Criterion for Peierls instability in quasi-one-dimensional metals

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(Submitted April 30, 1977; resubmitted July 6, 1977)
Pis'ma Zh. Eksp. Teor. Fiz. **26**, No. 5, 349–352 (5 September 1977)

The phonon spectrum of a strongly anisotropic quasi-one-dimensional metal is analyzed with allowance for the flexural oscillations of the individual chains, and a criterion is derived for the Peierls instability. It is shown that if the electron-phonon interaction is strong enough, the Peierls transition is "three-dimensional" and of first order. At the same time, a structural second-order phase transition accompanied by a softening of the low-frequency transverse-flexural phonon mode is possible.

PACS numbers: 63.20.Kr

Peierls instability in quasi-one-dimensional systems has been the subject of intenstive study of late.[1,2] In most theoretical papers devoted to the Peierls transition, the phonon spectrum is assumed to be three-dimensional and isotropic (see, e.g., [3–6]). Furthermore, in none of the studies, with the exception of,[4] has the Peierls instability been consistently analyzed within the framework of identical approximations of the electron-phonon and Coulomb interactions (the latter has most frequently been reduced to a renormalization of the coupling constant).

We consider in this paper the structural phase transition in quasi-one-dimensional conductors with strongly anisotropic electronic and elastic properties, and use as the basis a continual model of a metal,[4,7] wherein the lattice vibrations and the electron-density oscillations are described in a unified self-consistent manner. This model is valid in the case in question, since the period of the charge-density wave (CDW), which is equal to $L = \pi/2k_F$ (where k_F is the Fermi momentum), greatly exceeds the period of the initial lattice. Assuming the matrix element of the hops between the chains to be large enough[8] we use the self-consistent-field approximation[11] to calculate the critical temperature T_c of the Peierls transition.

The linearized system of equations for small elastic vibrations in an isotropic metal is given in[4,9]. For crystals having the tetragonal symmetry typical, in particular, of $K_2Pt(CN)_4Br_{0.3}3H_2O$(KCP),[10] the nonzero elastic moduli are

$$\left.\begin{aligned} &\lambda_{zzzz} \equiv \beta; \quad \lambda_{xxxx} = \lambda_{yyyy} \equiv \alpha_2; \quad \lambda_{xyxy} \equiv \alpha_1; \\ &\lambda_{xxzz} = \lambda_{yyzz} \equiv \nu; \quad \lambda_{xzxz} = \lambda_{yzyz} \equiv \mu; \quad \lambda_{xxyy} \equiv \delta. \end{aligned}\right\} \tag{1}$$

In quasi-one-dimensional crystals consisting of weakly coupled linear chains of atoms or molecules, the following condition is satisfied:

$$\beta >> \alpha_1, \alpha_2, \delta, \mu, \nu. \tag{2}$$

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In addition to the acoustic oscillations, it is necessary to take into account here also the flexural oscillations of individual chains.[11] In view of the weak (hopping) conductivity across the chains, the description of the electron-phonon interaction need only take into account the longitudinal component of the deformation potential $\Lambda_{zz} \equiv \Lambda_{\parallel}$. As a result, the dispersion equation for the phonon spectrum takes the form

$$\omega^6(\mathbf{k}) + B(\mathbf{k})\omega^4(\mathbf{k}) + C(\mathbf{k})\omega^2(\mathbf{k}) + D(\mathbf{k}) = 0. \tag{3}$$

