Energy-versus-Momentum Relation for the Piezoelectric Polaron

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We note that the intermediate-coupling theory for the piezoelectric polaron gives an energy-momentum relation that is quadratic for small P and asymptotes to a straight line with slope equal to the speed of sound at high P. We present arguments that indicate that this is the correct qualitative behavior for the piezoelectric polaron. We show, however, that if we modify the Hamiltonian so as to cut off the interaction with the long-wavelength phonons, there is a maximum momentum for which this type of solution exists.

I. INTRODUCTION

Interest in the piezoelectric polaron started when Mahan and Hopfield[1] pointed out that second-order perturbation theory at finite temperatures leads to an extremely anomalous energy-momentum $[E(P)]$ curve. Osaka[2] subsequently showed that the Tamm–Dancoff theory at finite temperature leads to a somewhat smoother curve.

In an attempt to understand the origin of these anomalies, the zero-temperature problem was investigated in a paper[3] that we will refer to here as I. It was noted in I that the structure in perturbation theory was caused by a degeneracy in the unperturbed energy levels. The natural thing to do then is degenerate perturbation theory, which turns out to be the Tamm–Dancoff theory in this problem. Unlike the analogous situation in the optical polaron,[4] the Tamm–Dancoff theory gives a qualitatively incorrect $E(P)$. It is explained in I that this is because the Tamm–Dancoff theory solves for a self-consistent $E(P)$ which locates the point of degeneracy incorrectly. The actual degeneracy occurs when the polaron can emit free phonons. In the case of acoustic phonons this point is directly determined by the polaron velocity $\vec{v}(P)=\vec{\nabla}_P E(P)$ rather than $E(P)$. When $v(P)>s$ (the speed of sound) the polaron can emit phonons, and when $v<s$ it cannot. Hence arguments in I, like those used for optical polarons,[4] suggest that if there is an anomaly in the $E(P)$ curve it will take place as $v(P)\to 1$.

It so happens that the intermediate-coupling theory treats the polaron velocity self-consistently, and that the zero of the energy denominator in this theory is determined directly by $v(P)$ rather than $E(P)$. One is then not surprised to find that this theory gives an $E(P)$ whose velocity stays below the speed of sound, and approaches it as $P\to\infty$.

It is not hard to picture that as the polaron approaches the speed of sound the enhancement of its electron-phonon interaction due to the degeneracy causes a large lattice deformation to form around the electron which in turn traps the electron and prevents it from going faster than the speed of sound. However, for this process to continue to large P (i.e., an appreciable fraction of the Brillouin zone) would imply some striking anomaly in the behavior of piezoelectric semiconductors, for which there seems to be no evidence.[5]

In Sec. II we argue that the intermediate coupling theory gives a qualitatively correct $E(P)$ for the ground state of the Hamiltonian given in Eqs. (1) and (2).

We expect that in addition to the low-lying polaron that we are discussing in this paper there is also a quasiparticle which behaves very much like a free electron. Such a quasiparticle could then account for much of the normal behavior of piezoelectric semiconductors. We have not yet obtained a theory which includes both a free-particle-like quasiparticle and the low-lying states that we are discussing here. The relative importance of these two types of states remains an important unsolved problem.

In Sec. III we note that the behavior of the polaron at high P depends on the interaction with very-long-wavelength phonons. If we modify the Hamiltonian so as to cut off the electron lattice interaction at large distances we find that the intermediate coupling theory applies only for $P<P_{cut}$, where P_{cut} is of the order of the momentum that a free electron has when it travels at the speed of sound. This is a very small momentum, and hence the theory applies only to a very small fraction of the Brillouin zone.

Since the crystal is of finite size, and for most experimental situations there are enough electrons in the conduction band to produce a screen length even smaller than the sample dimension, we feel that the predictions of the cut-off theory are more realistic. However, we still regard finding the correct solutions for the Hamiltonian [(1) and (2)] to be an interesting academic problem.

