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Self-consistent dielectric response of a quasi-one-dimensional metal at high frequencies

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We present the results of a calculation of the frequency- and wave-vector-dependent longitudinal dielectric function of a quasi-one-dimensional electron gas. The electrons are taken to be localized to the chains and both the tight-binding and free-electron extremes are considered along the chain axis. Local-field effects are included. Dispersion curves for plasmons and single-particle-excitation spectra are presented. We find that the plasmon modes are not Landau damped and that for long wavelengths these modes have eigenfrequencies ranging continuously from the usual three-dimensional plasma frequency for propagation along the chain axis to zero for propagation perpendicular to it. Finally, we discuss the effects these excitations should have on the optical properties. The absorption in the free-electron extreme contains both single-particle and plasmon contributions throughout the optical spectrum. In the tight-binding limit, the plasmon contributions persist to frequencies larger than the single-particle bandwidth. In no event is the absorption of the Drude form.

I. INTRODUCTION

It has long been recognized that the behavior of the one-dimensional electron gas is expected to differ dramatically from that of its three-dimensional counterpart. The fundamental instability of the metallic state,\(^1\) the preponderant influence of fluctuations\(^2\) and disorder,\(^3\) and the theoretical possibility of giant conductivities\(^4\) have been objects of occasional speculation for some time. This interest ceased to be merely academic with the development\(^5\) of a class of anisotropic conductors whose structures consist of parallel linear chains, along which conduction electrons propagate essentially in one dimension. These include organic compounds such as tetrathiofulvalene-tetracyanoquinodimethane (TTF-TCNQ), the best organic conductor known,\(^6,7\) and inorganic salts such as the "mixed-valence" platinocyanides.\(^8\)

Because of limitations on crystal size and quality, reliable experimental data on these systems were until lately rather scarce, and were confined largely to measurements of static and low-frequency characteristics. Recently, however, crystals of some of these materials have become available which are large enough for good optical measurements to be performed. It is found that at sufficiently high frequencies, the materials appear metallic in one dimension.\(^9,10\) This has encouraged interpretation of the data in terms of a simple Drude metal even at frequencies well above the nominal width of the conduction band.\(^9,8\) Unfortunately, the dielectric response of the one-dimensional electron gas has received much less attention at these frequencies than it has in the static limit, and better interpretations of the data have been rare.

To help clarify the situation, we evaluate here the frequency- and wave-vector-dependent dielectric response of a quasi-one-dimensional metal in the random-phase approximation. This is not to imply that we regard all of the materials in question as true metals: Their low-frequency properties are obviously much more complicated than that.\(^5,10-13\) Rather, we simply recognize that at sufficiently high frequencies, these properties can have little qualitative influence upon our results. We neglect exchange and the phonon dynamics, and we replace the highly inhomogeneous molecular-charge distribution by a uniform interchain background of dispersionless polarizability. What remains is a simple tractable model which must, in our view, be understood before the high-frequency response of the real materials can be interpreted properly. We consider our approximations too severe, however, to generate a reliable description of the static screening.

We investigate both the free-electron and tight-binding extremes of the one-dimensional conduction band. In both cases the random-phase approximation (RPA) as usual describes collective effects not present in a Hartree-Fock treatment of the problem. In particular, when the interchain Coulomb interaction is taken properly into account, we find that long-wavelength electron-density fluctuations on neighboring chains are correlated. These can be described in terms of plasma excitations whose frequencies depend upon the directions of their wave vectors as well as the magnitudes, and range continuously from the three-dimensional plasma frequency down to zero. (The same con-
clusion has been reached by Dzyaloshinskii and Kats. At sufficiently short wavelengths, the directional dependence of the frequency is lost. In no case does the plasmon spectrum ever merge with that of the single-particle excitations; there is no simple Landau damping.

Like the single-particle excitations, the plasmons may be coupled to a light field through interaction with phonons or lattice disorder. Unlike the isotropic case treated by Hopfield, this process in a quasi-one-dimensional metal leads to an absorption which in general exhibits no sharp threshold, but competes with the usual single-particle contribution throughout the optical spectrum. Nor is the single-particle absorption itself of the simple Drude form. We conclude that the Drude expressions are not appropriate vehicles for the interpretation of the optical properties of these systems, and that reasoning based on fits to these formulas over a narrow spectral range should be accepted with reserve.

The development of these ideas is presented in five parts. In Sec. II we derive a general self-consistent expression for the complex frequency- and wave-vector-dependent dielectric function of model quasi-one-dimensional metals. This result is valid within the random-phase approximation and local-field effects are included. This general formulation is used in Sec. III to discuss the properties of the quasi-one-dimensional electron gas in the free-electron and tight-binding extremes. The plasmon dispersion and single-particle excitation spectra are presented and convenient long-wavelength approximations for the dielectric function are given. In Sec. IV the application of these results to the optical-absorption spectra of the quasi-one-dimensional metals is discussed. Section V contains a brief summary.

II. GENERAL FORMULATION

We will often have need to refer to the components of a vector along the chain axis and will adopt the convention \( q = \vec{Q} \cdot \hat{e}_z \), \( k = \vec{K} \cdot \hat{e}_z \), etc., where \( \hat{e}_z \) is a unit vector in the chain direction. The component of a vector perpendicular to the chain axis we denote by \( \vec{q}_s \), \( \vec{k}_s \), etc.

Our treatment is simplified by modelling the system as a set of parallel conducting strands of finite diameter, regularly spaced in a square array and immersed in a medium of dispersionless background dielectric constant \( \epsilon_\infty \). We neglect the interstrand charge-transfer matrix elements, but retain the Coulomb potential experienced by an electron on one strand due to those on other strands as well as its own.

Throughout the paper we shall assume that the electric field (although not necessarily the polarization field) induced by a test charge in our model systems is purely longitudinal. This assumption reduces the dielectric function from a tensor to a scalar quantity, which depends on both the magnitude and direction of the wave vector. The longitudinal-field approximation is equivalent to neglect retardation, and is valid for three-dimensional wave vectors \( |\vec{Q}| > \Omega/c \), where \( \Omega \) is the frequency of interest and \( c \) the velocity of light. In this sense our work complements that of Bulaevskii and Kukharenko, who have treated the solutions to Maxwell’s equations for these systems in the extreme long-wavelength limit.

