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線性固體與電子－聲子交互作用

One-Dimensional Solid with Electron-Phonon Interaction

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ABSTRACT — We use the quasiparticle approach and the method of the equation of motion to study a one-dimensional solid with electron-phonon interaction. After transforming to a moving frame, an energy gap in the single electron energy spectrum with a width at $v=0$; $\Delta=16E_f e^{-1/\lambda}$ is obtained, which has the same form as in pairing theory. We also obtain a state in which the electrons will move with the phonons in an organized way. Thus the conductivity will increase extraodinary if v is sufficiently small. This result agrees with the experimental pheonomenon, which was observed recently in some one-dimensional organic solid.

I. Introduction

In recent years many efforts has been studied on the properties of one-dimensional system and a wide range of both inorganic and organic compounds which have characteristic one-dimensional metallic behavior have been discovered. [1-3] Ferraris et al have found some one-dimensional solid is metallike above the transition temperature T_c and insulatorlike below. Coleman et al [4] have observed an extradinary increase of conductivity at T_c for an organic solid TTF;TCNQ in which the conduction is primarily along chains of molecules in essentially one-dimension. It has long been well-known that an one-dimensional metal is inherently unstable with respect to charge or spin-density waves. [5-6] The localization of electronic wave function from a single band by electron-electron interaction [7] is not the only means of achieving a metal-to-insulator transition. One principal mechanism has been proposed by Peierls and developed in details by Adler and Brook. Peierls observed that a crystallic distortion would give rise to a lower symmetry with the result that additional one-electron band splitting would appear. If such splitting were to result in an energy gap at the Fermi energy, an insulating state would be obtained.

One-dimensional model of metals have been investigated by Fröhlich [7] and Overhauser [8], and recently by many others [9,10]. Fröhlich considered a one-dimensional model of a metal with electron-phonon interaction, and show that such a metal is (for sufficiently strong electron-phonon coupling) a superconductor. Overhauser also considered a one-dimensional metal, however, with direct electron-electron interaction. In Overhauser's model, there is also a transition to an ordered state at a relatively high temperature. The fluctuation effect has been studied by Allender, Bray and Bardeen[11] using a Ginsbury-Landau approach and they obtain a maximum normalized conductivity ratio of $\sigma/\sigma_{RT} \sim 10$ in agreement with the experimental value found by Coleman et al.

The reflectivity measurement by Jarobsen et al [12] has confirmed the existence of an energy gap in the electronic spectrum of some one-dimensional organic solid, which is of importance to understanding the origin of the remarkable transport properties of this solid. [2,4,13,14] The nonmagnetic ground state and nonenhanced susceptibility in the metallic state [15] demonstrated that Coulomb correlation effect plays a minor role. On the other hand the electron-phonon interaction was found experimentally to be strong. [16-17] Therefore, in this work we shall only consider the electron-phonon interaction.

Theoretical calculations on the one-dimensional system usually have been carried out by a self-consistent method, which is complicated. In this work we shall consider a one-dimensional system, using quasiparticle approach, by means of the method of equation of motion. Essentially a quasiparticle is simply an excitation of a many-body system, with some of the properties of a free particle. Historically the concept originated in Landau's theory of liquid He. Landau advanced arguments to make it seem plausible that the low-lying energy eigenstates of a gas of interacting Bosons could be in one-to-one correspondence with those of a gas of noninteracting Bosons. An important landmark in the evolution of the quasiparticle concept was the introduction by Fröhlich of the polaron model of an electron moving through an ionic crystal. As the idea of quasiparticle has developed, it has been used to many application, for example, the Debye theory of specific heat can be regared as a quasiparticle theory in which the quasiparticles are phonons, the quanta of excitation of the normal modes of a crystal lattice.

In this paper, we consider a one-dimensional system, which includes the electron-phonon interaction. After getting a coupled differential equations for the electron creation operators, we make a Bogoliubov's transformation to the quasiparticle states. We shall see that an energy gap will naturally appear in the quasiparticle energy spectrum. This result was also obtained by Fröhlich[11], but based on a physical argument proposed by Peierls [18] and using a complicated self-consistent method. We also find a state in which the electrons and associated lattice displacement move bodily through the lattice and thus carry an electric current. For completeness, we shall briefly review the method of equation of motion in the second section. In the third section we apply this method to a one-dimensional system and give a detailed calculations. Conclusion and discussion will be given in the final section.

