Motion of the Fröhlich mode and the conductivity of $NbSe_3$

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It is shown within the framework of a model with superposed parts of the Fermi surfaces that the contribution of the free Fröhlich mode to the conductivity near the transition temperature can, in principle, cancel out with a high degree of accuracy the decrease that occurs in the conductivity as a result of the appearance of a gap in the electron-energy spectrum. Such a behavior has been observed by Ong and Monceau [Phys. Rev. **B16**, 3443 (1977)] in $NbSe_3$.

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The quasi-one-dimensional conductor $NbSe_3$ undergoes two structural transitions at the temperatures $T_{c1} = 145$ K and $T_{c2} = 59$ K into incommensurate phases with wave vectors $\mathbf{q}_1 = (0; 0.243; 0)$ and $\mathbf{q}_2 = (0.5; 0.263; 0.5)$ respectively (for references, see Ong and Monceau's paper[1]). Both transitions greatly affect the conduction-electron subsystem, as evidenced by the presence of anomalies in the resistance in the neighborhood of the transition temperatures. The latter circumstance, as is now generally believed, provides a serious argument in favor of the electronic origin of the structural transitions themselves. Because of the quasi-unidimensional ($1D$) nature of the energy spectrum of the conduction electrons, it is reasonable to ascribe the structural transition to a Peierls-type instability caused by the approximate coincidence of some sections of the Fermi surfaces.

The lattice distortion, $u(\mathbf{r})$, in the new phase has the form

$$u = u_0 \cos(\mathbf{qr} + \varphi). \tag{1}$$

The arbitrary phase φ reflects the fact that the vectors $\mathbf{q}_1$ and $\mathbf{q}_2$ are incommensurable with the original high-temperature lattice constant, and, consequently, the associated electron and lattice-deformation wave could propagate along the direction of the chain, producing a new possible conductivity mechanism—the so-called Fröhlich conductivity. Lee *et al.*[2] have shown that such a mechanism could prove to be important for the properties of quasi-unidimensional materials, but that its manifestation in the conductivity of real materials is, probably, not certain because of the numerous effects of wave pinning by the impurities or imperfections of the crystal. It is, nevertheless, believed that the recent measurements of the non-Ohmic conductivity of $NbSe_3$ in the region of very weak electric fields and the high sensitivity of the anomalies in the resistance to the field frequency (see Ref. 1) were the first observation of the contribution of the Fröhlich mode to the conductivity of a quasi-unidimensional conductor.

It has already been argued[1] that the low electric-field strengths that wash out the anomalies in the resistance in the vicinity of the transition at T_{c1} or T_{c2} can be explained only if by chance some large characteristic dimension participates in the phenomenon. This observation led to ideas, according to which such a scale is provided by the weak pinning of the superlattice [or the charge-density wave (CDW)] on crystal defects. A qualitative analysis[3] attests the point of view of the authors of Ref. 1.

In the present paper, using a microscopic approach, we attempt to describe the anomalies in the resistance in the immediate neighborhood of the transition temperature. In our model description, we assume that the electron Fermi surfaces even though they are not plane surfaces because of the three-dimensional interchain tunneling nevertheless satisfy some "superposition criterion" that is sufficient for a Peierls instability to occur at low temperatures. The problem in question consists in the fact that, according to the experimental data,[1] in the presence of strong electric fields, or for conduction at "high" frequencies, the anomaly in the resistance at the transition temperature simply gets washed out without giving rise to an additional contribution with respect to the conductivity of the metal phase. We shall show that this result is indeed reproducible at least within the framework of the reasonably plausible model, although some increase in the conductivity below T_c as a result of an additional contribution from the traveling charge density wave might have been expected here. The "pinning-depinning" mechanisms per se will not be considered below (see Ref. 3).

