

Peierls effect in conducting polymers

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We consider the theory of quasi-one-dimensional conductors with combined Peierls effect, when the dielectric gap in the electron spectrum is the result of two effects: the fixed potential Δ_e of the main structure of the chain and the strain field Δ_i due to the interaction of the lattice with the electrons. We investigate two semi-phenomenological models that describe systems with either a single electron or a noninteger number electrons per molecule of the conducting chain. Certain microscopic models of specific compounds are also considered. Exact solutions are obtained for the periodic structures formed upon appearance of an excess electron density n . In the limit $n \rightarrow 0$ we obtain various charged solitons that act as impurity carriers in the considered systems. We discuss the observed properties of certain polymers (polyacetylene and its derivative, polyphenylene) and of compounds in which a charge-density wave develops against the background of a non-Peierls superstructure.

§1. INTRODUCTION

Most quasi-one-dimensional conductors are, at least at low temperatures, in the dielectric state. In most extensive classes of these substances (charge transfer complexes (CTC), the KCP, MX_3 families, and others, this transition is due as a rule to spontaneous breaking of the system symmetry, resulting in the appearance of a superstructure with a wave vector equal to the diameter of the Fermi surface $2k_F$. On the contrary, among the polymer materials this phenomenon (the Peierls effect) is observed only in the *trans*-(CH) $_x$ polymer. Moreover, in each class of matter, with exception of the MX_3 families, there exist materials that should have dielectric properties even in their basic structure, without formation of lattice structures. Such are all the semiconducting polymers except *trans*-polyacetylene (*cis*-polyacetylene, polydiacetylenes, polyphenylenes, polypyrroles, *et* others, see Fig. 1a–d. Among the CTC and KCP, such materials are those whose conducting properties are ensured by total charge transfer from divalent donors or acceptors to a conducting chain. Representatives of materials of this kind among the KCP are the so-called compounds with non-Peierls substructures,¹ and among the CTC this is the compound $(\text{DBTTF})_8(\text{SnCl})_6$ (Ref. 2). In these substances the chemical formula and the structure are such that the flat Fermi surface lie from the very outset on the boundaries of the Brillouin zone, so that a gap is produced on the Fermi surface also in the absence of the Peierls effect. However, the widths of the forbidden bands in the electron spectra of these substances turn out usually to be of the same order ($\sim 10^{-2}$ eV for CTC, $\sim 10^{-1}$ eV for KCP, and 1–2 eV for polymers) as in analogous substances with spontaneous metal–insulator transitions. This observation, as well as the theoretical models presented below, allow us to assume that in these substances the interaction of the electrons with the lattice exerts a substantial influence on the properties of the dielectric state, although, unlike in Peierls dielectrics, it is not its only cause. We shall call such quasi-one-dimensional substances dielectrics of the combined type, to distinguish them

from the Peierls dielectric and of the opposite limit of a dielectric with a rigid structure.

The deformable component in the lattice structure of a combined-type dielectric plays a substantial role because the Peierls state is characterized by strong self-trapping of the electron and hole excitations,³ which leads to formation of deep polarons and charge or spin solitons (see the review^{4,5}). The Peierls effect in dielectric of the combined type can also lead to strong self-trapping. The most interesting manifestation of these effects can be the existence of bipolarons^{5,6} and solitons with non-integer charge in systems of the type of Fig. 1b. In particular, doping or other methods of charge transfer should lead, just as in *trans*-polyacetylene (see the review⁷) to the appearance of nonparamagnetic carriers.

One can single out a few of the materials that are now of greatest interest and for which the available experimental data permit the construction of definite theoretical models. This is *cis*-polyacetylene (Fig. 1b), which is of interest because of the proximity of its structure to polyphenylene (Fig. 1b), for which data on the nonmagnetic nature of the carriers are available,⁸ and a family of crystalline polymer⁵-polydiacetylenes⁹ (Fig. 1c). The optical properties of *cis*- and *trans*-

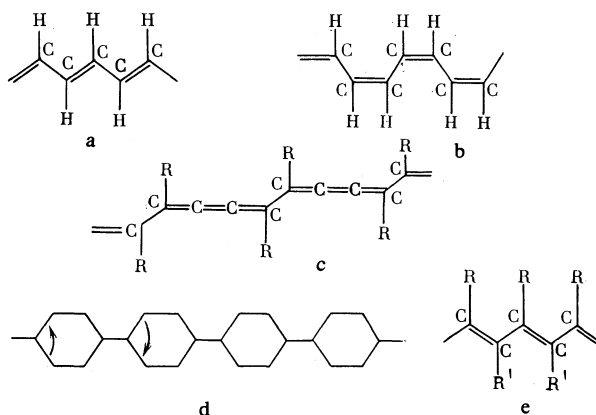


FIG. 1.

(CH)_x were compared in Refs. 4, 6, and 10. A highly interesting case would be that of "orthogonal combination," when the ground state of the system is doubly degenerate (Ref. 6).¹¹ This class can include *trans*-isomers of polyphylacetylene (R=H, R'-phenyl in Fig. 1e),¹⁰ as well as of the polymer (C₂HF_x with R=H, R'=F, the synthesis of which was discussed in the literature.¹¹

In most cases, with the exception of polyphenylene and polypyrrole, the gap E_g in the electron spectrum is small compared with the total width D of the π -electron band (or of the d -electron band for KCP). This permits the use of semiphenomenological continual models, whose parameters can be selected in accord with experimental data. Two characteristic cases can be singled out, designated hereafter as the model C and the transition model AC . The model C , introduced in Refs. 4 and 6, takes into account the interaction of the electrons in the vicinity of the center or of the edge of the Brillouin zone with one nondegenerate deformation mode. This case is most typical of polymers. The AC model describes systems in which the potentials of both the Peierls deformation and of the nonpeierls $2k_F$ superstructure are superimposed on the initial state with non-integer number of electrons per cell ($\rho_0 \neq 1, 2$). Such a system can be regarded as a charge-density wave in a special case of "single commensurability."

The electronic properties of systems describable by models C and AC differ noticeably; this makes it possible in principle to determine their adequacy from experimental data.

We investigate in this paper the ground state of systems described by the models C (§2) and AC (§3) at a finite electron density, and determine the character of the charge excitations. In §4 is given a microscopic derivation of the parameters of the continual model for certain concrete examples and are discussed the most interesting of the known compounds.

