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Yuriy Skorenkyy & Oleksandr Kramar

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Antiferromagnetic ordering and pseudogap in a model of quasi-1D organic superconductor electronic subsystem

Yuriy Skorenkyy and Oleksandr Kramar

Ternopil Ivan Puluj National Technical University, Ternopil, Ukraine

ABSTRACT

Model of electronic subsystem of Fabre salts $(TMTTF)_2X$ and Bechgaard salts $(TMTSF)_2X$ has been studied theoretically within Green function equation of motion approach with special attention to microscopical mechanism for antiferromagnetic order stabilization and pseudogap formation in the electronic spectrum. Conditions for pseudogap opening in the spectrum and anomalies in dispersion of current carriers are determined. Next-nearest-neighbor hopping is shown to break the perfect nesting property and destabilize antiferromagnetic state. On the basis of these results, experimental phase diagrams for quasi-one-dimensional organic superconductors $(TMTSF)_2X$ and $(TMTTF)_2X$ are interpreted.

KEYWORDS

quasi-1D organic superconductors; quasiparticle energy spectrum; pseudogap; antiferromagnetic ordering

Diversity of organic materials synthesized so far opens new prospects for industrial applications. Among those, organic compounds with electrical and magnetic properties similar to those of transition metal compounds attract much attention [1, 2]. However, distinctive feature of many organic conductors is their strongly anisotropic conductance. Theoretical description of this phenomenon within a microscopical model of electronic subsystem taking into account the combined effect of low dimensionality and strong electron correlations, latter causing, *inter alia*, the Mott insulator-to-metal transition [3] under the external pressure or doping [4], is an open problem. To provide such a description one has to deal, on the equal ground, with magnetic ordering and electron transfer, for which task the Hubbard model [5] has been successfully used for decades.

Psedogap phenomenon, which is a gap in the energy spectrum on a part but not all of the Fermi surface attracts attention due to its presence [6] in high temperature superconductors and also in quasi-one-dimensional organic superconductors like Fabre salts $(TMTTF)_2X$ (here TMTTF denotes tetrametyl-tetrathiafulvalene molecule, X stands for monovalent anion PF_6,AsF_6, ClO_4 or Br) and Bechgaard salts $(TMTSF)_2X$ (here TMTSF stands for tetrametyl-tetraselenfulvalene which differs from TMTTF by substitution of S by Se), which are superconducting for very low temperatures of a few Kelvins, but probably have the same mechanism of superconductivity as HTSC [1, 7]. This causes current interest in electronic spectrum properties of quasi-one-dimensional organic superconductors.

Strong electron correlations cause phase transitions in systems $(TMTSF)_2X$, namely metal-insulator and paramanget-antiferromagnet [2, 8, 9] transitions. To explain abnormal electric

CONTACT Yuriy Skorenkyy skorenkyy@tstu.edu.ua Ternopil Ivan Puluj National Technical University, 56 Ruska St, Ternopil, Ukraine

resistivity and magnetotransport [10, 11] of quasi-one-dimensional systems a notion of "hot spots" has been introduced. These are regions on Fermi surface for which characteristic amplitude of electron dispersion is large. On the basis of experimental data a few phenomenological models have been proposed, in which Fermi surface was divided in hot and cold regions. However the microscopic mechanism which causes occurrence of these peculiarities of electron dispersion on the Fermi surface was not specified. It is worth to note Bourbonnais' "g-ology" model [12] for interacting electrons on connected chains within renormalisation group approach. It was found that inhomogeneity of electron interference in sperconducting and antiferromagnetic channels in k-space leads to hot spots formation on Fermi surface at very low temperatures for $k_\perp = 0, \pm\pi$ ($k_\perp = \pm\pi/2$), where $k_\perp$ is a component of vawe vector transversal to chains. The latter result contradicts that of Mozes and McKenzie [11]. We would like to note that in paper [7] very small values of interchain electron hopping vere considered. In the case of very strong anisotropy the interference may indeed play a dominant role as it is decisive in one-dimensional case. However the method and results of work [7] are completely irrelevant to large interchain hopping and this regime has not been studied so far. Therefore, the mechanism for occurrence of hot spots and the pseudogap phenomenon in quasi-one-dimensional organic systems requires further investigations.