With the loss of translational invariance, the self-consistent calculation of the dielectric response is complicated considerably. In particular, the effect of the periodic lattice is to mix Fourier-potential components differing by reciprocal-lattice vectors \( \vec{G} \). The response to an applied potential of wave vector \( \vec{Q}_0 \) and frequency \( \Omega \) is no longer local, and must be specified at each of the wave vectors \( \vec{Q} = \vec{Q}_0 + \vec{G} \). Specifically, for an applied potential \( \psi_{\text{ext}}(\vec{Q}_0, \Omega) \), we seek the function \( \epsilon(\vec{Q}, \vec{Q}_0, \Omega) \) such that the components \( u_{\text{tot}}(\vec{Q}, \Omega) \) of the total potential are given by

\[
   u_{\text{tot}}(\vec{Q}, \Omega) = \frac{\psi_{\text{ext}}(\vec{Q}_0, \Omega)}{\epsilon(\vec{Q}, \vec{Q}_0, \Omega)}.
\]

For the remainder of the discussion, we shall take

\[
   \psi_{\text{ext}}(\vec{Q}, \Omega') = \psi_{\text{ext}}(\vec{Q}) \left[ \delta(\Omega - \Omega') + \delta(\Omega + \Omega') \right]
\]

and will suppress the frequency arguments.

The localized wave functions are

\[
   \psi_{n,1_z}(\vec{R}) = \left( \frac{1}{L^{1/2}} \right) e^{i \vec{r} \cdot \vec{l}_z} \Theta(\vec{r}_z - \vec{l}_z) \theta(\vec{l}_z - \vec{l}_z)
\]

free electron

\[
   \psi_{n,1_z}(\vec{R}) = \frac{1}{n^{1/2}} \left( \sum_{\vec{t}} e^{i \vec{t} \cdot \vec{r}} \right) \Theta(\vec{r}_z - \vec{t}_z)
\]

tight binding

where \( n \) is the number of unit cells on a chain and \( L \) is the chain length. Following our convention, \( \vec{r}_z \) is the perpendicular component of \( \vec{R} \) and \( \vec{r} \) the parallel component. The quantities \( \vec{l}_z \) and \( \vec{l} \) refer to perpendicular and parallel lattice vectors, respectively. The function \( \Theta(\vec{r}_z) \) is strongly peaked about \( \vec{r}_z = 0 \) and describes the localization of the electrons to the strands, and \( \psi \) is a Wannier function.

We define

\[
   \beta(\vec{k}_z) = \int e^{-i \vec{k}_z \cdot \vec{z}} \left| \Theta(\vec{z}) \right|^2 d^2 x_z, \quad \text{free electron}
\]

\[
   \beta(\vec{k}_z) = \int e^{-i \vec{k}_z \cdot \vec{z}} \left| \psi(\vec{x}) \right|^2 dx \int e^{-i \vec{k}_z \cdot \vec{z}} \left| \Theta(\vec{z}) \right|^2 d^2 x_z \quad \text{tight binding}
\]

The interaction Hamiltonian describing the interaction of the potential \( V_{\text{tot}}(\vec{R}, \vec{l}) \) with the electron
gas is given by

\[ H_{\text{int}} = \sum_{k, \bar{G}} \sum_{I_k} C^*_k I_k C_k I_k e^{i \bar{G} \cdot I_k} \times \sum_{Q} \psi_{\text{tot}}(Q + \bar{G}) \beta^*(Q + \bar{G}) \]  \hspace{1cm} (4)

The summation over \( k \) and \( \bar{G} \) runs over one Brillouin zone with the understanding that in the free-electron extreme the Brillouin zone is taken to have infinite extent along the chain axis. Similarly, the summation over reciprocal-lattice vectors \( G \) is taken to be two-dimensional for the free-electron extreme and three-dimensional in the tight-binding extreme. The quantity \( \psi_{\text{tot}}(Q) \) is the Fourier transform of \( \psi_{\text{tot}}(\bar{R}) \).

The induced charge density is calculated as usual to first order in \( H_{\text{int}} \) by determining the expectation value of the particle-density operator \( \rho(\bar{R}) \) for the perturbed ground state. The induced potential \( \psi_{\text{tot}}(\bar{R}) \) is then determined from Poisson's equation with charge density \( \psi(\bar{R}) \). The contribution from the polarization of the molecular cores is included by assuming an isotropic-background dielectric constant \( \epsilon_\infty \). Finally, we write

\[ \psi_{\text{tot}}(\bar{Q}) = \psi_{\text{ext}}(\bar{Q}) + \psi_{\text{fاد}}(\bar{Q}) + \psi_{\text{ext}}(\bar{Q}) \]  

and solve for \( \psi_{\text{tot}}(\bar{Q}) \) in terms of \( \psi_{\text{ext}}(\bar{Q}) \).

The equation thus obtained for \( \psi_{\text{tot}}(\bar{Q}) \) is

\[ \psi_{\text{tot}}(\bar{Q} + \bar{G}) = \frac{1}{\epsilon_\infty} \psi_{\text{ext}}(\bar{Q}) - \frac{4\pi e^2}{\epsilon_\infty} Q \chi(Q, \Omega) \beta(\bar{Q} + \bar{G}) \]  \hspace{1cm} (5)

where

\[ \chi(q, \Omega) = \chi(q, \Omega) - i \chi_0(q, \Omega) \]

is given by

\[ \chi_0(q, \Omega) = \frac{2\pi}{\Omega} \int \frac{n(k)}{[1 - n(k + q)]} \times [\omega_{k, \pm}^2 - (\Omega^2 - Q^2)] dk \]  \hspace{1cm} (6)

\[ \chi_0(q, \Omega) = \frac{2\pi}{\Omega} \int \frac{n(k)}{[1 - n(k + q)]} \times [\delta(\omega_{k, \pm} - \Omega) - \delta(\omega_{k, \pm} + \Omega)] dk \]  \hspace{1cm} (6)

\[ \omega_{k, \pm} = \left\{ \frac{1}{h} \right\} \left( E(k) - q \right) \]  \hspace{1cm} (6)

The term \( n(k) \) refers to the occupation number of the one-electron state with wave vector \( k \), and \( E(k) \) is the energy of this state. \( \Omega \) is the number of strands per unit area, and the quantity \( (4\pi e^2/\epsilon_\infty Q^2) \chi(q, \Omega) \) is just the usual Kramers-Heisenberg electronic polarizability.\(^{18}\)