§2. CONTINUAL MODEL C FOR A COMBINED STATE

The electronic properties of a one-dimensional dielectric with a narrow forbidden band E_g can be described on the basis of the effective Hamiltonian

$$\hat{H} = \begin{pmatrix} v\hat{k} & \Delta(x) \\ \Delta^*(x) & -v\hat{k} \end{pmatrix}, \quad \hat{k} = -i \frac{\partial}{\partial x}, \quad v = \text{const}, \quad (1)$$

which acts on the electron wave function $\psi = (\psi_+, \psi_-)$. (We use throughout a velocity $v = 1$.) In the homogeneous state $\Delta(x) = \bar{\Delta} = \text{const}$ and $E_g = 2\bar{\Delta}$.

The appearance of $\Delta(x) \neq 0$ in a Peierls dielectric is the result of spontaneous breaking of the system symmetry as a result of the interaction of the metal electrons with the lattice deformations—the Peierls effect. In a dielectric of the combined type, $\Delta(x)$ can be represented in the form (see Fig. 2a)

$$\Delta(x) = \Delta_e + \Delta_i(x)e^{i\varphi}, \quad \Delta_e = \text{const}, \quad \varphi = \text{const}. \quad (2)$$

Here Δ_e is the potential produced by the basic structure of the chain, for example by the rigid polymer skeleton of the bonds, $\Delta_i(x)$ is the contribution made to the potential $\Delta(x)$ by the deformation of the lattice stabilized by the interaction

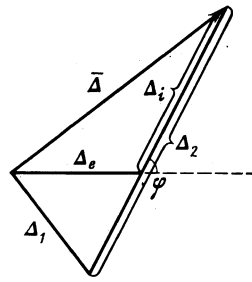


FIG. 2.

with the electrons, and φ is the phase difference between the matrix elements of the interacting electrons with potentials Δ_e and $\Delta_i(x)$.

The parameters Δ_e and φ are determined by the atomic structure of the initial lattice. For certain concrete examples, the values will be determined in §4 below. $\Delta_e = 0$ for a Peierls dielectric (e.g., *trans*-(CH)_x, see Fig. 1a).

The lattice deformation energy is determined only by the component $\Delta_i(x)$. Therefore the functional of the system energy should be of the form

$$W\{\Delta(x)\} = \sum_{E < \mu} E + \int \frac{\Delta_i^2(x)}{g^2} dx, \quad E = E\{\Delta(x)\}, \quad (3)$$

where g is the electron-phonon interaction constant, E are the eigenvalues of the operator (1), and μ is the chemical potential of the electrons. We introduce the functions $u_E(x)$, $v_E(x)$, $\Delta_2(x)$ and the parameter Δ_1 in accord with the formulas (see Fig. 2)

$$\begin{aligned} \Delta(x) &= [-i\Delta_1 + \Delta_2(x)]e^{i\varphi}, \\ \Delta_1 &= \Delta_e \sin \varphi, \quad \Delta_2 = \Delta_i(x) + \Delta_e \cos \varphi, \\ \psi_{\pm}(x) &= 2^{-1/2}(u_{\pm} \pm v_{\pm}). \end{aligned} \quad (4)$$

In terms of the variables U_e , v_E , Δ_1 and Δ_2 the functional of the energy (3) takes the form

$$W\{\Delta(x)\} = \int \left\{ \sum_{E < \mu} [-i(u_E^* v_E' + v_E^* u_E') + \Delta_1(u_E^* u_E - v_E^* v_E) + i\Delta_2(v_E^* u_E - u_E^* v_E)] + \frac{1}{g^2}(\Delta_2 - \Delta_e \cos \varphi)^2 \right\} dx, \quad (5)$$

where $u_E = u(E, x)$, $v_E = v(E, x)$, $f' = df/dx$.

The components u and v of the wave function satisfy the equations

$$u_E' - \Delta_2 u_E = i(E + \Delta_1)v_E, \quad v_E' + \Delta_2 v_E = i(E - \Delta_1)u_E. \quad (6)$$

From (6) follow equations for the functions $u_E(x)$ and $v_E(x)$:

$$\begin{aligned} u_E'' + (E^2 - \Delta_1^2 - p)u_E &= 0, \quad p = \Delta_2^2 + \Delta_2', \\ v_E'' + (E^2 - \Delta_1^2 - q)v_E &= 0, \quad q = \Delta_2^2 - \Delta_2' \end{aligned} \quad (7)$$

and the normalization conditions

$$\int u_E^* u_E dx = \frac{E + \Delta_1}{2E}, \quad \int v_E^* v_E dx = \frac{E - \Delta_1}{2E}. \quad (8)$$

Varying (5) with respect to $\Delta_2(x)$ we obtain the self-consistency condition

$$\frac{2}{g^2}[\Delta_2(x) - \Delta_e \cos \varphi] + \frac{1}{i} \sum_{\mathbf{r} < \mathbf{r}_0} (u_{\mathbf{r}}^* v_{\mathbf{r}} - u_{\mathbf{r}} v_{\mathbf{r}}^*) = 0. \quad (9)$$

The system of equations (6)–(9) determines the sought set of wave functions $u_E(x)$ and $v_E(x)$ and the deformation $\Delta_2(x)$. We shall show that, just as the basic models of the Peierls effect, this functional system reduces to an algebraic one and is thus solved on the class of the so-called finite-band potentials.¹⁴ The potentials p and q in the Schrödinger equations (7) satisfy the stationary Korteweg–de Vries (KdV) equation or one of the higher KdV equations.¹⁴

For the system ((6)–(9) to be solvable in the class of finite-band potentials it suffices that the system deformation energy [the last term of (5)] be representable in the form of a linear combination of integrals of the KdV equation.^{15–19} For the case of a Peierls dielectric ($\Delta_e = 0$) the deformation energy was of the form $W_{ph} \langle \Delta^2 \rangle / g^2$ and was equal, apart from a constant factor, to one of the KdV integrals. It was shown in Refs. 18 and 19 that the functional (5) at $\Delta_e = 0$ has a minimum on a potential with one (in E^2) forbidden band. In the model considered by us the deformation energy contains an additional term $I = \langle \Delta_2(x) \rangle$, which is not in the general case a KdV integral. We shall prove that nonetheless, in the single-band case the functional I is a KdV integral. In this case $p(x)$ and $q(x)$ satisfy the stationary KdV equation, and consequently $\Delta_2(x)$ satisfies the stationary modified KdV (MKdV) equation. The time-dependent MKdV equation takes in the Hamiltonian formulation the form

$$\frac{\partial \Delta_2}{\partial t} = \frac{\partial}{\partial t} \frac{\delta I_1}{\delta \Delta_2} = \frac{\partial}{\partial x} \frac{\delta I_0}{\delta \Delta_2}, \quad I_0 = \frac{1}{2} \langle p^2(x) \rangle,$$

where I_0 is the KdV integral. It follows from this equation that $\partial I / \partial t = 0$, i.e., I is an MKdV and a KdV integral. In the Appendix we shall find the relations between the integrals I_{-1} , I_0 , and I and derive equations for the extremum of the functional (3), without resorting to the coordinate form (9).