We have carried out a theoretical study of electronic subsystem of Fabre salt $(TMTTF)_2X$ and $(TMTSF)_2X$ with special attention to microscopical mechanism for antiferromagnetic order stabilization and pseudogap formation in the electronic spectrum. In these systems the overlap of wavefunctions of sites belonging to one chain is much larger than the interchain overlap, resulting in one-dimensional conductance in which electronic correlations play an essential role. In a dimerized stack of molecules [8], two nearest neighbour molecules are modeled by a single site. Hopping via anions to the other stack is characterized by far lower amplitude but still does not allow treatment of the system as purely one-dimensional. A quastion we would like to answer is whether in this quasi-one-dimensional system

For consistent description of electron correlations and correlated hopping of electrons we use the configurational representation of the microscopical model [13] taking into account translation processes with nearest-neighbors and next-nearest-neighbors hopping parameters between TMTSF (TMTTF) sites dependent on the applied external pressure or doping of anion subsystem as well as intra- and intersite Coulomb interactions and intersite exchange interaction.

Effective Hamiltonian [13] of the model takes into account the strong intra-site Coulomb, intersite exchange interaction of electrons and correlated hopping of electrons both along the chain (see Fig. 1) and in transversal direction.

$$
\begin{aligned}
H_{eff} &= H_0 + H_{tr} + H'_{tr} + H_{ex} + H'_{ex} + \tilde{H}_{ex} + \tilde{H}'_{ex},\\
H_0 &= -\mu \sum_{i\sigma} (X^{\sigma}{}_i + X_i{}^2) + U \sum_i X_i^2 + \frac{1}{2} N V_0 \kappa u^2,\\
H_{tr} &= \sum_{\langle ij\rangle\sigma} t_{ij}(n) X_i^{\sigma 0} X_j^{0\sigma} + \sum_{\langle ij\rangle\sigma} \tilde{t}_{ij}(n) X_i^{2\sigma} X_j^{\sigma 2},\\
H'_{tr} &= \sum_{\langle\langle il\rangle\rangle\sigma} t'_{il}(n) X_i^{\sigma 0} X_l^{0\sigma} + \sum_{\langle\langle il\rangle\rangle\sigma} \tilde{t}'_{il}(n) X_i^{2\sigma} X_l^{\sigma 2},\\
H_{ex} &= -\frac{1}{2} \sum_{\langle ij\rangle\sigma} J(ij) \left(\left(X_i^{\sigma} + X_i^2\right) \left(X_j^{\sigma} + X_j^2\right) + X_i^{\sigma\bar{\sigma}} X_j^{\bar{\sigma}\sigma} \right),
\end{aligned}
$$

$$H'_{ex} = -\frac{1}{2}\sum_{\langle\langle il\rangle\rangle\sigma} J'(il)\left(\left(X_i^{\sigma} + X_j^{2}\right)\left(X_l^{\sigma} + X_l^{2}\right) + X_i^{\sigma\bar{\sigma}}X_l^{\bar{\sigma}\sigma}\right),$$

$$\tilde{H}_{ex} = -\frac{1}{2}\sum_{\langle ij\rangle\sigma} \tilde{J}(ij)\left(X_i^{\sigma}X_j^{\sigma} - X_i^{\sigma\bar{\sigma}}X_j^{\bar{\sigma}\sigma}\right),$$

$$\tilde{H}'_{ex} = -\frac{1}{2}\sum_{\langle\langle il\rangle\rangle\sigma} \tilde{J}'(il)\left(X_i^{\sigma}X_l^{\sigma} - X_i^{\sigma\bar{\sigma}}X_l^{\bar{\sigma}\sigma}\right). \tag{1}$$

