The effect of umklapp processes on the dynamics of the Peierls–Fröhlich state

S. A. Brazovskii, I. E. Dzyaloshinskii, and S. P. Obukhov

L. D. Landau of Theoretical Physics, USSR Academy of Sciences
(Submitted October 11, 1976)
Zh. Eksp. Teor. Fiz. 73, 1550–1556 (April 1977)

When the period of the Peierls–Fröhlich wave is sufficiently closely commensurate with the lattice period, umklapp processes which can lead, in particular, to a "stepping" (pinning) of the wave become important. We calculate the static correlation functions and find the real and imaginary parts of the permittivity in the strong and weak pinning regimes.

PACS numbers: 77.20.+y

1. INTRODUCTION

Recently the effect of the anomalous Fröhlich conductivity (Binder and Zeller have given a survey of recent papers) in quasi-one-dimensional conductors which experience a Peierls transition has been widely discussed. It has been recognized recently that this effect is responsible for the existence of a steep maximum in the optical absorption spectra in the KCP and TTF–TCNO compounds. Lee, Rice, and Anderson give in the framework of the self-consistent field theory a clear microscopic derivation of the permittivity of a Peierls dielectric, and also considered qualitatively the effect of periodic and random inhomogeneities. They showed in agreement with (1) that the permittivity of a Peierls dielectric is the same as that of a free charged gas with a large effective mass m*:

\[ \epsilon(\omega) = -\frac{4\pi}{m^*} \]

which is caused by the existence of an optically active Goldstone mode. The violation of the translational invariance leads to the appearance of a gap in the spectrum of the oscillations (pinning) and guarantees a finite value of \( \epsilon(0) \). This fact stimulates an increased interest in a study of the effects of periodicity in one-dimensional systems which have been studied with an eye on static effects first of all by the present authors. The problem of the evaluation of the conductivity or of active optical losses in a Peierls dielectric has been studied in a Peierls dielectric state, and also considered systematically the problem of the properties of systems which have been studied with an eye on static effects first of all by the present authors. The problem of the evaluation of the conductivity or of active optical losses in a Peierls dielectric has been studied in a Peierls dielectric state, and also considered systematically the problem of the properties of systems which have been studied with an eye on static effects first of all by the present authors.

2. CORRELATION FUNCTIONS

1. We study the behavior of the correlation function \( K(x) \) of the relative longitudinal displacements \( d(x)/d_0 = \phi \) of a system of non-interacting filaments, where \( d_0 \) is the equilibrium displacement in a Peierls dielectric. The electron-phonon interaction has in the general case the form

\[ \mathcal{H}_{\text{ep}} = \sum_{\mathbf{p}, \mathbf{p}' \neq \mathbf{0}} g(p, p') \phi^* \mathbf{d} \cdot \mathbf{a}(\mathbf{p}) \mathbf{a}(\mathbf{p}'), \]

where \( \mathbf{q} = 2\pi/\mathbf{a} \) is the size of a cell in the reciprocal lattice. We note that the interaction parameter \( g(p, p') \) is independent of the magnitude \( q \) of the vector in the reciprocal lattice which participates in the umklapp process.

2. In the first instance we consider the simplest case where the diameter \( 2d_0 \) of the electron Fermi surface is close to \( q/2 \), i.e., when the number of electrons per elementary cell is close to unity. In the Hamiltonian the direct processes, \( n = 0 \), and the umklapp processes with \( n = 1 \) are then equally important. To describe the system in terms of long-wavelength deformations of the lattice it is convenient to introduce the field \( \phi(x) \) defining it through its Fourier component \( \phi_n \) as

\[ \phi(x) = \sum_n \phi_n e^{2\pi i n x/\mathbf{a}}. \]