II. Review of the Equation of Motion Method

Consider a system of noninteracting particles with a Hamiltonian

$$H_o = \sum_k \varepsilon_k a_k^+ a_k \quad , \tag{2-1}$$

where a_k^+ and a_k are the creation and annihilation operators for the system of Fermions. The Heisenberg equations of motion for a_k^+ and a_k will have the simple form

$$i\hbar \frac{da_k^+}{dt} = [a_k^+ , H_o] = \varepsilon_k a_k^+ \quad , \tag{2-2}$$

$$i\hbar \frac{da_k}{dt} = [a_k , H_o] = -\varepsilon_k a_k \quad . \tag{2-3}$$

Equations (2-2) and (2-3) are immediately integrable, to give

$$\begin{aligned} a_k^+(t) &= a_K^+(0) e^{-i\varepsilon_k t/\hbar} \\ a_k(t) &= a_k(0) e^{i\varepsilon_k t/\hbar} \end{aligned} \tag{2-4}$$

Thus the creation and annihilation operators a_k^+ and a_k are oscillating functions of time, with a frequency $\varepsilon_k/\hbar$.

If the interaction is included in the system, the Hamiltonian becomes

$$(2\text{-}5) \qquad H = \sum_k \varepsilon_k a_k^+ a_k + \frac{1}{2} \sum_{k,\ell,q} V_q \, a_{k+q}^+ \, a_k \, a_{\ell-q}^+ \, a_\ell \quad ,$$

then the equations of motion for a_k^+ and a_k are

$$(2\text{-}6) \qquad i\hbar \frac{da_k^+}{dt} = \varepsilon_k \, a_k^+ + \sum_{\ell,q} V_q \, a_{k+q}^+ \, a_{\ell-q}^+ \, a_\ell \quad ,$$

$$i\hbar \frac{da_k}{dt} = -\varepsilon_k a_k - \sum_{\ell,q} V_q \, a_\ell^+ \, a_{\ell-q} \, a_{k+q} \quad .$$

Thus the creation operator for a single excitation is coupled to a trilinear operator which in general acts to create a particle and a particle-hole pair. By formal integration of (2-6) and repeat iteration, we can generate an infinite regression of increasingly complicated expressions, involving multiple products of field operators at different times. However, this is not a profitable procedure, a more fruitful approach is to use some approximation to terminate the regression after at most a few steps. The simplest of these schemes is to rewrite the equation by seperating out those parts of the trilinear term that are proportional to a_k^+, i.e, with q=0, q=l-k

$$(2\text{-}7) \qquad \begin{aligned} i\hbar \frac{da_k^+}{dt} &= [\varepsilon_k + NV_o - \sum_q V_q N_q] a_k^+ + \sum_q V_q [a_{q+k}^+ \, a_{q+k} - N_q] a_k^+ \\ &\quad + \sum_{\substack{q \neq 0 \\ \ell \neq q+k}} V_q \, a_{k+q}^+ \, a_{\ell-q}^+ \, a_\ell \quad , \end{aligned}$$

where $N = \sum_\ell a_\ell^+ a_\ell$.

If we neglect the second fluctuation term and the third nonlinear term, we obtain a linear equation for a_k^+ and a_k

$$(2\text{-}8) \qquad i\hbar \frac{da_k^+}{dt} = \eta_k \, a_k^+ \quad ,$$

$$i\hbar \frac{da_k^+}{dt} = -\eta_k \, a_k \quad ,$$

where

$$(2\text{-}9) \qquad \eta_k = \varepsilon_k + NV_o - \sum_q V_q N_q \quad .$$

Thus (2-8) is also immediately integrable, to give

$$(2\text{-}10) \qquad a_k^+(t)=a_k^+(0)e^{-i\eta_k t/\hbar}$$

$$a_k(t)=a_k(0)e^{i\eta_k t/\hbar}$$

The self-consistency can be easily seen as follows: on substituting for the time-dependent field operators (2-10) in (2-9), the phase factors cancel, and the expression multiplying a_k^+ or a_k on the right-hand side of (2-8) is indeed independent of time.

Since the equations of motion for the a_k^+ and a_k operators have the same form as in the noninteracting system, thus a_k^+ and a_k behave as creation and annihilation operators for quasiparticles of energy η_k. From the structure of (2-9), it is seen that there is no energy gap. In order to obtain an energy gap as required for superconductor, the above treatment must be modified by making a Bogoliubov's transformation. It will be explained in the next section when we consider the case of one-dimensional system.