Here X^{kl} are transition operators of site from state $|l\rangle$ to state $|k\rangle$ (Hubbard operators) in configurational representation. Hamiltonian (1) terms have the following meaning. H_0 describes electron subsystem in localization limit (μ is chemical potential, U is Coulomb repulsion of two electrons in the same site), first sum in H_{tr} describes electron hoppings in the chain of TMTTF (TMTSF) molecules between sites occupied with single electron and empty sites with hopping integral $t_{ij}(n)$ (these processes form the lower Hubbard subband), second sum describes hoppings between singly and doubly occupied sites with parameter $\tilde{t}_{ij}(n)$ (these processes form the upper Hubbard subband), H'_{tr} describes the inter-chain electron hoppings with parameters $t'_{il}(n)$ and $\tilde{t}'_{il}(n)$, H_{ex} and H'_{ex} describe direct intersite exchange within one chain and between different chains, respectively; $\tilde{H}_{ex}$ describes effective exchange interaction within the chain with effective exchange integral $\tilde{J}(ij) = \frac{(\tilde{t}_{ij}(n))^2}{U}$, $\tilde{H}'_{ex}$ describes effective inter-chain exchange with integral $\tilde{J}'(il) = \frac{(\tilde{t}'_{il}(n))^2}{U}$ ($\tilde{t}_{il}$ and $\tilde{t}'_{il}$ are integrals of hole-doublon pair creation at translation along the TMTTF molecules chain and between the chains, respectively). In this model, application of the external pressure or chemical doping can be easily modeled by respective renormalization of intersite parameters. Hopping integrals also depend on electron concentration due to correlated hopping of electrons [14] which reduces the hopping probability for occupied sites. Anisotropy of transport characteristics can be described by dimensionless parameter $g = t'_{il}(n)/t_{ij}(n)$ (g may depend on the external pressure or chemical composition). For $(TMTSF)_2X$ with anions PF_6, ClO_4 magnitudes of conductivities along crystallographic directions *a:b:c* are in $10^5{:}10^3{:}10^0$ proportion [11].

We study the dependence of the energy spectrum, obtained with use of generalized projection procedure [13, 14] within Green function method, on wave vector to determine conditions for pseudogap opening in the spectrum and possible occurrence of carrier dispersion singularities ("hot spots"). Energy spectrum of the model obtained with use of this procedure is given by formula

$$E^{\sigma}_{1,2}(\vec{k}) = -\mu + \frac{1}{2}\sum_{x}\left(\alpha_x^{\sigma} t_{\vec{k}}(n) + \beta_x^{\sigma}(\vec{k})\right) - zJ_{eff} \mp \frac{1}{2}\sqrt{(K^{\sigma})^2 + 4\alpha'^{\sigma}_{s}\alpha'^{\sigma}_{p}\left(t'_{\vec{k}}\right)^2} \tag{2}$$

where $t(\vec{k})$ is the hopping integral in $\vec{k}$-representation, subscript x can take values p or s, depending on which of two antiferromagnetic sublattices electron currently sits,

$$K^{\sigma} = \left(\alpha_s^{\sigma} - \alpha_p^{\sigma}\right) t_{\vec{k}}(n) + \beta_s^{\sigma} - \beta_p^{\sigma} + zJ_{eff}, \tag{3}$$

α_x^{σ} and α'^{σ}_{x} are band correlation narrowing factors and $\beta_x^{\sigma}(\vec{k})$ are spin-dependent subband shifts, zJ_{eff} is effective antiferromagnetic exchange integral (direct ferromagnetic exhange may be neglected in the system under consideration). Magnitudes of band correlation narrowing

factors α_x^σ and α'^σ_x can be obtained using the quasi-classical approach for calculation of averages, providing the type of magnetic ordering is known. Spin-dependent subband shifts $\beta_x^\sigma(\vec{k})$ are to be calculated is self-consistent way.

One can see from spectrum (2) that instabilities of one-dimensional quasiparticle current can be caused by electron hoppings between next-nearest chains of molecules. It leads to Fermi-liquid behavior of electronic subsystem or the onset of ordering. Neglecting the correlated hopping in the considered model we reproduce characteristic features of models [12, 15], spectrum (2) resembles corresponding results of papers [12, 16, 17] is high-temperature paramagnet phase and papers [18] in high-temperature antiferromagnet phase. The energy spectrum similar to spectrum (2), has been obtained in paper [18]. This similarity is a formal consequence of analogous calculation technique. Spectrum obtained in paper [18] also describes opening of the energy gap, however of completely different nature, caused by ordering in anion subsystem. A principal distinction of our result from the paper [18] is our ability to describe perfect nesting property removal for Fermi surface.