II. PIEZOELECTRIC POLARON

In this section we will discuss the properties of the Hamiltonian,

$$H=\tfrac{1}{2}p^2+\sum_q (a_q^\dagger a_q+\tfrac{1}{2})q + \sum_q Q(q)\,(a_q+a_{-q}^\dagger)\,e^{i\vec{q}\cdot\vec{r}}\,. \qquad (1)$$

For the piezoelectric polaron we use

$$Q(q)=(4\pi\alpha/Vq)^{1/2}\,. \qquad (2)$$

The unit of energy is ms^2 and the unit of length is $\hbar/ms$, where s is an average speed of sound. The operators a_q and $a_q^\dagger$ annihilate and create phonons of the one mode that replaces the three acoustic modes, and $\vec{r}$ and $\vec{p}$ are the electron position and momentum. The volume is V and α is the coupling constant.[3,6]

The total momentum operator

$$\vec{\mathcal{P}}=\vec{p}+\sum_q a_q^\dagger a_q\vec{q}$$

commutes with H, and hence the eigenstates of H can be chosen so that they are also eigenstates of $\vec{\mathcal{P}}$.

In the intermediate-coupling theory[7] one uses a trial wave function

$$\psi_P=e^{i\vec{P}\cdot\vec{r}}e^{-S}|0\rangle\,; \qquad (3)$$

$$S=\sum_q i(d_q a_q e^{i\vec{q}\cdot\vec{r}}+\text{H.c.})\,. \qquad (4)$$

The parameters d_q are chosen to minimize the expected value of H. This expected value $E(P)$ is given by

$$E(P)=\tfrac{1}{2}P^2-\tfrac{1}{2}[P-v(P)]^2 - \sum_q \frac{Q^2(q)}{q-\vec{q}\cdot\vec{v}(P)+\frac{1}{2}q^2}\;, \qquad (5)$$

where $\vec{v}(P)=\vec{P}-\sum\vec{q}d_q^2$ is determined by the transcendental equation

$$\vec{v}(P)=\vec{P}-\sum_q \frac{Q^2(q)(\vec{q})}{[\vec{q}\cdot\vec{v}(P)-\frac{1}{2}q^2-q]^2}\;. \qquad (6)$$

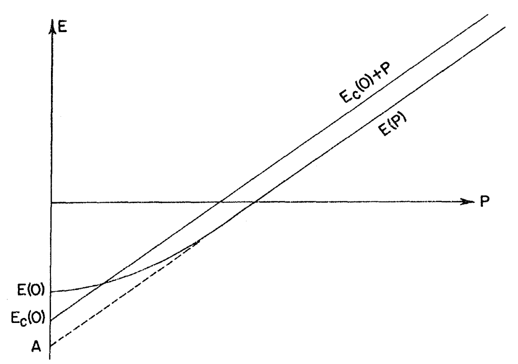

FIG. 1. $E(P)$ compared with the energy of a state composed of a stationary polaron plus a free phonon. As long as $E_c(0)>A$, $E(P)$ will be lower at high P. Units are chosen so that $\hbar=s=m=1$.

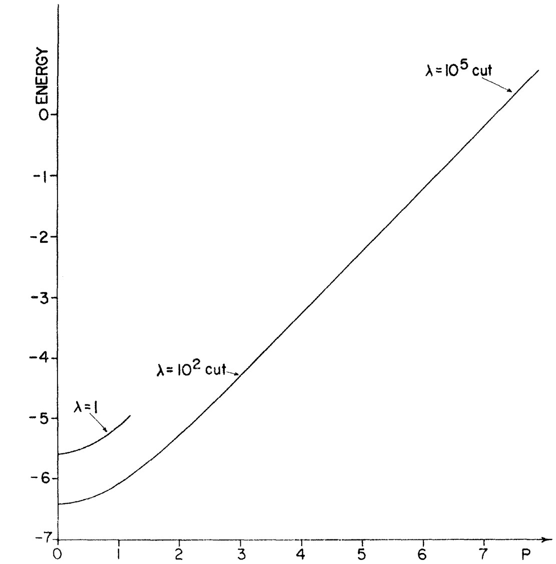

FIG. 2. $E^s(P)$ using $Q^{(2)}$ in H, and $\alpha=1$. For $\lambda>10^2$ the curve is indistinguishable from $E(P)$. Units are chosen so that $\hbar=s=m=1$.