If we take \( \psi_{\text{ext}}(\bar{Q}) = \psi_{\text{ext}}(\bar{Q} + \bar{G}) \), (5) describes an infinite set of coupled linear equations for \( \psi_{\text{tot}}(\bar{Q}) \), \( \psi_{\text{tot}}(\bar{Q} + \bar{G}) \), ... that must be solved in order to determine the dielectric constant \( \epsilon(\bar{Q}, \bar{Q} + \bar{G}) \). The solution can be obtained by observing that the summation in (5) is periodic in \( \bar{Q} \) with a period of a reciprocal-lattice wave vector. We define \( \bar{Q} = \bar{Q} + \bar{G}_1 + \bar{G}_2 \), where \( \bar{Q}_1 \) and \( \bar{Q}_2 \) lie in the first Brillouin zone and \( \bar{G}_1 \) and \( \bar{G}_2 \) are reciprocal-lattice vectors. The final result is

\[ \psi_{\text{tot}}(\bar{Q} + \bar{G}) = \frac{\psi_{\text{ext}}(\bar{Q})}{\epsilon_\infty - (4\pi e^2/\epsilon_\infty Q^2) \chi(q, \Omega)} \delta_{\bar{Q}, \bar{Q} + \bar{G}_1 + \bar{G}_2} \]  \hspace{1cm} (7)

Equation (7) could have been obtained by taking the electronic wave functions in (2) to be nonlocalized three-dimensional anisotropic tight-binding wavefunctions. Using these functions, Hayashi and Shimizu\(^\text{20}\) have investigated dielectric screening in transition metals and have obtained identical results for the dielectric function.

For \( |\bar{G}| \) sufficiently small, the short-wave-length modulation of the electronic density by the lattice is unimportant, and (7) reduces to the local (in \( Q \) space) RPA result. Here the lattice affects \( \epsilon(\bar{Q}, \Omega) \) only in that it modifies the electron-dispersion relation and, therefore, \( \chi(q, \Omega) \). Only the \( \bar{G} = 0 \) term contributes to \( U(\bar{Q}) \) and, taking \( \beta(\bar{Q}) = 1 \), (7) becomes

\[ U(\bar{Q}) = 4\pi e^2 \sum_{\bar{G}} \frac{1}{|\bar{Q} + \bar{G}|^2} |\beta(\bar{Q} + \bar{G})|^2 \]  \hspace{1cm} (7a)

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\[ U(\bar{Q}) = \frac{\psi_{\text{ext}}(\bar{Q})}{\epsilon_\infty - (4\pi e^2/\epsilon_\infty Q^2) \chi(q, \Omega)} \delta_{\bar{Q}, \bar{Q} + \bar{G}_1 + \bar{G}_2} \]  \hspace{1cm} (8)

### III. EXPLICIT RESULTS

#### A. Density-density response function \( \chi(q, \Omega) \)

1. **Free electron**

   If the energies of the one-electron states have the one-dimensional free-electron form \( \delta^\text{FE}(q) = h^2 q^2/2m \), we find from Eq. (6) the real and imaginary parts of \( \chi^\text{FE}(q, \Omega) \):

\[ \chi_1^\text{FE}(q, \Omega) = \frac{\pi M}{\hbar^2 q} \ln \left| \frac{\Omega^2 - \omega_+^\text{FE}(q)^2}{\Omega^2 - \omega_-^\text{FE}(q)^2} \right| , \]

\[ \chi_2^\text{FE}(q, \Omega) = \frac{\pi M}{\hbar^2 q} , \text{ if } \omega_+^\text{FE}(q) \leq \Omega \leq \omega_-^\text{FE}(q) \]  \hspace{1cm} (9)

\[ \chi_2^\text{FE}(q, \Omega) = 0 , \text{ otherwise,} \]
where the frequencies
\[ \omega_k^\pm(q) = \left| \frac{kr^2}{2m} \pm qv_F \right| \] (10)
represent the maximum and minimum energies for excitation of an electron-hole pair with wave vector \( q \), and \( v_F \) is the Fermi velocity.

We observe that in the static limit, (9) contains the logarithmic divergence at \( q = 2k_F \) representing the Peierls instability of a one-dimensional metal:
\[ \chi_0^\pm(q, 0) = \frac{2\pi n}{\pi \hbar q} \ln \left| \frac{q + 2k_F}{q - 2k_F} \right| . \]

Our neglect of exchange, however, probably precludes a meaningful discussion of the static

The evaluation of the density-response function proceeds as in the free-electron case. The results are

\[ x_1^{TB}(q, \Omega) = -\left( \frac{2\pi n}{\pi \hbar a} \right) \left\{ \frac{1}{[\Omega^2 - \omega_0^2(q)]^{1/2}} \right\} \tan^{-1} y(q, \Omega) , \]
\[ = -\left( \frac{2\pi n}{\pi \hbar a} \right) \left\{ \frac{1}{[\omega_0^2(q) - \Omega^2]^{1/2}} \right\} \ln \left[ \frac{1 + y(q, \Omega)}{1 - y(q, \Omega)} \right] , \]
\[ x_2^{TB}(q, \Omega) = \left( \frac{2\pi n}{\pi \hbar a} \right) \left\{ \frac{1}{[\omega_0^2(q) - \Omega^2]^{1/2}} \right\} , \]
\[ = 0 \]

where
\[ y(q, \Omega) = \frac{2\omega_0(q)}{\Omega^2} \left[ \frac{\Omega^2 - \omega_0^2(q)}{\Omega^2 - \omega_0^2(q) + \sin\frac{\Omega}{2} qa} \right] , \]
\[ \omega_0(q) = \left( \frac{W}{\hbar} \right) \left| \sin \frac{\Omega}{2} qa \right| , \]
and
\[ \omega_1^{TB}(q) = \omega_0(q) \left| \sin a \left( k_F \mp \frac{1}{2} | q | \right) \right| , \]
\[ \omega_1^{TB}(q) = \omega_0(q) \left| \sin a \left( k_F + \frac{1}{2} | q | \right) \right| , \]
\[ \omega_2^{TB}(q) = \omega_0(q) \left| \sin a \left( k_F \pm \frac{1}{2} | q | \right) \right| , \]
\[ = \omega_0(q) , \]
and where \( 0 \leq \tan^{-1} y \leq \pi \). In the static limit, the logarithmic divergence again appears at \( q = 2k_F \):
\[ x_1^{TB}(q, 0) = \frac{2\pi}{\pi \hbar \omega_0(q) a} \left[ \frac{\sin k_F a + \sin \frac{1}{2} qa}{\sin k_F a - \sin \frac{1}{2} qa} \right] . \]

B. Effective Coulomb potential \( U(\hat{Q}) \)

In many respects, localizing the electrons to thin strands has the effect of replacing the usual bare Coulomb potential \( 4\pi e^2/q^2 \) by an effective Coulomb potential \( U(\hat{Q}) \), as defined in Eq. (7). For this reason, the behavior of \( U(\hat{Q}) \) is of interest.

