Frölich conductivity at temperatures comparable with the Debye temperature

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The conductivity of a quasi-one-dimensional system in a Peierls-Frölich state is considered at relatively high temperatures, amounting to 50–300 K for real system. It is shown that the interaction of the Frölich mode with acoustic-branch phonons should determine the damping of the Frölich mode for the frequencies of the far-infrared band. In the microwave region, the frequency-independent damping is produced by umklapp and by interaction between chains with participation of thermal phonons.

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1. INTRODUCTION

One of the pressing topics of physics of quasi-one-dimensional systems has been in recent years the investigation of effects connected with the existence of charge-density waves (CDW). A detailed investigation of these effects was carried out in KCP (Ref. 1) and in TTF-TCNQ.[2] The existence of CDW having a period incommensurate with the period of the main structure manifests itself, besides in the structural data, also in an unusually high static permittivity and in a specific frequency dependence of the optical conductivity in the far infrared (the so-called Fröhlich conductivity[3]). The totality of these effects is due to the existence of an optically active phase mode of the CDW and to the approximate translational invariance of the system.[4]

The final low-frequency limit of the permittivity is determined by the pinning of the CDW phase by disturbances of the translational invariance of the host structure—by interaction with the impurities and by umklapp processes.[4] This limit is reached for the investigated substances in the microwave band. At higher frequencies the permittivity should be determined by the interaction between the phase mode of the CDW and various excitations of the system. Its damping due to interactions with the excitations of the CDW proper (phase and amplitude) and with the thermally activated electrons was considered by Dzyaloshinskiĭ and the author.[5] The interaction with the electrons turns out to be insignificant in the entire region of existence of the CDW. The anharmonicities of the CDW proper are significant in the low-temperature region at finite frequencies. The corresponding damping decreases more rapidly than the frequency itself, and thus cannot be responsible for the finite value of the static conductivity.

At higher temperatures (the scale is a temperature T_0 of the order of the amplitude oscillations of the CDW) the proper anharmonicities should give way to interaction with excitations of higher frequency—the normal phonons. The temperatures T_0 for the investigated substances lie somewhat lower than the Debye temperatures $\overline{\omega}$ and amount to $\lesssim 100$ K, which coincides with the temperatures at which three-dimensional effects manifest themselves. Therefore an investigation of the region $T > T_0$ in quasi-one-dimensional systems is of greatest importance in the analysis of the regions of the specifically one-dimensional behavior in the vicinities of the points of three-dimensional phase transitions. The condition $T \gg T_0$ determines also whether a classical description of the CDW is admissible. This must be borne in mind in particular when comparing the experimental data with theoretical results on the pinning of one-dimensional CDW, obtained within the framework of classical physics.

This paper deals with effects due to the interaction of CDW with thermal phonons in quasi-one-dimensional structures. Account will also be taken of the interactions of incommensurate CDW with the main structure and of the interaction between the chains.

2. INTERACTION OF OPTICALLY ACTIVE CDW MODE WITH THERMAL PHONONS

1. The anomalous electric properties in a system with CDW in the absence of pinning are connected with the existence of an optically active low-frequency mode.[4] The main effect of its interaction with the thermal phonons stems from the triple anharmonicities that are determined mainly by the interaction of the phonons via the electron subsystem. The result of the interaction is determined by the polarization insets shown in the diagram of Fig. 1 for the permittivity. The solid lines in this figure correspond to electrons of the ground state, dashed ones to thermal phonons, and the dash-dot line is the line of the optically active mode and carries the low frequency of the external electric field.

A physically realistic picture of the phonomena must account for the interaction of the CDW and of the phonons with the main structure of the chain, and for the interactions between the chains. It will be more convenient for this purpose to consider the system in terms of a two-fluid picture: the optically active mode corresponds to slow changes of the CDW phase $\chi(x, t)$ (Ref. 4); the shorter-wavelength thermal modes contain an explicit local dependence on $\chi(x, t)$ in the expressions for their normal coordinates in terms of the atomic displacements $d(x, t)$; this dependence leads to nonlinear effects that are exactly equivalent to the anharmonicities of Fig. 1.

2. We consider now a one-dimensional Peierls dielec-

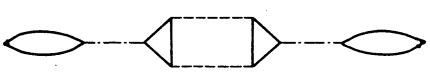

FIG. 1.

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tric at temperatures comparable with the Debye energy, but small compared with the gap 2Δ in the electron spectrum. The displacements of the unit cells of the lattice relative to the main structure can be represented in two forms:

$$d(x,t)=\mathrm{Re}\{d_0\exp\{i2p_Fx\}[\exp\{i\chi(x,t)\}+\Phi(x,t)]\}, \quad (1a)$$

$$d(x,t)=\mathrm{Re}\{d_0\exp\{i[2p_Fx+\chi(x,t)]\}[1+\varphi(x,t)]\}. \quad (1b)$$

Here d_0 is the equilibrium deformation in a Peierls dielectric, the phase $\chi(x,t)$ characterizes the large-scale distortions of the super-structure (or the CDW) with wave vectors $|q|\ll\xi_0^{-1}=\Delta/v_F$, and the fields Φ or φ describe excitations with $|q|\gg\xi_0^{-1}$. We shall henceforth make throughout a natural distinction between these two scales, so that the redundance of the description in the form (1a) and (1b) will not be significant.

The characteristic scale of the quantities φ and Φ as a result of the thermal and quantum fluctuations can be estimated at

$$\langle\varphi^2(x,t)\rangle\approx\langle\Phi^2(x,t)\rangle\sim\lambda_0\frac{\varepsilon_F}{\Delta}\max\left\{\frac{T}{\Delta},\frac{\bar\omega}{\Delta}\right\}, \quad (2)$$

where $\lambda_0=g^2/\pi v_F$ is the dimensionless constant of the interaction of the electrons with the Debye phonons, and $\bar\omega=\bar\omega(2p_F)$. In TCNQ the value[6] $\lambda_0\lesssim 0.1$ is much less than the total interaction constant $\lambda=[\ln(\varepsilon_F/\Delta)]^{-1}$; in KCP we have $\lambda_0\approx\lambda\approx 0.4$.