By differentiating (5) we can see the $\vec{v}(P)=\vec{\nabla}_P E(P)$ and hence is the polaron velocity.

It is important to note that since ψ_P is an eigenfunction of $\mathcal{P}$ with eigenvalue P, then $E(P)$ is an upper bound to the correct eigenvalue of H for each value of P. We will call the correct ground-state energy $E_c(P)$.

The functions $E(P)$ and $v(P)$ can be evaluated in closed form and are reported in the Appendix of I. The curve $E(P)$ is plotted in Fig. 1. Although $E(P)$ starts out quadratic, its slope stays below 1 (the speed of sound in our units) and approaches 1 as $P\to\infty$. It is clear that since $E(P)$ is an upper bound, the correct ground-state energy cannot be a quadratic function of P because the quadratic must eventually cross the linear curve. This suggests that $E_c(P)$ has the same qualitative behavior as $E(P)$ but lies slightly below it. It is possible that $E_c(P)$ could be sublinear at large P but we do not think it very likely. Remember there is a plausible physical picture for a polaron being trapped at the speed of sound. The same thing happens to a moving-point imperfection in an elastic medium.[8] Moreover, the energy-crossing arguments in I suggest this kind of curve.

There are two possible objections that we would like to discuss here, both of which amount to the suggestion that $E_c(P)$ is not the energy of a polaron.

(i) There is the possibility that when $P\to 1$, ψ_P does not describe a polaron anymore, but something more like a free phonon with wave vector $\approx P$ and a small momentum electron. The strongest argument against this is that the form of the

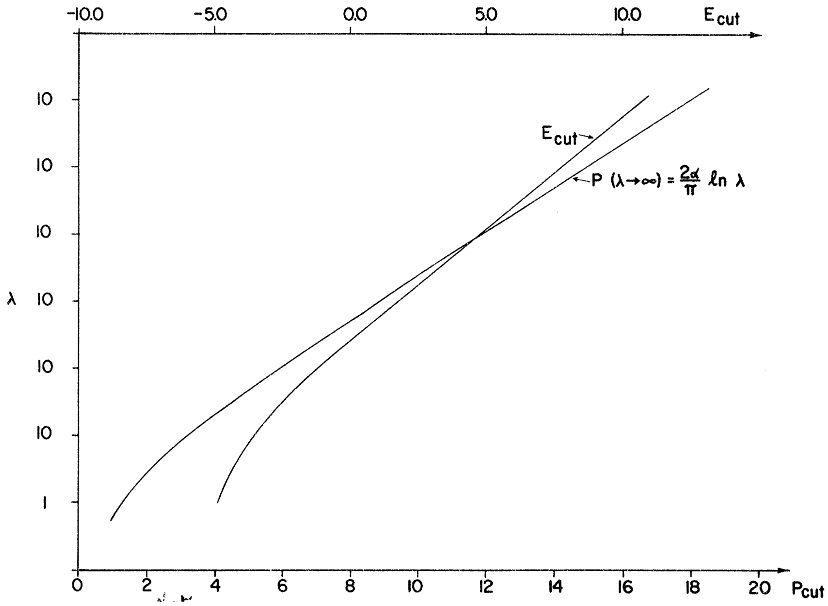

FIG. 3. Cut-off momentum and energy, P_{cut} and E_{cut}, as a function of cut-off length λ. Units are chosen so that $h = s = m = 1$.

intermediate-coupling wave function ψ_P is that of a distorted lattice around the electron, i.e., a polaron, not a free phonon.

