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## Dynamics of conversion of conduction electrons into a collective charge-density-wave current

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The exactly solvable model which describes the dynamics of transformation of conduction electrons into nonlinear charge-carrying excitations of charge-density waves in quasi-one-dimensional Peierls-Frohlich conductors is formulated and studied by the inverse scattering transformation method. The pair of self-trapped conduction electrons transform into a charged  $2\pi$  kink localized in a single conducting chain and surrounded by dipoles in neighboring chains. [S0163-1829(96)04123-9]

The commonly accepted model for the Peierls-Frohlich quasi-one-dimensional charge-density-wave (CDW) conductors relates their unusual transport properties, i.e., nonlinear conductivity, memory effects, etc., to the phase of the CDW order parameter,  $\Delta \exp(i\varphi)$ , where the modulus  $\Delta$  is the Peierls energy gap in a quasiparticle spectrum, and  $\varphi$  governs the dynamics of the CDW condensate (the collective excitations of the occupied valence band). The phase dynamics description has been successful in explaining bulk characteristics (see, e.g., Refs. 1–3). However, one principal aspect of the CDW physics, viz. the problem of interaction of the current-carrying CDW-phase deformations with conduction electrons, remains unresolved and still arouses controversial explanations.

The basic feature of quasi-one-dimensional CDW conductors such as e.g., TaS<sub>3</sub>, is the instability of conduction electrons against self-trapping, and conversion into the valence band where electrons transform into additional collective charge carriers. Self-trapping is connected with local gap deformations in conducting chains. The potential barrier for the self-trapping is  $\sim \Delta$ , the time interval of the gap deformation is of order of  $\omega^{-1}$  ( $\omega$  is the frequency of the Peierls phonons which is of order of the Debye frequency),<sup>4–6</sup> and the interchain interaction is of order  $T_C$  ( $T_C$  is the temperature of the Peierls transition). In a weakly coupled array of highly conducting chains,  $T_C \ll \Delta$ , the self-trapping of electrons occurs in individual chains independently, and the charge transformations proceeds in two steps, each characterized by its own time scale  $t_i$ : transfer of conduction electrons into the valence band in a single chain,  $t_1 \sim \omega^{-1}$ , and formation of a collective charge carrier in this chain,  $t_2 \gg t_1$ . The time  $t_2$  is the intrinsic scale of the CDW-phase Hamiltonian at  $\Delta = \text{const}$ .<sup>3–5</sup>

The collective CDW charge  $\rho$ , and the current  $j$ , densities in a single chain are related to phase gradients via the Frohlich relations

$$\rho = \frac{e}{\pi} \frac{\partial \varphi}{\partial x}, \quad j = -\frac{e}{\pi} \frac{\partial \varphi}{\partial t}. \quad (1)$$

When  $q$  electrons are converted into the CDW condensate, the phase acquires a local deformation with the net phase shift

$$\delta\varphi = \varphi(x = \infty) - \varphi(x = -\infty) = q\pi. \quad (2)$$

The final stage of conversion is aggregation of individual phase deformations in different chains to a three-dimensional array,  $t \sim t_3 \gg t_2$ . The time  $t_3$  can be attributed to a long-range Coulomb interaction.<sup>5</sup>

Such arrays are responsible for generation of an excess voltage observed in numerous experiments on mesoscopic CDW samples (see, e.g., Refs. 7–9) and were studied in a series of phenomenological theoretical models<sup>5,10–13,7</sup> in which the problem of the charge transformation dynamics remained unresolved. To our knowledge, the only paper in which an attempt was made to devise a self-trapping mechanism in a one-dimensional (1D) CDW conductor was Ref. 6. It has been shown in Ref. 6 that at low temperatures  $T \ll T_C$ , the transfer of conduction electrons to the valence band occurs via instantons which split from a conduction band a level occupied by two electrons, and push this level toward the valence band. An instanton interpolates between the initial state,  $|in\rangle \equiv |2 \text{ conduction electrons}, \varphi = \text{const}, \Delta = \text{const}\rangle$ , and the final one,  $|f\rangle \equiv |0 \text{ conduction electrons}, \delta\varphi_{in} = 2\pi, \Delta = \text{const}\rangle$ , and describes the self-trapping stage of conversion or the nucleation of a phase-slip-center (PSC). The instanton mechanism is most efficient at a metal-CDW interface,<sup>6</sup> because the instanton action exponentially grows with the distance.