It is clear that \( \phi_n \) are \( \delta \) functions, i.e., the field \( \phi(x) \) is real. The free phonon Hamiltonian

\[ \mathcal{H}_{\text{ph}} = \sum_{\mathbf{q}, \mathbf{p} \neq \mathbf{0}} \omega_{\mathbf{q}} \mathbf{a}_{\mathbf{q}}^\dagger \mathbf{a}_{\mathbf{q}} + \sum_{\mathbf{q}, \mathbf{p} \neq \mathbf{0}} \omega_{\mathbf{q}} \mathbf{a}_{\mathbf{q}}^\dagger \mathbf{a}_{\mathbf{q}} + \sum_{\mathbf{q}, \mathbf{p} \neq \mathbf{0}} \omega_{\mathbf{q}} \mathbf{a}_{\mathbf{q}}^\dagger \mathbf{a}_{\mathbf{q}+\mathbf{p}} + \sum_{\mathbf{q}, \mathbf{p} \neq \mathbf{0}} \omega_{\mathbf{q}} \mathbf{a}_{\mathbf{q}}^\dagger \mathbf{a}_{\mathbf{q}+\mathbf{p}} \]

can be expressed analytically in terms of \( \phi(x) \) by virtue of the condition

\[ \frac{\partial \omega_{\mathbf{q}}}{\partial q} = 2\pi q. \]
We can transform the interaction Hamiltonian (1) into
\[ \mathcal{H}_{\text{int}} = \sum_{q} \frac{1}{2} \dot{q}^2 + \frac{1}{2} (\dot{q}^2 + \dot{x}^2) + \frac{1}{2} f(x) \Delta (x - \dot{x}^2 \Delta/8x). \]

We have thus obtained a Hermitian Hamiltonian relative to the real field \( x \). This means that the Lagrangian (or, in the classical limit, the free energy functional) of the field \( x \) must be an even function of the quadratic \( a/ax \). For a weak electron-phonon interaction the dependence of the Lagrangian on the gradients is given by the electron polarizability \( n(q) \) which is a maximum for \( q = 2p_f \).\(^2\)

If \( |q/2 - 2p_f| < \frac{\pi}{4} \), we get, expanding \( \sum_l \frac{f_l(x)}{\sqrt{2}} \) close to the maximum with \( q = 2p_f \pm \frac{\pi}{2} \), where \( \nu = m / 2 \).

In the temperature range where the interaction between the filaments can be neglected the correlation function is

\[ K(x) = \cos \frac{Q}{2} x \exp \left( \frac{1}{R_{\text{c}}} \right). \]

For a real field the correlation radius \( R_{\text{c}}(T) \) increases exponentially when the temperature is lowered (see, e.g., Fig. 3).

3. We consider the case of higher-order commensurability, when \( 2p_f = \frac{Q}{m} \), where \( m \) is an integer and \( m > 2 \). For instance, in KPC crystals the diameter of the electron Fermi surface \( 2p_f \approx \frac{Q}{4} \) and in TTF-TCNQ crystals, the value of \( 2p_f \) is close to \( \frac{Q}{4} \). Introducing the umklapp processes leads to a change in the Lagrangian of the phase of the lattice displacements which was introduced earlier.\(^1\)

Introducing the wavevectors reckoned from the point \( \frac{Q}{2} \) we get

\[ L = \frac{1}{2} \left( \frac{\partial f(x)}{\partial x} \right)^2 + \frac{1}{2} \left( \frac{\partial f(x)}{\partial x} \right)^2 + \frac{b}{2} \Delta (x - \dot{x}^2 \Delta/8x), \]

where \( \Delta = T_{\text{c}}(x) \) is the gap in the spectrum of the Peierls dielectric, \( g \) is the coupling constant for moments corresponding to the umklapp process.

In the classical region \( \Delta \gg T \gg \alpha \Delta/v \) we can neglect\(^1\) the dependence of \( x \) on the static time \( \tau \). In that case the problem of evaluating the correlation function \( K(x) = \exp \left( \frac{1}{R_{\text{c}}} \right) \) is solved by the Feynman method. Following the papers by Vaks et al.\(^1\) and Scalapino et al.\(^1\) we find

\[ K(x) = \cos \frac{Q}{2} x \exp (-|x|/R_{\text{c}}); \]

\[ R_{\text{c}} = \frac{Q}{2} \ln \left( E_f - E_0 \right), \quad Q = \frac{Q}{2} \ln \left( E_f - E_0 \right), \]

where \( R_{\text{c}} \) and \( E_0 \) are the ground state and the first excited state of the "Hamiltonian"

\[ H = \frac{2\pi f(x)}{v} \frac{\partial f(x)}{\partial x} + \frac{v}{2} \frac{\partial f(x)}{\partial x} + \frac{\alpha}{v} f(x)^2 \quad \text{for} \quad |x| > 2p_f. \]

where \( T_{\text{c}} = \frac{\pi}{2} / \alpha \) is the characteristic pinning temperature.