As is well known (see Refs. 4 and 5), a lattice becomes unstable at a sufficiently low temperature if the electron spectrum satisfies the relation

$$\varepsilon(\mathbf{p}+\mathbf{q}) = -\varepsilon(\mathbf{p}), \tag{2}$$

where $\mathbf{q}$ is the instability wave vector. The simplest case of a plane Fermi surface (the Peierls instability in one-dimensional conductors) is at the same time the most complicated for a theoretical investigation, which is due to the decisive role of the fluctuations in one-dimensional systems.[6] Fortunately, in the case of $NbSe_3$ there are numerous experimental indications that its properties can be understood in a model that does not exhibit all these specific one-dimensional complications. Indeed, x-ray measurements[7] have shown that, for the two transitions corresponding to the wave vectors $\mathbf{q}_1$ and $\mathbf{q}_2$, the fluctuations are small, and well-defined Bragg reflections appear in each phase just below the transition temperature. The observation of Shubnikov-de Haas oscillations in the magnetoresistance[8]

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shows that the Fermi surfaces, being anisotropic and quite small in the low-temperature phase, have a well-pronounced three-dimensional character. Consequently, it can be assumed that the Fermi surfaces of the carriers in $NbSe_3$, while retaining the one-dimensional features characteristic of a filamentary conductor, are at the same time appreciably deformed by the three-dimensional effects of electron tunneling between the filaments. Consequently, the Eq. (2) for some vector **q** can be satisfied only approximately. In other words, the coincidence of the Fermi surfaces is not complete and this leads to a semimetal picture at low temperatures. Whether or not a structural transition occurs under such conditions depends on the strength of the electron-phonon interaction. For a fixed strength of the latter, the transition temperature evidently decreases with enhancement of the three-dimensional effects.

Let $\varepsilon_+(\mathbf{p})$ ($\varepsilon_-(\mathbf{p})$) be the electron energy on the right (left) hand side of the Fermi surface (the electron is assumed to have zero energy at the Fermi level). The distortion of the lattice with the wave vector **q** removes the degeneracy (2), giving the well-known form for the new electron spectrum:

$$\varepsilon_{1,2}(\mathbf{p}) = \eta(\mathbf{p}) \pm (\xi^2 + \Delta^2(\mathbf{p}))^{1/2}. \qquad (2')$$

Here $\varepsilon = v(p - k_F)$ is the distance from one of the two Fermi surfaces [say, on the right-hand side of $\varepsilon_+(p_\parallel, p_\perp) = v(p_\parallel - k_F) + \eta_+(p_\perp)$], $2\eta(\mathbf{p}) = \varepsilon_+(\mathbf{p}) + \varepsilon_-(\mathbf{p}+\mathbf{q})$, where $\eta(\mathbf{p})$ can be regarded as a quantity that varies only along the Fermi surface and $\Delta(\mathbf{p}) = d_{ep}|u_0|$ is the "gap", proportional to the lattice-distortion amplitude $|u_0|$ and the electron-phonon deformation potential d_{ep}, in the new electron spectrum.

The properties of $\eta(\mathbf{p})$ and $d_{ep}(\mathbf{p})$ depend on the actual band structure, and will not be specified in this paper. As to $\Delta(T)$ [i.e., $u_0(T)$], its temperature dependence is usually determined by minimizing the total elastic energy. Regardless of the specific model, the value of the transition temperature T_c is of the order of $\Delta(T=0)$; for $|T - T_c| \ll T_c$ we have $\Delta^2(T) \propto T_c(T_c - T)$ [see Eq. (14) below].

After these observations of a general character, let us proceed to compute the temperature dependence of the conductivity (near T_c). According to Ong and Monceau,[1] $NbSe_3$ is a good conductor with a highly temperature-dependent resistance (the residual resistance is low in good samples). Consequently, scattering by the impurities does not play any significant role in the relaxation processes at $T \sim T_c$. Above T_{c1} the resistance in the metal phase obeys a linear law with a high accuracy [$\rho \propto T^{0.98}$ (Ref. 1)]. This fact speaks in favor of the electron-phonon scattering mechanism. As to the static defects, they are responsible for the nonlinear part of the current-voltage characteristic.[1,3] Below we restrict ourselves to only two limiting situations: the superlattice is completely fixed, or, in its turn, the pinning can be completely neglected. In its general form, the electric current contains contributions from both the single-particle excitations with the new energy (2′) and the running charge-density wave.

