PHYSICAL REVIEW B VOLUME 25, NUMBER 12 15 JUNE 1982

Electron-libron modes in a quasi-one-dimensional conductor

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(Received 16 December 1981)

A model calculation is done so as to study the conduction-electron excitation spectrum for a solid consisting of equally spaced atoms which are free to rotate about their centers of mass. In this model, the atoms are assumed to be electrically neutral dipoles of spin $\frac{1}{2}$. Owing to the Coulomb interaction between an electron and a libron (the quantum of libration) on neighboring lattice sites, it is shown that the system possesses a bound *excitonic state* consisting of an electron and a libron. The quasiparticle spectrum for the excitons consists of two branches $\omega_k^{(\pm)}$ which are labeled by the wave number k. The gap function Δ_k, which is a measure of the electron-libron pairing, and a parameter η, determining the orientational order for the atomic system, are determined by a set of coupled equations. We study the conditions under which there are electron-libron modes by doing numerical calculations for various values of the parameters.

I. INTRODUCTION

For several years there has been considerable interest in quasi-one-dimensional and two-dimensional solids.[1] In this paper we are concerned with the study of electron-libron excitations in a one-dimensional model and we show that such excitations are well defined, with the use of a formalism which we have presented recently.[2,3]

In the quasi-one-dimensional charge-transfer salts such as TTF-TCNQ (tetrathiafulvalene tetracyanoquinodimethane) the molecular degrees of freedom can be classified into translations, librations, and intramolecular distortions.[4] All three types of motion have received some attention, with the librational modes being discussed after a suggestion by Morawitz[5] that they may play a role in the Peierls transition. Merrifield and Suna[6] showed that there would not be a Peierls transition if the coupling is quadratic in the libron amplitude. However, Morawitz avoided this difficulty by assuming electronic degeneracy. Weger and Friedel[7] showed that it is possible to understand the crystallographic phase transitions in TTF-TCNQ by considering mechanisms which give rise to both librational and translational motion of the molecules. In their formalism they assumed that the lattice distortions which produce the N$-$S bond are dominant and consequently the libration about the axis perpendicular to the molecular plane plays a crucial role in their theory. Gutfreund, Weger, and co-workers[8] have also assumed that this mode gives rise to the main scattering effects which govern the resistivity at high temperature. Their Hamiltonian thus contains a term which couples this mode to the conduction electrons. This coupling which is dependent on the size and shape of the molecules gives rise to a modulation of the transfer integral and of the Madelung potential as well. With the use of a model for the lattice vibrations in a molecular crystal, Conwell[9] has made a detailed study of the contribution of librons (as well as phonons) to the transport properties of one-dimensional conductors.

In our recent papers in which we studied libron modes in crystals, we assumed that the molecules are *free* to rotate with angular momentum $S=1$, and thus our formalism is applicable to crystals with *large* amplitude librations. The angular momentum is thus a good quantum number for this model and we proceed by working with these eigenstates. In a solid material such as TTF-TCNQ the amplitude of libration is small, between 1° and 4° at low temperature, and thus for such materials it would not be accurate to treat the angular momentum as a good quantum number in our model. However, for small amplitude librations there is still a coupling of the H_{CQ} (i.e., electron-quadruple) or H_{CD} (i.e., electron-dipole) type [see Eqs. (1.6) and (1.7) of Ref. 3] when the molecules are tilted with respect to the chain axis, as they are in TTF-TCNQ. The crystallographic studies by Johnson and Watson[10] for crystalline compounds formed from combinations of the tetrathiafulvalene molecule and the iodide salts of the radical cation and dication show that the rotations for $(\mathrm{TTF})_7\mathrm{I}_5$ are indeed large. The molecules are perpendicular to the chain axis and there is no tilt.

