Universal Field-Induced Charge-Density-Wave Phase Diagram: Theory versus Experiment

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We suggest a theory of field-induced charge-density-wave phases, generated by high magnetic fields in quasi-low-dimensional conductors. We demonstrate that, in layered quasi-one-dimensional conductors, the corresponding critical magnetic field ratios are universal and do not depend on any fitting parameter. In particular, we find that $H_1/H_0 = 0.73$, $H_2/H_0 = 0.59$, $H_3/H_0 = 0.49$, and $H_4/H_0 = 0.42$, where H_n is a critical field of a phase transition between the field-induced charge-density-wave phases with numbers n and $n + 1$. The suggested theory is in very good qualitative and quantitative agreement with the existing experimental data in α-$(ET)_2KHg(SCN)_4$ material.

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The high magnetic field properties of organic conductors and superconductors have been intensively studied [1,2] since the discovery of the so-called field-induced spin-density-wave (FISDW) phase diagrams [3,4]. Phase transitions from the metallic to FISDW phase were successfully explained in terms of the 3D → 2D dimensional crossovers [1,5–11]. In particular, the metal-FISDW phase transition line was calculated in Refs. [5–7], whereas a free energy of the FISDW phases was evaluated for all ranges of temperatures and magnetic fields in Refs. [8,9]. In addition, the so-called three-dimensional quantum Hall effect, experimentally observed in the FISDW phases [1–4], was theoretically explained in Refs. [10,11].

A related phenomenon—the so-called field-induced charge-density-wave (FICDW) phase diagram—was anticipated in Refs. [5,12] and recently experimentally discovered in the α-$(ET)_2KHg(SCN)_4$ conductor [13–18]. Although originally the FICDW phases were predicted to exist due to electron-electron interactions [12], later it was shown [19] that they naturally appeared in a physical picture, where only electron-phonon interactions were taken into account. Note that the phase diagram, suggested in Ref. [12], depends on many parameters such as details of electron-electron interactions, temperature, and anisotropy ratios of a quasi-one-dimensional (Q1D) electron spectrum. In addition, according to Ref. [12], the FICDW phases are always mixed with the FISDW ones. The above-mentioned circumstances make it almost impossible to test the theory [12] and to compare it with the existing experiments [13–18]. In a model [19] based on electron-phonon interactions, there are no FICDW-FISDW mixing effects, but the analysis [19] is oversimplified and, as we stress below, is not in quantitative agreement with the experimental data.

The main goal of our Letter is to suggest a universal theory of the FICDW phase diagram, which does not depend on details of electron-electron and electron-phonon interactions as well as on temperature and details of a Q1D electron spectrum. In particular, we suggest a model, based on electron-phonon interactions, for a general form of a layered Q1D spectrum. We demonstrate that the critical magnetic fields ratios $H_1/H_0 = 0.73$, $H_2/H_0 = 0.59$, $H_3/H_0 = 0.49$, and $H_4/H_0 = 0.42$ (where H_n is a critical field of a phase transition between the FICDW phases with numbers n and $n + 1$) do not depend on any parameter and calculate them. A comparison of the present theory with the experiments [14–18] shows not only qualitative but also quantitative agreement. This justifies the validity of our approach and indicates, in particular, that the electron-electron interactions and FICDW-FISDW mixing effects [12] are not very important.

Let us consider the most general layered Q1D electron spectrum, linearized near its two Fermi surface (FS) sheets,

$$\begin{aligned} \epsilon^{\pm}(\mathbf{p}) &= \pm v_F(p_x \mp p_F) + t_y^0(p_y a_y) + t_z^0(p_z a_z), \\ t_y^0(p_y a_y) &= 2t_y \cos(p_y a_y \pm \alpha), \\ t_z^0(p_z a_z) &= 2t_z \cos(p_z a_z \pm \beta), \end{aligned} \tag{1}$$

which obeys the so-called "nesting" condition [1,2],

$$\begin{aligned} \epsilon(\mathbf{p} + \mathbf{Q_0}) + \epsilon(\mathbf{p}) &= 0, \\ \mathbf{Q_0} &= [2p_F, (\pi - 2\alpha)/a_y, (\pi - 2\beta)/a_z]. \end{aligned} \tag{2}$$