Worth noting, spectrum (2) has a perfect nesting property (see papers [19–21]) $E(\vec{k}+\vec{Q}) = -E(\vec{k})$ only for $g = 0$. At nonzero values of anisotropy parameter g only a part of Fermi surface can be nested onto Fermi surface with translation by vector $\vec{Q}$, which property is known as imperfect nesting. Nesting property is characteristic for Fermi surface in antiferromagnetic state, deviations from perfect nesting cause instabilities of the antiferromagnetic ordering. For system $(TMTSF)_2NO_3$ at ambiant pressure and temperature 10 K the spin-density-wave vector is very close to optimal nesting vector which leads to pseudogap opening on the Fermi surface followed by transition to antiferromagnet at $T<10$ K [22]. Similarly, in $(TMTSF)_2PF_6$ the transition from antiferromagnetic insulating state to paramagnetic metallic one, for which activation energy and critical temperature can be lowered by applying the external pressure, is accompanied by change of nesting rate for Fermi surface [23].

Fermi surface form is important for explaining [24] minima of angular dependence of magnetoresistance at some angles (Lebed magic angles). The property of Fermi surface imperfect nesting can explain, for example, strong temperature dependence of critical field for $(TMTSF)_2ClO_4$ [25] and $(TMTSF)_2NO_3$ [17]. Our result (2) allows correction of linear spectra of papers [17, 24, 25] by taking into account the strong Coulomb interaction.

Expression in square root in the spectrum (2) depends on sublattice magnetization m, thus on temperature. It leads to a temperature-dependent gap in energy spectrum, which fact is principally important for explanation of anomalous quantum Hall effect in spin density wave state induced by strong magnetic field [26]. For consistent study of this effect, our Hamiltonian is to be generalized by taking into account strong (quantizing) magnetic field explicitly.

At low temperature and ambient pressure the studied systems show antiferromagnetic properties. In the case of antiferromagnetic ordering an expression for concentration of electrons with spin σ reads as (model rectangular density of electronic states is used):

$$n_{s\sigma} = \frac{n - n_{\bar{\sigma}}}{2w} \int_{-w}^{w} \left(\frac{A_\varepsilon^\sigma}{\exp(E_1^\sigma/kT)+1} + \frac{B_\varepsilon^\sigma}{\exp(E_2^\sigma/kT)+1} \right) d\varepsilon, \tag{4}$$

where

$$A_\varepsilon^\sigma = \frac{1}{2}\left(1 - \frac{K^\sigma}{\sqrt{(K^\sigma)^2 + 4\alpha'^\sigma_s \alpha'^\sigma_p \left(t'_{\vec{k}}\right)^2}}\right), \quad B_\varepsilon^\sigma = 1 - A_\varepsilon^\sigma.$$

This expression allows us to obtain an equation for chemical potential calculation $n_{s\uparrow} + n_{s\downarrow} = n$, and the sublattice magnetization $n_{s\uparrow} - n_{s\downarrow} = m$.

For the antiferromagnetic state one has

$$\alpha_i^\sigma = \alpha'^\sigma_i = \frac{(2-n+\eta_\sigma\eta_i m)^2 + n^2 - m^2}{(2-n+\eta_\sigma\eta_i m)},\ \alpha'^\sigma_s\alpha'^\sigma_p = \frac{\left((2-n)^2+n^2\right)^2 - 4m^2(2-n)^2}{4\left((2-n)^2-m^2\right)}$$

$$\beta_i^\sigma = \frac{w(n)(1-n)(n-\eta_\sigma\eta_i m)}{(2-n+\eta_\sigma\eta_i m)},$$

where $\eta_\sigma = 1$ at $\sigma = \uparrow$ and $\eta_\sigma = -1$ at $\sigma = \downarrow$, $w(n)$ is unperturbed half-bandwidth, dependent on electron concentration due to taking into account the correlated hopping [14].