It follows from (2) that at $\Delta\ll\varepsilon_F$ the fluctuations of the lattice displacements can exceed the equilibrium displacement d_0. The electrons, however are acted upon only by phonons with momenta k in the region $|k\pm 2p_F|\lesssim\xi_0^{-1}$. The gap in the electron spectrum is therefore well defined:

$$\langle(\Delta-\Delta_0)^2\rangle/\Delta_0^2\sim T/\Delta_0.$$

The Lagrange function of the system in the representation (1a) is $\mathcal{L}=\mathcal{L}_1+\mathcal{L}_{11}$, where

$$\mathcal{L}_1\{\chi\}=\sum_q\left\{\frac{1}{2v}[\dot\chi_q\dot\chi_{-q}-u^2q^2\chi_q\chi_{-q}]+\frac{1}{\pi}e\chi_qE_{-q}\right\},$$

$$\mathcal{L}_{11}\{\Phi,\chi\}=\sum_q\frac{1}{v}[\dot\Phi_q^*\dot\Phi_q-\omega_q^2(\Phi_q^*\Phi_q+e^{-2i\chi}A_q\Phi_q\Phi_{-q}+e^{2i\chi}A_q\Phi_q^*\Phi_{-q}^*)], \quad (3)$$

with u the phase velocity of the CDW, $v=2\pi u^2/v_F=2\pi v_Fm/m^*$, e the electron charge, W the external electric field, $m/m^*=(\lambda^2/4\lambda_0)(\bar\omega/\Delta)^2$,

$$\omega_q^2=\bar\omega^2(2p_F+q)[1-\lambda_0\ln(p_F^2/q^2)],$$
$$A_q=\lambda_0\ln(q^2\xi_0^2)/(q\xi_0)^2, \quad |q|\xi_0\gg 1, \quad (4)$$

where $\bar\omega(k)$ is the nonrenormalized spectrum of the phonons, m is the optical mass of the electrons, and m^* is the so-called CDW mass.[4]

The quantities A_q are the amplitudes of the phonon umklapps on the wave vector $4p_F$ because of their interaction with the superstructure. They split the spectrum of the Kohn anomaly (4), and in the ordered phase, when $\langle e^{2i\chi}\rangle\neq 0$, they ensure second Bragg reflections.

We change now to the representation (1b) and write

$$\varphi(x,t)=a(x,t)+ib(x,t),$$

where a and b are real functions,

$$\mathcal{L}_{11}\{a,b;\chi\}=\frac{1}{2v}\sum_q[|\dot a_q-\dot\chi b_q|^2+|\dot b_q+\dot\chi a_q|^2$$
$$-\omega_a^2(q)|a_q|^2-\omega_b^2(q)|b_q|^2+i\Omega_q^2(b_{-q}a_q-a_{-q}b_q)]; \quad (5)$$
$$\omega_{a,b}^2(q)=\tfrac12(\omega_q^2+\omega_{-q}^2)(1\pm A_q),\quad \Omega_q^2=\tfrac12(\omega_q^2-\omega_{-q}^2).$$

The quantity $\Omega_q^2\approx 2\bar s q(\omega_q/\bar\omega)^2$, where $\bar s=\partial\omega_{2p_F+q}/\partial q$ is the renormalized speed of sound at $q=0$, reflects the asymmetry of the Kohn anomaly.

Since $\bar s\neq 0$, the terms $\omega_{a,b}^2(q)$ cross and it is necessary to transform to the variables α_q and β_q:

$$a_q=U_q\alpha_q+iV_q\beta_{-q},\ b_q=U_q\beta_q+iV_q\alpha_{-q},$$
$$U_{-q}=U_q,\quad V_{-q}=-V_q.$$

The Hamiltonian (5) is diagonalized

$$U_q^2,\ V_q^2=\frac12(1\pm B_q),\quad B_q=\frac12\left[A_q^2+\left(\frac{\omega_q^2-\omega_{-q}^2}{\omega_q^2+\omega_{-q}^2}\right)^2\right]^{-1/2}.$$

The total picture of the spectrum is shown in Fig. 2 for the doubled Brillouin zone of the superstructure. We obtain

$$\mathcal{L}_{11}\{\alpha,\beta,\chi\}=\frac{1}{2v}\sum_q[|\dot\alpha_q|^2+|\dot\beta_q|^2-\omega_\alpha^2(q)|\alpha_q|^2$$
$$-\omega_\beta^2(q)|\beta_q|^2+2B_q(\alpha_q\dot\beta_{-q}-\beta_q\dot\alpha_{-q})\dot\chi+\dot\chi^2(|\alpha_q|^2+|\beta_q|^2)], \quad (6)$$

where

$$\omega_{\alpha,\beta}(q)=\tfrac12(\omega_a^2+\omega_b^2)\pm[\tfrac14(\omega_a^2-\omega_b^2)^2+\Omega^4]^{1/2}$$
$$=\tfrac12(\omega_q^2+\omega_{-q}^2)[1\pm(2B_q)^{-1}].$$

The last two terms in (6) characterize the Coriolis and the centrifugal forces acting on the rotator (α,β) in a state coherent relative to the CDW. The Coriolis forces should be small compared with the noncentral forces $\sim(\omega_\alpha^2-\omega_\beta^2)$ that cause the rotator to rotate. To this end we must have

$$\dot\chi<\delta=\min\{\omega_\alpha(q)-\omega_\beta(q)\}\sim\bar\omega(\bar s\Delta/v_F\bar\omega)^{3/4},$$

which corresponds to an upper bound $v_e<\bar s(\Delta/\varepsilon_F)^{2/3}$ on the electron drift velocity in the CDW. This condition is satisfied at any real current density.