(ii) In the case of the optical polaron, the lowest state at high P is clearly a free optical phonon with momentum P and a polaron with momentum zero. So it is natural to suggest that in the case of acoustic phonons the lowest state at high P may be composed of a polaron with $P=0$ and an acoustic phonon with momentum P. The energy of this state is $E_c(0)+P$, and is shown in Fig. 1. Note that if $E_c(0)$ is sufficiently smaller than $E(P)$, this state will be the lowest. However, it is possible to show[9] that $E_c(0) > A$ for a wide range of α, where A is the point indicated in Fig. 1. It is easy to see that for small α and large P, we have $E_c(0) + P > E(P)$. To show this we note that as $\alpha \to 0$, we expect $E_c(0) \propto -\alpha$, but $A \to -\frac{1}{2}$. Therefore $E_c(0) > A$ and the free-phonon state will be higher than ψ_P.

We conclude that $E(P)$ is an upper bound to the ground state of (1), $E_c(P)$, for each P; $E(P)$ is the energy of a polaron; $E_c(P)$ is most likely a polaron; and $\nabla_P E_c(P)$ most likely approaches the speed of sound as $P \to \infty$.

However, we will see in Sec. III that the fact that $E(P)$ continues to approach a straight line even for very large P is a property of the form of $Q(q)$ used in Eq. (2). Almost any modification of $Q(q)$ will lead to a spectrum that stops at some critical P where the slope has become 1.

III. CUT-OFF INTERACTION

If we transform the sum in Eq. (6) to an integral we can see that when $v \to 1$ the integral is dominated by the region near $q=0$. Hence the interaction with the long-wavelength phonons is responsible for the large-P behavior. This means that the interaction of the electron with distant parts of the lattice is of primary importance.

It is natural to wonder what would happen to $E(P)$ if we modified the Hamiltonian so as to cut off the interaction with the long-wavelength phonons. We have done this in two ways, replacing $Q(q)$ either by

$$Q_1^2(q) = \frac{\omega}{q^2 + 1/\lambda^2}$$

or

$$Q_2^2(q) = \frac{q^2\omega}{(q^2 + 1/\lambda^2)^2} \quad .$$

$Q_1(q)$ represents the simplest form that cuts off as $q \to 0$ and $Q_2(q)$ is the form that corresponds to the interaction that would arise from Debye screening.

We find that when $\lambda \gg 1$, both forms of Q lead to an $E^s(P)$ almost identical to $E(P)$ except that the curve stops at $P = P_{cut}$ and $P_{cut} \cong 1 + (2\alpha/\pi) \ln|4\lambda|$ for both Q's.

Hence we get an $E(P)$ which has an anomaly at the speed of sound but which is a solution for only a small part of the Brillouin zone. Numerical calculations of $E^s(P)$ are presented in Figs. 2–4.

It is interesting to note that for an electron interacting with acoustic phonons with the deformation potential, $Q(q) \propto q^{1/2}$, and one obtains an $E(P)$ curve similar to $E^s(P)$. In no case (involving acoustic phonons) can the intermediate-coupling theory give a polaron traveling faster than the speed of sound.

Although these cut-off dispersion relations are more plausible than $E(P)$ (Fig. 1) we cannot use the fact that they are an upper bound to eliminate