System of equations for magnetization and chemical potential can be rewritten in the form

$$m = \frac{m}{4w}\int_{-w}^{w} d\varepsilon \left(f(E_1) + f(E_2)\right) - \frac{m(2-n)}{4w}\int_{-w}^{w} d\varepsilon \frac{L(\varepsilon)}{\sqrt{D}}\left(f(E_1) - f(E_2)\right),$$

$$n = \frac{2-n}{2wn}\int_{-w}^{w} d\varepsilon \left(f(E_1) + f(E_2)\right) - \frac{m^2(2-n)}{4w}\int_{-w}^{w} d\varepsilon \frac{L(\varepsilon)}{\sqrt{D}}\left(f(E_1) - f(E_2)\right), \quad (5)$$

where $f(E)$ is Fermi distribution function,

$$L = \left(1 - \frac{n^2-m^2}{(2-n)^2-m^2}\right)t_{\vec{k}} - \frac{4w(1-n)}{(2-n)^2-m^2} + zJ_{eff},$$

$$D = (K_\varepsilon^\sigma)^2 + 4\alpha'^\sigma_s\alpha'^\sigma_p g^2\varepsilon^2. \quad (6)$$

Equations (5) allows calculation of the sublattice magnetization as a function of temperature, external pressure and model parameters.

The chemical potential can be calculated numerically from the equation

$$\frac{2n}{2-n} = 2 - \frac{\Theta_N}{\alpha_1 w}\ln\left(\frac{\exp\left(\frac{\mu^*}{\Theta_N}\right) + \exp\left(\frac{\alpha_1 w}{\Theta_N}\right)}{\exp\left(\frac{\mu^*}{\Theta_N}\right) + \exp\left(-\frac{\alpha_1 w}{\Theta_N}\right)}\right) - \frac{\Theta_N}{\alpha_2 w}\ln\left(\frac{\exp\left(\frac{\mu^*}{\Theta_N}\right) + \exp\left(\frac{\alpha_2 w}{\Theta_N}\right)}{\exp\left(\frac{\mu^*}{\Theta_N}\right) + \exp\left(-\frac{\alpha_2 w}{\Theta_N}\right)}\right), \quad (7)$$

here $\Theta_N = kT_N$, $\mu^* = \mu - \beta^* + nzJ_{eff}$, $\alpha_{1,2} = (1 \mp g)\frac{2-2n+n^2}{2-n}$, $\beta^* = \frac{w(1-n)n}{2-n}$.

In quasi-one-dimensional superconductors transitions occur at temperatures close to 10 K so one can neglect temperature dependence of chemical potential and use its ground state value

$$\frac{\mu}{w} = \left(1-g^2\right)\frac{(3n-2)(2-2n+n^2)}{(2-n)^2} - \frac{(1-n)n}{2-n} + \frac{n}{2}\frac{zJ_{eff}}{w}. \quad (8)$$

With use of expression (8) one can numerically calculate dependences of Neel temperature on model parameters and pressure at electron concentration $n = 0.5$, which is the case of $(TMTSF)_2X$.

One can see from figures 2 and 3 that antiferromagnetic ordering of TMTSF (TMTTF) stacks is stabilized by the effective inter-stack exchange. At very low temperature and under pressure which causes superconductind transition in $(TMTSF)_2PF_6$ local magnetic moments develop. Similar picture is realized in the other coumpounds of this family, in particular, $(TMTSF)_2NO_3$ at ambient pressures up to temperature of 10 K, when antiferromagnetic

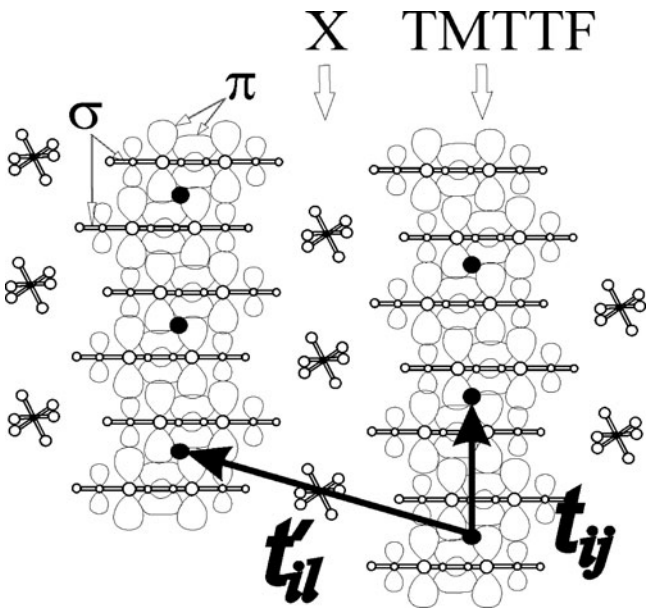

Figure 1. Spatial structure of $(TMTTF)_2X$. π- and σ-orbitals are shown. Hopping integrals t_{ij} within a chain and t'_{il} between chains are indicated.

ordering stabilizes. The external pressure leads to T_N decrease, superconducting transition occurs under 24 kbar pressure at temperature 50 mK [22].