Allowance for the term splitting is meaningful only in those cases when the splitting exceeds the corresponding phonon line widths Γ_q. This is indeed the case. The ternary and quaternary anharmonicities in the region of the Kohn anomaly yield for the relaxation frequencies $\Gamma_q^{(3)}$, $\Gamma_q^{(4)}$

$$\frac{\Gamma_q^{(3)}}{\bar\omega}\sim\frac{T\bar\omega}{\Delta^2}\frac{1}{(\xi_0q)^4},\quad \frac{\Gamma_q^{(4)}}{\bar\omega}\sim\frac{T^2}{\Delta^2}\frac{1}{(\xi_0q)^2}.$$

It is obvious that $\Gamma_q^{(3)},\Gamma_q^{(4)}\ll A_q\bar\omega<\omega_\alpha(q)-\omega_\beta(q)$. Such a definite conclusion concerning the behavior of the relaxa-

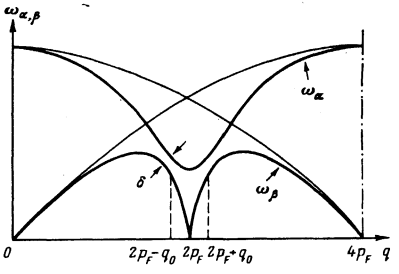

FIG. 2.

tion frequencies is possible only because in the region of the Kohn anomaly the phonon interactions via the electron subsystem exceed their interactions via the lattice anharmonicities.

Calculations on the basis of the Hamiltonian (6) in the approximation linear in $\dot{\chi}$ correspond exactly to the diagram of Fig. 1. The equations of motion for χ, α, and β take the form

$$\frac{d}{dt}\left\{\dot{\chi}\left[1+\sum_q B_q(|\alpha_q|^2+|\beta_q|^2)\right]\right\}+\sum_q[\ddot{\beta}_q\alpha_{-q}-\ddot{\alpha}_q\beta_{-q}]=\frac{1}{\pi}evE, \tag{7a}$$

$$\ddot{\alpha}_q+\omega_\alpha^2\alpha_q=B_q[\ddot{\chi}\beta_q+2\dot{\chi}\dot{\beta}_q], \tag{7b}$$

$$\ddot{\beta}_q+\omega_\beta^2\beta_q=-B_q[\ddot{\chi}\alpha_q+2\dot{\chi}\dot{\alpha}_q]. \tag{7c}$$

The first sum in (7a) determines the increase of the CDW mass due to the phonon dragging. According to the estimate (2), it may be not small. The second term in (7a) leads to resonant absorption of phase oscillations with frequency $\omega=\omega_\alpha(q)-\omega_\beta(q)\geqslant\delta$. The quantity δ can be roughly estimated at 10–20 cm^{-1}, corresponding to the lowest frequencies at which the relaxation of the low-frequency optical mode has been investigated.[1,2]

For the permittivity we get

$$\varepsilon(\omega)=\varepsilon_\infty\left[1-\frac{m}{m^*(T)}\frac{\omega_p^2}{\omega^2+i\omega\gamma}\right] \tag{8}$$

$$\frac{m^*(T)}{m^*(0)}=1+\lambda_0\frac{\varepsilon_F}{\Delta}\frac{q_m}{p_F}\max\left\{\frac{\bar{\omega}}{\Delta},\frac{T}{\Delta}\right\}.$$

Here ω_p is the plasma frequency, and q_m is the maximum wave vector of the phonons dragged by the CDW and is determined by the transverse dispersion w of their spectrum on account of the interaction between the chains (see Sec. IV below). In the case of strong anisotropy we have $w\ll\delta$ and $q_m\approx p_F$, and at $\Delta\ll\varepsilon_F$ there exists for (8) a region of linear temperature dependence, $m^*(T)\sim T$, $T>\bar{\omega}$.

The damping γ_{ph} is calculated from the formula

$$\gamma_{ph}(\omega,T)=\frac{4}{\pi}v\frac{m^*(0)}{m^*(T)}\frac{\omega^2}{n(\omega)}\int dq\, n(\omega_\beta(q))[1+n(\omega_\alpha(q))]\times\delta(\omega+\omega_\beta(q)-\omega_\alpha(q)).$$

a) At $\omega\gg\delta$ we have $\omega_\alpha(q)-\omega_\beta(q)\approx 2A_q$ and

$$\gamma_{ph}(\omega,T)=2\pi\frac{m}{m^*(T)}\frac{\Delta}{T}n(\tilde{\omega})[1+n(\tilde{\omega})](2\tilde{\omega}\omega)^{1/2}, \tag{9}$$

where $\tilde{\omega}=\omega_{a,b}(\tilde{q})$ with $\tilde{q}$ such that $A_{\tilde{q}}=\omega$. We always have $\gamma\ll\omega$, so that the optical conductivity at $T>\bar{\omega}$ is

$$\sigma(\omega,T)\sim\varepsilon_\infty\frac{\omega_p^2}{\tilde{\omega}^2}\left[\frac{m}{m^*(T)}\right]^2\Delta T\left(\frac{\tilde{\omega}}{2\omega^3}\right)^{1/2}. \tag{9a}$$

b) At $\omega\approx\delta$ we have $\omega_\alpha(q)-\omega_\beta(q)=\delta+6\bar{s}(|q|-q_0)^2/q_0$, where $q_0\approx(\omega_{q0}^2/\bar{s}\bar{\omega}\xi_0^2)^{1/2}\sim\xi_0^{-1}(p_F\xi_0)^{1/3}\sim p_F(p_F\xi_0)^{-2/3}$, i.e., $\xi_0^{-1}\ll q_0\ll p_F$. The points $\pm q_0$ are shown in Fig. 2 by dashed vertical lines. If the spectrum of the Kohn anomaly is described approximately by the logarithmic law of the weak-coupling model $\omega_q^2=\bar{\omega}^2\lambda\ln(q^2\xi_0^2)$, then $\omega_{q0}=\bar{\omega}/3^{1/2}$. The damping is given by

$$\frac{\gamma_{ph}(\omega,T)}{\delta}=\frac{1}{3\cdot 2^{1/2}}\frac{v_F}{\bar{s}}\frac{m}{m^*(T)}n_{q_0}[1+n_{q_0}]\frac{\delta}{T}\left(\frac{\delta}{\omega-\delta}\right)^{1/2}. \tag{10}$$

At $T>\bar{\omega}$, assuming that $m^*(T)\gg m^*(0)$, we get

$$\frac{\gamma_{ph}(\omega,T)}{\delta}=c\left(\frac{\delta}{\omega-\delta}\right)^{1/2},\quad c\sim\left(\lambda_0\frac{m^*(0)}{m}\frac{\bar{s}}{v_F}\right)^{1/2}\sim 1. \tag{10a}$$

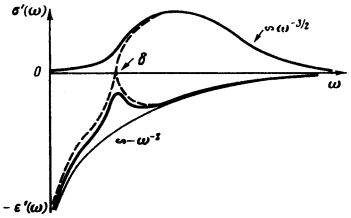

FIG. 3.