The molecules of mesogenic compounds are elongated and the liquid-crystalline phase of these substances is characterized by long-range order in the orientation but not in the position of the molecules. The orientational order parameter deviates from unity (perfect order) owing to the thermal motion of the molecules. In the case of complete disorder, the order parameter is zero. A distinction is made between nematic, cholesteric, and smectic crystalline phases.[11,12] These can best be characterized by considering a liquid single crystal, i.e., a region over which the long-range order is ideal, apart from the thermal fluctuations. In particular, in the smectic phase one finds that in addition to orientational order there is a *partial* positional order, with the molecular centers of mass arranged in equidistant planes. There are two different possibilities for smectic phases. In one case the molecular centers do not have any long-range order within the layers, and thus correspond to a two-dimensional liquid. In the second case the layers are built up regularly so that the positions of the molecular centers lie on a two-dimensional lattice. If a conducting liquid crystal could be made which has a smectic-like phase with the molecules being regularly spaced along a linear chain, then we feel that this would be a system in which to look for electron-libron excitations of the type discussed in this paper.

Recently, we suggested that a collective mode could arise in a semiconductor from a bound state between a conduction electron and an exciton which is an intermediately bound electron-hole pair.[13] The qualitative difference between the Hamiltonian of Ref. 13 which produces an electron-exciton coupling and the term in our Hamiltonian in Sec. II which gives rise to an electron-libron coupling, is that the former is linear in the electron density while the latter is quadratic in this operator. Despite this difference the methods we use to solve these two problems are similar.

In Sec. II we describe the model Hamiltonian. Section III contains a calculation of the electron-libron Green's functions and some numerical results.

II. FORMULATION OF THE THEORY

In this section we describe the model Hamiltonian for our system. For *simplicity* all considerations due to electron spin are ignored and we assume that the system is described by

$$H \equiv \sum_{ll'} t(ll')\alpha_l^\dagger \alpha_{l'} + H_{DD} + H_{eD} \,, \tag{2.1}$$

where $t(ll')$ is the hopping energy of an electron from lattice site $R_{l'}^0$ to lattice site R_l^0. $\alpha_l, \alpha_l^\dagger$ are the destruction and creation operators for an electron at the lattice site R_l^0. The rotating molecules interact via the dipole-dipole interaction H_{DD}. This interaction is three dimensional and is given by

$$H_{DD} \equiv \tfrac{1}{2} \sum_{ll'} V_{ll'}(\vec{\Omega}_l, \vec{\Omega}_{l'}) \tag{2.2a}$$

with the dipole-dipole interaction given by

$$V_{ll'} \equiv -v_{ll'} \sum_{MN} C(112;MN) Y_{1M}(\vec{\Omega}_l) \times Y_{1N}(\vec{\Omega}_{l'}) Y_{2,M+N}(\vec{\Omega}_{ll'}) \,, \tag{2.2b}$$

and

$$v_{ll'} \equiv \frac{\mu^2}{R_{ll'}^3} \left[\frac{4\pi}{3}\right] \left[\frac{24\pi}{5}\right]^{1/2} . \tag{2.2c}$$

Here, μ is the dipole moment, $R_{ll'}$ is the distance between lattice sites at l and l', and $\vec{\Omega}_l$ is the orientation of the molecule at the lattice site l relative to the crystal axis. Also, $\vec{\Omega}_{ll'}$ is the angle between molecules at l and l', $C(112;MN)$ is a Clebsch-Gordan coefficient, and Y_{1M} is a spherical harmonic. Transforming to a coordinate system where the z_l axis of each molecule is along the symmetry axis of that molecule at l, we have

$$Y_{1M}(\vec{\Omega}_l) = \sum_m \mathscr{D}_{Mm}^{(1)*}(\alpha_l, \beta_l, \gamma_l) Y_{1m}(\vec{\omega}_l) \,, \tag{2.3}$$

where $\alpha_l, \beta_l, \gamma_l$ are Euler angles, $\mathscr{D}_{Mm}^{(1)}$ is the transformation matrix, and $\vec{\omega}_l$ is the orientation of the molecule at R_l^0 relative to the local frame of reference. In terms of local-tensor operators, we have