The subsequent evolution of the PSC has not yet been investigated, and it is the aim of this paper to put forward a qualitative microscopic theory of transformation of a trapped electron pair into intrinsically nonlinear phase excitations of a CDW. The scenario of charge transformation is as follows: at time  $t \sim \omega^{-1}$ , a self-trapped pair of electrons creates the initial phase profile  $\delta\varphi_{in} = 2\pi$  ( $|f\rangle$  state) localized on a scale of the Peierls coherence length,  $l_{\text{trap}} \sim \xi_0 = \hbar V_F / \Delta$ , the length  $\xi_0$  not being the intrinsic scale of the phase Hamiltonian;<sup>3,4</sup> thus  $\delta\varphi_{in}$  serves as the initial condition to the phase motion equations. To solve the Cauchy problem, we formulate an exactly solvable model of phase dynamics in a cluster of next-to-nearest-neighbor chains and apply the inverse scattering transformation (IST) method. It appears that over the time  $t_2 \sim \Delta / (\omega T_C)$ , the initial condition  $\delta\varphi_{in}$  transforms into a charged  $2\pi$  kink with a core of order  $\hbar V_F / T_C$  localized in the same chain and surrounded by dipoles in all the other chains.

For definiteness, let us take the zero temperature and study the conversion of two electrons to a cluster of  $Z$  near-

est CDW chains which are oriented along the most conducting  $x$  direction and occupy the semiaxis  $x \geq 0$ . The phase Lagrangian takes the form<sup>3,4</sup>

$$L = \frac{1}{\pi \hbar V_F} \left\{ \frac{\Delta^2 \left( \frac{\partial \varphi_0}{\partial t} \right)^2}{\omega^2} + Z \frac{\Delta^2 \left( \frac{\partial \varphi}{\partial t} \right)^2}{\omega^2} - \frac{\hbar^2 V_F^2}{4} \left( \frac{\partial \varphi_0}{\partial x} \right)^2 - Z \frac{\hbar^2 V_F^2}{4} \left( \frac{\partial \varphi}{\partial x} \right)^2 + 2Z T_C^2 \cos(\varphi_0 - \varphi) \right\}, \quad (3)$$

where  $\varphi_0$  denotes the phase in a central chain, and  $\varphi$  denotes phases in the nearest ones. At  $t=0$  two electrons pass to a central chain:  $\varphi_0(x, t=0) \neq 0$  and  $\varphi(x, t=0) = 0$ .

The Cauchy problem must be supplemented with boundary conditions at  $t=0$ , see Eqs. (5) below. The initial moment  $t=0$  to the Cauchy problem is defined as the one when the phase configuration  $\delta\varphi_{\text{in}}$  escapes from under the autolocalization potential barrier with zero velocity,<sup>6</sup>

$$\frac{\partial \varphi}{\partial t} (t=0) = 0. \quad (4a)$$

As the result of the energy conservation in the process of self-trapping, we obtain

$$2\Delta = \int dx \frac{\hbar V_F}{\pi} \left( \frac{\partial \varphi_0}{\partial x} \right)^2, \quad (4b)$$

and hence

$$\frac{\partial \varphi_0}{\partial x} (t=0) \sim \frac{1}{\xi_0}.$$

The compatibility of this initial condition to the Lagrangian (3) needs special comment. The Lagrangian (3) is actually valid when phase gradients are small compared to  $\xi_0^{-1}$ , when  $|\partial\varphi/\partial x| \sim \xi_0^{-1}$ , the modulus and phase dynamics cannot be separated. Thus, when introducing the above initial condition to the phase dynamics equations solely, we suppose the existence of an intermediate region which we cannot describe analytically where the product  $\xi_0 |\partial\varphi_0/\partial x|$  is numerically but not parametrically small.