We define the boundaries of the electron spectrum $E_1 < E_2 < E_3$ as shown in Fig. 3. The wave functions $u_E(x)$ and $v_E(x)$ and the potentials $p(x)$ and $q(x)$ can be expressed¹⁴ in terms of a function $\gamma(x)$ defined in the forbidden band (E_2^2, E_3^2). The function $\gamma(x)$ is determined from the equation

$$\gamma'(x) = \pm 2[-R(\gamma)]^{1/2}, \quad (10)$$

$$E_2^2 \leq \gamma \leq E_3^2,$$

where

$$R(\gamma) = (\gamma - E_1^2)(\gamma - E_2^2)(\gamma - E_3^2).$$

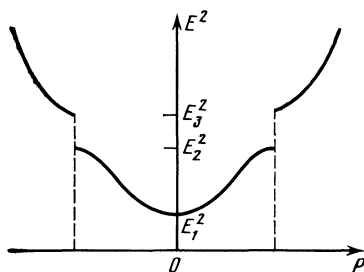


FIG. 3.

The solution of (10) is

$$\gamma(x) = E_3^2 - (E_3^2 - E_2^2) \operatorname{sn}^2(x(E_3^2 - E_2^2)^{1/2}, r), \quad (11)$$

$$r = [(E_3^2 - E_2^2) / (E_3^2 - E_1^2)]^{1/2},$$

where $\operatorname{sn}(\xi, r)$ is the Jacobi elliptic sine.

The potentials $q(x)$ and $p(x)$ are expressed in terms of the function $\gamma(x)$ in the following manner:

$$q(x) = E_1^2 + E_2^2 + E_3^2 - \Delta_1^2 - 2\gamma(x), \quad (12)$$

$$p(x) = E_1^2 + E_2^2 + E_3^2 - \Delta_1^2 - 2\gamma(x + x_0). \quad (13)$$

The parameter x_0 will be determined below. From (12) and (13) we can obtain $\Delta_2(x)$:

$$\Delta_2(x) = [E_1^2 + E_2^2 + E_3^2 - \Delta_1^2 - \gamma(x) - \gamma(x + x_0)]^{1/2}. \quad (14)$$

Following the method of Ref. 14, we obtain for the wave functions $u_E(x)$ and $v_E(x)$ expressions that satisfy the normalization condition (8):

$$v_E(x) = \left(\frac{E - \Delta_1}{2E} \right)^{1/2} \left[\frac{E^2 - \gamma(x)}{A_E} \right]^{1/2} \times \exp \left\{ \pm i R^{1/2}(E^2) \int_0^x \frac{dy}{E^2 - \gamma(y)} \right\}, \quad (15)$$

$$u_E(x) = c \left(\frac{E + \Delta_1}{2E} \right)^{1/2} \left[\frac{E^2 - \gamma(x + x_0)}{A_E} \right]^{1/2} \times \exp \left\{ \pm i R^{1/2}(E^2) \int_{x_0}^{x+x_0} \frac{dy}{E^2 - \gamma(y)} \right\},$$

where $c = \text{const}$ and $|c| = 1$. The normalization factor A_E is equal to

$$A_E = \int [E^2 - \gamma(x)] dx = \langle E^2 - \gamma(x) \rangle L, \quad (16)$$

where L is the length of the chain. Substituting the expression for $\gamma(x)$ from (11) in (16) we obtain

$$A_E = L [E^2 - E_1^2 - (E_3^2 - E_1^2) E(r) / K(r)], \quad (17)$$

where $K(r)$ and $E(r)$ are complete elliptic integrals of the first and second kind.

We obtain now the value of the parameter x_0 . We note that it follows from (6) and (15) that

$$\Delta_2(x) = - \frac{v_E'(x)}{v_E(x)} \Big|_{E=-\Delta_1} = \frac{u_E'(x)}{u_E(x)} \Big|_{E=\Delta_1} = \frac{R^{1/2}(\gamma) + R^{1/2}(\Delta_1^2)}{\gamma - \Delta_1^2}. \quad (18)$$

Substituting (18) in (14) we have after simple transformations

$$x_0 = \frac{1}{k} F \left[\arcsin \frac{k}{(E_3^2 - \Delta_1^2)^{1/2}}, r \right], \quad k = (E_3^2 - E_1^2)^{1/2}. \quad (19)$$

We obtained a periodic solution for the deformation $\Delta_2(x)$ [Eqs. (14) and (18)], with a period

$$L_0 = 2K(r) / k. \quad (20)$$

The wave functions $u_E(x)$ and $v_E(x)$ are characterized by a wave vector or by a momentum $P(E^2)$. From (15) we obtain

$$P(E^2) = \frac{1}{L_0} \int_0^{L_0} \frac{R^{1/2}(E^2)}{E^2 - \gamma(x)} dx = \left\langle \frac{R^{1/2}(E^2)}{E^2 - \gamma(x)} \right\rangle$$

$$= k \left\{ E(v, r') + \left[\frac{E(r)}{K(r)} - 1 \right] F(v, r') \right\}, \quad (21)$$

$$v = \arcsin[(E^2 - E_1^2)/(E_2^2 - E_1^2)]^{1/2}, \quad r'^2 = 1 - r^2.$$

For density of states we obtain from the general theory¹⁴

$$\frac{dP}{dE^2} = \frac{A_E}{2LR^{1/2}(E^2)}. \quad (22)$$

The $E^2(P)$ spectrum is shown in Fig. 3. There are two allowed bands with respect to E^2 : $E_1^2 < E^2 < E_2^2$ and $E_3^2 < E^2$. From (21) it follows here that

$$P(E_1^2) = 0, \quad P(E_2^2) = \pi k / 2K(r). \quad (23)$$

In addition we have $P(E_3^2) = P(E_2^2)$.