The divergences in (10) and (10a) are connected with the approximation of the one-dimensional dispersion law. They are cut off at $\omega-\delta\approx w$. It follows from (10a) that in the region of the edge of the interphonon absorption we have $w<\omega-\delta<\delta$ and $\gamma_{ph}(\omega,T)>\omega$, i.e., a transition is possible into a dissipative regime with a decrease of the optical conductivity:

$$\sigma(\omega,T)\approx\frac{3}{2\pi}\varepsilon_\infty\frac{\omega_p^2}{\delta^2}\frac{\bar{s}}{v_F}\times\frac{T}{n_{q_0}[1+n_{q_0}]}\left(\frac{\omega-\delta}{2\delta}\right)^{1/2}.$$

c) At $\omega\lesssim\delta$ expression (10) contributes to $\mathrm{Re}\varepsilon(\omega)$. At $\omega<\delta$ the coherent phonons do not influence any more the relaxation of the CDW motions. In this region it is necessary to take into account the interactions of the phonons with the main structure and the transitions between the chains.

The general frequency dependences $\sigma'(\omega)=\mathrm{Re}\sigma(\omega)$ and $\varepsilon'(\omega)=\mathrm{Re}\varepsilon(\omega)$ is shown in Fig. 3 with account taken of the smoothing action of the transverse dispersion. It is assumed that $\delta^2<\omega_p^2 m/m^*(T)$.

In concluding this part we emphasize once more that only region a) takes place at $w>\delta$. The real values of these parameters are most likely to be comparable.

3. INTERACTION OF INCOMMENSURATE CDW WITH THE MAIN STRUCTURE OF THE STRING VIA THERMAL PHONONS

1. The interaction of the superstructure and of the phonons with the main periodic structure of the string is expressed in terms of umklapp processes at the commensurate points Ql/n, where Q is the period of the reciprocal lattice of the main structure, and l and n are integers. A measure of the incommensurability of the structure is the wave number $q_n=n2p_F-lQ$. It is proposed that the points $2p_F\pm q_n/2$ lie within the limits of the Kohn anomaly, but the q_n are too large to allow static pinning of the CDW in the absence of phonons. This means[7] that $|q_n|>C_n^{1/2}/\xi_0$, where $n>2$,

$$C_n=\frac{1}{2\pi}\frac{g_U}{\lambda g_N}\left(\frac{\Delta}{\varepsilon_F}\right)^{n-2},$$

g_N and g_U are the constants of electron-phonon interaction with and without conservation of the quasimomentum, and at $n=3$ and 4 we have $\tilde{\varepsilon}_F=\varepsilon(3p_F)-\varepsilon(p_F)$.

Under these conditions the interaction of the CDW with the main structure occurs with participation of two-phonon processes. They correspond to the Hamiltonian

$$\mathcal{H}_U=C_n\frac{\Delta^2}{v_F}[\exp\{i[q_n x+n\chi(x,t)]\}\varphi^2(x,t)+\text{c. c.}]. \tag{11}$$

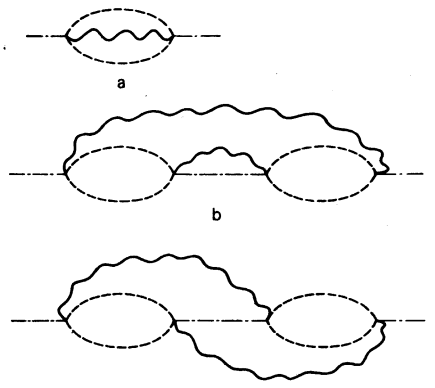

FIG. 4.

We must use the coherent representation (1b) if the influence of $\mathcal{H}_U$ is small compared with the term splitting $\omega_\alpha(q)-\omega_\beta(q)$, i.e.,

$$C_n\Delta^2/v_F \ll A_q\omega_q^2/v,$$

whence

$$q^2\xi_0^2 \gg (g_N/g_U)(\varepsilon_F/\Delta)^{n-2}.$$

This inequality is always satisfied at $n>4$. In the case $n=4$ this inequality is violated at $|q|\sim p_F$, i.e., outside the limits of the Kohn anomaly, a factor of no importance to us. In the case $n=3$ there exists a limiting vector $q_m\sim(p_F/\xi_0)^{1/2}$, i.e., $\xi_0^{-1}\ll q_m\ll p_F$. For phonons with $|q|>q_m$ it is necessary to use the representation (1a) and regard their interaction with the superstructure, $\sim A_q$, as a perturbation. This question will be particularly important in Sec. 4. The case $n=2$ is singular. The umklapp processes must be taken into account here from the very outset together with the normal ones.[7] At $|q_n|<\xi_0^{-1}$ the CDW is always commensurate and the Fröhlich effect vanishes.

The total Hamiltonian (5) or (11), just as (3), can be diagonalized in the phonon modes at constant χ. Now, however, the parameters of the diagonal Hamiltonian will depend explicitly on χ, and not only on $\dot\chi$, as in (5). Expanding in the small deviations $\chi=\chi_0+\chi_1$, which indeed ensure a finite damping of the oscillations of the phase χ_1 at low frequencies. It is simpler, however, to regard (11) as a perturbation.