Keeping the above in mind, we present the model complete set of boundary conditions in a physically plausible form

$$\frac{\partial \varphi_0}{\partial x} (t=0) \sim \frac{1}{\xi_0}, \quad \frac{\partial \varphi}{\partial x} (t=0) = 0, \quad (5a)$$

$$\int_0^\infty dx \frac{\partial \varphi_0}{\partial x} (t=0) = \delta\varphi_{\text{in}} = 2\pi, \quad (5b)$$

$$\varphi_0(x=0, t=0) = -2\pi, \quad \varphi_0(x \Rightarrow \infty, t=0) = 0, \quad (5c)$$

$$\frac{\partial \varphi_0}{\partial t} (t=0) = \frac{\partial \varphi}{\partial t} (t=0) = 0, \quad (5d)$$

and introduce new variables  $\eta = \varphi_0 - \varphi$  and  $\chi = \varphi_0 + Z\varphi$ . Now the motion equations read

$$\frac{\partial^2 \chi}{\partial \tau^2} - \frac{\partial^2 \chi}{\partial y^2} = 0, \quad (6a)$$

$$\frac{\partial^2 \eta}{\partial \tau^2} - \frac{\partial^2 \eta}{\partial y^2} + (Z+1) \sin \eta = 0, \quad (6b)$$

where

$$\tau = 2T_C \frac{\omega}{\Delta} t, \quad y = \frac{4T_C}{\hbar V_F} x.$$

it is convenient to make the analytical continuation to the whole axis  $-\infty < x < \infty$ , recalling when necessary that the physically significant results should be studied finally on the semiaxis  $0 \leq x < \infty$ , and represent the boundary conditions in a model form

$$\frac{\partial \chi}{\partial \tau} (\tau=0) = \frac{\partial \eta}{\partial \tau} (\tau=0) = 0, \quad (7a)$$

$$\frac{\partial \chi}{\partial y} (\tau=0) = \frac{\partial \eta}{\partial y} (\tau=0) = f(y), \quad (7b)$$

where

$$f(y) = \begin{cases} 2f_0 = \frac{\pi\Delta}{2T_C}, & |y| < \frac{4T_C}{\Delta} = l_0 \\ 0, & |y| > l_0. \end{cases} \quad (8)$$

The charge conservation imposes the topological constriction

$$\int_0^\infty dy \left( \frac{\partial \varphi_0(y, \tau)}{\partial y} + Z \frac{\partial \varphi(y, \tau)}{\partial y} \right) = \int_0^\infty dy \frac{\partial \chi(y, \tau)}{\partial y} = 2\pi. \quad (9)$$

The solution of the linear equation (6a) takes the form

$$\chi(y, \tau) = \frac{1}{2} [F(y - \tau) + F(y + \tau)] - 2\pi, \quad (10)$$

where

$$F(z) = \begin{cases} 2\pi + \chi(z, 0), & z > 0 \\ -\chi(|z|, 0) - 2\pi, & z < 0. \end{cases} \quad (11)$$