$$Y_{1m}(\vec{\omega}_l) = c_m L_1^m(l) \,, \tag{2.4a}$$

$$c_0 = c_{-1} = -c_{+1} = \left[\frac{3}{4\pi}\right]^{1/2} . \tag{2.4b}$$

Substituting Eq. (2.4) into (2.3) and then making use of the result for $Y_{1M}(\vec{\Omega}_l)$ in Eq. (2.2), we obtain

$$H_{DD} = -\sum_{ll'} \sum_{mn} \Gamma_{mn}(ll') L_1^m(l) L_1^n(l') \,, \tag{2.5}$$

where

$$\Gamma_{mn}(ll')\equiv\tfrac{1}{2}v_{ll'}c_mc_n\sum_{MN}C(112;MN)Y_{2,M+N}(\vec{\Omega}_{ll'})\mathscr{D}^{(1)*}_{Mm}(\alpha_l,\beta_l,\gamma_l)\mathscr{D}^{(1)*}_{Nn}(\alpha_{l'},\beta_{l'},\gamma_{l'})\ . \tag{2.6}$$

From the properties of the rotation matrix[14]

$$\mathscr{D}^{(1)*}_{mm'}(\alpha_l,\beta_l,\gamma_l) =(-1)^{m+m'}\mathscr{D}^{(1)}_{-m,-m'}(\alpha_l,\beta_l,\gamma_l)\ , \tag{2.7}$$

we obtain

$$\Gamma_{-m,-n}(ll')=(-1)^{m+n}\Gamma^*_{mn}(ll')\ , \tag{2.8}$$

$$\Gamma_{mn}(ll')=\Gamma_{nm}(l'l)\ . \tag{2.9}$$

The interaction between an electron and the rotating molecule may also be expressed in terms of spherical-tensor operators. We have

$$H_{eD}=\sum_{ll'}\lambda(ll')L^0_1(l)\rho(l')\ , \tag{2.10}$$

where $\rho(l)$ is the electron-density operator in the Wannier representation and $\lambda(ll')$ is the coupling between a molecule and the conduction electrons. Collecting the terms we rewrite the Hamiltonain of Eq. (2.1) as

$$\begin{aligned}H=&\sum_{ll'}t(ll')\alpha^\dagger_l\alpha_{l'}\\&-\sum_{ll'}\sum_{mn}\Gamma_{nm}(ll')L^m_1(l)L^n_1(l')\\&+\sum_{ll'}\lambda(ll')L^0_1(l)\rho(l')\ .\end{aligned} \tag{2.11}$$

For a molecule of spin S, we may express the spherical-tensor operators in terms of the spin components:

$$\begin{aligned}&L^\pm_1(l)=S^\pm_l/\sqrt{2f_1},\\&L^z_1(l)=S^z_l\sqrt{f_1},\\&f_1\equiv S(S+1)\ .\end{aligned} \tag{2.12}$$

Writing $a_l\equiv S_l^+$, $a_l^\dagger\equiv S_l^-$, we have

$$S^z_l=\tfrac{1}{2}-n_l\ , \tag{2.13}$$

$$[a_l,a_{l'}]_-=[a^\dagger_l,a^\dagger_{l'}]_-=0\ , \tag{2.14a}$$

$$[a_l,a^\dagger_{l'}]_-=(1-2n_l)\delta_{ll'}\ , \tag{2.14b}$$

where $n_l\equiv a^\dagger_l a_l$. The electron ($\alpha$) and libron ($a$) operators *anticommute* with each other and the electron operators obey the usual anticommutation relations. In terms of the libron operators Eq. (2.11) is rewritten as

$$\begin{aligned}H=&\sum_{ll'}t(ll')\alpha^\dagger_l\alpha_{l'}-\tfrac{1}{3}\sum_{ll'}\{\Gamma_{00}(ll')(1-2n_l)(1-2n_{l'})+[2\Gamma_{+1+1}(ll')a_la_{l'}+\text{ H.c}]\\&+[2\Gamma_{+1-1}(ll')a_la^\dagger_{l'}+\text{ H.c}]+[2\sqrt{2}\Gamma_{+10}(ll')a_l(1-2n_{l'})-\text{ H.c.}]\}\\&+\frac{1}{\sqrt{3}}\sum_{ll'}\lambda(ll')(1-2n_l)\rho(l')\ ,\end{aligned} \tag{2.15}$$