As a result of studying (4) for the case \( m = 4 \) we obtain the following results about the behavior of the correlation functions when the temperature is lowered (see also\(^1\)).

1. Large incommensurability, \( |x| \gg T_{\text{c}} \)

a) \( T \gg v |x| \gg T_{\text{c}} \):

[Formula]

b) \( T \ll v |x| \ll T_{\text{c}} \):

[Formula]

2. Small incommensurability, \( |x| \ll T_{\text{c}} \)

a) \( T \gg T_{\text{c}} \):

[Formula]

b) \( T \ll T_{\text{c}} \):

[Formula]

In the latter case we have with exponential accuracy a typical pinning at the "rational" point \( \frac{Q}{m} \).

The results obtained above are fully applicable if the pinning temperature lies in the classical region \( T_{\text{c}} \gg \alpha \Delta/v \), i.e., if the condition \( T_{\text{c}} \gg T \gg \alpha \Delta/v \) can be satisfied. When the temperature is lowered further we go in that case over into the region \( T_{\text{c}} \ll T \gg \alpha \Delta/v \) where the temperature dependence of the phase \( \chi \) becomes important:

\[ \chi(x) \approx \eta_s(x) + \frac{1}{2} \sum_{n,m} \eta_s(x)^n \eta_s(x)^m |n,m| \approx \eta_s(x)^2. \]

The inequality \( \alpha \Delta / v \ll T \) shows that the inhomogeneous term in (2) appreciably affects only the component \( \eta_s \) which does not oscillate with time while one can accomplish the functional integration over the component \( \eta_s \) by considering only the quadratic part of the Lagrangian (3). As a result of the averaging over \( \eta_s \) the non-quadratic part of (2) and, hence, of the Hamiltonian (4) be-
comes distorted. These distortions can be estimated as the uncertainty \( \Delta x_0 \propto (\Delta \phi_0)^2 \) appearing in the coordinate eigenfunctions of the operator (5). A simple analysis shows that the effect of \( \Delta \phi_0 \) on \( x_0 \) can be neglected, if \( T \ll (uT_v/\ln u/vT) \), which, apart from a logarithmic factor is exactly the same as the limitation introduced above. Using the result of an earlier paper\(^{63} \) we get in the region mentioned

\[
K(x) = \left\{ \frac{2 \Delta \phi_0}{x} \right\} \exp \left( \frac{-2\Delta \phi_0}{x} \right) \left[ 1 + \frac{x}{2\Delta \phi_0} \right] \, K(x),
\]

(7)

where \( c = 1 \) and \( K(x) \) is given by Eqs. (4) to (6).

When \( |x| \ll u/T \) the correlation function (7) has the scaling form (cf.\(^{63} \))

\[
K(x) = \left( \frac{x}{2\Delta \phi_0} \right)^{-1} \exp \left( -\frac{x}{2\Delta \phi_0} \right) K_0(x),
\]

(8)

and when \( x \gg R_T \) gives an exponential decrease:

\[
K(x) = \left( \frac{x}{2\Delta \phi_0} \right)^{-1} \exp \left( -\frac{x}{2\Delta \phi_0} \right) K_0(x).
\]

When the temperature is lowered further, \( T \ll uT_v/v \) or when pinning from the very beginning is quantal (\( T \ll u\phi/v \)) the spatial and temporal correlations of the phase \( x \) are given by the quantal propagator \( D_\phi \) from Sec. 3 (see Eq. (16) below). Using \( H_0 \), we get from standard calculations (cf.\(^{63} \))

\[
K(x) = e^{\left( \frac{x}{2\Delta \phi_0} \right)^{-1} \exp \left( -\frac{x}{2\Delta \phi_0} \right) K_0(x)},
\]