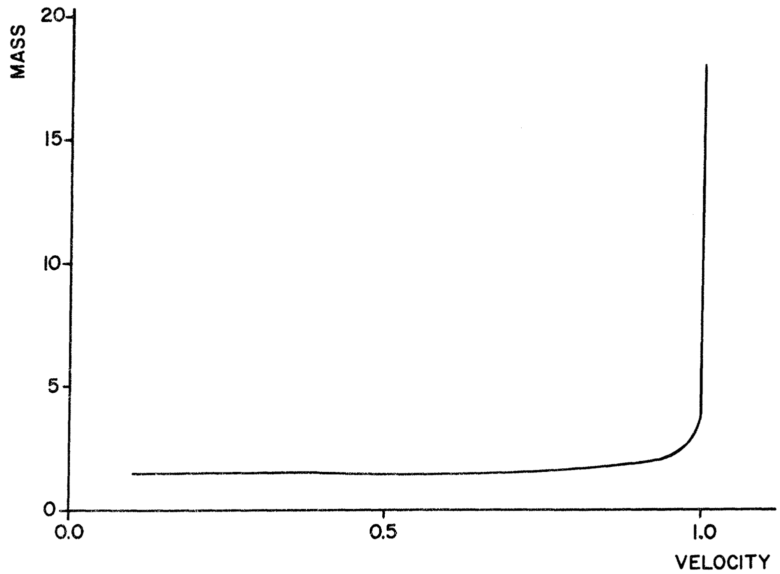

FIG. 4. Polaron effective mass as a function of velocity. Units are chosen so that $\hbar = s = m = 1$.

a quadratic-energy–versus–momentum dependence.

APPENDIX

In I and in Sec. II of this paper we use

$$Q(q) = (4\pi\alpha/V)^{1/2}\,(1/q^{1/2}) \,,$$

which leads to the energy-momentum relation $E(P)$ shown in Fig. 1. We will modify the Hamiltonian by using different forms for $Q(q)$. A way of characterizing a polaron interaction is in the strong-coupling theory where the parameters of the system enter only through the function $Q^2(q)/\omega(q)$.

For both the optical and piezoelectric polarons, where the interaction is essentially Coulombic,

$$Q^2/\omega \propto 1/q^2 \,.$$

For both the deformation-potential optical and acoustic interactions, which are short range, we have

$$Q^2/\omega \propto q^0 \,.$$

(i) A simple choice of a cut-off interaction is then

$$\frac{Q_1^2(q)}{\omega(q)} = \frac{1}{q^2 + 1/\lambda^2} \,.$$

(ii) If we take into account the electron screening of the electron-lattice interaction, we have

$$\frac{Q_2^2(q)}{\omega(q)} = \frac{q^2}{(q^2 + 1/\lambda^2)^2} \,.$$

In the limit of large λ the energy-momentum relationships obtained from these two techniques are the following:

Case 1:

$$E_L = \frac{P^2}{2} - \frac{|\vec{P}(v) - \vec{v}|^2}{2} + \frac{\alpha}{2\pi v}\left(\Lambda - \frac{\tan^{-1}\lambda q_m}{2\lambda} \ln\left|\frac{\Lambda - v}{\Lambda + v}\right| - v\ln|\Lambda^2 - v^2| - \ln\left|\frac{1-v}{1+v}\right| + v\ln|1 - v^2|\right)$$

$$+ \frac{\alpha}{2\lambda v}\left(\tfrac{1}{2}\ln\frac{1+P^2}{1+n^2} + \ln\frac{\Lambda+v}{\Lambda-v}\right) + \frac{2\alpha}{\pi}\left(\tan^{-1}P\ln P - \tan^{-1}n\ln n + \int_n^P \frac{\tan^{-1}\chi}{\chi}\,d\chi\right) ,$$

$$\vec{P} = \vec{v} + \frac{2\alpha}{\pi v^2}\left(\Lambda\ln\frac{\Lambda - v}{\Lambda + v} + \frac{P^2}{1+P^2}\ln P - \frac{n^2}{1+n^2}\ln n\right) - \frac{\alpha}{4\lambda v^2}\ln\left|\frac{1+n^2}{1+P^2}\right| + \frac{\alpha}{V}\left(\frac{n}{1+n^2} + \frac{P}{1+P^2}\right)$$