In Ref. 3 Lee and Rice make the assumption that the mobility of the charge-density wave is due to the interaction of the electrons largely with the so-called phase mode (i.e., with the acoustic mode that appears simultaneously with the superlattice in the new phase). Such a process could be important at very low temperatures, but at high temperatures, and particularly in the vicinity of the transition, all the thermal phonons should be taken into account in the study of the friction for the CDW (i.e., of the CDW kinetics). By assumption, because of the three-dimensionality of the transition, the role of the soft phonons amounts to that of small fluctuations. Below we shall, for simplicity, consider only the vicinity of T_{c1}. The set of inequalities used has the following form

$$\eta(\mathbf{p}) \sim T \gg \Omega \gg 1/\tau;\ \Delta, \qquad (3)$$

where Ω is the characteristic phonon frequency and $1/\tau \propto T$ is the electron-phonon relaxation time.

The method of kinetic equations ceases to be simple for the system in the new phase; this is particularly true for the processes of relaxation of the order parameter (the lattice distortion and its phase φ). This aspect of the problem reminds us of a similar problem in the theory of nonequilibrium phenomena for superconductors. We shall follow the method of analytic continuation of thermodynamic responses developed in Ref. 9. For example, in order to find the electric current that is linear in the electric field, it is necessary to write down[9] the responses to both the applied field and the field-induced perturbation of the order parameter. In its turn, the change in the order parameter (1) can be found from the balance between the two responses, each of which represents a driving force produced by the field, and the frictional force.

The two equations are schematically shown in Fig. 1. All the lines are labeled by two indices (±), which represent the diagonal and off-diagonal elements of the matrix Green function,

$$\hat{G}_z = \begin{pmatrix} G_{++} & G_{+-} \\ G_{-+} & G_{--} \end{pmatrix}, \qquad (4)$$

describing the electron subsystem in the presence of the deformation (1), which mixes the (±) sides of the Fermi surface. The upper line in Fig. 1 carries the sum frequency z; the lower line, the frequency $z - \omega_Q$, where ω_Q is the field frequency. To compute the conductivity in a constant field, it is necessary to retain only the first nonzero terms in ω_Q.

In the method proposed in Ref. 9, we begin by writing down the responses in Fig. 1 in the Matsubara representation ($z = iz_n = i(2n+1)\pi T$; $\omega_Q = i\omega_n = i \cdot 2n\pi T$) and then continue all the expressions to the physical-frequency axis. The result of the analytic continuation is expressed by the relation

$$T\sum_{\varepsilon} \hat{G}^{(1)}_{\varepsilon,\varepsilon-\omega} = -\int_{-\infty}^{+\infty} \frac{dz}{4\pi i} \left\{ \mathrm{th}\left(\frac{z-\omega}{2T}\right) \hat{G}^{(R)}_z \hat{h}_\omega \hat{G}^{(R)}_{z-\omega} \right.$$

$$\left. - \mathrm{th}\left(\frac{z}{2T}\right) \hat{G}^{(A)}_z \hat{h}_\omega \hat{G}^{(A)}_{z-\omega} + \left[\mathrm{th}\left(\frac{z}{2T}\right) - \mathrm{th}\left(\frac{z-\omega}{2T}\right) \right] \hat{G}^{(R)}_z \hat{h}_\omega \hat{G}^{(A)}_{z-\omega} \right\}, \qquad (5)$$

where $\hat{G}^{(R)}$ and $\hat{G}^{(A)}$ are the retarded and advanced functions (4):

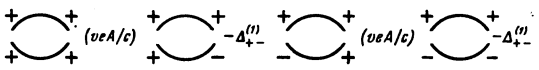

FIG. 1. Diagrammatic representation of the responses in the expressions for the current and the lattice distortion.

$$\hat{G}_z = \frac{1}{(z+\eta)^2-\xi^2-\Delta^2}\begin{pmatrix} (z+\eta+\xi), & \Delta_{+-} \\ \Delta_{-+}, & (z+\eta-\xi) \end{pmatrix}. \tag{6}$$