\( U(\hat{Q}) \) depends on the radius of a conducting strand through the "form factor" \( \beta(\hat{k}) \). For wave vectors perpendicular to the chain axis, \( \beta(\hat{k}) \) is just the Fourier transform of the function describing the localization of the electron to the strands \( \Theta(\hat{r}_i) \).

If the strands have radius \( \rho_i \), it is clear that the effect of \( \beta(\hat{k}) \) in Eq. (7a) is to cut the summation off at a value of \( \vec{g}_i \), roughly given by \( \vec{g}_i = 1/\rho_i \).

Since the sum is two dimensional, it diverges logarithmically for small \( \rho_i \). This corresponds to the logarithmic divergence of the potential energy of a line charge.

**2. Tight binding**

We now consider the extreme in which the conduction electrons are taken to occupy a single one-dimensional tight-binding band, well isolated in energy from any other bands in the system. The one-electron energies are \( \mathcal{E}^{TB}(k) = \frac{1}{2} W (1 - \cos ka) \), where \( W \) is the bandwidth and \( a \) is the unit-cell dimension along the chain axis. The single-particle excitations are given by
\[ \omega_{\pm}(k) = \left( \frac{W}{\hbar} \right) \sin \left( \frac{1}{2} qa \right) \sin \left( k a \pm \frac{1}{2} qa \right) . \]

The evaluation of the density-response function proceeds as in the free-electron case. The results are

\[ \chi_0^{TB}(q, 0) = \frac{2\pi n}{\pi \hbar q} \ln \left| \frac{q + 2k_F}{q - 2k_F} \right| . \]

We will take the functions \( \Theta(\hat{r}_i) \) and \( \psi(\hat{r}) \) to be Gaussians of radius \( \rho_i \) and \( \rho \), respectively:
\[ \Theta(\hat{r}_i) = \left( \frac{1}{\pi \rho_i^2} \right) e^{-r_i^2 / \rho_i^2} , \]
\[ \psi(\hat{r}) = \left( \frac{1}{\pi a^2} \right) e^{-r^2 / a^2} . \]

With these definitions, \( \beta(\hat{k}) \) is given by

\[ \beta(\hat{k}) = e^{-i\mathcal{E}_k^2 / 4} , \]

\[ \beta(\hat{k}) = e^{-i\mathcal{E}_k^2 / 4} e^{-i\mathcal{E}_k^2 / 4} . \]

We will take the parameter \( \rho_i \) to be much less than the interchain spacing so that the charge-transfer matrix element between chains vanishes, and we will take \( \rho \sim a \).

Figure 1 shows plots of the effective Coulomb potential \( U(\hat{Q}) \) in the tight-binding extreme for
three different strand radii, as well as a plot of the usual Coulomb potential $4\pi q^2/Q^2$. In this figure, $Q$ is along the chain axis. The divergence of $U(Q)$ for small strand radii can be seen here. Also, it appears that for strand radii $\sim 1$ Å, and for $Q$ along the chain axis, the usual Coulomb potential represents a good approximation to the effective potential, at least out to the Brillouin-zone edge.

Because of the periodicity of $U(Q)$, the similarity between the two potentials ends as the wave vector passes outside of the first Brillouin zone. The effects of this periodicity can be seen in Fig. 2. Here $U(Q)$ is shown for a fixed strand radius of 1 Å and for wave vectors making angles $\theta$ with the chain axis of 0°, 60°, and 80°. The ordinate in the figure is $q = |Q| \cos \theta$, and $Q$ is taken to lie in the $xz$ plane. The oscillatory behavior of the 60° and 80° curves results from the periodicity of $U(Q)$. As $Q_1$ passes through a perpendicular reciprocal-lattice vector, $U(Q)$ has the same value it would have if $Q_1 = 0$, and it therefore must meet the $\theta = 0°$ curve.

C. Local forms $\varepsilon(Q, \Omega)$

As discussed in Sec. II, the dielectric function becomes local in $Q$ space when $|\bar{Q}| \ll |\bar{G}|$ for all $\bar{G}$. Additionally, in this region $\varepsilon(Q, \Omega)$ can be written in a particularly transparent form.

1. Free electron

From Eqs. (8)-(10), we obtain in the free-electron extreme,

$$\varepsilon_{1}^{\text{FE}}(Q, \Omega) = \varepsilon_{\infty} \left(1 + \frac{\omega_{p}^{2} \cos^{2} \theta}{\omega^{2} - \omega_{*}^{2}} \ln \left| \frac{\omega^{2} - \Omega^{2}}{\omega_{*}^{2} - \Omega^{2}} \right| \right),$$

$$\varepsilon_{i}^{\text{FE}}(Q, \Omega) = \frac{1}{2} \pi \varepsilon_{\infty} \frac{\omega_{p}^{2} \cos^{2} \theta}{\omega^{2} - \omega_{*}^{2}}, \quad \omega_{\ast} \leq \Omega \leq \omega_{*}$$

$$= 0,$$ otherwise

where $\theta$ is the angle between $Q$ and the chain axis, and we have employed the shortened notation $\omega_{*} = \omega_{*}^{\text{FE}}(q)$. As expected for a one-dimensional system, the dielectric function (15) diverges at the upper and lower thresholds for single-particle excitation.

We identify the parameter

$$\omega_{p}^{2} = \frac{8\pi e^{2} v_{F}}{\epsilon_{\infty} m} = \frac{4\pi N e^{2}}{\epsilon_{\infty} m}$$

as the squared plasma frequency, with $N$ the volume electron density. To examine its physical significance, we consider the long-wavelength limit of (15):

$$\varepsilon_{1}^{\text{FE}}(0, \Omega) = \varepsilon_{\infty} \left[1 - \left(\frac{\omega_{p}^{2}}{\Omega^{2}}\right) \cos^{2} \theta \right].$$

FIG. 1. Effective Coulomb potentials (solid line) in the tight-binding extreme for three strand radii. Also shown (dashed line) is the usual bare Coulomb potential. The wave-vector direction is along the chain axis ($\theta = 0°$), $\rho = a = 1.5$ Å, and $k_F = 5 \times 10^7$ cm$^{-1}$.