With the aid of (23) we find that the total number of states (with allowance for the spin) in each of the allowed bands $-E_2 < E < E_1$ and $E_1 < E < E_2$ is equal to

$$n = (2/\pi)P(E_2^2) = k/K(r). \quad (24)$$

Equation (24) yields the first relation between the three parameters E_1 , E_2 , and E_3 . Substituting in (9) the expression for $\Delta_2(x)$ from (18), for $v_E(x)$ from (15) and for $u_E(x)$ from (6) and using (10), we obtain the equation

$$J_1 \gamma(x) + J_2 = 0, \quad (25)$$

where

$$J_1 = \frac{1}{g^2} + \sum_{E < \mu} \frac{E^2 - \Delta_1^2}{EA_E}, \quad (26)$$

$$J_2 = \frac{\Delta_e \cos \varphi}{g^2} + \sum_{E < \mu} \frac{R^{1/2}(\Delta_1^2)}{EA_E}. \quad (27)$$

Equation (25) is identically satisfied if $J_1 = J_2 = 0$. The summation in (26) and (27) is with the aid of (22). The sum in (26) diverges logarithmically. We regularize it by subtracting from the equation $J_1 = 0$ a like equation with the parameter values

$$E_1 = E_2 = \Delta_1 = 0, \quad E_3 = \Delta_0, \quad \Delta_0 \sim \epsilon_F e^{-1/\lambda}, \quad (28)$$

corresponding to the homogeneous state in the Peierls model ($\Delta_e = 0$) with the same coupling constant λ and with an occupation number $n = 0$. Here $2\Delta_0$ is the gap in the Peierls dielectric. As a result we get from (26) and (27) the relations

$$F(\beta, t) - \frac{\Delta_e \cos \varphi}{\lambda} \frac{kE_2}{[-R(\Delta_1^2)]^{1/2}} = 0, \quad (29)$$

$$(E_3^2 - \Delta_1^2)F(\beta, t) - (E_3^2 - E_2^2)\Pi(\beta, r', t) + \frac{1}{2} E_2 k \ln \frac{\Delta_0^2}{E_3^2 + E_2^2 - E_1^2} = 0, \quad (30)$$

where $F(\beta, t)$ and $\Pi(\beta, r', t)$ are elliptic integrals of kind I and III respectively,

$$\beta = \arcsin \frac{E_2}{E_3}, \quad t = \frac{E_3}{E_2} r', \quad r' = \left(\frac{E_2^2 - E_1^2}{E_3^2 - E_1^2} \right)^{1/2}. \quad (30a)$$

Equations (24), (29), and (30) determine the spectrum param-

eters E_1 , E_2 , and E_3 in terms of the constants Δ_0 , Δ_e , φ , λ as functions of the electron density n .

The ground-state energy $W_0(n)$ is determined by the value of the functional (3) if relations (24), (29), and (30) are satisfied. Using (22), we write $W_0(n)$ in the form

$$L^{-1}[W_0(n) - W_0(0)] = \left\langle \frac{(\Delta_2 - \Delta_e \cos \varphi)^2}{g^2} \right\rangle - \frac{[(\bar{\Delta}^2 - \Delta_1^2)^{1/2} - \Delta_e \cos \varphi]^2}{g^2} - \frac{1}{\pi} \int_{E_1^2}^{E_2^2} \frac{\langle E^2 - \gamma(x) \rangle E dE^2}{R^{1/2}(E^2)} + \frac{2}{\pi} \int_0^{P_m} (P^2 + \bar{\Delta}^2)^{1/2} dP, \quad (31)$$

where $2\bar{\Delta}$ is the gap in the homogeneous state of the considered combined Peierls model.

In (31) we subtracted from the energy (5) the value of the energy of the same system at $n = 0$. In the calculation of (31) account must be taken of the connection between the cutoff momentum P_m and the cutoff energy E_m (see Ref. 18). It can be easily obtained, e.g., from Eq. (7):

$$E_m^2 = P_m^2 + \langle P + \Delta_1^2 \rangle = P_m^2 + E_3^2 + E_2^2 - E_1^2 - 2(E_3^2 - E_1^2) \frac{E(r)}{K(r)}. \quad (32)$$

Calculations yield

$$L^{-1}W_0(n) = \frac{1}{\pi} \frac{F(\beta, t)}{E_2 k} [E_1^2 E_2^2 - \Delta_1^2 (E_3^2 + E_2^2 - E_1^2) + 2\Delta_1^2 k E(r) n] + \frac{1}{\pi} E_2 k E(\beta, t) + \frac{2kE(r)}{\pi} - \frac{2\Delta_e \cos \varphi}{g^2 K(r)} \frac{[-R(\Delta_1^2)]^{1/2}}{E_3^2 - \Delta_1^2} \Pi \left[\frac{E_3^2 - E_1^2}{E_3^2 - E_1^2}, r \right] + \frac{3}{2\pi} \left(E_1^2 - E_2^2 - \frac{1}{3} E_3^2 \right) + \text{const.} \quad (33)$$

Here $\Pi(\alpha, r)$ is a complete elliptic integral of kind III.

We obtain the distribution of the charge density

$$\rho(x) = e \sum_{E < \mu} (v_E^* v_E + u_E^* u_E), \quad (34)$$

where e is the electron charge. Substituting in (34) Eqs. (10), (15), and (22) we get

$$\rho(x) - n = \frac{eK(r')}{\pi} \frac{E_3^2 - E_2^2}{k} \left\{ \text{sn}^2(kx, r) + \text{sn}^2[k(x+x_0), r] - \frac{2}{r^2} \left[1 - \frac{E(r)}{K(r)} \right] \right\} - \frac{\Delta_1 (E_3^2 - E_2^2)}{g^2 [-R(\Delta_1^2)]^{1/2}} \times \{ \text{sn}^2[k(x+x_0), r] - \text{sn}^2(kx, r) \}. \quad (35)$$

Equations (10), (15), (18), (19), (24), (30), (33), and (35) determine the principal physical characteristics of the system. At $\Delta_e = 0$ they go over the corresponding expressions obtained earlier¹⁶ for the Peierls model.