To solve the Cauchy problem for the sine-Gordon (SG) Eq (6b), we use the IST.<sup>14</sup> Within the IST approach, the SG equation is related to the linear scattering problem

$$\hat{L}(\eta, \eta_{\bar{\tau}}; \lambda) \Psi(\xi, \bar{\tau}, \lambda) = 0 \quad (12)$$

for the auxiliary spinor Jost function  $\Psi(\xi, \bar{\tau}, \lambda) = \begin{pmatrix} \Psi_1 \\ \Psi_2 \end{pmatrix}$ , where  $\lambda$  is the spectral parameter which takes real positive values, and  $\xi = y/(\sqrt{Z+1})$  and  $\bar{\tau} = \tau/(\sqrt{Z+1})$ . The operator  $\hat{L}$  takes the form

$$\hat{L} = \hat{I} \frac{\partial}{\partial \xi} - \frac{i}{2} \left[ \left( \lambda - \frac{1}{4\lambda} \cos \eta \right) \hat{\sigma}_3 - \frac{\hat{\sigma}_2}{4\lambda} \sin \eta + \frac{\hat{\sigma}_1}{2} (\eta_\xi - \eta_{\bar{\tau}}) \right], \quad (13)$$

where  $\hat{\sigma}_\alpha$  ( $\alpha=1, 2$ , and 3) are the Pauli matrices, and  $\hat{I}$  is the unit matrix.

To construct the solution, one has to calculate the Jost coefficients and to find zeros of the reflection coefficient.<sup>14</sup> Consider the eigenfunctions  $\Psi(\xi, \lambda)$  with the asymptotes at  $\xi \rightarrow -\infty$ ,

$$\Psi_- = \begin{pmatrix} 0 \\ e^{-i\Lambda\xi} \end{pmatrix}, \quad (14)$$

where  $\Lambda = \lambda - (1/4\lambda)$ , and, at  $\xi \rightarrow \infty$ ,

$$\Psi_+ = \begin{pmatrix} b(\lambda)e^{i\Lambda\xi} \\ a(\lambda)e^{-i\Lambda\xi} \end{pmatrix}. \quad (15)$$

The functions  $b(\lambda)$  and  $a(\lambda)$  are the Jost coefficients; zeros of  $a(\lambda)$  define the solutions to the SG equation.

To calculate the initial Jost coefficients, in (13) we set  $\cos \eta(\xi, 0) = 1$ , which is justified by the inequality  $T_c \ll \Delta$ . We obtain

$$a(\lambda, 0) = \exp(i\Lambda l_0) \left( \cos kl_0 - i \frac{\Lambda}{k} \sin kl_0 \right), \quad (16)$$

$$b(\lambda, 0) = i \frac{f_0}{k} \sin kl_0, \quad (17)$$

where  $k = \sqrt{\Lambda^2 + f_0^2}$ .

The spectral equations  $a(\lambda, 0) = 0$  was analyzed in Ref. 15, where it has been shown that the condition  $l_0 f_0 = \pi$  [see Eq. (8)] corresponds to a pair of kinks moving in opposite directions. The kink on the semiaxis  $x \geq 0$  is the only one that is physically relevant for us. Its asymptote at  $t \rightarrow \infty$  is

$$\eta(x, t) = -4 \tan^{-1} \exp \left\{ - \frac{x - Vt}{d \sqrt{Z+1} \left( 1 - \frac{V^2}{C_0^2} \right)^{1/2}} \right\}, \quad (18)$$

where  $d = \hbar V_F / 2T_C$ ,  $C_0 = V_F \hbar \omega / \Delta$ , and

$$V = C_0 \left\{ 1 - \frac{\frac{8\sqrt{Z+1} T_C}{\pi} \frac{\Delta}{\Delta}}{1 + \left[ 1 - \left( \frac{8\sqrt{Z+1} T_C}{\pi} \frac{\Delta}{\Delta} \right)^2 \right]^{1/2}} \right\}. \quad (19)$$

For the parameters of TaS<sub>3</sub>,<sup>2</sup>  $V \approx 0.98 C_0$ .