2. The calculation of the homogeneous damping $\gamma(\omega)$ of the CDW phase oscillations is carried out in accord with diagram 4a, where the wavy line denotes the Green's function

$$K_n(x,t)=\langle\!\langle \exp\{in\chi(x,t)\}\,|\exp\{-in\chi(0,0)\}\rangle\!\rangle. \tag{12}$$

We leave out diagrams with overlaps, such as Figs. 4b and 4c. This corresponds to the condition $\omega\tau_n\gg1$ or $\gamma\tau_n\gg1$, where τ_n is the relaxation time in (12), For noninteracting chains we have $\tau_n^{-1}=\pi n^2(u/v_F)T$.

The diagram-technique rules[8] yield directly

$$\gamma_U(\omega)=\frac{1}{2}n^2C_n^2v^3\int\frac{dqdkd\Omega}{(2\pi)^3}S_n(k,\Omega)\left\{\frac{n[\omega_\alpha(q_n/2-q/2-k)]}{\omega_\alpha(q_n/2-q/2-k)}\right.$$
$$\times\frac{(1+n[\omega_\alpha(q_n/2+q/2)])}{\omega_\alpha(q_n/2+q/2)}\delta\left[\omega_\alpha\left(\frac{q_n}{2}+\frac{q}{2}\right)-\omega_\alpha\left(\frac{q_n}{2}-\frac{q}{2}-k\right)+\Omega-\omega\right]$$
$$+\frac{n[\omega_\beta(q_n/2-q/2-k)](1+n[\omega_\beta(q_n/2+q/2)])}{\omega_\beta(q_n/2-q/2-k)\,\omega_\beta(q_n/2+q/2)}\delta\left[\omega_\beta\left(\frac{q_n}{2}+\frac{q}{2}\right)\right.$$
$$\left.\left.-\omega_\beta\left(\frac{q_n}{2}-\frac{q}{2}-k\right)+\Omega-\omega\right]\right\}. \tag{13}$$

Here $S_n(k,\omega)$ is the correlation function (structure factor) corresponding to the Green's function (12).

The calculation result does not depend on the form of $S_n(k,\omega)$ if $\bar\omega\tau_n\gg1$,

$$\gamma_U(\omega)=\frac{\pi^2}{2}n^2C_n^2\left(\frac{m}{m^*}\right)^3\frac{\Delta^4}{T}\left\{\frac{v_F/s_\alpha}{\omega_\alpha^2(q_n/2)\,\mathrm{sh}^2[\omega_\alpha(q_n/2)/T]}\right.$$
$$\left.+\frac{v_F/s_\beta}{\omega_\beta^2(q_n/2)\,\mathrm{sh}^2[\omega_\beta(q_n/2)/T]}\right\},\quad s_{\alpha,\beta}=\frac{\partial\omega_{\alpha,\beta}(q)}{\partial q}\Big|_{q=q_n/2}. \tag{14}$$

Formulas (13) and (14) are valid if $\xi_0^{-1}\ll q_n/2<q_m$. The cases $q_n\ll\xi_0^{-1}$ and $q_n/2>q_m$ are considered in the Appendix. At

$$(\Delta/\varepsilon_F)^2\approx\bar\omega/\Delta\approx(m/m^*)^{1/n}\approx10^{-1},\quad n=3,\ 4$$

and $T\approx\bar\omega$ we obtain from (14) $\gamma_U\approx1$–$10\ \mathrm{cm}^{-1}$. At $T>\bar\omega$ in a realistic situation we have $\bar\omega\tau_n\ll1$. Consequently, the energy conservation law becomes immaterial in formula (13) and the integration with respect to q is over the entire band. The principal role is now plaid by the non-dragged phonons (1a) with $|q|>q_n$. Their effect is considered in the second section of the Appendix.

4. CDW DAMPING BY INTERACTIONS BETWEEN NEIGHBORING STRINGS

1. As indicated in the Introduction, in the considered region BI different strings can be naturally regarded as uncorrelated. Therefore, in the coherent-mode representation (1a), the phonon-exchange Hamiltonian contains an explicity dependence on the phases of the chian chains:

$$\mathcal{H}_{ex}=\frac{1}{2v}\sum_{i,j,q}W_{ij}^2\Phi_{q,i}^*\Phi_{q,j}, \tag{15a}$$

or

$$\mathcal{H}_{ex}=\frac{1}{2v}\sum_{i,j,q}W_{i,j}^2\{\cos(\chi_i-\chi_j)\,[a_{q,i}a_{-q,j}+b_{q,i}b_{-q,j}]$$
$$+\sin(\chi_i-\chi_j)\,[a_{q,i}b_{-q,j}-a_{-q,j}b_{q,i}]\}. \tag{15b}$$

In view of the anisotropy of the phonon spectrum at $w\ll\bar\omega$ we have

$$\sum_j W_{ij}^2\sim w\bar\omega.$$

At not too large values of $q<q_m$ the condition $w/\bar\omega<A_q$, $q<q_m$.may be satisfied. If $w/\bar\omega<(\Delta/\varepsilon_F)^2(\lambda_0/\lambda)$, then this condition is satisfied for all q. In these cases α_q and β_q are as before normal modes, and the Hamiltonian (15b) can be regarded as a perturbation.

In analogy with the calculations of Sec. 3 we must take into account the diagonal element of the self-energy part $\sum_{ii}(k,\omega)$, shown in Fig. 5a. The vertical zigzig lines represent the interaction W_{ij}^2 between the chains. The off-diagonal elements $\sum_{ii}(k,\omega)$ which are produced when the external lines in the inset of Fig. 5 are added

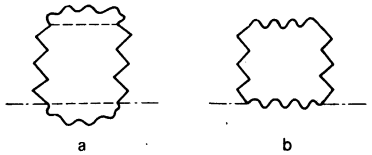

FIG. 5.

on different levels, vanish in the absence of correlation between the chains.