[Here $+(-)$ stands for the right (left) sheet of Q1D FS (1); p_F and v_F are the Fermi momentum and Fermi velocity, respectively; t_y and t_z are overlapping integrals between electron wave functions; $p_F v_F \gg t_y \gg t_z$; α and β are some phase shifts; $\hbar \equiv 1$.] It is well known [1,2,5–9,12,19] that the so-called Peierls instability for "nested" FS (1) results in the appearance of a density wave ground state. Below, we consider a CDW ground state in accordance with the existing experimental data in α-$(ET)_2KHg(SCN)_4$ material [13–18].

If we take into account a small (but finite) nonlinearity in a Q1D electron spectrum (1) along the conducting chains, then we obtain the following electron spectrum:

$$\begin{aligned} \epsilon^{\pm}(\mathbf{p}) &= \pm v_F(p_x \mp p_F) + t_y(p_y a_y), \\ t_y(p_y a_y) &= 2t_y \cos(p_y a_y \pm \alpha) + 2t_y' \cos(2p_y a_y \pm 2\alpha), \end{aligned} \tag{3}$$

with small "antinesting" term $2t_y' \cos(2p_y a_y \pm 2\alpha)$, where $t_y' \sim t_y^2/(p_F v_F) \ll t_y$. [Note that, in Eq. (3), we use a 2D model electron spectrum, since we suggest that $t_y \gg t_z$. In this case, the CDW and FICDW phases always correspond to an ideal nesting vector (2) along the z axis since the corresponding antinesting term is too small: $t_z' \sim t_z^2/(p_F v_F) \ll t_y'$.] The antinesting term in Eq. (3) is known to decrease a stability of the CDW ground state, and, therefore, at high pressures (i.e., large enough values of t_y'), the metallic phase has to be restored [1,2,5–9].

At first, let us discuss the FICDW phase formation, using qualitative arguments. For this purpose, we consider a Q1D electron spectrum (3) in the presence of an external magnetic field, applied along the z axis,

$$\mathbf{H} = (0, 0, H), \qquad \mathbf{A} = (0, Hx, 0). \tag{4}$$

To obtain the electron Hamiltonian in a magnetic field (4) from the spectrum (3), we use the Peierls substitution method $p_x \to -i(d/dx)$, $p_y \to p_y - (e/c)A_y$, and take into account the Pauli spin-splitting effects

$$\left[\pm v_F\left(-i\frac{d}{dx} \mp p_F\right) + t_y\left(p_y a_y - \frac{\omega_c}{v_F}x\right) - \mu_B \sigma H\right] \times \Psi_\epsilon^\pm(x, p_y, \sigma) = \delta\epsilon \Psi_\epsilon^\pm(x, p_y, \sigma), \tag{5}$$

where $\sigma = +1(-1)$ for spin up (down), $\omega_c = e v_F H a_y/c$, and $\delta\epsilon = \epsilon - \epsilon_F$.

It is important that Eq. (5) can be solved and the corresponding wave functions can be determined analytically:

$$\Psi_\epsilon^\pm(x, p_y, \sigma) = \exp(\pm i p_F x)\exp\left(\pm i\frac{\delta\epsilon}{v_F}x\right)\exp\left(\pm i\frac{\mu_B\sigma H}{v_F}x\right) \times \exp\left[\mp\frac{i}{v_F}\int_0^x t_y\left(p_y a_y - \frac{\omega_c}{v_F}u\right)du\right]. \tag{6}$$