The solution (18) itself satisfies the topological condition

$$\delta\eta = \eta(x \rightarrow \infty, t) - \eta(x=0, t) = 2\pi, \quad (20)$$

which means that the electric charge of self-trapped electrons remains localized in a central chain at arbitrary time. Indeed, combining the Eqs. (20) and (9), we obtain

$$\delta\varphi_0 = 2\pi \quad \text{and} \quad \delta\varphi = 0. \quad (21)$$

All the other chains contain dipoles with charges

$$q_{\text{dip}} = \pm \frac{2e}{Z+1}; \quad (22)$$

the length of a dipole is proportional to  $l_{\text{dip}} \sim \delta V = C_0 - V$ . The phase distribution for TaS<sub>3</sub> ( $Z=2$ ) is shown schematically in Fig. 1.

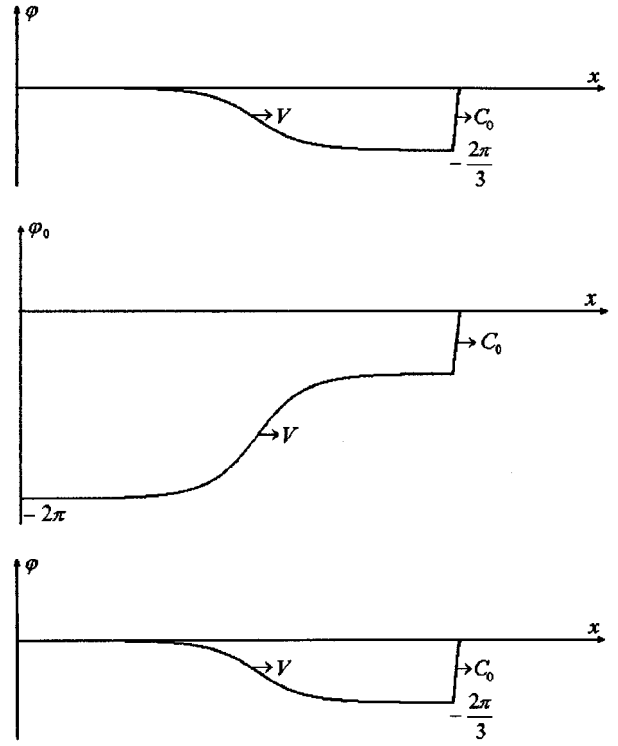


FIG. 1. Phase distribution in central and neighboring chains,  $Z=2$ .

The solution of the Cauchy problem presented above, though physically transparent, leaves a certain feeling of discontent for two reasons: it is accessible only for a model boundary condition (7b) and (8). In addition, the IST method cannot give even an estimate of the time at which the asymptote (18) is reached.

Below, we show qualitatively that the principal results (21) are, in fact, insensitive to a specific shape (8) of  $f(x)$ , and depend only on the topological constrictions (5b) and (5c). We also show that the time of the steady-kink-velocity formation is of the order  $\omega^{-1}$ , and that it depends weakly on the shape of the initial phase profile. The time of a  $2\pi$ -kink steady profile formation is of the order of  $\Delta/\omega T_C \gg \omega^{-1}$ .

Consider the one-parameter two-soliton solution to the SG equation<sup>16</sup>

$$\eta_R(\xi, \tau) = -4 \tan^{-1} \left( \frac{\cosh \left( \frac{U\tau}{\sqrt{1-U^2}} \right)}{U \sinh \left( \frac{\xi}{\sqrt{1-U^2}} \right)} \right), \quad (23)$$

where  $U = V/C_0$ , which obeys the initial conditions

$$\begin{aligned} \eta_R(\xi = +0, \tau = 0) &= -2\pi, \\ \eta_R(\xi \rightarrow +\infty, \tau = 0) &= 0. \end{aligned} \quad (24)$$