At $\Delta_e \neq 0$ the orthogonal case $\varphi = \pi/2$ is singled out. In this case, as already indicated, the system is symmetric with respect to the substitution $\Delta_i \rightarrow -\Delta_i$, just as at $\Delta_e = 0$. From the self-consistency condition it follows at $\varphi = \pi/2$ that $E_1 = \Delta_1 = \Delta_e$, and the equation for the deformation

$\Delta_2(x)$ takes the simple form:

$$\Delta_2(x) = (E_3^2 - E_2^2)^{1/2} s^{1/2} \operatorname{sn} \left[x \left(\frac{E_3^2 - E_2^2}{s} \right)^{1/2}, s \right], \quad (36)$$

$$r = \frac{2s^{1/2}}{s+1}$$

which corresponds to a domain structure in a system with doubly degenerate ground state.

In the low density limit $n \rightarrow 0$ we can obtain the expressions of Ref. 6 for spinless excitations. The self-consistency conditions (24), (29), and (30) go over into the relations

$$\frac{\pi \Delta_e \cos \varphi}{g^2} = (\bar{\Delta}^2 - \Delta_1^2)^{1/2} \ln \frac{\bar{\Delta}}{\Delta_0}, \quad (37)$$

$$\ln \frac{\bar{\Delta}^2}{\Delta_0^2} = \frac{E_0^2 - \Delta_1^2}{E_0 (\bar{\Delta}^2 - E_0^2)^{1/2}} \arcsin \frac{E_0}{\bar{\Delta}}, \quad (38)$$

where $\bar{\Delta} = E_3, E_0 = E_1 = E_2$.

Equation (37) determines the value of the gap in the homogeneous state. Equation (38) determines the position of the local level E_0 . Separating from the expression (33) for the energy the term proportional to the density n , we obtain the chemical potential or the excitation energy per electron:

$$\mu = \frac{\delta W}{\delta n} = \frac{2}{\pi} [\bar{\Delta}^2 - E_0^2]^{1/2} + 2 \arcsin \left(\frac{E_0}{\bar{\Delta}} \right) \frac{\Delta_1^2}{\pi E_0} + \frac{2 \Delta_e \cos \varphi}{g^2} F \left[\left(\frac{\bar{\Delta}^2 - E_0^2}{\bar{\Delta}^2 - \Delta_1^2} \right)^{1/2}, r \right]. \quad (39)$$

In the general case at $\varphi \neq \pi/2$, $\Delta_e \neq 0$ the periodic structure (11) and (18) describes as $n \rightarrow 0$ a lattice of solitons of the bipolaron type, with characteristic dimension

$$x_0 = \frac{1}{(\bar{\Delta}^2 - E_0^2)^{1/2}} \ln \frac{(\bar{\Delta}^2 - E_0^2)^{1/2} + (\bar{\Delta}^2 - \Delta_1^2)^{1/2}}{(E_0^2 - \Delta_1^2)^{1/2}} \quad (40)$$

and with density $n/2$, and hence with activation energy $E_{\text{rot}} = 2\mu$. From (35) we find readily that the bipolaron charge, as expected is $e^* = 2e$. It is remarkable, however, that at $\varphi \neq 0$ the distribution of the charge is asymmetric and the bipolaron has a dipole moment

$$\mathcal{P}_{\text{rot}} = (e_+ - e_-) x_0, \quad e_{\pm} = e \left[1 \pm \frac{2}{\pi} \frac{\Delta_1}{E_0} \arcsin \frac{E_0}{\bar{\Delta}} \right], \quad (40a)$$

$$e^* = e_+ + e_- = 2e.$$

The presence of the dipole moment is natural, for in the tight-binding limit the system considered by us goes over into a chain of molecules with heterogeneous chemical bond. In the orthogonal case $\varphi = \pi/2$ we have $x_0 \approx 1/n$ as $n \rightarrow 0$, and the deformation (36) describes a lattice of kinks with period $1/n$, i.e., the kink activation energy, as a result of (39), is equal to

$$E_k = \mu = \frac{2}{\pi} (\Delta_0^2 - \Delta_e^2)^{1/2} + \frac{2}{\pi} \Delta_e \arcsin \frac{\Delta_e}{\Delta_0}. \quad (41)$$

From (35) we readily calculate the charge of the kink:

$$e^* = \frac{2}{\pi} e \arcsin \frac{\Delta_e}{\Delta_0}, \quad (42)$$

which turns out to be fractional and dependent on the pa-

rameters of the medium. (The remainder of the charge is uniformly distributed with a density $(e - e^*)/L$ that does not depend, in contrast to the local charge (42), on the kink position²⁰.) Only this fraction e^*/e of the elementary charge can contribute to the current. Equations (41) and (42) agree with the results of Refs. 6 and 20.

The foregoing results go over into the equations given without proof in Ref. 6, if one introduces the parameter β or α connected with E_0 by the relations

$$E_0 = \bar{\Delta} \cos \beta = [\bar{\Delta}_e^2 \sin^2 \varphi + (\bar{\Delta}^2 - \Delta_e^2 \sin^2 \varphi) \operatorname{ch}^{-2} \alpha]^{1/2}.$$

We recall also that according to the results of Refs. 4 and 6 the spin excitations of a system with $n = 0$ constitute, at $\varphi \neq \pi/2$, polarons with spin $1/2$ and charge $\pm e$. At $\varphi = \pi/2$ the spin excitations are solitons of the kink with a singly filled level $E_0 = \Delta_1$. They have a spin $1/2$ and a fractional charge $\pm (2/\pi)e \arcsin(\Delta_e/\Delta_1)$.

In conclusion we dwell briefly on the parameters of the homogeneous ground state ($n = 0$). It follows from (38) that in the orthogonal case ($\varphi = \pi/2$) the following relation always holds

$$\bar{\Delta} = (\Delta_e^2 + \Delta_1^2)^{1/2} = \Delta_0,$$

i.e., the value of the gap with its value Δ_0 for a Peierls dielectric with the same coupling constant. Consequently $\Delta_i \neq 0$ and spontaneous dimerization takes place only if $\Delta_e < \Delta_0$.

At $\varphi = \pi/2$ we always have $\Delta_i \neq 0$, and in the weak binding limit ($\lambda \ll \cos \varphi$) we have $\Delta_i > \Delta_e$, i.e., at a sufficiently narrow forbidden band the Peierls effect becomes decisive.