III. Calculations on One-Dimensional System

Consider a system of N electrons in linear chain coupled to phonons, the Hamiltonian of this system may be written as

$$(3\text{-}1) \qquad H=\sum_k \varepsilon_k a_k^+ a_k + \sum_w \hbar w s\, b_w^+ b_w + H_{int} \quad ,$$

$$(3\text{-}2) \qquad H_{int}= i \sum_k \sum_{w>0} D_w [a_{k+w}^+ a_k (b_w+b-w^+) - a_k^+ a_{k+w} (b_w^+ + b_{-w})] .$$

where a_k^+ and a_k are the creation and annihilation operators for electrons, b_w^+ and b_w are the creation and annihilation operators for phonons, s is the velocity of sound and D_w is the interaction constant.

One might think that if the interaction parameter D is sufficiently small that H_{int} can be treated as a small perturbation. It can be easily shown by calculating the self-energy of a vibrational quantum of wave number w that this is not true because when $w \simeq w_o = 2k_o$, where k_o is related to the Fermi energy $E_f=\hbar^2 k_o^2/2m$, then the self-energy diverges so that the perturbation theory cannot be applied. Therefore we shall consider the Hamiltonian to be divided into two parts

$$H = H_o + H' \tag{3-3}$$

$$H_o = \sum_k \varepsilon_k a_k^+ a_k + \sum_{w>0} \hbar w_s (b_w^+ b_w + b_{-w}^+ b_{-w}) + i \sum_k D[a_{k+w_o}^+ a_k (b_{w_o} + b_{w_o}^+) - a_k^+ a_{k+w_o} (b_{w_o}^+ + b_{-w_o})] \tag{3-4}$$

$$H' = i \sum_k \sum_{w>0}' D_w [a_{k+w_o}^+ a_k (b_w + b_{-w}^+) - a_k^+ a_{k+w} (b_w^+ + b_{-w})] \tag{3-5}$$

where the prime on the sum means the term $w=w_o$ is omitted. The momentum of the whole system is

$$P = \sum_k \hbar k \, a_k^+ a_k + \sum_{w>0} \hbar w (b_w^+ b_w - b_{-w}^+ b_{-w}) \tag{3-6}$$

It can be shown that this momentum operator commutes with H, this implies that P is a constant of motion. Thus instead of considering H_o, we shall consider the operator

$$H_o - vP = \sum_k (\varepsilon_k - \hbar k v) a_k^+ a_k + [\hbar w_o (s-v) b_{w_o}^+ b_{w_o} + \hbar w_o (s+v) b_{-w_o}^+ b_{-w_o}] + i \sum_k D[(b_{w_o} + b_{-w_o}^+) a_{k+w_o}^+ a_k - (b_{w_o}^+ + b_{-w_o}) a_k^+ a_{k+w_o}] , \tag{3-7}$$

where v is quantized with the periodic boundary condition

$$mv/\hbar = 2\pi n/L \quad ,$$

with n be integer and L be the length of the linear chain.

Starting from the equation of motion for the creation operator a_k^+

$$i\hbar \frac{da_k^+}{dt} = [a_k^+ , H_o - vP] \quad . \tag{3-8}$$

Using (3-7), it is easily shown that

$$-i\hbar \frac{da_k^+}{dt} = (\varepsilon_k - \hbar k v) a_k^+ + iD[(b_{w_o} + b_{w_o}^+) a_{k+w_o}^+ - (b_{w_o}^+ + b_{-w_o}) a_{k-w_o}^+] . \tag{3-9}$$

Similarly, we obtain

$$i\hbar \frac{da_k}{dt} = (\varepsilon_k - \hbar k v) a_k + iD[(b_{w_o} + b_{-w_o}^+) a_{k-w_o} - (b_{w_o}^+ + b_{-w_o}) a_{k+w_o}] , \tag{3-10}$$

(3-19) $$\alpha_k^+ = u_k\, a_k^+ + v_k a_{k-w_o}^+ \quad ,$$

with

(3-20) $$u_k^2 + v_k^2 = 1 \quad ,$$

and α_k^+ satisfies

(3-21) $$-i\hbar\frac{d\alpha_k^+}{dt} = \eta_k^o\, \alpha_k^+ \quad .$$

Substitute (3-19) into (3-21) and make use of (3-16) and (3-17), we obtain

(3-22) $$u_k\varepsilon_k + 2i\, E_f\beta v_k = u_k\eta_k^o \quad ,$$

(3-23) $$v_k\varepsilon_{k-w_o} - 2i\, E_f\beta^*\, u_k = v_k\eta_k^o \quad .$$

From these equations, we have

(3-24) $$\eta_{k\pm}^o = 2E_f\left[\frac{k^2}{2k_o^2} - \frac{k}{k_o} + 1 \pm \sqrt{\left(\frac{k-k_o}{k_o}\right)^2 + |\beta|^2}\right] ,$$