$$+ \frac{\alpha}{\pi\lambda v}\left[(\ln n)\left(\tan^{-1}n - \frac{n}{1+n^2}\right) + (\ln P)\left(\tan^{-1}P - \frac{P}{1+P^2}\right)\right] + \frac{\alpha}{2\pi\lambda v^2}\int_{2\lambda(1-v)}^{2\lambda(1+v)} \frac{\tan^{-1}\chi}{\chi}\,d\chi ,$$

where

$$\Lambda = \tfrac{1}{2}q_m + 1 \,, \quad n = 2\lambda(1 - v) \,, \quad P = 2\lambda(1 + v) \,.$$

Case 2:

$$E_L = \frac{P^2}{2} - \frac{[\vec{P}(v)-\vec{v}]^2}{2} + \frac{\alpha 2}{\pi v}\left(\Lambda \ln\frac{\Lambda - v}{\Lambda + v} - v\ln|\Lambda^2 - v^2| - \ln\left|\frac{1-v}{1+v}\right| + v\ln|1-v^2|\right)$$

$$+\frac{\alpha}{\pi}\,\frac{1}{\lambda v}\,\frac{1}{2}\left(\frac{\lambda q_m}{1+\lambda^2 q_m^2} - 3\tan^{-1}\lambda q_m\right)\ln\frac{\Lambda - v}{\Lambda + v} + \left(\frac{\alpha}{4\pi v\lambda^2} + \frac{3\alpha}{8\lambda v}\right)\left(\ln\frac{1+P^2}{1+n^2}\right) + \frac{\alpha}{4\lambda v}\left(\frac{n^2}{1+n^2} - \frac{P^2}{1+P^2}\right)$$

$$+\frac{\alpha}{\pi v\lambda}\,\frac{1}{2}\left[\ln n\left(3\tan^{-1}n - \frac{n}{1+n^2}\right) - \tan^{-1}n\right] - \frac{\alpha}{\pi v\lambda}\,\frac{1}{2}\left[\ln P\left(3\tan^{-1}P - \frac{P}{1+P^2}\right) - \tan^{-1}P\right]$$

$$+\frac{3}{2}\,\frac{\alpha}{\lambda\pi v}\int_{2\lambda(1-v)}^{2\lambda(1+v)} \frac{\tan^{-1}\chi}{\chi}\,d\chi\,,$$

with

$$P = v + \frac{2\alpha}{\pi v^2}\left[\Lambda\ln\frac{\Lambda - v}{\Lambda + v} + \frac{P^2}{1+P^2}\ln P - \frac{n^2}{1+n^2}\ln n + (1-v)\left(\frac{\ln P}{1+P^2} + \frac{\ln n}{1+n^2}\right)\right]$$

$$+\frac{\alpha}{2\pi\lambda v^2}\left[\ln n\left(3\tan^{-1}n - \frac{n}{1+n^2}\right)\right] - \frac{\alpha}{2\pi\lambda v^2}\left[\ln P\left(3\tan^{-1}P - \frac{P}{1+P^2}\right)\right]$$

$$+\frac{2\alpha}{\pi v}\left(\frac{n^2}{(n^2+1)^2}\ln n - \frac{P^2}{(P^2+1)^2}\ln P\right) + \frac{3}{2}\,\frac{\alpha}{4\lambda v^2}\ln\frac{1+P^2}{1+n^2} + \frac{\alpha}{4\lambda v^2}\left(\frac{n^2}{1+n^2} - \frac{P^2}{1+P^2}\right)$$

$$+\frac{3\alpha}{2\pi\lambda v^2}\int_{2\lambda(1-v)}^{2\lambda(1+v)} \tan^{-1}\chi\,d\chi\,,$$

where

$$\Lambda = \tfrac{1}{2}q_m + 1\,,\quad n = 2\lambda(1-v)\,,\quad P = 2\lambda(1+v)\,.$$

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