From figure 4, in which the quasi-particle spectrum is shown, it follows that the system's conductance is essentially one-dimensional.

Study of the quasi-particle spectrum calculated within the Green function method proves that the strong electron correlations lead to realization of the conductor-to-insulator and paramagnet-antiferromagnet phase transitions. In the insulator state, energy gap opens in the energy spectrum (see Fig. 5) on Fermi surface or part of it. For the latter case, we obtained the

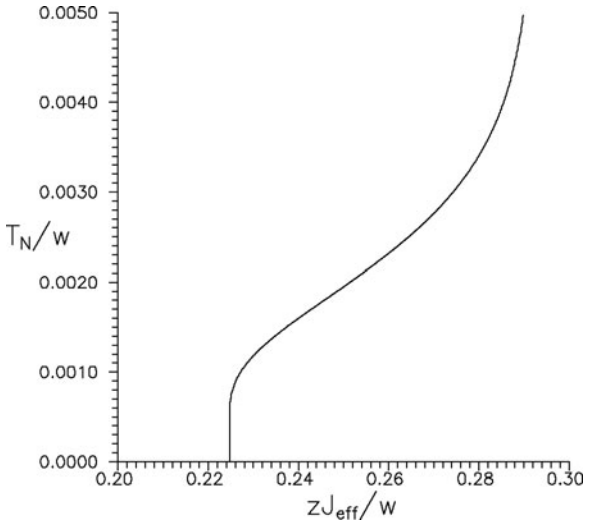

Figure 2. Neel temperature dependence on the effective exchange integral at asymmetry parameter value $g = 0.1$.

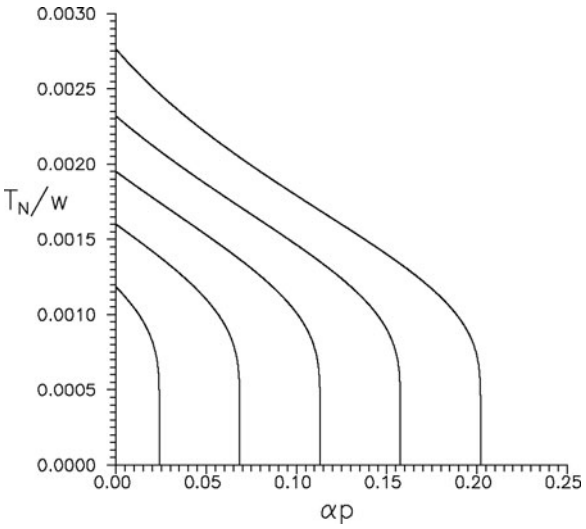

Figure 3. Neel temperature dependence on the lattice deformation at different values the effective exchange integral and $g = 0.1$. Up to down, $zJ_{eff}/w = 0.27, 0.26, 0.25, 0.24, 0.23$.

energy gap dependence on the wave vector. The perfect nesting property is removed mainly by inter-chain hopping and also by correlated hopping of electrons which causes the concentration dependence of transfer integrals.

The energy spectrum (2) allows the calculation of the effective masses of current carriers $m^* = (\frac{\partial^2 E_1(k)}{\partial k^2})^{-1}$, $\tilde{m}^* = (\frac{\partial^2 \tilde{E}_2(k)}{\partial k^2})^{-1}$, where $E_1(k)$ is the energy spectrum of current carriers in the lower $(\sigma\text{–}0)$ subband and $E_2(k)$ – in the upper $(\uparrow\downarrow\ -\sigma)$ subband. From Fig. 6 one can see that effective mass is strongly anisotropic in this model, its maxima correspond to the Brilloin zone "hot spots" and their occurrence is due to antiferromagnetic order. In our opinion, this result is in favor of the suggestion of an antiferromagnetic spin-pairing mechanism for the Bechgaard salts (see [27]).