FIG. 2. Effective Coulomb potentials in the tight-binding extreme for off-axis wave-vectors. Strand radius is 1 Å, $\rho = a = 1.5$ Å, $k_F = 5 \times 10^7$ cm$^{-1}$, and $q = |Q|\cos\theta$. 
Setting (17) equal to zero, we find that there exists a set of long-wavelength plasmons whose frequency depends upon their directions of propagation:
\[ \omega(Q) = \omega_* \cos \theta. \]

This observation can also be made in the tight-binding extreme and it will be discussed in more detail in the subsection on elementary excitations.

2. Tight binding

Particularly convenient are the forms for the dielectric function resulting from the special case of a half-filled band, \( k_F = \pi / 2a \). Using Eq. (11) and (12) in (8) we obtain for this case, with no further approximations,
\[
\epsilon_1^T(Q, \Omega) = \epsilon_0 \left[ 1 - \frac{\omega_0^2 \cos^2 \theta}{\Omega^2 - \Omega^2_{\text{pl}}} + \frac{\sin^2 \frac{1}{2} \Omega a}{\Omega^2_{\text{pl}}} \tan^{-1} \left( \frac{\Omega^2 - \Omega_{\text{pl}}^2}{\Omega^2 - \Omega_{\text{pl}}^2} \right) \right], \quad \Omega > \omega_0,
\]
\[
\epsilon_2^T(Q, \Omega) = \frac{\omega_0^2 \cos^2 \theta}{2 \Omega^2_{\text{pl}}} + \frac{\sin^2 \frac{1}{2} \Omega a}{\Omega^2_{\text{pl}}} \tan^{-1} \left( \frac{\Omega^2 - \Omega_{\text{pl}}^2}{\Omega^2 - \Omega_{\text{pl}}^2} \right), \quad \Omega < \omega_0,
\]
\[
\epsilon_2^T(Q, \Omega) = 0, \quad \omega_0 \leq \Omega < \omega_0.
\]

where we have again used the shortened notation \( \omega_* = \omega_*^T (q) \).

From this equation, we see that \( \epsilon_1^T (Q, \Omega) \) is discontinuous at \( \Omega = \omega_* \). It approaches a finite limit as \( \Omega \) approaches \( \omega_0 \), from below, and a divergent limit from above. This is a general result for all \( |q| > |2k_F - \pi / a| \). For \( |q| < |2k_F - \pi / a| \), \( \epsilon_1^T (Q, \Omega) \) diverges in a symmetric manner about \( \Omega = \omega_* \). It always diverges symmetrically about \( \Omega = \omega_* \). The long-wavelength behavior of \( \epsilon^T (Q, \Omega) \) is quite analogous to that of \( \epsilon_1^R (Q, \Omega) \).

D. Elementary excitations

We next discuss the plasmon dispersion and the single-particle excitation spectrum for the two extremes of tight-binding and free-electron behavior along the chain axis. In both cases, the plasmon frequency for long wavelength is given by
\[
\omega_p^2 (Q) = \omega_*^2 \cos^2 \theta + \ldots,
\]
\[
\omega_p^2 = \frac{8 \pi e^2 v_F}{\epsilon_* \hbar}.
\]

There exists a set of long-wavelength plasma oscillations with frequency dependent on the angle of propagation with respect to the chain axis. Physically, these may be regarded as consisting of charge oscillations propagating along each chain with the same wavelength, but stepped in phase from chain to chain so that the direction of propagation is off axis. The polarization of such modes is mixed and angle dependent, changing from a purely longitudinal plasmon with the usual three-dimensional plasma frequency \( \omega_p \), at \( \theta = 0^\circ \), to a purely transverse mode with zero frequency at \( \theta = \frac{1}{2} \pi \). This result has been noted by Dzyaloshinskii and Katoh and by Bulaevskii and Kukharenko; we emphasize that it results entirely from the coupling of charge density on different chains through the interchain Coulomb interaction.

The angular dependence of the frequency follows readily from simple considerations. For plasmon propagation along the chain axis, the Coulomb restoring forces are also along the axis, and the forces keeping the electrons localized to the chains do not come into play. The equation of motion for the electron gas is the same as in the isotropic case and the plasma frequency is given by the usual three-dimensional result. For plasmon propagation at angles oblique to the chain axis, however, the Coulomb restoring forces have a component perpendicular to the axis. This component is cancelled by the forces keeping the electrons localized to the chains, so that the net restoring force on the plasmon charge-density wave is reduced. It is easy to show that the resulting plasmon frequency is proportional to \( \cos \theta \). Another effect of these localizing forces is to give the plasmon a partial transverse character.

For shorter wavelengths, the dielectric function is not local so that the general result given by Eq. (7) must be used to determine the plasmon spectrum. The plasmon frequency \( \omega (Q) \) is determined by requiring a finite response for an infinitesimal applied field of wave vector \( Q \) and frequency \( \omega (Q) \). As can be seen from (7), this condition is met by requiring that
\[
\chi(Q, \omega) = -\frac{1}{\omega} \chi(Q, \omega) U(Q) - 0.
\]

This equation defines the plasmon dispersion. It is of the same form as the usual isotropic RPA result except that the bare Coulomb potential \( 4 \pi e^2 / Q^2 \) is replaced by the effective Coulomb potential \( U(Q) \).

As usual, the single-particle excitation spectrum is determined by requiring that the dielectric func-
tion have a nonvanishing imaginary part. We now present explicit results for the plasmon dispersion and the single-particle excitation spectrum throughout the Brillouin zone.