where we have used the symmetry properties of Γ_{mn} in Eqs. (2.8) and (2.9). In calculating our Green's functions in the next section we use the form of the Hamiltonian in Eq. (2.15). We emphasize that we ignore all effects due to electronic spin. In the calculations in Sec. III, we make use of the fact that $\Gamma_{+1+1}(ll')$ is much less than $\Gamma_{00}(ll')$ and $\Gamma_{+1-1}(ll')$ in most cases for fixed l and l', and so set Γ_{+1+1} equal to zero in our calculations. This is done for convenience but it can be included easily.

III. DISPERSION RELATION FOR ONE-DIMENSIONAL ELECTRON-LIBRON MODES

In this section we discuss the response of the one-dimensional conductor to an external perturbation. We use a Green's-function method similar to that used in the theory of superconductivity.[15] This method was recently used by the authors in a theory for biexciton and electron-exciton complexes in *insulators,* where the excitons are formed from intermediately bound electron-hole pairs.[13,16]

Since we are interested in a bound state between an electron and a libron, the Green's function which we evaluate is $\langle\langle a_l(t);\alpha_{l'}^\dagger(0)\rangle\rangle$. In this notation, $\langle\langle ;\rangle\rangle$ denotes a retarded Green's function and the time dependence of the operators is in the Heisenberg representation. This *libron-electron* Green's function is coupled to the *electron* Green's function

$$\langle\langle \alpha_l^\dagger(t);a_{l'}(0)\rangle\rangle \ .$$

To calculate the equations of motion of these functions we commute a_l and $\alpha_l^\dagger$ in turn with the Hamiltonian (2.15). Making use of the algebra in Eq. (2.14) and the commutation relations for the electron operators, we obtain the following set of coupled equations within our decoupling scheme

$$[a_l,H]_-\approx\tfrac{4}{3}\eta\sum_{l'}\left[\Gamma_{00}(ll')a_l-\tfrac{1}{2}[\Gamma_{+1-1}(l'l)+\Gamma^*_{+1-1}(ll')]a_{l'}-\frac{\sqrt{3}}{2}\lambda(ll')\langle\alpha_{l'}a_l\rangle\alpha_{l'}^\dagger\right] , \tag{3.1}$$

$$[\alpha_l^\dagger,H]_-\approx-\sum_{l'}t(ll')\alpha_{l'}^\dagger-\frac{1}{\sqrt{3}}\eta\sum_{l'}\lambda(ll')\alpha_l^\dagger+\frac{2}{\sqrt{3}}\sum_{l'}\lambda(ll')\langle a_{l'}^\dagger\alpha_l^\dagger\rangle a_{l'} \ . \tag{3.2}$$

In deriving these equations, we have used the symmetry properties of Γ_{mn} in Eqs. (2.8) and (2.9) and we have replaced the dipolar operator by its thermal average:

$$\eta\equiv1-2\langle n_l\rangle \ . \tag{3.3}$$

Here η is independent of lattice site since all sites are equivalent. We have also ignored higher-order terms which describe libron-libron scattering effects. This approximation is expected to be valid in the coherence approximation where scattering effects are supposed to be negligible. The last terms on the right-hand side of Eqs. (3.1) and (3.2) describe the pairing between an electron and a libron located at different lattice sites of the crystal.

Since it is more convenient to do our calculations in momentum space, we use Eqs. (3.1) and (3.2) in our equations of motion of our Green's functions and then Fourier transform with respect to time and space. We obtain (set $\hbar=1$)

$$(\omega-\xi_k)\langle\langle a_k;\alpha_{-k}\rangle\rangle+\eta\Delta_k\langle\langle\alpha_{-k}^\dagger;\alpha_{-k}\rangle\rangle=0 \ , \tag{3.4}$$

$$(-\omega-E_k)\langle\langle\alpha_{-k}^\dagger;\alpha_{-k}\rangle\rangle+\Delta_k^*\langle\langle a_k;\alpha_{-k}\rangle\rangle=1 \ , \tag{3.5}$$