(9)

where \( K_0 \) is a Macdonald function and \( \omega_p \) the "pinning frequency";

\[
u_p = 2\pi \omega_p T/v.
\]

When \( |x| \ll \omega_p /\omega_p \) we have again the scaling Eq. (9) and when \( |x| \gg \omega_p \)

\[
K(x) = \left( \frac{x}{2\Delta \phi_0} \right)^{-1} \exp \left( -\frac{x}{2\Delta \phi_0} \right) K_0(x).
\]

(10)

Equations (9) and (10) show that in the quantal region there appears a new correlation radius—the "pinning radius":

\[
R_p = \omega_p /\omega_p T/v.
\]

(11)

When \( |x| \gg R_T \) Eq. (10) gives "long-range order"—the average value of the cosine of the phase (displacement);

\[
\langle \cos x \rangle = \left( \frac{x}{2\Delta \phi_0} \right)^{-1} \exp \left( -\frac{x}{2\Delta \phi_0} \right).
\]

(12)

Of course, the statement about long-range order is an approximate one. It is the consequence of the fact that we used the harmonic approximation when calculating \( D_\phi \). Taking the anharmonic terms in (2) into account leads to sound oscillations in the system of the pinned charge-density wave and to moving walls and solitons described by the appropriate sine-Gordon equation. These effects destroy the long-range order at any sufficiently low temperature (cf.\(^{63} \)).

3. FRÖHLICH CONDUCTIVITY UNDER WEAK PINNING CONDITIONS

In our earlier paper\(^{63} \) we showed rigorously that the optical properties of the one-dimensional system considered contain for \( T \ll T_{ph} \) a Fröhlich contribution to the permittivity \( \varepsilon^\omega \) and we calculated the anomalous optical conductivity accompanying it. In the present paper we shall study these quantities taking the periodicity of the system into consideration.

We first consider the weak pinning region \( T \gg T_p \), when we can use perturbation theory to take into account the inhomogeneous term in (2). The anomalous part of the permittivity can be expressed\(^{63} \) in terms of the analytical continuation of the Green function \( D_\phi (x, x') \) of the phase operators \( x \) considered as a Bose field,

\[
\varepsilon^\omega (x, u) = -\frac{1}{x} e^{\nu_p x} D_\phi (x, u).
\]

(13)

The first terms in the expansion in powers of \( x \) can easily be determined by writing \( (D_\phi (x, x')) \) in (13)

\[
\frac{\partial^2 D_\phi (x, x')}{\partial x \partial x'} = \frac{2x}{x^2 - \Delta \phi_0^2} \left[ \frac{\partial^2 D_\phi (x, x')}{\partial x \partial x'} \right]
\]

or

\[
\exp \left( \frac{x}{2\Delta \phi_0} \right)^{-1} \exp \left( -\frac{x}{2\Delta \phi_0} \right) K_0(x).
\]

Expanding in \( \nu_p \) and using the formula \( (\Delta \phi_0 = 0) \)

\[
\exp \left( \frac{1}{2} \sum \Delta \phi_0 D_\phi (x, x') D_\phi (x, x') \right)
\]

to evaluating the resulting averages we get an expression of the form

\[
D_\phi (x) = D_\phi (x) + v^\omega \sum \Delta \phi_0 D_\phi (x) D_\phi (x)
\]

or, in Fourier components,

\[
D_\phi (q, \omega) = D_\phi (q, \omega) + v^\omega \sum \Delta \phi_0 D_\phi (q, \omega) [K_0 (0, 0) - K_0 (q, \omega)],
\]

(14)

where according to (3)

\[
D_\phi (q, \omega) = e^{\nu_p q} \frac{1}{v} e^{\nu_p q}.
\]

To second order in \( \nu_p \) the function \( K_n \) is equal to

\[
K_n (q, \omega) = \frac{1}{v} e^{\nu_p q} (\exp \left( \nu_p q \right) - 1).
\]