In the case of purely longitudinal oscillations ($k_{\perp} = 0$) the free term is $D(\mathbf{k}) \equiv 0$, so that the Peierls-instability condition, which corresponds to the vanishing of the frequencies as a result of the logarithmic singularity of the polarization operator of the one-dimensional electron gas at the point $k_z = 2k_F$, takes the form $C(\mathbf{k}) = 0$. This leads to an expression for the critical temperature of the Peierls transition

$$T_{c\parallel} \cong E_F \exp\{-1/g_{\parallel}\}; \qquad g_{\parallel} = \nu_F \frac{2N_0\Lambda_{\parallel} + \dfrac{(\Lambda_{\parallel}k_F)^2}{\pi e^2} - \beta}{\left(N_0 + \Lambda_{\parallel}\dfrac{k_F^2}{\pi e^2}\right)^2}, \tag{4}$$

where ν_F is the state density on the Fermi surface, E_F is the Fermi energy, and N_0 is the concentration of the conduction electrons. It must be emphasized that if the screened Coulomb interaction is consistently taken into account, then the principal terms of order $\Pi_{\parallel}(2k_F) \approx \nu_F \ln(E_F/T)$ are cancelled in $C(\mathbf{k})$, so that a structural second-order phase transition and the onset of a longitudinal CDW with $\mathbf{q} = \{0, 0, 2k_F\}$ are possible only under the condition (cf.[41]):

$$2N_0\Lambda_{\parallel} + \frac{(\Lambda_{\parallel}k_F)^2}{\pi e^2} - \beta > 0. \tag{5}$$

The condition of "three-dimensional" structural instability of a crystal lattice at $k \neq 0$ (but $k_{\perp}^2 \ll k_z^2$) corresponds to the appearance of complex roots of Eq. (3), and reduces accurate to terms $\sim (k_{\perp}/k_z)^4$ and $(\gamma^2 k_z^2/\beta\rho)^2$, where ρ is the density of the crystal and γ^2 is proportional to the flexural rigidity of the chain of atoms,[11] to the approximate equality $B(\mathbf{k}) \cong 0$, from which we get:

$$T_c \cong E_F \exp\{-1/\nu_F V\}; \tag{6}$$

$$V = \frac{\left[2N_0\Lambda_{\parallel} + \dfrac{(\Lambda_{\parallel}k_F)^2}{\pi e^2} - \beta\right] - \left(\dfrac{k_{\perp}}{2k_F}\right)^2\left[a_1 + a_2 + \mu - \dfrac{(\Lambda_{\parallel}k_F)^2}{\pi e^2}\right]}{N_0 + \left(\Lambda_{\parallel}\dfrac{k_F^2}{\pi e^2}\right)^2 + \left(\dfrac{k_{\perp}}{2k_F}\right)^2\left[2\left(N_0 + \Lambda_{\parallel}\dfrac{k_F^2}{\pi e^2}\right)^2 + \left(\dfrac{k_F^2}{\pi e^2}\right)^2\right]}. \tag{7}$$

A phase transition into the dielectric state is possible only under the condition $V > 0$, which is a generalization of the criterion (5). Inasmuch as none of the roots of (3) vanishes if $B(\mathbf{k}) = 0$, the lattice restructuring takes place jumpwise, corresponding to a first-order phase transition. This conclusion agrees with

experiments[11] according to which the giant Kohn anomaly in KCP never leads to a complete softening of the lattice ($\omega \neq 0$), while in TTF-TCNQ the phase transition at $T = 38$ K is accompanied by hysteresis.[12] As follows from[7], that the condition

$$(\Lambda_{\parallel} k_F)^2/\pi e^2 - a_2 - \mu > 0 \quad , \tag{8}$$

the coupling constant V increases with increasing $k_{\perp}$, so that the three-dimensional CDW, which is observed both in KCP and in TTF-TCNQ,[1,2] is favored.

In addition to the structural transitions considered above, if $D(\mathbf{k}) = 0$, one second-order phase transition is also possible at the point $k_z = 2k_F$ at $k_{\perp} \neq 0$, and is accompanied by a vanishing of the frequency of the transverse-flexural oscillation mode. The critical temperature of this transition is

$$T_c' \cong E_F \exp\left\{ - \frac{\beta (2k_F)^2 + \mu k_{\perp}^2}{\nu_F \Lambda_{\parallel}^2} \right\} . \tag{9}$$

It appeared that this situation takes place in KCP, where the structural transition at $T \approx 120$ K[13] was seen to be followed by a jump of Young's modulus at $T \approx 35$ K.[14]

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