As a result we get

$$\gamma_{ex}(\omega)=\frac{v}{\omega}\Sigma_{ii}(0,\omega)=\frac{\pi}{4}zW^4\frac{v}{T}\int\frac{dq\,dk_1\,dk_2}{(2\pi)^3}$$
$$\times\frac{d\omega_1\,d\omega_2}{\omega(q+k_1)\,\omega(q+k_2)}[1+n_{q+k_1}]n_{q+k_2}S_1(k_1,\omega_1)S_1(k_2,\omega_2)$$
$$\times\delta(\omega-\omega_1-\omega_2-\omega(q+k_1)+\omega(q+k_2)), \qquad (16)$$
$$zW^4=\sum_j W_{ij}^4$$

(z is the number of nearest neighbors).

Bearing $s_q \ll u$ in mind, we get

$$\gamma_{ex}(\omega)=\frac{\pi^2}{2}zW^4\frac{u^2}{v_F}B(T)\int d\omega_1 S_1(\omega_1)S_1(\omega-\omega_1),$$

where

$$B(T)=\int\limits_{|q|<q_m}\frac{dq/2\pi}{[\omega(q)\operatorname{sh}(\omega(q)/2T)]^2}=b(T)\frac{T^2}{\bar\omega^4}q_m, \qquad b(T)\sim 1,$$
$$S_1(\omega)=\int\frac{dk}{2\pi}S_1(k,\omega)=\frac{1}{\pi}\frac{\tau_1}{1+(\omega\tau_1)^2}.$$

Ultimately

$$\gamma_{ex}(\omega)=\frac{\pi}{4}z\left(\frac{W}{\bar\omega}\right)^4\frac{m}{m^*}T\tau_1 b(T)v_F q_m=\frac{1}{4}z\left(\frac{W}{\bar\omega}\right)^4\left(\frac{m}{m^*}\right)^{1/2}b(T)v_F q_m. \qquad (17)$$

We see that for uncorrelated strings, i.e., at $T\gg T_c$, the damping depends little on the temperature. The presence of the factor τ_1 in the intermediate part of formula (17) can attest to the increase of the damping in the vicinity of T_c. In this region, however, a more detailed investigation is needed.

2. At not too strong an anisotropy, when $w/\bar\omega > (\lambda_0/\lambda)(\Delta/\varepsilon_F)^2$, part of the phonons with $|q|>q_m$, satisfy the condition $A_q < w/\bar\omega$. In this case the normal modes must be chosen primarily with allowance for the transitions between the strings (15). This means that the noncoherent modes $\Phi_{q,i}$ (1a) are now the normal ones. The Lagrangian of the system is determined by formulas (3) and (15a). The terms proportional to A_q in (3) must be regarded as perturbations. They contain an explicit dependence on $\chi(x,t)$ and therefore lead to a finite low-frequency damping.

Calculations similar to those of Item 1 of this section yields a result corresponding to diagram 4a with functions A_q in the vertices:

$$\gamma(\omega)=\frac{v}{T}\iint\limits_{|q|>q_m}\frac{dk\,dq}{(2\pi)^2}A_q A_{q-k}\omega_q\omega_{q-k}(1+n_q)n_{q-k}$$
$$\times S_2(k,\omega+\omega(q-k)-\omega(q))=\frac{1}{\pi}d(T)\left(\frac{m}{m^*}\right)^{1/2}(A_{q_m})^2 v q_m, \qquad (18)$$

where the function $d(T)\sim 1$ is defined by

$$d(T)q_m(A_{q_m})^2T^2=\int\limits_{|q|>q_m}\frac{dq}{2\pi}A_q^2\omega_q^2 n_q(1+n_q).$$

Comparing the results (17) and (18) we see that both regions $|q|<q_m$ and $|q|>q_m$ make contributions of the same order to the damping $\gamma(\omega)$. Therefore the uncertainty of the transition region $|q|\approx q_m$ is not essential.

In the case of weak anisotropy of the spectrum and at a low coupling constant of the Debye phonons, when $w/\bar\omega > \lambda_0$, all the normal modes correspond to the representation (1a). In this case the integral in (18) converges on $q\approx\xi_0^{-1}$ and we must put $q_m\to\xi_0^{-1}$ and $A_{qm}\to\lambda_0$. We obtain, as seen from (18), the largest value of $\gamma=\gamma_{max}$ that is possible for the given λ_0:

$$\gamma_{max}\sim\lambda_0^2(m/m^*)^{1/2}\Delta. \qquad (19)$$

This situation can correspond to TCNQ compounds. Putting, according to the estimates of Rice *et al.*,[8] $\lambda_0^2\approx 10^{-3}$, $m/m^*\approx 10^{-2}$, and $\Delta\approx 10^3\,\mathrm{cm}^{-1}$, we obtain $\gamma\approx 10^{-1}\,\mathrm{cm}^{-1}$.

We note that it follows from the quasi-onedimensionality condition $T_c\ll\Delta$ (T_c is the temperature of the three-dimensional transition) that $W/\bar\omega\ll\lambda^2/\lambda_0$, since the lower bound of T_c is estimated at $T_c > W(m^*/m)^{1/2}$ and $m^*/m\approx 4\Delta^2\lambda_0/\lambda^2\bar\omega^2$.[6] (We took into account the elastic interaction between the strings. Coupling via electron tunneling can also be significant.) Therefore the case (19), which corresponds to extremely weak dragging of the phonons by the CDW motion, can occur only at $\lambda_0\ll\lambda$.

3. The result obtained in this part must be compared with the effect on the direct interaction of the CDW on various strings. The interaction energy is given by

$$\mathcal{H}_c=\sum_{i,j}V_{ij}\cos(\chi_i-\chi_j), \qquad (20)$$

where $V_{ij}\approx aT_c^2/zv_F$, a is a number of the order of unity, T_c is the temperature of the three-diagonal transition. In the region $T>T_c$ the chains are not correlated, and their interaction leads to a finite damping γ_c of the oscillations of the CDW phase. With exception of a narrow region near T_c, at $T>T_c$ the condition $\tau_1\gamma_c\ll 1$ will be satisfied everywhere. Therefore, in analogy with the other cases considered above, it suffices to take into account the diagram of Fig. 5b for the self-energy part.