The perturbation Hamiltonian has the form

$$\hat{h}_\omega = \begin{pmatrix} (veA_\omega/c), & -\Delta_{+-\omega}^{(1)} \\ -\Delta_{-+\omega}^{(1)}, & -(veA_\omega/c) \end{pmatrix}. \tag{7}$$

Equation (5) for $\hat{G}^{(1)}$ preserves the diagram structures of Fig. 1 and, consequently, has a form suitable for the computation of a number of relaxation processes. Such a procedure was used earlier[9] only for scattering on static impurities. Let us formulate without detailed derivation the analogous rules for electron-phonon scattering in the limiting case $T \gg \Omega$. Essentially, in this case the thermal phonons create for the electron motion some disordered potential that varies slowly compared to the electron-energy scale, T. Scattering by this potential gives corrections to the electron Green functions (Fig. 2a) or the loops (Figs. 2b and 2c). The principal difference between this procedure and the diagram technique[9] for impurity scattering consists in a change in the factor corresponding to a dashed line between two crosses in Fig. 2:

$$\mathrm{cth}\,(\omega/2T)\ \mathrm{Im}\,(D_\omega^R - D_\omega^A) = 2g_{ep}^2 T\omega_0(\mathbf{k})\,\delta(\omega^2-\omega_0^2(\mathbf{k})), \tag{8}$$

where g_{ep} is the dimensionless electron-phonon interaction constant.

In other words, scattering by the phonons is not elastic, but quasielastic, with energy transfer $\omega_0(\mathbf{k}) \sim \Omega \ll T$. Near the transition temperature $\Omega \gg \Delta$, and all the corrections of the type shown in Fig. 2c can be discarded. In the simplest isotropic-scattering variant, the relaxation rate, $1/\tau = 2\pi g_{ep}^2 T$, does not depend on the location of the electron momentum on the Fermi surface, and to obtain the final results it is sufficient to simply compute the diagram of Fig. 1, replacing the G matrices by their expressions (6) with $z \to z \pm i/2\tau$. Near T_c, the time τ does not differ from its value in the metallic phase.

As noted above, in the course of the computations we retain only the terms linear in the external frequency ω, thus making the electric field a constant field:

$$i\omega A_\omega/c = E \text{ and } -i\omega\varphi^{(1)} = \dot{\varphi}.$$

With allowance for (3), we can significantly simplify the evaluation of (5), since the first two (regular) terms make contributions when $z \sim \varepsilon \sim T$, whereas the main contribution from the integration in the products $\hat{G}^{(R)}\hat{G}^{(A)}$ corresponds to the neighborhoods of the poles of Eq. (6). Consequently, this last term describes the

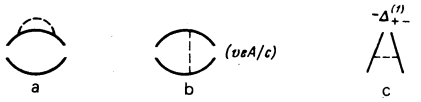

FIG. 2. Corrections to the diagrams in Fig. 1 as a result of the electron-phonon scattering processes.

kinetics of the carriers, and normally make the dominant contributions to all the transport coefficients.

After obvious, but somewhat tedious computations, we obtain the following expression for the longitudinal current ($\mathbf{j} \| \mathbf{1}$):

$$\begin{aligned} j = {} & e^2 vES(2\pi)^{-2}\{2\tau/\pi - \langle R^{-1}\rangle(\Delta\tau)^2/T(4(\Delta\tau)^2+1)^{1/2}\} \\ & \dot{\varphi} eS(2\pi)^{-2}\{\langle\psi_+''+\psi_-''\rangle\Delta^2/16\pi^3T^2 \\ & -\langle R^{-1}\rangle\Delta^2\tau/2T(4(\Delta\tau)^2+1)^{1/2}\}. \end{aligned} \tag{9}$$