1. Free electron

Following (20) and using the free-electron result for \( \chi \), the plasmon dispersion is given in this extreme by

\[
\omega^2(\mathbf{q}) = \omega_{\text{PE}}^2(q)^2 + \frac{1}{2} [\omega_{\text{PE}}^2(q)^2 - \omega_{\text{FE}}^2(q)^2] \times \left[ \coth \left( \frac{\pi \hbar^2 q}{2m \mathbf{U}(\mathbf{q})} \right) \right],
\]

(21)

The long-wavelength approximation to the plasmon frequency given by (19) is obtained immediately upon expansion in powers of \( q \):

\[
\omega^2(\mathbf{q}) = \omega^2_{\text{FE}}(q)^2 + q^2 v_F^2 + q^4 v_F^4 / 4\hbar^2 + \cdots .
\]

(22)

The first two terms are the same as obtained by Dzyaloshinskii and Kats.\(^{14}\)

The dispersion curves for strand radii of 0.5 and 3.0 Å are shown in Fig. 3. The plasmon propagation direction is along the chain axis for these curves. In Fig. 4, the plasmon dispersion is shown for a fixed strand radius of 3 Å, but for propagation angles of 0° and 60°. The single-particle excitation spectrum is also shown in Figs. 3 and 4. The “hole” in the single-particle spectrum is a general feature of one-dimensional electron bands; it would not be present with isotropic threedimensional bands. As a result of this hole, low-energy single-particle excitations are allowed only for \( q = 0 \) and for \( q = 2k_F \).

An unusual feature of the one-dimensional plasmon-dispersion curves is that they never enter the region of single-particle excitations. In contrast to isotropic metals, the plasmons in quasi-one-dimensional metals are not Landau damped. That this must be the case in the free-electron extreme can be seen immediately from (21). Remembering that \( \coth x = 1 \) for all positive \( x \), it follows that \( \omega(Q) \geq \omega_{\text{FE}}^2(q) \), the maximum single-particle excitation frequency. As we shall show, this same conclusion can be drawn in the tight-binding extreme.

Although only the \( \theta = 0° \) and \( \theta = 60° \) dispersion curves are shown in Fig. 4, there actually are an infinity of such curves, one for each propagation direction. Because of the oscillatory nature of \( \mathbf{U} (\mathbf{q}) \), the off-axis plasmon-dispersion curves also oscillate. For large propagation angles with respect to the chain axis, these curves may oscillate between the \( \theta = 0° \) plasmon curve and the top of the single-particle excitation spectrum. The period of the oscillation is \( (2\pi / b) \cot \theta \), where \( b \) is the spacing between strands.

2. Tight binding

To determine the plasmon dispersion in this extreme, we proceed as before, using the tight-binding \( \chi \) in (20). The result is a transcendental equation for \( \omega(Q) \) which can only be solved numerically. We will present dispersion relations determined in this manner for two choices of the strand radius.

Even without an analytic solution, there are some general observations that can be made con-
cerning the nature of the excitations. For example, in the small-\( Q \) limit, Taylor expansions can be used to determine that

\[
\omega^2(\mathbf{Q}) = \omega_p^2 \cos^2\theta + \left( \frac{\epsilon_F}{a^2} - \frac{1}{12} \omega_p^2 \cos^2\theta \right) \times q^2 a^2 + \ldots .
\]

(23)

In this limit, the plasmons exhibit the same angular dependence as in the free-electron extreme. As \( q \) increases, the dispersion is severely modified by the band structure, and the plasmon frequency may rise or fall depending on \( \theta \) and on the relative magnitudes of \( W \) and \( \epsilon_\omega \).

As in the free-electron extreme, the plasmons are not Landau damped by single-particle excitations. That this must be the case can be seen by noting from (11) that \( \chi_T^{1B} \) is singular at \( \Omega = \omega_T^{1B}(q) \) and that the sign of \( \chi_T^{1B} \) in this region is such that the quantity

\[
f(\mathbf{Q}, \Omega) = \epsilon_\omega - U(\mathbf{Q}) \chi_T^{1B}(q, \Omega),
\]

is negative. Since \( f(\mathbf{Q}, \Omega) \) must approach \( \epsilon_\omega \) for large \( \Omega \), it must pass through zero for some \( \Omega > \omega_T^{1B}(q) \).

The transcendental equation for the plasmon dispersion has been solved numerically for a number of specific cases, and the results for two of these are shown in Figs. 5 and 6. Figure 5 shows the plasmon-dispersion curves for a strand radius of 1 Å and for propagation angles of 0° and 60°. Figure 6 shows similar results, but for a strand radius of 3 Å. Also shown in these figures is the single-particle-excitation spectrum. In both figures, a half-filled conduction band was assumed. Both the plasmon dispersion and the single-particle excitation spectrum must be periodic with period of reciprocal-lattice vector.

Although only two curves are shown for each figure, there is a different dispersion curve for each propagation angle. As in the free-electron extreme, the oscillatory nature of \( U(\mathbf{Q}) \) causes an oscillatory behavior for the off-axis dispersion curves. In analogy with the free-electron extreme, the dispersion curves for all propagation angles fill the space between the top of the single-particle excitation spectrum and the \( \theta = 0^\circ \) plasmon-dispersion curve.

IV. OPTICAL PROPERTIES

In a translationally invariant electron gas, neither plasmon nor single-particle excitations can contribute to the optical absorption.\(^{16,21}\) In the quasi-one-dimensional metals, translational invariance is broken in two ways: by the filamentary structure itself, and by static or thermal disorder. These two factors give rise to very different mechanisms for optical absorption, and it is important that we distinguish between them.

The first type of absorption process has been studied in detail by Bulaevskii and Kukharenko,\(^{17}\) and will be discussed briefly here. In a uniaxial conductor, the polarization of off-axis plasmons is partially transverse, and direct coupling to \( p \)-polarized light is possible in certain optical geometries. This possibility has not appeared explicitly in the results of Secs. II and III because of our neglect of retardation. When retardation is included, it is found\(^{17}\) that for extremely long wave-
lengths \((Q < \omega_p / c)\), the plasmon dispersion is split into two branches, separated by an angle-dependent gap within which light polarized along the chain axis does not propagate. In appropriate configurations the result is a region of high reflectivity in the vicinity of the upper-branch threshold \(\omega_p\). The effect has recently been observed experimentally by Bräsch.\(^\text{22}\)

As \(Q\) is increased, the lower branch of the dispersion smoothly joins \(\omega(Q) = \omega_p \cos \theta\), the leading term of the results of Sec. III. Presumably this branch can be directly excited, and its angular dependence verified, by coupling through a frustrated internal reflectance device such as those employed by Otto\(^\text{23}\) and by Barker\(^\text{24}\) in their studies of surface excitations. For the delicate materials in question such an experiment may be a preferable alternative to electron energy-loss measurements, and we suggest it as a means of testing directly some of the conclusions of the present paper.