§3. THE AC TRANSITION MODEL

We consider in this section systems with a combined Peierls effect which develops on a chain with a noninteger number ρ_0 of electrons per molecule: $\rho_0 \neq 0, 1, 2$.

In such systems there should appear a charge density wave (CDW) with a wave vector

$$q_0 = 2k_F^{(0)} = (\pi/a_0) \rho_0,$$

where a_0 is the period of the undeformed chain. We shall be interested in cases when simultaneously with the CDW the chain is modulated by a fixed periodic perturbation having the same wave vector q_0 . As a rule this situation arises in systems where the charge transfer to the conducting chain is from chains of divalent ions whose density is $\rho_0/2$, i.e., the period coincides with $2\pi/q_0$.

Regarding both sources of the superstructure as perturbations, we can separate the deformation $\rho(x)$ and the electron wave functions $\psi(x)$:

$$\zeta(x) \propto \Delta(x) \exp(iq_0 x) + \Delta^*(x) \exp(-iq_0 x),$$

$$\psi(x) = \psi_+(x) \exp(iq_0 x/2) + \psi_-(x) \exp(-iq_0 x/2).$$

The system energy functional should be of the form

$$W_{AC} = W_A + \delta W_{AB} + \delta W_{AC}, \quad (43)$$

where

$$W_A = \sum_{E < \mu} E + \int \frac{|\Delta|^2}{g^2} dx, \quad (44)$$

$$\delta W_{AB} = \frac{1}{2g^2} \int [B\Delta^2 + B^*\Delta'^2] dx, \quad B = |B|e^{2i\theta} = \text{const}, \quad (45)$$

$$\delta W_{AC} = \frac{1}{g^2} \int [C\Delta + C^*\Delta'] dx, \quad C = |C|e^{i\alpha} = \text{const}. \quad (46)$$

The energy levels E in (44) should be determined as the eigenvalues of the Hamiltonian (I), where the matrix element $\Delta(x)$ should be regarded as an arbitrary complex quantity, and the constants B and C are microscopically determined complex quantities. The appearance in the functional (43) of the invariants δW_{AB} and δW_{AC} in addition to the main functional W_A of the Peierls-Fröhlich model is due to the existence of a non-Peierls superstructure, as a result of which q_0 becomes the reciprocal-lattice vector.

Obviously, the condition that the lattice be stable prior to the turning on of the electron-phonon interaction imposes the restriction $|B| < 1$. Considering the increment δW_{AB} which corresponds to the model of "weak twofold commensurability,"²¹ we see that the electrons interact with two phonon modes Δ_+ and Δ_- , to which correspond different interaction constants λ_+ and λ_- :

$$\Delta(x) = e^{-i\theta}(\Delta_+ + i\Delta_-), \quad \lambda_{\pm}^{-1} = \lambda^{-1}(1 \pm B), \quad \lambda = g^2/\pi v. \quad (47)$$

It is easy to verify that at $|B| \gtrsim \lambda$ the interaction with the electrons is substantial only for the deformation Δ_- , which is characterized by the largest coupling constant λ_- . The deformation Δ_+ remains fixed and is determined from the condition that the lattice part of the functional (43) be a minimum. As a result we return to the model C (§2) with the parameters

$$\Delta_+ = \Delta_e \cos \varphi = \frac{\cos(\alpha - 2\beta)}{1 + |B|^{-1}}, \quad \Delta_- \sin \varphi = \frac{\sin(\alpha - 2\beta)}{1 - |B|^{-1}}.$$

For a weak non-Peierls superstructure $|B| \ll 1$ and $|C| \sim |B|^{1/2}$, so that both phonon modes Δ_+ and Δ_- are now significant. The small perturbations (45) and (46) can affect only the phase of the deformation $\Delta(x)$:

$$\Delta(x) = \Delta_0 e^{i\chi(x) - \alpha}, \quad \Delta_0 \sim e_{\mathcal{F}} e^{-1/\lambda} = \text{const}.$$

In the functional W_A it suffices here to take into account only the changes due to the gradients $\chi(x)$ of the phases. Bearing in mind that $|C| \sim |B|^{1/2}$, we can neglect the term δW_{AB} and obtain the effective functional

$$W_{AC} \approx \int dx \left[\frac{v}{4\pi\lambda} \left(\chi'^2 - \frac{1}{2\pi} \mu \chi' - \frac{1}{l^2} \cos \chi \right) \right], \quad l^2 = \frac{\lambda v}{8|C|\Delta_0}. \quad (48)$$

The chemical potential μ is determined from the specified density n of the supplementary electrons

$$n = (\rho - \rho_0)/a_0 = \langle \chi' \rangle / 2\pi. \quad (49)$$

The ground state and the elementary excitations of the functional (48) are well known (see, e.g., Ref. 22). At small n the state of the system is characterized by a lattice of solitons with a period $n/2$. The charge excitations of the system at $n = 0$ are individual solitons with a phase shift $\pm 2\pi$, a charge $\pm 2e$, and an energy

$$E_s = 2\mu|_{n=0} = \frac{2^{\frac{1}{2}} v}{\pi l}.$$

We consider now the question of spin excitation in the AC model. We recall that^{3,20} in model A the spin excitation is an uncharged amplitude soliton with reversal of the sign of $\Delta(x)$ in the region $\sim \xi_0 = v/\Delta_0$. It is easily seen that in this region the perturbations δW_{AB} and δW_{AC} are negligible in terms of the parameters $|B|/\lambda \ll 1$ and $|C|/\Delta_0 \ll 1$. However, the influence of the perturbations δW_{AC} becomes substantial at large distances, since this term admits in the ground state of only phase changes that are multiples of 2π , whereas a phase shift π takes place in the vicinity of the soliton core. As a result the phase χ should become equalized at large distances, and the final form of the soliton becomes

$$\Delta(x) = \Delta_0 \text{th} \frac{x-x_0}{\xi_0} e^{i\chi(x) - \alpha}, \quad \chi(\pm\infty) = 2\pi n.$$

In analogy with the influence of the ordering between chains, investigated in Ref. 20, we can determine the distribution of the phase $\chi(x)$ from the extremum condition for the functional (48) with boundary conditions

$$\chi(0) = \pi/2, \quad \chi(-\infty) = 0, \quad \chi(+\infty) = 0, \quad 2\pi.$$

We find that at $x < 0$

$$\chi(x) = -4 \arctg \left[\text{tg} \frac{\pi}{8} e^{x/l} \right], \quad (50a)$$

and at $x > 0$ two cases are possible:

$$\chi(x) = -\chi(-x), \quad \chi(x) = 2\pi + \chi(-x). \quad (50b)$$

In the case (50a) the soliton acquires a charge e concentrated in tails of size $l \gtrsim \xi_0$. In case (50b) the soliton as a whole remains neutral, and the charges $\pm e/2$ in the left and right tails of the solitons cancel each other.