Note that since $t_y(y) = t_y(y + 2\pi)$ is a periodic function of y and since $\int_0^{2\pi} t_y(y)dy = 0$, then the last exponential function in Eq. (6) has to be a periodic function of x with a period $2\pi v_F/\omega_c$. Therefore, the wave functions (6) can be rewritten in a form of the Fourier series:

$$\Psi_\epsilon^\pm(x, p_y, \sigma) = \exp(\pm i p_F x)\exp\left(\pm i\frac{\delta\epsilon}{v_F}x\right) \times \exp\left(\pm i\frac{\mu_B\sigma H}{v_F}x\right) \times \sum_{n=-\infty}^{+\infty} A_n(p_y)\exp\left(i\frac{\omega_c n}{v_F}x\right). \tag{7}$$

As it directly follows from Eq. (7), 2D electron spectrum (3) in a magnetic field (4) becomes pure 1D and corresponds to an infinite number of 1D FS, located near $p_x \simeq p_F$ and $p_x \simeq -p_F$,

$$\delta\epsilon^\pm(p_x) = \pm v_F(p_x \mp p_F) + n\omega_c - \mu_B\sigma H, \tag{8}$$

where n is an integer quantum number. Electron spectrum (8) is shown Fig. 1.

Note that a metallic phase with 1D spectrum (8) is unstable with respect to the CDW phase formation because of its 1D nesting properties. Since the FICDW instability corresponds to a pairing of an electron near p_F and a hole near $-p_F$ (and vice versa) with the same spins, then we expect that possible projections along the x axis of the FICDW wave vectors are quantized at low enough temperatures (see Fig. 1):

$$Q_x^n = 2p_F \pm 2\mu_B H/v_F + n(\omega_c/v_F), \qquad \pi T \le \omega_c, \tag{9}$$

where the quantization of the electron spectrum (8) is important. Therefore, at low temperatures, we expect a competition between the quantized FICDW order parameters (9) and have to choose the order parameter corresponding to the highest transition temperature.

Below, we consider a problem about a formation of the FICDW phases due to electron-phonon interactions by means of the Feynman diagram technique [20,21]. In particular, we consider the FICDW order parameter in the following form:

$$\Delta(x, y) = \exp(iQ_x x)\exp(iQ_y y) + \text{c.c.}, \qquad Q_x = 2p_F + q_x, \qquad Q_y = (\pi - 2\alpha)/a_y + q_y \tag{10}$$

(where c.c. stands for a complex conjugated quantity), which allows us to take into account deviations of the FICDW nesting vector from its ideal value (2) both along the x and y axes. In a mean field approximation, a phase transition temperature between the metallic and FICDW phases is defined by the so-called electron polarization operator [20,21]

$$\frac{1}{g^2} = -\int_0^{2\pi}\frac{d(p_y a_y)}{2\pi}\sum_\sigma T\sum_{\omega_n}\int_{-\infty}^{+\infty} dx_1 g_{--}^\sigma(i\omega_n; x, x_1; p_y - Q_y) g_{++}^\sigma(i\omega_n; x_1, x; p_y)\exp[iq_x(x - x_1)], \tag{11}$$

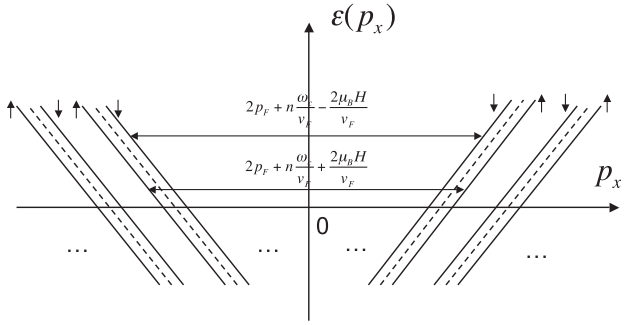

FIG. 1. A schematic view of the quantized electron spectrum (8) near $p_x \simeq p_F$ and $p_x \simeq -p_F$. There exist an infinite number of 1D Fermi surfaces, characterized by quantum number n, with each of them being split due to an electron spin. As a result, at low enough temperatures, there exists a competition between an infinite number of nesting vectors, corresponding to Eq. (9).