In its turn, for $\varphi = 2k_F u$, where u is the velocity of the superlattice, we obtain the relation

$$\begin{aligned} 0 = {} & eE\{-v\Delta_{+-}\langle\psi_+''+\psi_-''\rangle/4\pi^2T^2 \\ & +2\langle R^{-1}\rangle v\Delta_{+-}\tau/T(4(\Delta\tau)^2+1)^{1/2}\} \\ & +\dot{\varphi}\{\langle R^{-1}\rangle\Delta_{+-}/T - \langle R^{-1}\rangle\Delta_{+-}/T \\ & +\langle R^{-1}\rangle\Delta_{+-}/T(4(\Delta\tau)^2+1)^{1/2}\}. \end{aligned} \tag{10}$$

The angular brackets in (9) and (10) denote averaging of the corresponding expressions over one side of the Fermi surface:

$$\langle\ldots\rangle = S^{-1}\int d^2p_\perp \ldots$$

In Eqs. (9) and (10) we have used the following notation:

$$\begin{aligned} & R = \mathrm{ch}^2(\eta/2T); \\ & \psi_+'' = \psi''(1/2+i\eta/2\pi T), \quad \psi_-'' = \psi''(1/2-i\eta/2\pi T) \end{aligned} \tag{11}$$

(ψ is the derivative of the logarithm of the gamma function). We have

$$\psi_+''+\psi_-'' = \begin{cases} -28\zeta(3), & \eta \ll 2\pi T \\ 2(\eta/2\pi T)^{-2}, & \eta \gg 2\pi T \end{cases}. \tag{11'}$$

As to Eq. (10), it is, to a certain extent, of a symbolic nature. Indeed, (10) is applicable only in the limit of a completely free (uncoupled) charge-density wave. In weaker fields it is necessary to consider the significantly more complex problem of superlattice drift in the presence of pinning forces. In the opposite case of very weak fields (the charge density wave is fixed), we can set $\varphi = 0$ in the expression, (9), for the current.

Thus, for this last case we obtain directly the following anomaly in the resistance:

$$\sigma = \sigma_0\{1 - \pi\Delta^2\tau\langle R^{-1}\rangle/2T(4(\Delta\tau)^2+1)^{1/2}\}. \tag{9'}$$

The additional term in the resistance is linear in $T_c - T$ in the vicinity of T_c ($\Delta\tau \ll 1$), but its dependence on temperature becomes more critical when $\Delta\tau \gg 1$.

In principle, it would not be so difficult to write down a general expression for the conductivity at lower temperatures. We believe that to do this within the framework of the present paper would be pointless, since it would then be necessary for us to draw on more detailed information about the band structure in $NbSe_3$. Nevertheless, it is clear that the conductivity has a minimum if the new phase is, by assumption, semimetallic in character. It can be seen from Eq. (9') that the magnitude of the anomaly in the resistance is very sensitive to the parameter η.

Let us turn to the opposite limiting case of the free motion of the superlattice. For $\eta = 0$

$$\sigma = \sigma_0\{1 + (7\zeta(3)/2\pi^3)(\Delta/T)^2\}, \tag{12}$$

i.e., the running mode increases the conductivity, which was intuitively assumed. This increase is not

large if $T\tau \gg 1$. Denoting the quadratic—in Δ—corrections in Eqs. (9′) and (12) respectively by $\delta\sigma_-$ and $\delta\sigma_+$, we see that

$$|\delta\sigma_-| = (\pi^4/7\zeta(3))\, T\tau\, |\delta\sigma_+|. \qquad (12')$$

In this sense our result is in accord with the Ong-Monceau experiments,[1] in which the "uncoupled" wave simply effaces the anomaly in the resistance at T_c by exactly compensating for it.