In the usual optical geometry (the light normally incident and polarized with its electric field along the chain axis) direct coupling to the plasmons does not occur; direct coupling to the single-particle excitations does not of course occur in any geometry. Optical absorption in these cases requires some additional source of momentum,\(^\text{16,21}\) such as the phonons, static disorder,\(^\text{41}\) and/or the periodicity of the lattice. For three-dimensional metals this is the familiar mechanism responsible for the infrared single-particle absorption and, in favorable cases,\(^\text{26}\) a weak plasmon absorption\(^\text{16,21}\) with a threshold at \(\omega_p\). In the quasi-one-dimensional case, we shall find that the single-particle absorption is not precisely of the Drude form, and that the plasmon absorption is qualitatively different, and potentially more important, than it is in three dimensions.

A. Free-electron extreme

Where the electrons are free in one dimension, the treatment of the optical absorption is particularly straightforward. For simplicity of discussion we take the longitudinal dielectric function to be local in \(Q\) space. In this approximation our model certainly resembles a Drude metal more closely than it would if the mixing of potential components by the periodic lattice were taken into account. Nevertheless, we shall find that even in this local approximation, the absorption does not obey the Drude formula. Clearly, our assumption of a local dielectric function will not be valid in all cases, but the effect of the corrections will be to aggravate the deviations from simple Drude behavior.

We consider the diagonal component of the imaginary transverse dielectric tensor along the direction of the chain axis. To second order in \(V^2\), the coupling to the momentum sink, the optical absorption in our local approximation may be written as\(^\text{16,21}\)

\[
\Omega \varepsilon_2^p(\Omega) = \frac{e^2}{m^2 \Omega} \sum_{\xi} V^2 \frac{\Omega^2}{2} \frac{q}{|\varepsilon(\Omega_{\xi})|^2} \frac{1}{\varepsilon(\Omega_{\xi})}. \tag{24}
\]

Equation (24) contains both single-particle and plasmon contributions; we consider these in turn, using the local RPA results of Sec. III to approximate the longitudinal loss function. Our results are to be compared with the absorption of a Drude metal carried to the same order of perturbation theory:

\[
\Omega \varepsilon_2^p(\Omega) = \frac{e^2}{m^2 \Omega} \Omega^2 \tau, \tag{25}
\]

where \(\tau\) is the electronic relaxation time.

1. Single-particle absorption

Using (15), we find that the single-particle contribution to (24) is

\[
\Omega \varepsilon_2^{sp}(\Omega) = \frac{e^2}{4\hbar^2 \omega_p^2} \sum_{\xi} V^2 \frac{q}{Q^2} \frac{1}{|\varepsilon(\Omega_{\xi})|^2}, \tag{26}
\]

where the notation \(\varepsilon(\Omega_{\xi})\) indicates the sum is to be taken over those values of \(\Omega\) for which single-particle excitations exist at a given \(\omega\). For want of detailed information about \(V^2\) we shall not evaluate the full expression (26) here. Instead, we consider its asymptotic behavior.

We examine first the limit \(\hbar \Omega \gg \hbar \omega_p\), \(\epsilon_p\) where \(\delta_p = \delta(k_p)\). Here departures from (25) arise primarily because the semiclassical derivation of the Drude formula,\(^\text{19}\) which treats the current essentially as a small oscillation of the Fermi surface, cannot account accurately for transitions from deep within the Fermi sea. After replacing \(V^2\) by its average value at large \(Q\), we find

\[
\Omega \varepsilon_2^{sp}(\Omega) \to \frac{1}{\hbar^2 \Omega} \frac{1}{\delta_p} (\text{const}) \Omega^{3/2} + \Omega^{7/2} \tag{27}
\]

Analogous effects are predicted\(^\text{26}\) and observed\(^\text{25}\) in three dimensions.

In the opposite limit, \(\hbar \Omega \ll \hbar \omega_p\), Eq. (24) remains valid so long as \(\Omega \tau \gg 1\), where \(\tau\) is the smallest important relaxation time. These two conditions are compatible within our free-electron model. At these frequencies the available phase space is severely restricted by the "hole" in the one-dimensional single-particle spectrum (Fig. 3) and the only terms in the sum (26) are from \(q \sim 2k_p\). The result is approximately

\[
\Omega \varepsilon_2^{sp}(\Omega) \to \frac{(\text{const.})}{\hbar / \tau = \hbar \omega_p = \hbar / \delta_p} \Omega^2 \ln \left[ (\Omega / \delta_p)^\frac{1}{2} \right] \times [1 + O(\hbar^2 \Omega^2 / \delta_p^2)]. \tag{28}
\]

This form rises more slowly with decreasing \(\Omega\) than does (25).

In the intermediate region, \(\Omega \varepsilon_2^{sp}(\Omega)\) is a reason-
able approximation to (25) over a limited-frequency region. It is in precisely this region, however, that the plasmon absorption considered in Sec. IV A 2 is strongest, so that the Drude formula is never really obeyed.

2. Plasmon absorption

Superimposed upon the single-particle absorption is the contribution to (24) of the plasmon pole of the loss function. From (15), the strength of the pole is given by

$$\text{Im} \frac{1}{\epsilon(Q, \Omega)} \bigg|_{\Omega \sim \omega_p} = \frac{(Q^2 - \omega_p^2) (\omega_p^2 - \omega_\epsilon^2) \pi}{2 \epsilon_\infty \epsilon_\infty \omega_p^2 \cos \theta} \times 5(\Omega - \omega(Q)).$$

For $Q = 0$ and $\theta = 0$, (29) exhausts the longitudinal $f$ sum rule,

$$\int_0^\infty \Omega \text{Im} \left[ 1/\epsilon(Q, \Omega) \right] d\Omega = \frac{\pi}{2} (\omega_p^2 / \epsilon_\infty)$$

just as in the three-dimensional case. For $q = \omega_c$ or $\theta = \frac{\pi}{2}$, the total plasmon oscillator strength is vanishingly small.

Because of their angular dependence and the absence of Landau damping, the plasmon eigenfrequencies in the free-electron extreme extend continuously from zero to infinity. The resulting optical absorption therefore extends in principle throughout the spectral range.

We can compute the second-order contribution to this absorption by substituting (29) into (24). For $Q \rightarrow 0$, we have from Eq. (21) and Fig. 3 that $\omega(Q) - \omega_p^2 (q)$ and the absorption is negligible. It is also negligible for $Q = 0$, where the conditions $Q - \omega_p^2 (q), \theta = \frac{\pi}{2},$ and $Q = 0$ outweigh the factor $\omega^3$ in (24). Between these extremes we find an extended shape of the absorption, the detailed shape of which depends on the functional form of $V^2$. In general, it exhibits a very broad maximum centered in the vicinity of $\omega_p$, where the pole (29) is strongest and the plasmon density of states large.