where

$$\Delta_k\equiv\frac{2}{\sqrt{3}}\frac{1}{N}\sum_{k'}\lambda(k')\langle\alpha_{k'-k}a_{k-k'}\rangle \tag{3.6}$$

in the energy-gap function for electron-libron bound pairs. N is the total number of atoms on the chain, with lattice spacing a_0 and the wave numbers are in the first Brillouin zone $-\pi/a_0\le k\le\pi/a_0$. In Eqs. (3.4) and (3.5) we have also introduced the notation

$$\xi_k\equiv\tfrac{2}{3}\eta[2\gamma_{00}(0)-\gamma_{+1-1}(k)-\gamma^*_{+1-1}(-k)] \ , \tag{3.7}$$

$$E_k\equiv\epsilon_k+\frac{1}{\sqrt{3}}\lambda(0) \ . \tag{3.8}$$

Here $\gamma_{mn}(k)$, $\lambda(k)$, and ϵ_k are the Fourier transforms of $\Gamma_{mn}(ll')$, $\lambda(ll')$ and $t(ll')$, respectively. ξ_k is, of course, the rotational energy for a molecule and ϵ_k the band energy for a free electron. In deriving Eqs. (3.4) and (3.5) we have ignored the possibility of a libron and an electron scattering off each other into different momentum states.

The two sets of coupled algebraic equations (3.4) and (3.5) may easily be solved to give the results,

$$\langle\langle a_k;\alpha_{-k}\rangle\rangle=-\eta\frac{\Delta_k}{(\omega-\xi_k)(-\omega-E_k)-\eta\,|\Delta_k|^2} \ , \tag{3.9}$$

$$\langle\langle\alpha_{-k}^\dagger;\alpha_{-k}\rangle\rangle=\frac{\omega-\xi_k}{(\omega-\xi_k)(-\omega-E_k)-\eta\,|\Delta_k|^2} \ . \tag{3.10}$$

The thermal average $\langle\alpha_{-k}a_k\rangle$ in the gap function of Eq. (3.6) may be obtained by taking the difference of the Green's function $\langle\langle a_k;\alpha_{-k}\rangle\rangle$ across the branch cut along the real axis and integrating over frequency,

$$\langle \alpha_{-k} a_k \rangle = -2 \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} f_0(\omega) \mathrm{Im} \langle\langle a_k ; \alpha_{-k} \rangle\rangle \,. \tag{3.11}$$

Here $f_0(\omega) \equiv (1+e^{(\omega - E_F)})^{-1}$ where T denotes temperature and units with $k_B = 1$ are used. E_F is the Fermi energy. Making use of Eq. (3.9) in Eq. (3.11) and then substituting the result $\langle \alpha_{-k} a_k \rangle$ into Eq. (3.6), we obtain the result for Δ_k:

$$\Delta_k = \frac{4}{\sqrt{3}} \eta \frac{1}{N} \sum_{k'} \lambda(k') \Delta_{k-k'} \times \frac{f_0(\omega_{k-k'}^{(+)}) - f_0(\omega_{k-k'}^{(-)})}{(\omega_{k-k'}^{(+)} - \omega_{k-k'}^{(-)})} \,, \tag{3.12}$$

where

$$\omega_k^{(\pm)} \equiv \tfrac{1}{2} \{ (\xi_k - E_k) \pm [(\xi_k + E_k)^2 - 4\eta \,|\, \Delta_k \,|^2]^{1/2} \} \tag{3.13}$$

are the excitation energies for the electron-libron bound states.