According to (10), the result for finite η is qualitatively the same, although the conductivity increases even more slowly and the sublattice mobility decreases:

$$u = -(evE\tau/k_F)\{1-(\langle\psi_+''+\psi_-''\rangle/\langle R^{-1}\rangle)(4(\Delta\tau)^2+1)^{1/2}/8\pi^3 T\tau\}. \qquad (13)$$

A curious property of the last expression is that the factor $\psi_+''+\psi_-''$ could change its sign at sufficiently large values of η. There is no physical contradiction here, as follows from the form of the equation determining, on the one hand, the theoretical dependence of T_c on the parameter η and, on the other, the magnitude of the "gap" $\Delta(T)$ in the vicinity of the transition curve $T_c(\eta)$:

$$\ln(T_c/T_{c0}) = \psi(1/2) - \langle\psi(1/2+i\eta/2\pi T_c) + \psi(1/2-i\eta/2\pi T_c)\rangle + (\Delta^2/16\pi^2 T_c^2)\langle\psi_+''+\psi_-''\rangle. \qquad (14)$$

Evidently, the combination $\psi_+''+\psi_-''$ also enters into the fourth-order term of the phenomenological expression of the Landau theory of second-order phase transitions. If this coefficient changes its sign at $\eta/2\pi T = 0.306$, then the transition becomes a first-order transition. Our analysis assumed the change in the quantities at the transition point to be small. In $NbSe_3$ the two transitions into the new phase are continuous.[1,7]

In Ref. 10, applying a mathematical analog of our model (for fixed η) to the problem of the superconductivity of a magnetized metal,[11] Mnatsakanov and one of the present authors also show that the wave vector $\mathbf{q}$ at which overlapping of the Fermi surfaces occurs changes at sufficiently low temperatures. It would be tempting to ascribe the second transition, at T_{c2}, to this phenomenon. However, following Ref. 7, we believe that another explanation is more probable: to wit, that the two transitions in $NbSe_3$ are connected with different systems of niobium filaments. Because of the nature of the crystallographic symmetry, the unit cell of $NbSe_3$ has, as is well known,[1,7] two sets of nonequivalent filaments. The band properties of the electrons of the two sets of filaments probably do not differ too greatly from each other. Nevertheless, this may prove to be sufficient for Peierls transitions to occur in the two systems of filaments separately as a result of interaction with different active phonons. (Apparently, the Coulomb interaction between the filaments also plays a role in the second transition at $T = T_{c2}$, since the corresponding vector $\mathbf{q}_2$ corresponds to a doubling of the lattice constant in the transverse direction.[12])

The most important question is whether there are other carriers in $NbSe_3$ besides the carriers on the Nb chains, i.e., whether there is overlap with other bands. The large magnitude of the anomalies in the resistance, as well as the fact that the vector $2(\mathbf{q}_1+\mathbf{q}_2)$ coincides to within the experimental error with the reciprocal-lattice vector,[1)] indicates that the answer to this question is in the negative. Assuming on the basis of the values of the resistance near T_{c1} and T_{c2} (Ref. 1) the value $n \sim 10^{21}$ cm^{-3} for the carrier-number density, we estimate that $(1/\tau)_{c1} \sim (1/\tau)_{c2} \sim 10^{14}$ sec^{-1}. Of course, in this case the inequalities (3) are violated, and it is difficult to compare the expression (9) directly with the experimental curves for the resistance[1] near T_{c1} and T_{c2}, but the main anomalies are undoubtedly accurately described, and in this respect our results are apparently not dependent upon the refinement of the calculation.

Let us, in conclusion, explain that the found anomalies in the expressions (9) and (10) are connected primarily with those electrons whose energies lie close to the gap that opens up in the energy spectrum, and whose density of states consequently have singularities. The relative number of such carriers at finite temperatures (and for $\Delta \sim 1/\tau$) is proportional to Δ/T.

The main result of the paper consists in the fact that, in the approximation (3), the decrease in the conductivity near T_c as a result of the decrease in the number of carriers upon the formation of a gap in the spectrum coincides with a high degree of accuracy [see (12′)] with the greatest possible contribution that can be made to the conductivity by the traveling Frölich mode. Therefore, comparing this result with the data of Ref. 1, we can conclude that the interpretation of these data as proof of the existence of a Fröhlich mechanism of conductivity is probably correct. Nevertheless, it would be desirable to perform experiments in stronger fields, or at higher frequencies, in order to observe the small excess contribution (12), or (13), of this conductivity mechanism as well.

[1)] That is, the integral number of carriers per unit cell.

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