We can estimate the total strength of this absorption by exploiting the transverse sum rule,

$$\int_0^\infty \Omega \epsilon_\parallel(\Omega) d\Omega = \frac{1}{2} \pi \epsilon_\infty \omega_p^2.$$

Some caution is required in evaluating the contribution to (31) of a second-order expression like (24). The difficulty is that (24) amounts to an expansion of the absorption in powers of $(\Omega \tau)^4$, and the integration (31) includes the low-frequency region where such an expansion is no longer valid. For the present problem, however, we observe from (29) that the plasmons contribute little oscillator strength at low frequencies, so that for the free-electron extreme ($\omega_p \tau \gg 1$) we expect (24) to provide a reasonably reliable estimate of the total plasmon contribution to (31).

We make this estimate by replacing $V^2$ in (24) by its average, which we approximate using values of $\tau$ deduced from the dc conductivities of real-one-dimensional conductors assuming a free-electron density of states. Using the parameters $\tau = 3 \times 10^{-16}$ sec, $\tau = 10^{14}$ cm$^{-2}$, and a linear electron density of 1 per 3 A, we find that the plasmons contribute about 15% of the total oscillator strength. This can represent a significant departure from the Drude formula beyond the deviation predicted for the single-particle contribution.

B. Tight binding extreme

The actual quasi-one-dimensional conductors are narrow-band systems. If their dc conductivities at room temperature are regarded as metallic, one deduces electronic mean free paths of the order of one lattice constant, or relaxation rates $\hbar/\tau$ of the order of the bandwidth $W$. This situation approaches the threshold condition for localization of the electronic wave function by strong scattering. For such materials the often-invoked free-electron theory is, to say the least, distinctly suspect.

1. Single-particle absorption

The intraband single-particle absorption is confined to energies $\hbar \Omega \approx W$. Since in these materials $W \approx \hbar/\tau$, this absorption occurs entirely within a region of the optical spectrum where a perturbation expansion in powers of $(\Omega \tau)^4$ is not valid, and the second-order expression (24) does not apply. Here we shall not undertake a detailed discussion of the optical response, but shall content ourselves with a few general observations.

In this situation, very substantial departure from Drude behavior is not only expected on physical grounds, but required by the $f$ sum rule. In particular, for sufficiently narrow bands the optical absorption must be peaked at some nonzero frequency. To demonstrate, imagine a single-band metal of plasma frequency $\omega_p$ and dc conductivity $\sigma(0) = \epsilon_\infty \omega_p^2 \tau / 4 \pi$. If plasmon contributions are neglected, the absorption $\Omega \epsilon_\parallel(\Omega) = 4 \pi \sigma(\Omega)$ is cut off at the bandwidth $W$. If $\sigma(\Omega)$ is nowhere to exceed $\sigma(0)$, the total strength of the absorption, $\int_0^\infty \Omega \epsilon_\parallel(\Omega) d\Omega$, can be no larger than $4 \pi \sigma(0) W / \hbar = \epsilon_\infty \omega_p^2 \tau W / \hbar$. But the sum rule (31) sets the integral equal to $\frac{1}{2} \pi \epsilon_\infty \omega_p^2$. It follows that $\sigma(\Omega)$ must rise above $\sigma(0)$ and pass through a maximum if $W < \frac{1}{2} \pi (\hbar / \tau)$. Even for somewhat larger values of $W$, the deviations from simple Drude behavior are strong.

In the quasi-one-dimensional conductors, a very broad strong peak is, in fact, observed in the infrared. The sum-rule arguments do not, of course, provide an explanation; they simply remind us that the condition $W \tau \approx \hbar$ means that the conduction electrons are strongly coupled, and that a corresponding optical absorption is expected. It requires a thorough study of the individual mate-
rials involved to determine the extent to which the actual mechanism involves electron correlations, the electron-phonon coupling, or, in appropriate cases, direct transitions between states weakly localized by structural disorder.

2. Plasmon absorption

The sum-rule arguments discussed above will in general be altered somewhat by any low-lying interband transitions and by the plasmon absorption. In narrow-band systems a calculation of the latter is complicated by the same problems as arise for the single-particle absorption. It is worth remarking, however, upon the interesting case where the band is so narrow that $\hbar \omega_p > W$. From the formula (19) with $\kappa \sim 10^{14}$ cm$^{-2}$ and $\epsilon_{\omega} \sim 2.3$, we find that this occurs for a half-filled band when $W < 0.75$ eV -- a condition which is probably fulfilled in the organic quasi-one-dimensional conductors. When the difference between $\hbar \omega_p$ and $W$ is substantial, we have above $W$ a continuum of "free" plasmon excitations which cannot easily decay into single-particle states even by phonon emission. They should give rise to optical absorptions which persist to energies larger than $W$. A possible experimental example of such behavior will be discussed in the following paper.

V. CONCLUSION

We emphasize again that our calculations are not intended to represent faithfully the detailed high-frequency behavior of real quasi-one-dimensional conductors. Rather, we have pursued the implications of the simple models upon which much of the current theoretical work and interpretation of experiments has been based. Within these models, we have found that the longitudinal dielectric response and elementary-excitation spectrum are qualitatively different from their counterparts in isotropic three-dimensional metals, and that as a result the usual assumptions about the form of the high-frequency optical absorption are unreliable.

When such assumptions fail even for these simple models, the optical properties of the complicated real materials must be interpreted with care. The present work gives a qualitative indication of some of the new features to be expected (such as an extended region of plasmon absorption), but its utility for direct quantitative predictions is limited. We have neglected, for example, the interband transitions which lie just above the intraband absorption in the organic materials and may even overlap it in the mixed-valence platinum chain salts. Also potentially serious is our failure to take better account of the short-range part of the electron-electron interaction. Except at short wavelengths, this neglect probably has little effect upon the plasmon dispersion, but it can alter substantially the single-particle spectrum in narrow-band systems. Finally, we remark that the inter-chain coupling in the quasi-one-dimensional organic semimetals such as TTF-TCNQ (which consist of two kinds of conducting chains, with the electron population on one equal to the hole population on the other) will be rather different from that considered here. Nevertheless, we regard the present work as a minimal first step toward an understanding of the high-frequency behavior of these materials.

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15. This conclusion has been reached independently by W. A. Little (private communication).
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