TABLE I. Theoretical and experimental [18] values of the critical fields ratios for different pressures.

Critical fields	H_1/H_0	H_2/H_0	H_3/H_0	H_4/H_0
Theory	0.73	0.59	0.49	0.42
$P = 4$ kbar	0.77	0.59	0.40	...
$P = 3.5$ kbar	0.74	0.57	0.37	...
$P = 3$ kbar	0.75	0.56	0.40	...

where g is an electron-phonon coupling constant and ω_n is the Matsubara frequency.

Note that Green functions of electrons near p_F and $-p_F$, $g^{\sigma}_{++}(\ldots)$ and $g^{\sigma}_{--}(\ldots)$, respectively, can be determined from the corresponding electron wave functions (6) and spectrum (8) [21]. After substitution of the Green functions into Eq. (11) and some calculations, we obtain the following equations, which determine the transition temperature to the FICDW phases (10):

$$T_{\mathrm{FICDW}} \simeq \omega_c \exp\left[-\frac{1}{g_{\mathrm{eff}}(t_y')g_{\mathrm{eff}}(H)}\right],$$
$$g_{\mathrm{eff}}(t_y') = \frac{1}{2\ln(t_y'/t_y^*)},$$
$$g_{\mathrm{eff}}(H) = \mathrm{MAX}_{n,q_y}\langle\cos[\phi(x,p_y,q_y)+nx]\rangle_{x,p_y}, \quad (12)$$
$$\phi(x,p_y,q_y) = -\frac{4t_y(q_y a_y)}{\omega_c}\sin(x/2)\cos(p_y a_y) + \frac{4t_y'}{\omega_c}\sin(x)\cos(2p_y a_y),$$

with the quantized x component of the wave vector

$$q_x = \pm 2\mu_B H/v_F + n(\omega_c/v_F). \quad (13)$$

(Here MAX_{n,q_y} denotes a maximization procedure over two components of the FICDW wave vector: the integer quantum number n and continuous variable q_y [see Eqs. (10) and (13)], whereas $\langle\ldots\rangle_{x,p_y}$ stands for an averaging procedure over the variables x and p_y.) Note that a metallic phase is supposed to be stable at $H = 0$, which means that $t_y' > t_y^*$ in Eq. (12), where t_y^* is a value of the parameter t_y', corresponding to a CDW phase transition at $H = 0$ and $T = 0$. The FICDW transition temperature (12) is calculated with the so-called logarithmic accuracy, where we use the following inequalities: $T \ll \omega_c$ and $t_y' \ll t_y$.

We point out that Eq. (12) is different in several important aspects from the typical results (e.g., Refs. [9,22]) of the existing theories of the FISDW phases. First of all, we take into account strong pressure dependence of the FICDW transition temperatures [i.e., function $g_{\mathrm{eff}}(t_y')$], which is not done in Refs. [9,22]. Second, the destructive spin-splitting effects against the FICDW phases decrease function $g_{\mathrm{eff}}(t_y')$ by the factor $1/2$ in Eq. (12). The third (most important) difference is that we retain in the phase $\phi(x, p_y, q_y)$ in Eq. (12) only terms of the order of t_y' and disregard all terms of the order of $(t_y')^3/t_y^2 \ll t_y'$. The above-mentioned approximation makes the suggested theory to be an universal one. Indeed, as directly seen from Eq. (12), the function $g_{\mathrm{eff}}(H)$ depends only on the ratio t_y'/ω_c, and, thus, all critical magnetic fields are proportional to a value of the parameter t_y'. Therefore, their ratios are universal and do not depend on any fitting parameter, in contrast to all previous theories of the FISDW and FICDW phases (e.g., Refs. [9,12,22]).