We obtain

$$\operatorname{Im}\Sigma_c(\omega)=zV^2\frac{1}{n(\omega)}\int S_1(k,\omega')S_1(-k,\omega-\omega')\frac{dk\,d\omega'}{(2\pi)^2}, \qquad (21)$$
$$zV^2=\sum_j V_{ij}^2=aT_c^4/zv_F^2, \qquad a\sim 1.$$

Substituting at $T\gg T_c$

$$S_1(x,t)=\exp\{-[|t-x/u|+|t+x/u|]/2\tau_1\}$$

with $\tau_1^{-1}=\pi Tu/v_F$, we get at $\omega\tau_1\ll 1$

$$\gamma_c=\frac{4}{\pi}\frac{a}{z}\frac{u}{v_F}\frac{T_c^4}{T^3}. \qquad (21')$$

It is seen from (21′) that

$$\gamma_c\tau_1=\frac{4}{\pi^2}\frac{a}{z}\left(\frac{T_c}{T}\right)^4\ll 1. \qquad (22)$$

Consequently, the calculation procedure is correct.

At $T\approx T_c$ we have

$$\gamma_c\approx 10^{-1}T_c\gg 1\,\mathrm{cm}^{-1},$$

but at $T\gg T_c$ the value of γ_c decreases rapidly in proportion to T^{-3}. This dependence is characteristic of temperature-induced destruction of pinning of any type [cf. (A4)]. Therefore at room temperature the Fröhlich conductivity is limited only by processes in which thermal phonons take part.

5. CONCLUSION

1. We have considered the damping of an optically active Fröhlich mode because of its interactions with thermal excitations. The mechanisms considered set in, depending on the frequency, in the temperature interval $T_0 < T < \bar{\omega}$ and become fully effective at $T > \bar{\omega}$. We recall that usually $\bar{\omega} \lesssim 100$ K, and the experimentally investigated region is $T < 300$ K, which is the minimum splitting of the phonon terms in the presence of a superstructure. Actually we should have $\delta \sim 10-20$ cm^{-1}, which likewise corresponds to discrimination between the data obtained in the far infrared[1,2] and the microwave[9] bands. At $\omega > \delta$ the decisive processes are the energy transfer from the Fröhlich mode to the subsystem of the high-frequency phonons dragged by the CDW motion (Sec. 2). In this case the damping γ_{ph} and the optical conductivity are determined by formulas (9) and (9a). In the vicinity $\omega \approx \delta$ at strong anisotropy of the phonon spectrum $w < \delta$ a transition of the Fröhlich mode into the dissipative regime is possible, accompanied by a decrease of the values of $\sigma(\omega)$ and $\varepsilon(\omega)$ (Fig. 3). At $\omega < \delta$ the damping of the Fröhlich mode in an isolated chain is determined by the interaction with the main structure. In the commensurate case it sufficies to take into account the superstructure itself [γ_p—formula (A.4)]. In the case of substantial incommensurability, the structure and the superstructure interact with phonon emission or absorption to compensate for the difference between wave vectors [Sec. 3, (13), (14), and (A.7)].

In the region $T > T_c$ an important role is played by damping due to the interaction of the neighboring chains (Sec. IV). The damping is determined either by exchange of the phonons dragged by the CDW motion [γ_{ex}, formula (17)], or by the direct coupling of the CDW with the neighboring strings [γ_c, formula (21′)].

In all cases an important role was played by the division of the thermal excitations into two subsystems: phonons (1b) in the region of the Kohn anomaly with $|q| < q_m$ and the other phonons (1a) with $|q| > q_m$. The quantity $q_m (q_m \gg \xi_0^{-1})$ should as a rule be determined by the transverse width w of the phonon spectrum. If the period of the superstructure is close to twofold or threefold commensurability, then the interaction with the main structure can also determine q_m. The phonons (1a) interact with the CDW via umklapp processes on the superstructure. Their contribution to the damping (18) coincides, apart from a factor, with the contribution (17) of the phonons (1b). At a specially small interaction parameter λ_0 of the electrons with the phonons of the acoustic branch, a situation is possible wherein all the phonons are of type (1a). The damping is then given by (20).

It is important to note that at $T > T_c$ the phonons (1b) are not directly observable in inelastic-scattering experiments. Their spectrum is smeared out to the width $\gamma_1 = \tau_1^{-1}$ of the structure factor $S_1(k, \omega)$. At $T \gg T_c$ we have $\gamma_1 = \pi T u / v_F$. Simple calculation shows that at $T \gg T_c$ this smearing leads to a Lorentz line profile. This result agrees qualitatively with the temperature dependence of the line profile of the amplitude oscillations in KCP.[10]

I note in conclusion that discussions of the considered questions with L. P. Gor'kov, I. E. Dzyaloshinskiĭ, and E. I. Rashba were most influential in the development of this research.

APPENDIX

1. The damping of the CDW phase oscillations were investigated in Sec. 3 for an interval where the incommensurability parameter q_n lies in the region $\xi_0^{-1} \ll |q_n|/2 < q_m$, where q_m is the dividing line between states of type (1a) and (1b).

At $|q_n| \ll \xi_0$ the first terms (α) in the curly brackets of (13) and (14) remain in force if α is regarded as a mode of the amplitude oscillations:

$$\omega_\alpha(q) = (u/v_F)[8\Delta^2 + {}^2/_3 v_F^2 q^2]^{1/2}, \quad |q| \ll \xi_0^{-1}.$$

It is more natural to express the coefficient C_n in (11) in terms of the pinning frequency ω_p:

$$\omega_p^2 = 2n^2 C_n v \Delta^2 / v_F.$$

From (14) we obtain directly

$$\gamma_U = \frac{3}{32}\frac{1}{n^2}\left(\frac{m^*}{m}\right)^{1/2}\left(\frac{\omega_p}{\omega_0}\right)^3 \frac{\omega_p}{u|q_n|} T \qquad (A.1)$$

[when account is taken of the coupling between the strings, we have $|q_n| > T_c/v_F$ in (A.1)].