The equation-of-motion method may again be applied to calculate the *libron-libron* Green's function $\langle\langle a_k ; a_k^\dagger \rangle\rangle$. Using the result in Eq. (3.1), one finds that $\langle\langle a_k ; a_k^\dagger \rangle\rangle$ is coupled to the Green's function $\langle\langle \alpha_{-k}^\dagger ; a_k^\dagger \rangle\rangle$. Therefore, using Eqs. (3.1) and (3.2) we obtain two sets of coupled equations which may be solved easily. In particular we have

$$\langle\langle a_k ; a_k^\dagger \rangle\rangle = -\eta \frac{\omega + E_k}{(\omega - \xi_k)(-\omega - E_k) - \eta \,|\, \Delta_k \,|^2} \,, \tag{3.14}$$

where, we emphasize, all effects arising from the scattering of an electron off a libron into different momentum states are neglected. Taking the difference of the Green's function $\langle\langle a_k ; a_k^\dagger \rangle\rangle$ in Eq. (3.14) across the branch cut along the real axis and integrating over frequency, we obtain from the definition of η in Eq. (3.2) another equation for η and Δ_k.

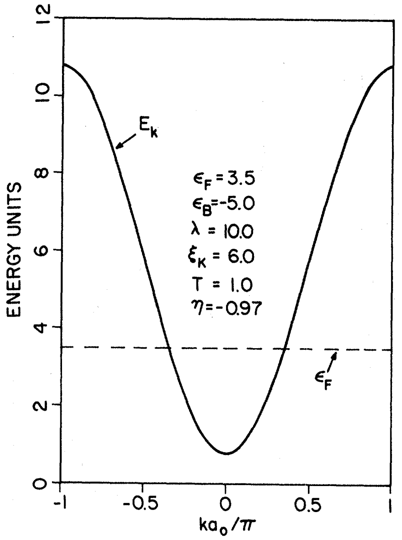

FIG. 1. Electron-band energy as a function of wave vector. Δ_0 is the unit of energy.

For an undistorted linear chain we have $\Delta_k = \Delta_0 \cos ka_0$. In our calculation we use $\Delta_0 = 0.02$ eV and set $\lambda(k) = \lambda \cos ka_0$, $\epsilon_k = \epsilon_B \cos ka_0$, which correspond to nearest-neighbor electron-libron coupling, and electron hopping between nearest-neighbor lattice sites, respectively. For the sake of convenience we ignore the k dependence of ξ_k defined in Eq. (3.7), by setting this quantity equal to a constant. All energies as well as temperature are measured in terms of Δ_0.

Figures 1 through 4 show plots of the electron-band energy and the two branches of the electron-libron excitation spectrum as a function of wave vector. The energy of the lower $\omega_k^{(-)}$ branch shows more variation with k than that of the upper $\omega_k^{(+)}$ branch of the excitation spectrum. The value of the ordering parameter η must lie between

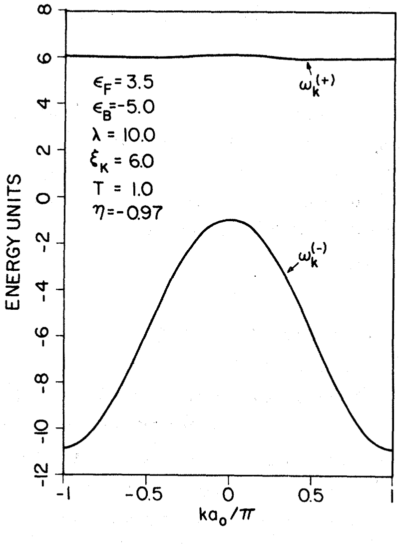

FIG. 2. Plot of the two branches $\omega_k^{(\pm)}$ of the electron-libron excitation spectrum for the same values of the parameters used in Fig. 1. Δ_0 is the unit of energy.