§4. MICROSCOPIC MODELS FOR CERTAIN COMPOUNDS

We consider in this section simple microscopic models for certain important cases, and deduce from them the phenomenological models investigated in §§2 and 3. The most interesting are the systems in which the spontaneous lattice deformation is via displacement of the molecule positions, and the external fixed potential is due to the nonequivalence of the lattice sites. Such systems are divided into two types: substances with one electron per molecules, $\rho_0 = 1$, shown in Fig 2e, and substances with fractional number ρ_0 , e.g.,² the compound $(\text{DBTTF})_8(\text{SnCl}_6)_3$ with $0 = 5/4$. (The anions SnCl_6 give up two electrons each to the conducting stack of DBTTF molecules.)

In the tight-binding approximation such a system can be described by an energy functional of the displacements u_n :

$$W = W\{u_n\} = \sum_{n < m} \varepsilon + \frac{k}{2} \sum_n (u_{n+1} - u_n)^2, \quad (51)$$

$$e\psi_n = -(t_n \psi_{n+1}^* \psi_n + \text{c.c.}) + v_n \psi_n, \quad t_n = t[1 - \alpha(u_{n+1} - u_n)]. \quad (51a)$$

In the cases of interest to us, the molecular potentials v_n are fixed and periodic with a period $2/\rho_0$. In the case of small forbidden bands one can transform to the slowly varying complex functions $c(n)$ and $\psi_I(n)$, writing

$$\begin{aligned}
t_n &= t_0 + C(n) \exp(i\pi\rho_0 n) + \text{c.c.}, \\
\psi_n &= \psi_+(n) \exp(i\pi\rho_0 n/2) + \psi_-(n) \exp(-i\pi\rho_0 n/2), \\
V(n) &= V_0 + V_1 \exp(i\pi\rho_0 n), \quad V_0, V_1 = \text{const.}
\end{aligned} \quad (52)$$

In the approximation (52) we obtain from (51a) an equation for the eigenvalues with the Hamiltonian (2) at

$$\begin{aligned}
E &= \varepsilon + 2t \cos(\pi\rho_0/2) - 2V_0, \quad v = 2t \sin(\pi\rho_0/2), \\
\Delta &= 2V_1 - 2C(n) \exp(-i\pi\rho_0/2).
\end{aligned} \quad (53)$$

The functional (51) takes the form

$$W = \sum_{\varepsilon < \mu} \varepsilon + \frac{1}{g^2} \int dn |C(n)|^2, \quad g = \frac{\alpha t}{k^{1/2}}. \quad (54)$$

At $\rho_0 = 1$ the functions $C(n)$ and V_1 are real, and we obtain from (1), (53), and (54) the functional of the model C in the orthogonal case $\varphi = \pi/2$ with $\Delta_1 = 2V_1 = \text{const}$, $\Delta_2 = 2C(n)$.

For fractional values of ρ_0 the functions $C(n)$ is arbitrary and complex, while V_1 is a complex constant whose phase depends on the shifts of the ion sublattices and of the conducting chains. As a result we obtain the functional (43)–(46) of the AC model at $B = 0$ and $C = 2V_1/g^2$. A finite value of B is obtained only when fourth-order anharmonicities are taken into account.

§5. CONCLUSION

In the present paper and in Refs. 4 and 6 we investigated the ground state and the elementary excitations of systems with combined Peierls effect. It is based on the assumption that the assumed combination of internal and external breaking of the symmetry of the ground state of a quasi-one-dimensional conductor. The concept of a combined state can be useful also in an analysis of systems where the spontaneous breaking of the symmetry is not tracked explicitly, such as polymers of the polyphenylene and polypyrrole. The effect considered is expected to be strong in all cases when the optical gap in the spectrum is small compared with the total width of the electron band. An exception is the special case of orthogonal mixing, when the internal symmetry breaking (dimerization of the bonds) is spontaneous, but takes place only if the electron–phonon interaction constant exceeds a certain threshold or, equivalently, the external symmetry breaking (dimerization of the sites) is weak enough. This is precisely why the combined state may also not be realized in the as yet little investigated physical compounds such as the polymers of Fig. 1d. In the general case the response to the external component Δ_e should be strong and tend to increase the full gap. This result agrees with data on isomers of polyacetylene, where $2\Delta_0 = 1.45$ eV in *trans*-polyacetylene and $2\bar{\Delta} = 2.05$ eV in *cis*-polyacetylene. In the case of polydiacetylenes, the gap $2\bar{\Delta} = 1.6$ eV is too close to the value of the gap in *trans*-polyacetylene, since it must be borne in mind that the field Δ_e is in this case certainly stronger than in the case of *cis*-polyacetylene. It is possible that the structure of the π band in polydiacetylene differs substantially from that in polyacetylene, so that the estimate of $2\Delta_0$ for polydiacetylene from the gap in *trans*-polyacetylene is not

valid. For example, the interaction constant can decrease because of the rigidity produced by the three-dimensional ordering of the large radical groups R —Fig. 1c.

The most general attribute of the combined state can be taken to be that no spin is possessed by the extrinsic carriers, namely the bipolarons in the general model C , the kinks in the orthogonal case, and the 2π -phase solitons in the AC model. In the last two cases there should also be observed, in analogy with spin diffusion in *trans*-(CH) $_x$, a particularly high degree of anisotropy of the mobility, due to the topological nature of the carriers. The intrinsic carriers of both the current and the spin in the general model C should be normal polarons.^{4,6} In the orthogonal case, the intrinsic and extrinsic charge carriers coincide, and the spin carriers [kinks with fractional charge (42)] differ somewhat in energy. The distinguishing property of the AC model is the smallness of the charge-carrier activation energy compared with that of the spin carriers ($2\Delta_0/\pi$) and with the half-width Δ_0 of the optical gap. The agreement of the activation energies in the paramagnetic susceptibility and in the conductivity, observed² in the compound (DBTTF) $_8$ (SnCl $_6$) $_3$, indicate that the model C is more likely to be adequate than AC . This means that the periodic potential of the ion sublattice is not small as assumed in the microscopic model of §3.