To connect  $U$  with the initial space derivative, put  $\eta_R(\xi = l_0, \tau = 0) = -\pi$ , which gives

$$l_0 = \sqrt{1-U^2} \tan^{-1} \frac{1}{U}. \quad (25)$$

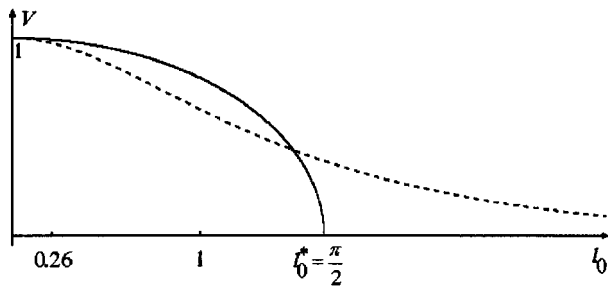


FIG. 2. Dependence of the kink velocity on the initial condition. Solid line: result of the IST analysis; dashed line: probe function (23).

For TaS<sub>3</sub> parameters,  $l_0 \approx 0.26$ , we obtain  $V \approx 0.96C_0$ , in excellent agreement with the IST results (Fig. 2). It is convenient to define the time of establishing the asymptotically steady velocity,  $\tau_\infty$ , as the one when the time derivative of a profile at a point  $\xi$  equals  $U/2$ , i.e.,

$$\left. \frac{d\xi}{d\tau} \right|_{\tau=\tau_\infty} = \frac{U}{2} = \frac{\sinh\left(\frac{U\tau_\infty}{\sqrt{1-U^2}}\right)}{\left[1 + \left(\frac{\cosh\left(\frac{U\tau_\infty}{\sqrt{1-U^2}}\right)}{U}\right)^2\right]^{1/2}}. \quad (26)$$

From (26) we obtain

$$\tau_\infty = \frac{\sqrt{1-U^2}}{U} \operatorname{arc} \sinh\left(\frac{1+U^2}{3}\right)^{1/2}. \quad (27)$$

Combining (27) and (25), we arrive at  $\tau_\infty \sim l_0$  ( $t_\infty \sim 1/\omega$ ).

Mention should be made, at least qualitatively, of how three-dimensional effects modify the results obtained within

the framework of the model (3) which, although it takes into account the interaction with neighboring chains, is essentially a one-dimensional one. As our model (3) mathematically is closely related to the model of a crowdion in a 3D elastic medium, we can use the results for crowdion.<sup>17</sup>

Numerical analysis<sup>17</sup> shows the existence of two scales in the core of a localized charge density  $\rho$ , resulting from the nonlocal nature of the model. At large distances  $x \gg l_C \sim 30d$  ( $d = \hbar V_F/2T_C$ ), the exponential decrease of the density  $\rho$  changes to the power-law relationship  $\rho \sim (l_C/x)^3$ .

Whereas the density peculiar to a charged chain is of order of  $\rho_0 \sim e/d$ , the charge densities in neighboring chains are of order of  $\rho_* \sim e/d10^{-2}$ , and the scale of a dipole field is of order of  $l_d \sim 10d$ . Furthermore, the dynamics of a charge formation in a 3D case is more complicated than in a 1D model. There are two different characteristic velocities in a 3D medium: the longitudinal one  $C_0$ , and the transverse one  $C_\perp \sim (T_C/\epsilon_F)C_0 \ll C_0$ . The charge moving with the velocities  $V$  in the interval  $C_\perp < V < C_0$  produces the Cherenkov radiation with the radiative force:<sup>18</sup>

$$F \sim (V^2 - C_\perp^2) \left(\frac{T_C}{\epsilon_F}\right)^4. \quad (28)$$

It follows from Eq. (28) that the charge which comes into being with the initial velocity  $V \equiv C_0$  loses its velocity to the point  $C_\perp$  over distance  $l_h \sim \xi_0(\epsilon_F/T_C)^3$  during the time  $t_h \sim 1/\omega(\epsilon_F/T_C)^3$ . This radiative effect can be speculatively related to the ‘‘narrow band noise’’ problem.<sup>1,2</sup> Finally, note that in a case of a static kink, the dipole field in neighboring chains transforms to a quadrupole one.<sup>17</sup>

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