The second term (β) in (13) or (13a) must be supplemented with approximations of higher order in $(uq_n\tau_n)^{-1}$. The result corresponds to direct interaction of the CDW with the main structure, an interaction described by the Hamiltonian

$$\mathcal{H}_p = \frac{\omega_p^2}{2n^2 v}[1 - \cos(n\chi + q_n x)]. \qquad (A.2)$$

We are interested in the region of weak pinning $T \gg T_p \sim \omega_p v_F / u$, which was investigated previously[7] for the quantum region $T \ll T_0$. At $T \gg T_0$ it is merely necessary to modify the form of the structure factor $S_n(k, \omega)$. According to Ref. 7,

$$\Sigma_p(k, \omega) = \frac{\omega_p^2}{8n^2 v^2}[K_n(q_n, 0) - K_n(k + q_n, \omega)], \qquad (A.3)$$

where $K(x, t)$ is defined by formula (12). In the preceding paper[7] the corresponding expression was derived as a correction to the Green's function of the phase, so that its applicability was restricted to the region $\gamma \ll \omega$. However, arguments similar to those advanced when the diagrams were selected in the present paper show that this correction can also be regarded as the self-energy part of (A.3). Consequently, the result that follows is valid also at $\gamma > \omega$, provided that $\gamma\tau_n \ll 1$.

Substituting (12) in (A.3) we get

$$\gamma_p = \frac{v}{\omega}\Sigma_p(0, \omega) = \frac{1}{\pi}\sin\left(\pi n^2 \frac{u}{v_F}\right)\frac{\omega_p^4}{[(q_n u)^2 + \tau_n^{-2}]^2} T. \qquad (A.4)$$

If $uq_n\tau_n \gg 1$, then (A.4), just as (A.3), yields $\gamma \sim T$, i.e., $\sigma \sim T^{-1}$. The relative value of γ_p/γ_U can be different.

2. If $|q_n|/2 > q_m$, then the resonant modes must be chosen in the representation (1a). We must use in lieu of (11)

$$\tilde{\mathcal{H}}_U = C_n \frac{\Delta^2}{v_F}[\exp\{iq_n x + (n-2)\chi(x, t)\}\Phi^2(x, t) + \text{c.c.}]. \qquad (A.5)$$

Accordingly, we must replace $S_n(k,\Omega)$ in (13) by $S_{n-2}(k,\Omega)$, and use ω_q in place of $\omega_{\alpha,\beta}(q)$. The ω_q spectrum is not symmetrical about $q=0$ (the point $2p_F$). In place of the resonance points $\pm q_n/2$ we have the points $-k_n$ and q_n-k_n, so that

$$\omega_{-k_n}=\omega_{q_n-k_n}.$$

Outside the term repulsion region $q\approx q_0$ we can put

$$\omega_q\approx\begin{cases}\omega_\beta(q) & q<0\\ \omega_\alpha(q) & q>0\end{cases},$$

$|q|\gg\xi_0^{-1}$, so that the transitions are between the α and β terms. In place of (14) we get at $\bar{\omega}\tau_{n-2}\gg1$

$$\gamma_U(\omega)=\pi^2(n-2)^2C_n^2\left(\frac{m}{m^*}\right)^3\frac{\Delta^4}{T}\frac{v_F}{s(q_n-k_n)-s(-k_n)}\times\frac{1}{\omega^2(-k_n)\mathrm{sh}^2[\omega(-k_n)/T]}. \tag{A.6}$$

Formulas (14) and (A.6) give relations that agree qualitatively. At a temperature $T>\bar{\omega}$ we must consider the case $\bar{\omega}\tau_{n-2}<1$, i.e.,

$$\bar{\omega}<\pi(n-2)^2(u/v_F)T.$$

In accord with the discussion at the end of Sec. 3 we obtain in place of (A.6)

$$\gamma_U\approx\pi^2(n-2)^2C_n^2\left(\frac{m}{m^*}\right)^3\left(\frac{\Delta}{\bar{\omega}}\right)^4T\tau_{n-2}v_F\bar{Q}, \tag{A.7}$$

where

$$\bar{Q}=\bar{\omega}^4\int\frac{dq}{\omega_q^4}\sim Q,$$

Q is the reciprocal-lattice vector. In (A.7) we must take into account all values of n, and the optimal is $n=3$. We obtain the following estimate:

$$\gamma_U\geqslant10^{-4}v_FQ\approx(1-10)\ \mathrm{cm}^{-1} \tag{A.8}$$

independently of the temperature. Activation of the intramolecular oscillations can result in an increase of (A.8) with increasing temperature.

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Optical-orientation anisotropy produced in semiconductors by quadrupole splitting of the spin levels of the lattice nuclei

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The onset of crystal anisotropy of the magnetic depolarization of recombination radiation under optical-orientation conditions is considered theoretically. The influence of the anisotropy of the nuclear field on the behavior of the average spin of the excited electrons is analyzed on the basis of general considerations. The concrete model chosen to describe this anisotropy is quadrupole splitting of the nuclear spin levels. Calculations for an external magnetic field much stronger than the local field produced at the nucleus by the neighboring nuclei agree with the experimental data in the corresponding region of magnetic-field values.

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It was recently observed that the crystal anisotropy exerts a substantial influence on the optical orientation of electrons in semiconductors of the gallium-arsenide type.[1-5] The shape of the plot of the magnetic depolarization of the recombination radiation (the Hanle curve) turned out to be strongly dependent on the orientation of the external magnetic field relative to the crystal axes. The crystal anisotropy manifests itself particularly strongly in the hysteresis observed by Novikov and Fleĭsher[1,4] when the exciting-light beam is directed along the [001] axis and the magnetic field is located in the (001) plane of the crystal. In this geometry, the hysteresis exists in a narrow region of angles near a field direction along the [110] axis.

The anisotropy of the Hanle curve was interpreted as a manifestation of the quadrupole splitting of the spin levels of the lattice nuclei, which influences the dynamic polarization of the nuclei by oriented electrons. It is known that dynamic polarization of the nuclei leads to

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