To obtain the analytical continuation of the function \( K_n (q, \omega) \) as before\(^{63} \) for \( m = 1 \). As a result we get the following expression for the conductivity for \( q = 0 \) and \( u \ll T_p \):

\[
e^\omega (q) = \frac{v^\omega}{\pi} \ln D_\phi (x) = \nu_p (u)
\]

\[
+ \nu_p \left[ \frac{1}{v} e^{\nu_p q} \right] + \nu_p \left[ \frac{1}{v} e^{\nu_p q} \right] + \nu_p \left[ \frac{1}{v} e^{\nu_p q} \right]
\]

where we have evaluated the contribution \( a_0 (u) \) before,\(^{63} \) for \( \nu_p = \nu_p q /v. \)
According to (9) and the results of Ref. 10 we get for \( \sigma(t) \) in some limiting cases:

1) when \( \omega \ll T_\nu \)

\[
\sigma(t) = i\alpha' \left( \frac{\omega}{\nu} \right)^{1/2} \frac{T}{\bar{\epsilon}} \left( \frac{\omega^2}{\nu^2} \right)^{1/2} \frac{1}{\nu \omega^4 \bar{\Gamma}(1/2)}.
\]

2) when \( \omega T/\nu \ll \omega \ll T \)

\[
\sigma(t) = i\alpha' \left( \frac{\omega}{\nu} \right)^{1/2} \frac{T}{\bar{\epsilon}} \left( \frac{\omega^2}{\nu^2} \right)^{1/2} \frac{1}{\nu \omega^4 \bar{\Gamma}(1/2)}.
\]

4. FRÖHLICH CONDUCTIVITY UNDER STRONG PINNING CONDITIONS

In the low-temperature region \( T \ll T_\nu \) the important values of the phase \( \chi \) lie in the vicinity of the minima of the pinning potential energy in (2). The probabilities for a transition between different minima are exponentially small and are not important for real frequencies. Hence, we can expand the potential energy in power of \( \chi \). The second-order terms then determine the gap in the phase oscillation spectrum and, hence, the final value of the anomalous permittivity

\[
\sigma_{\text{an}}(\omega) = \frac{1}{\chi \omega} e^{t \omega^4 + \omega^2},
\]

where \( \omega^2(q) = \omega^2 + \omega^2_{\text{an}} \).

4.1 Phenomenological expressions for the ground state

The fourth- and higher-order anharmonic terms guarantee the damping of the phase oscillations and determine the anomalous optical absorption. The magnitude of the damping \( \Gamma(\omega) \) is given by an expression which is similar to that from our earlier paper\(^{25}\)

\[
\Gamma(\omega) = \frac{1}{2} \left[ \left( \frac{\omega}{\nu} \right)^{1/2} \frac{T}{\bar{\epsilon}} \left( \frac{\omega^2}{\nu^2} \right)^{1/2} \frac{1}{\nu \omega^4 \bar{\Gamma}(1/2)} \right] \sum \frac{1}{12 \nu^2 \bar{\epsilon}^2} \sum \alpha_{\text{an}} \left( \bar{\epsilon} \alpha_{\text{an}} \right) \sum \alpha_{\text{an}} \left( \bar{\epsilon} \alpha_{\text{an}} \right) \alpha_{\text{an}} \left( \bar{\epsilon} \alpha_{\text{an}} \right) \alpha_{\text{an}} \left( \bar{\epsilon} \alpha_{\text{an}} \right) \alpha_{\text{an}} \left( \bar{\epsilon} \alpha_{\text{an}} \right). \tag{18}
\]

An investigation shows that always \( \omega^2(q) < \omega^2 + \omega^2_{\text{an}} \) and, hence, we can, in agreement with (15), put

\[
\sigma(t) = \frac{1}{\chi \omega} e^{t \omega^4 + \omega^2} \sigma(\omega^2 + \omega^2_{\text{an}}).
\]