(3-25) $$|u_{k\pm}|^2 = \pm\frac{1}{2}\,\frac{k_o - k \pm k_o\left[\left(\frac{k-k_o}{k_o}\right)^2 + |\beta|^2\right]^{1/2}}{k_o\left[\left(\frac{k-k_o}{k_o}\right)^2 + |\beta|^2\right]^{1/2}} ,$$

(3-26) $$v_{k\pm} = \frac{i\beta^* k_o}{k - k_o \pm k_o\left[(k-k_o/k_o)^2 + |\beta|^2\right]^{1/2}} \quad .$$

Next we shall calculate the second order correction. (3-14) and (3-15) become

(3-27) $$-i\hbar\frac{da_k^+}{dt} = \varepsilon_k a_k^+ + 2i\, E_f(\beta a_{k+w_o}^+ - \beta^*\, a_{k-w_o}^+) \quad ,$$

(3-28) $$i\hbar\frac{da_{k-w_o}}{dt} = \varepsilon_{k-w_o} a_{k-w_o}^+ + 2i\, E_f(\beta a_k^+ - \beta^*\, a_{k-2w_o}^+) .$$

Now we also make a transformation

$$r_k^+ = u_k a_k^+ + v_k a_{k-w_o}^+ + t_k a_{k+w_o}^+ + s_k a_{k-2w_o}^+ \quad , \tag{3-29}$$

with

$$u_k^2 + v_k^2 + t_k^2 + s_k^2 = 1 \quad , \tag{3-30}$$

and

$$-i\hbar \frac{dr_k^+}{dt} = (\eta_k^o + \Delta\eta_k) r_k^+ \quad . \tag{3-31}$$

From (3-29) and (3-31) and using (3-22), (3-23), (3-27), (3-28) we can obtain

$$\begin{aligned} \varepsilon_k + w_o t_k + 2i E_f \beta u_k &= \eta_k t_k \quad , \\ \varepsilon_{k-2w_o} s_k - 2i E_f \beta^* v_k &= \eta_k s_k \quad , \\ 2i E_f \beta s_k &= v_k \Delta\eta_k \quad , \\ -2i E_f \beta^* t_k &= u_k \Delta\eta_k \quad . \end{aligned} \tag{3-32}$$

The solutions which are corrected up to second order in β are

$$t_k = -\frac{i\beta u_k k_o}{2(k+k_o)|u_k|} \quad , \quad \Delta\eta_k = -\frac{E_f|\beta|^2 k_o}{(k+k_o)} \quad , \text{ if } 0<k<k_o \quad , \tag{3-33}$$

$$s_k = \frac{i\beta^* k_o v_k}{2(3k_o-k)|v_k|} \quad , \quad \Delta\eta_k = -\frac{E_f|\beta|^2 k_o}{(3k_o-k)} \quad , \text{ if } k_o<k<2k_o \quad . \tag{3-34}$$

Now let us consider (3-11), after average the whole equation over the electron states we introduce a new constant operator f:

$$b_{w_o} = B_{w_o} + f \quad , \tag{3-35}$$

where

$$f\hbar w_o(s-v) = i D \langle \Sigma_k a_k^+ a_{k+w_o} \rangle \quad . \tag{3-36}$$

Substituting (3-35) into (3-11) and using (3-36), we get

$$(3\text{-}37) \qquad i\hbar \frac{dB_{w_o}}{dt} = \hbar w_o (s-v) B_{w_o} \ .$$

Therefore $B^+_{w_o}$ is an oscillatory function of time with frequency $w_o(s-v)$ and we can obtain

$$(3\text{-}38) \qquad \langle b_{w_o} \rangle = \frac{iD}{\hbar w_o (s-v)} \langle \sum_k a_k^+ a_{k+w_o} \rangle \ .$$

Similarly we have

$$(3\text{-}39) \qquad \langle b_{-w_o} \rangle = - \frac{i D}{\hbar w_o (s+v)} \langle \sum_k a_{k+w_o}^+ a_k \rangle \ .$$

Next we calculate:

$$(3\text{-}40) \qquad \langle \sum_{k'} a_{k'+w_o}^+ a_{k'} \rangle = \frac{1}{N} \sum_k \langle 0 | r_k \sum_{k'} a_{k'+w_o}^+ a_{k'} r_k^+ | 0 \rangle \ ,$$

where k goes over all occupied electron states. It is easily to obtain by using (3-29) to (3-34):

$$\langle 0 | r_k \sum_{k'} a_{k'+w_o}^+ a_{k'} r_k^+ | 0 \rangle = \begin{cases} \frac{i\beta^*}{2} \left[\frac{1}{\sqrt{|\beta|^2 + (k-k_o/k_o)^2}} + \frac{k_o}{k+k_o} \right] & \text{if } 0<k<k_o \ , \\ \frac{i\beta^*}{2} \left[\frac{1}{\sqrt{|\beta|^2 + (k-k_o/k_o)^2}} + \frac{k_o}{3k_o - k} \right] & \text{if } k_o<k<2k_o \ . \end{cases}$$