Equation (12) and its numerical analysis are the main results of this Letter. The distinct feature of Eq. (12) is that the ratios of the FICDW magnetic critical fields [i.e., phase transition fields to the FICDW phases with different quantum numbers (13)] do not depend on any parameter. Numerical calculations of the effective coupling constant $g_{\mathrm{eff}}(H)$ in Eq. (12) for the value of the parameter $t_y' = 4.5\omega_c(H = 1\ \mathrm{T})$ are presented in Fig. 2, where each FICDW phase is characterized by some quantum number n in Eq. (13) (see the figure caption). The calculated ratios $H_1/H_0 = 0.73$, $H_2/H_0 = 0.59$, $H_3/H_0 = 0.49$, and $H_4/H_0 = 0.42$ (where H_n is a critical field of a phase transition between the FICDW phases with numbers n and $n+1$) are compared with the experimental data [18] in Table I. As it follows from the table, there is an excellent agreement between the calculated values H_1/H_0 and H_2/H_0 and the measured ones. As to the measured ratio $H_3/H_0 \simeq 0.4$, it is in satisfactory agreement with the corresponding calculated value $H_3/H_0 = 0.49$. On the other hand, we cannot exclude [23] that, in the experiments [18], in fact, the fourth critical field H_4 was measured instead of the third one H_3. This would give an excellent agreement with the corresponding calculated value $H_4/H_0 = 0.42$.

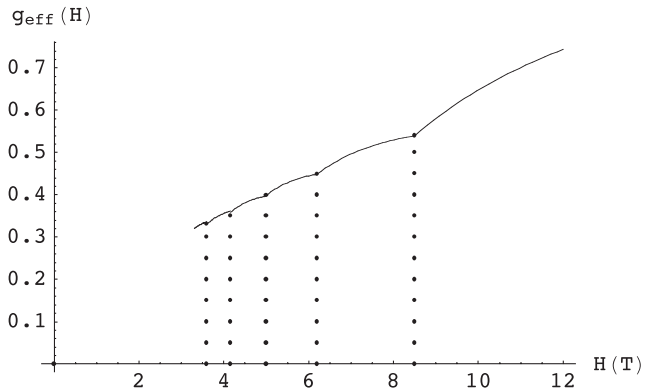

FIG. 2. Numerically calculated effective coupling constant $g_{\mathrm{eff}}(H)$, which defines the metal-FICDW phases transition temperature [see Eq. (12)], is shown by a solid line. Phase transitions between different FICDW phases, characterized by different quantum numbers n in Eq. (13), are shown by dotted lines. Phase $n = 0$ corresponds to $H > 8.5$ T;phase $n = 1$—8.5 T $> H >$ 6.2 T; phase $n = 2$—6.2 T $> H >$ 5 T; phase $n = 3$—5 T $> H >$ 4.15 T; phase $n = 4$—4.15 T $> H >$ 3.6 T; phase $n = 5$—3.6 T $> H$.

Another important property of Eq. (12) is that the phase transition temperature is the same for two wave vectors, corresponding to signs (+) and (−) in Eq. (13).

In our opinion, a very good correspondence between the results of the present theory and the experimental data [14–18] is a strong argument in favor of our model, based on electron-phonon interactions. On the other hand, we point out that the previous simplified model [19] is not in a quantitative agreement with the existing experiments. Indeed, we have numerically analyzed Eq. (11) of Ref. [19] and found that, in the framework of the simplified model, $H_1/H_0 = 0.55$, $H_2/H_0 = 0.38$, and $H_3/H_0 = 0.29$, which is in obvious disagreement with the experimental data [18] (see Table I). Therefore, it is crucial to maximize the FICDW phase transition temperature (12) over two components of the wave vector (10), q_x and q_y, which is not done in Ref. [19]. We note that the following inequalities: $T \ll \omega_c$ and $t_y' \ll t_y$, are used for the derivation of Eqs. (12) and (13). Therefore, we do not take into account the finite temperature effects, described in Refs. [24,25] for the case of the FISDW phases. The next step in our studies will be to suggest a relative universal theory of the FISDW phase diagram and to compare its results with the existing experimental data. This problem will be considered in detail elsewhere [26].

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