APPENDIX

We derive below the self-consistency conditions (26) and (27) by the method of Refs. 17 and 19. The derivation is based on the known spectral properties of a Schrödinger equation with a periodic potential.¹⁴ We present the necessary results. For Eqs. (7) there exists the following expansion of the momentum P of the wave function u_E or v_E in powers of $1/E$ as $E \rightarrow \infty$:

$$P = E + 2 \sum_{n=0}^{\infty} \frac{(-1)^{n+1} I_{n-1}}{(2E)^{2n+1}}, \quad (A.1)$$

where the functionals I_n are known also as integrals of the KdV equation. We shall need some of them:

$$2I_{-1} = - \int U dx, \quad 2I_0 = \int U^2 dx, \quad U = p(x), \quad q(x). \quad (A.2)$$

For the variation δP of the momentum for an m -band potential (m is the number of forbidden bands) with band boundaries E_j^2 we have the formula

$$\frac{\delta P}{\delta u} = - \prod_{s=1}^m [E^2 - \gamma_s(x)] / 2R^{1/2}(E^2),$$

$$R(E^2) = \prod_{j=1}^{2m+1} (E^2 - E_j^2), \quad (A.3)$$

where each function $\gamma_s(x)$ has definition region in the s th forbidden band. On the other hand, from (A.1) as $E \rightarrow \infty$ we have

$$\delta P = - \frac{\delta I_{-1}}{E} + \frac{\delta I_0}{4E^3} + \dots \quad (A.4)$$

It is stated that the $m+1$ integrals I_0, \dots, I_m are independent. For a one-band potential ($m = E_j^2 = E_1^2, E_2^2, E_3^2$) we

obtain from (A.2)–(A.4)

$$R(\varepsilon) = (\varepsilon - E_1^2)(\varepsilon - E_2^2)(\varepsilon - E_3^2), \quad \varepsilon = E^2, \quad (\text{A.5})$$

$$\delta P = [\delta I_{-1}(-2\varepsilon + s) + \frac{1}{2}\delta I_0] / 2R^{1/2}(\varepsilon), \quad (\text{A.6})$$

$$s = E_1^2 + E_2^2 + E_3^2, \quad 2I_{-1} = - \int (\Delta_1^2 + \Delta_2^2) dx, \quad \frac{\delta I_{-1}}{\delta \Delta_2} = -\Delta_2, \quad (\text{A.7})$$

$$2I_0 = \int (\Delta_2^4 + \Delta_2'^2 + 2\Delta_2^2 \Delta_1^2 + \Delta_1^4) dx, \quad \frac{\delta I_0}{\delta \Delta_2} = 2\Delta_2^2 - \Delta_2'' + 2\Delta_1^2 \Delta_2. \quad (\text{A.8})$$

The self-consistency conditions are obtained by varying the energy functional (3) at fixed periods, i.e., at specified numbers of states in each allowed band:

$$\delta W = - \frac{2}{\pi} \int \delta P dE + \delta W_{ph} = 0, \quad (\text{A.9})$$

$$\delta W_{ph} = \delta \int \frac{(\Delta_2 - \Delta_e \cos \varphi)^2}{g^2} = \frac{2}{g^2} \left[\int \Delta_2 \delta \Delta_2 dx - \Delta_e \cos \varphi \int \delta \Delta_2 dx \right]. \quad (\text{A.10})$$

For the problem to be integrable, the variation of the deformation energy (A.10) must be represented as a linear combination of the variations of the integrals I_{-1} and I_0 , which by virtue of (A.3) determine the first term in (A.9). To do this we use the fact that at $m = 1$ the potential U should satisfy the stationary KdV equation¹⁴ and consequently the deformation Δ_2 should satisfy the MKdV equation:

$$\Delta_2'' - 6\Delta_2 \Delta_2' + A\Delta_2' = 0. \quad (\text{A.11})$$

Using (A.11) we have

$$\Delta_2'' - 2\Delta_2^3 + A\Delta_2 = B. \quad (\text{A.12})$$

For the constants A and B we easily obtain the expressions

$$A = 2(s - 3\Delta_1^2), \quad B = 4R^{1/2}(\Delta_1^2). \quad (\text{A.13})$$

With the aid of (A.7), (A.8), (A.12), and (A.13) we obtain

$$-\delta I_0 - 2(s - 2\Delta_1^2)\delta I_{-1} = 2R^{1/2}(\Delta_1^2)\delta I. \quad (\text{A.14})$$

The relation (A.14) establishes a linear connection between I_1 and the integrals I_0 and I_{-1} in the particular case of a single-band potential. Substituting (A.14) and (A.7) we obtain the following expression for the variation of the deformation energy:

$$\delta W_{ph} = - \frac{2\delta I_{-1}}{g^2} + \frac{\Delta_e \cos \varphi}{g^2} \frac{\delta I_0 + 2(s - 2\Delta_1^2)\delta I_{-1}}{2R^{1/2}(\Delta_1^2)}. \quad (\text{A.15})$$

Substituting (A.7) and (A.15) in (A.9) we get

$$\frac{1}{\pi} \int \frac{\delta I_{-1}(-2E^2 + s) + \frac{1}{2}\delta I_0}{2ER^{1/2}(E^2)} dE^2 = \frac{2}{g^2} \left[-\delta I_{-1} + \Delta_e \cos \varphi \frac{\delta I_0 + 2(s - 2\Delta_1^2)\delta I_{-1}}{4R^{1/2}(\Delta_1^2)} \right]. \quad (\text{A.16})$$

Equating in (A.16) the coefficients of the independent variations δI_{-1} and δI_0 we obtain the self-consistency conditions

$$\frac{2\pi\Delta_e \cos \varphi}{g^2} = R^{1/2}(\Delta_1^2) \int \frac{dE^2}{ER^{1/2}(E^2)}, \quad \frac{2\pi}{g^2} = \int \frac{E^2 - \Delta_1^2}{ER^{1/2}(E^2)} dE^2. \quad (\text{A.17})$$

¹The results of the paper of S. A. Brazovskii and N. N. Kirova⁶ for this special occasion recently have been introduced in the papers of M. J. Rice and E. J. Mele¹² and A. Jackiw and G. Semenoff.¹³

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