The model, used in this paper for description of Fabre and Bechgard salts which have a quarter-filled band, can also be applied to ¾ filled band of $(o\text{-}DMTTF)_2X$ salts with (X = Cl, Br, I) [28].

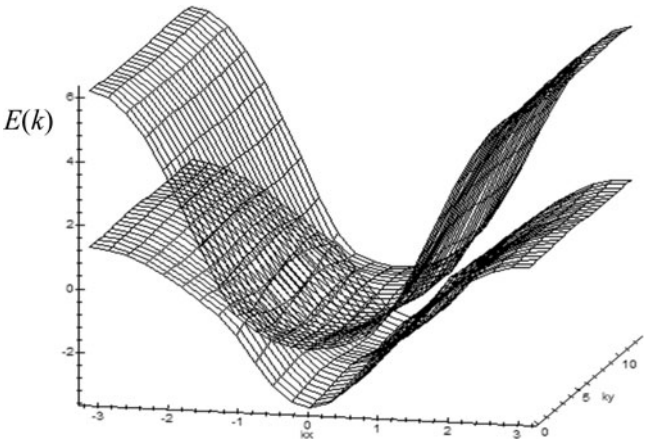

Figure 4. Quasi-1D character of the energy spectrum. Values of the model parameters used are $n = 0.5$, $m = 0.4$, $g = 0.1$, $zJ_{eff}/w = 0.2$.

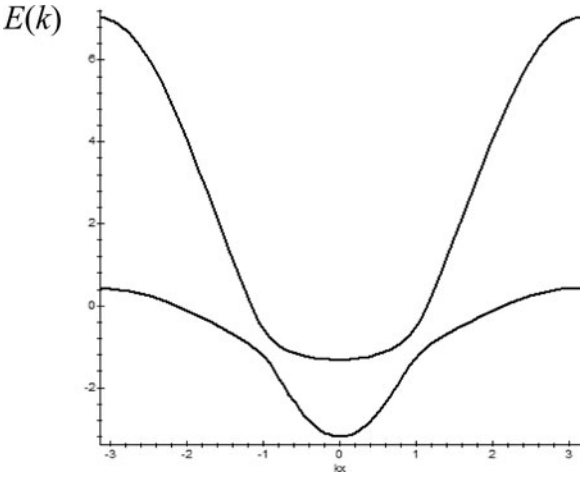

Figure 5. Pseudogap in the energy spectrum of system with antiferromagnetic ordering.

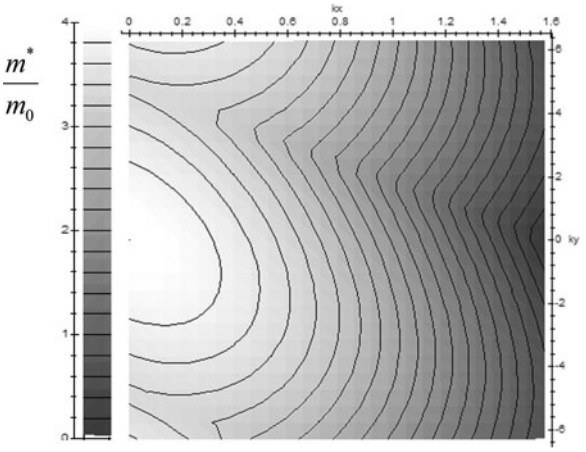

Figure 6. Effective mass asymmetry in the antiferromagnetically ordered state of $TMTSF_2PF_6$ model.

The model used in this paper is quite general to cover wide range of features peculiar to anisotropic molacular conductor and remains analytically treatable, in particular, allows analytical calculation of energy spectrum, conductivity and effective masses of current carriers as well as study of stabilization of various types of ordering in the electronic subsystem of a molecular conductor. The obtained results allow to attribute magnetic ordering and hot spots in $(TMTSF)_2X$ and $(TMTTF)_2X$ to inter-stack effective exchange. The correlated hopping of electrons decreases antiferromagnetic tendencies, however contributes to asymmetry of transport characteristics with respect to half-filling [28].

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