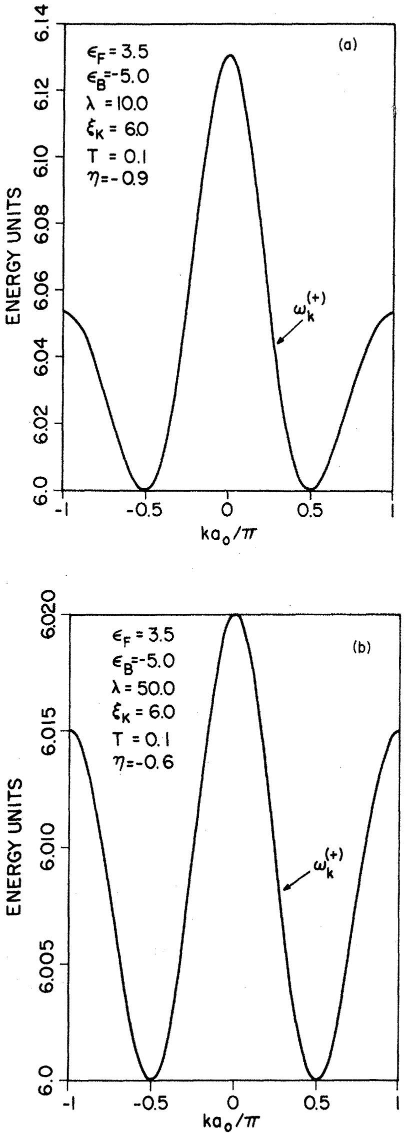

FIG. 3. Upper branch $\omega_k^{(\pm)}$ of the electron-libron excitation spectrum is plotted for two different values of the coupling constant λ. The Fermi energy (ϵ_F), the electron bandwidth (ϵ_B), the libron energy (ξ_k), and the temperature (T) are the same in (a) and (b). Δ_0 is the unit of energy.

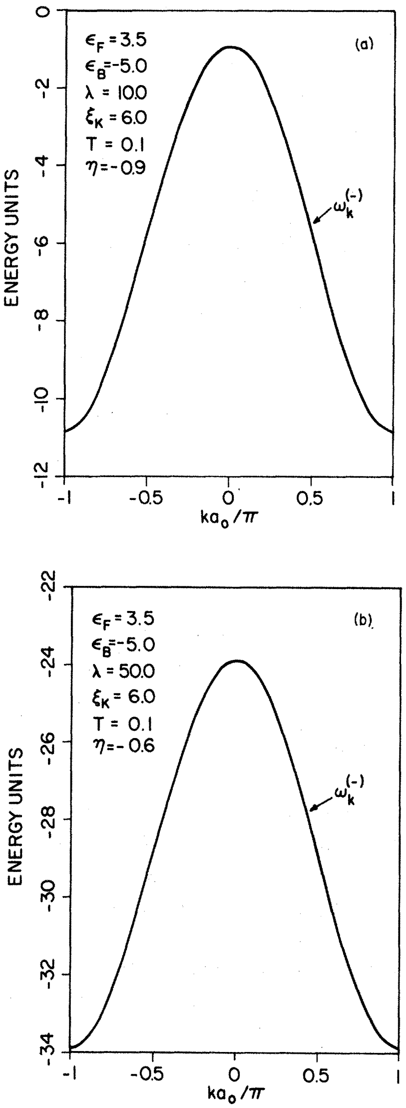

FIG. 4. Same as Fig. 3 but with the lower branch $\omega_k^{(-)}$ being plotted.

0 and -1. Keeping all other parameters fixed, a state of total disorder ($\eta = 0$) is achieved by increasing the coupling parameter λ. For the electron and libron energies which we have chosen, numerical calculation shows that the excitation spectrum at $T = 0$ differs from its value at $T = 0.1$ (in units) of Δ_0 by only a few percent because of the presence of the exponential in the Fermi distribution function. Comparing the values of η in Figs. 2 and 3(a), we see that this parameter is not very sensitive to temperature as well. From our results in Figs. 3 and 4, we find that for $-\eta$ to be reduced from 0.9 to 0.6, λ has to be increased by a factor of 5. Therefore, since our results are not very sensitive to changes in either temperature or electron-libron coupling, we feel that their experimental verification is feasible.

IV. FINAL REMARKS

The model calculation which we have presented here should be understood as the first steps towards a general model theory for librons with large amplitudes in crystals which are electrically conducting. This microscopic treatment in terms of a model whose molecules have rigidly fixed centers of mass illuminates some essential features of a more general approach. A generalization to incorporate phonons into the present model would be desirable. We hope that the future will see a bridging between this and other models, at least in some simple cases, as well as this calculation and experiment.

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