Taking care of the spin state, we replace

$$(3\text{-}43) \qquad \sum_k \to 2 \frac{L}{2\pi} \int_{-k_o}^{k_o} dk \ .$$

Thus

$$(3\text{-}44) \qquad \langle \sum_k a_{k+w_o}^+ a_k \rangle = \frac{i\beta^*}{4} \left(\ell n \frac{\sqrt{1+|\beta|^2}+1}{\sqrt{1+|\beta|^2}-1} + 2\ell n 2 \right) \approx \frac{i\beta^*}{2} \ell n \frac{4}{|\beta|} \ ,$$

using $|\beta|^2 < 1$

(3-39) gives

$$(3\text{-}45) \qquad \langle b_{-w_o} \rangle = \frac{\beta^* D}{2\hbar w_o (s+v)} \ln \frac{4}{|\beta|} \quad .$$

Similarly we get

$$(3\text{-}46) \qquad \langle b_{w_o} \rangle = \frac{\beta D}{2\hbar w_o (s-v)} \ln \frac{4}{|\beta|} \quad .$$

From (3-18), we have

$$(3\text{-}47) \qquad \beta = [\frac{D\beta}{2\hbar w_o (s-v)} + \frac{D\beta}{2\hbar w_o (s+v)}] \ln \frac{4}{|\beta|} \frac{D}{2E_f} \quad .$$

This gives

$$(3\text{-}48) \qquad |\beta| = 4e^{-\frac{1}{\lambda}(1-\frac{v^2}{s^2})} \quad ,$$

where

$$(3\text{-}49) \qquad \lambda = \frac{2\hbar w_o E_f S}{D^2} \quad .$$

From the electron energy spectrum of (3-24), an energy gap occurs at $k=k_o$:

$$(3\text{-}50) \qquad \Delta = 4E_f |\beta| \quad .$$

At v=0, we get

$$(3\text{-}51) \qquad \Delta = 16E_f e^{-1/\lambda} \quad .$$

With this energy gap it might seems that the interaction between electron and phonon transform the system into insulator as $T \to 0$. In fact, by introducing the quasiparticle concept there exists state in which the electrons and associated lattice displacement move bodily through the lattice and thus carry an electric current, J=eNv. The movement of the lattice displacement will lead to a fluctuation of each ion arround its average position and thus does not contri-

bute to the current. The inertia of this moving system is then equal to the inertia of all electrons augmented by the inertia due to the motion of the lattice displacement. Elastic scattering of individual electron which normally leads to the residual resistance is impossible if v is sufficient small.

IV. Conclusion and Discussion

We have considered a system of linear chain with interaction between electron and phonon. The electron-electron interaction in neglected due to the fact that such interaction is indeed very small in the organic metal found recently. By a quasiparticle condiseration, there exists states in which the electrons and associated lattice displacements will move through the solid in an organized way, and therefore the conductivity increases extradinary. The quasiparticles can be considered as the electrons moving along with the phonons, the resulting entity behaves in many ways like a free particle. Its charge is of course the electronic charge, but its mass differs from that of the free electron. In fact, it can be proved that the inertia of this moving system is equal to the inertia of all electrons and the inertia due to the motion of lattice displacement. This result agrees with that of Fröhlich using a complicated self-consistent method.

We also find an energy gap at v=0: $\Delta = 16E_f e^{-1/\lambda}$ This is of the same form as the expression for the gap in the pairing theory, with the Fermi energy E_f replacing the phonon cut-off $K_B\theta_D$. Thus at lower temperatures, only the states below the gap are occupied, giving a current flow J=Nev. If E_k is the energy of the electron in a frame moving with the electron, the energy in the rest frame of the crystal is $E_k+\hbar kv$. When $\hbar kv$ becomes greater than Δ, it becomes favorable for electrons to be scattered to the next higher band, decreasing the current.

The mechanism for the paraconductivity, not superconductivity below T_c as found by Coleman et al, is still unknown. Several groups of people have tried to study this phenomena by using the fluctuation effect.[9,10,11] The results are still in disagreement with experimental values. It is clear that considerable experimental and theoretical works are required to clarify this point.

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