*-iv_F\dfrac{d}{dx} \end{pmatrix}\begin{pmatrix} f_S^+ \\ f_S^- \end{pmatrix}=0, \tag{13}$$

$$\begin{pmatrix} \epsilon_A+\square^*+iv_F\dfrac{d}{dx}, -\Delta^*(x) \\ -\Delta(x),\epsilon_A+\square^*-iv_F\dfrac{d}{dx} \end{pmatrix}\begin{pmatrix} f_A^+ \\ f_A^- \end{pmatrix}=0. \tag{14}$$

Equations (13) and (14) describe two bands of one-dimensional S and A electrons interacting with SDW potential (3) with Fermi energies of S and A electrons being shifted by $\delta(\mu_{S(A)})=\pm\square^*$.

It will be recalled that Eqs. (13) and (14) are valid for electrons with small energies, $|\epsilon|\ll\omega_c$. We also suppose that $\Delta(x)$ is a smooth function of the variable x on the scale $x_H\sim v_F/\omega_c$ and that $\max|\Delta(x)|\ll\omega_c$, $H\gg H_0$ [i.e., $\max|\square^*(H)|\ll\omega_c$]. It is obvious that these inequalities are fulfilled in high fields, $H\gg 10$ T, since $H_0\simeq 10$ T and $\Delta\simeq 10$ K. Under the above-mentioned conditions, Eq. (13) [(14)] determines the energy spectrum of $S(A)$ electrons.

It is important that we are interested in the difference between the energy of the SDW state and the energy of metallic one, $\delta W(H)=W_{\mathrm{SDW}}(H)-W_{\mathrm{met}}(H)$. Due to the logarithmic divergence of the energy of the 1D Peierls state, the quantity $\delta W(H)$ is defined both by electrons with small energies, $|\epsilon_{S(A)}|\ll\omega_c$, and by electrons with large energies, $|\epsilon_{S(A)}|\gg\omega_c$. It has been shown[4] that the contribution of electrons with energies $|\epsilon|\gg\omega_c$ leads only to the renormalization of the constant of electron-electron (''e-e'') interactions:

$$\frac{1}{g^*}=\frac{1}{g}-\ln\left(\frac{\Omega}{\omega_c}\right)>0, \tag{15}$$

where g is a constant of ''e-e'' interactions responsible for SDW pairing, g^* is a renormalized constant of ''e-e'' interactions, and Ω is the cutoff energy. Therefore, we do not need to know the energy spectrum at $|\epsilon|\sim\omega_c$ if we want to determine $\delta W(H)$ with logarithmic accuracy.

Using Eqs. (13)–(15), we can derive the difference between the energy of the SDW state and the energy of metallic one:

$$\delta W(H)=\int_{-\infty}^{+\infty}\frac{|\Delta(x)|^2}{g^*}dx+\sum_{-\omega_c'<\epsilon_S<0}\epsilon_S-\sum_{-\omega_c<\epsilon_S^0<0}\epsilon_S^0$$
$$+\sum_{-\omega_c'<\epsilon_A<0}\epsilon_A-\sum_{-\omega_c<\epsilon_A^0<0}\epsilon_A^0. \tag{16}$$

(Here $\epsilon_{S(A)}$ is the energy of the $S(A)$ electrons in a SDW state [Eq. (3)], $\epsilon_{S(A)}^0$ is the energy of $S(A)$ electrons in a metallic state [i.e., when $\Delta(x)=0$]; the cutoff energy in a SDW, ω_c', differs from one in a metallic state, ω_c, in order to keep the same numbers of electrons in all summations in Eq. (16) (see Ref. 16).)

From a mathematical point of view, the problem of minimizing of the functional (16) is equivalent to the problem of minimizing of the energy of 1D conductor with two nonequivalent chains in the presence of lattice distortion[16]. The potential, $\Delta(x)$, minimizing energy (16) is known to be a superstructure of SSH solitons.[16,17] In the incommensurate case an isolated soliton is known to carry spin, but it does not carry charge.[16] It is possible to prove that in our particular case solitonic superstructure $\Delta(x)$ has two characteristic scales, $x_1\sim v_F/|\square^*|$ and $x_2\sim v_F/\max|\Delta(x)|$, and thus $\Delta(x)$ is a smooth function of the variable x on the scale $x_H\sim v_F/\omega_c$, as was suggested. We stress that, unlike harmonic SDW, the SSH solitonic superstructure opens two gaps in the electron spectrum to minimize the total energy.

Taking advantage of the mathematical analogy between our problem and the problem of Ref. 16, we can investigate the function $\delta W(H)$ for arbitrary H. Below we present expressions for FISDW energy gap, $\Delta(H)$, and magnetic moment, $M(H)$, in the case when $\max|\Delta(x)|\ll\square\sqrt{\omega_c/t_b}$:

$$\Delta(H)=\frac{\Delta_0^2(2\pi t_b)^{1/2}}{\Box\omega_c^{1/2}|\cos(4t_bc/eHv_Fb^*)|},\tag{17}$$

$$M(H)H=-\frac{16\Delta_0^4t_b^2\sin(4t_bc/eHv_Fb^*)}{\Box^2\omega_c^2\cos^3(4t_bc/eHv_Fb^*)},\tag{18}$$

where $\Delta(x)=\Delta_0$ is the solution of the problem (16) in the case $\Box^*=0$. [Note that Eqs. (17) and (18) are valid when $|4t_bc/eHv_Fb^*-\pi/2-\pi n|\gg\Delta_0 t_b^{1/2}/\Box\omega_c^{1/2}\ll 1$, whereas in the narrow vicinities of the zeroths of $\cos(4t_bc/eHv_Fb^*)$, $|4t_bc/eHv_Fb^*-\pi/2-\pi n|\simeq\Delta_0 t_b^{1/2}/\ \Box\omega_c^{1/2}\ll 1$, small islands of the uniform Peierls-Frohlich state have to exist.]

From Eqs. (17) and (18) it follows that energy gap and magnetic moment are periodic functions of $1/H$ in accordance with experiments.[7–9]

To summarize, a ground state of the organic conductor $(\mathrm{TMTSF})_2\mathrm{ClO}_4$ at high magnetic fields, $H\gg H_0\sim 10$ T, is constructed for the first time. The solitonic ground state suggested by us explains the observation of ''rapid magnetic oscillations'' both in a specific heat[5] and a magnetic moment[6,7] as well as the existence of an anomalously strong resistive oscillations at $H>27$ T[8]. We stress that our calculations are strictly valid at $H\gg 10$ T and thus we cannot pretend to describe some exotic experimental features[7] observed in moderate magnetic fields.

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