At high temperatures, \( T \gg \omega \), we have

\[
\sigma(t) = \frac{1}{\chi \omega} e^{t \omega^4 + \omega^2} \frac{T}{\bar{\epsilon}} \left( \frac{\omega^2}{\nu^2} \right)^{1/2} \frac{1}{\nu \omega^4 \bar{\Gamma}(1/2)}.
\]

At low temperatures, \( T \ll \omega \), but high frequencies, \( \omega \gg \omega \),

\[
\sigma(t) = \frac{1}{\chi \omega} e^{t \omega^4 + \omega^2} \frac{T}{\bar{\epsilon}} \left( \frac{\omega^2}{\nu^2} \right)^{1/2} \frac{1}{\nu \omega^4 \bar{\Gamma}(1/2)}.
\]

This kind of behavior continues right up to the threshold for the decay of one phason into three when \( \omega = 3\omega \), where

\[
\sigma(t) = \frac{1}{\chi \omega} e^{t \omega^4 + \omega^2} \frac{T}{\bar{\epsilon}} \left( \frac{\omega^2}{\nu^2} \right)^{1/2} \frac{1}{\nu \omega^4 \bar{\Gamma}(1/2)}.
\]

At low temperatures, \( T \ll \omega \), but high frequencies, \( \omega \gg \omega \),

\[
\sigma(t) = \frac{1}{\chi \omega} e^{t \omega^4 + \omega^2} \frac{T}{\bar{\epsilon}} \left( \frac{\omega^2}{\nu^2} \right)^{1/2} \frac{1}{\nu \omega^4 \bar{\Gamma}(1/2)}.
\]
Electron-phonon interaction in one-dimensional systems in the adiabatic approximation

V. Ya. Krivnov and A. A. Ovchinnikov

L. Ya. Karpov Physical-Chemical Research Institute

Submitted October 13, 1976


A one-dimensional Fermi gas interacting with lattice vibrations is considered in the adiabatic approximation. It is shown that the system should be a superconductor at small electron-phonon coupling constants $g^2$, in accord with the results of Fröhlich. The critical velocity at which the superconducting state is stable with respect to scattering by impurities is found. The extreme case of large values of $g^2$ is investigated, and it is shown that in this case the problem reduces to the analysis of a system of almost noninteracting polarons. It is also shown that for the lattice Fermi gas the ground state corresponds to the Peierls state with a deformed lattice, and that the system is a dielectric.

PACS numbers: 74.20.−-z

As is well known, the one-dimensional system of electrons interacting with a lattice, considered by Fröhlich in 1954, was the first example of a model for which the dependence of the energy on the constant of the electron-phonon interaction bore a non-analytic character. It was later made clear, however, that the nature of the ground state of the electron-phonon system is completely different in the three-dimensional and one-dimensional cases. According to Ref. 1, in the one-dimensional case the system is in the so-called Peierls state, the characteristic feature of which is the presence of strongly excited lattice vibrations with wave number $q = 2\pi p$, in whose periodic field the electrons move. Here the system of electrons and lattice displacements can move with the velocity $v_0$. On the basis of arguments similar to Landau's well-known argument, Fröhlich identified these states at $v = 0$ with the superconducting states. On the other hand, Allender, Bray, and Bardeen, [1] state that at $p = 1$ this system is a typical dielectric ($\rho = n/N$, where $n$ is the number of electrons and $N$ is the number of centers in the lattice).

The purpose of the present work is the study of the phase states of a one-dimensional electron-phonon system as a function of $\rho$ in the adiabatic limit (we note that both the early work of Fröhlich [1] and the work of Ref. 2 were carried out in just this approximation). We shall show that the analysis of Fröhlich is valid only at $\rho^2 < 1$. At these values of the parameters of the system, we investigate the stability of the superconducting states relative to scattering by impurities and calculate the critical velocity $v_0$. It will be shown that in the presence of a strong electron-phonon interaction, $\rho^2 > 1$, polaron states appear in the system. So far as the lattice Fermi gas is concerned, in this case the electron-phonon interaction forms a Peierls state with a distorted lattice and, as will be shown, the system is a dielectric.