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Strong coupling theory of the moving piezoelectric polaron*

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We have applied the strong coupling polaron theory to the piezoelectric polaron. We obtain an energy-momentum relation which is quadratic at small P, and approaches a straight line with slope equal to the speed of sound at large P.

I. INTRODUCTION

It has previously been shown[1] that the intermediate coupling theory of the piezoelectric polaron leads to an anamolous energy-momentum relation. This relation $E_I(P)$ is quadratic for small P, and then approaches asymptotically a straight line with slope equal to the speed of sound. We have shown[1] that this anomaly is caused by a degeneracy in the unperturbed energy levels, and that the degeneracy should occur when the polaron velocity is equal to the speed of sound. Since the intermediate coupling theory treats the polaron velocity (rather than the energy) self-consistently, and hence locates the degeneracy in a careful way, we consider its results to be quite convincing. Moreover, the fact that the intermediate coupling theory is an upper bound to the correct ground state for each P, adds support[2] to the idea that there is an anomaly in $E(P)$. [The more usual quadratic $E(P)$ would have to eventually cross $E_I(P)$, which is only linear at large P.] It has also been shown that the intermediate coupling theory gives a polaron whose velocity is limited by the speed of sound in the following cases: for finite temperatures[3]; when the anisotropy of the crystal is included[4]; and even when the interaction is replaced by one that is screened.[5]

These anomalies in the piezoelectric problem are analogous to structure that occurs in the optical polaron near the threshold of phonon emission.[6] The same type of energy-crossing arguments that we used in the optical-polaron problem[6] give what we feel is quite strong support for the intermediate coupling results in the piezoelectric problem.[1]

A plausible physical picture of this effect is as follows: As the electron velocity approaches the speed of sound, the electron-phonon interaction increases because of the degeneracy, and hence the electron is surrounded by an increasingly large lattice deformation. As the momentum P grows, the energy of the polaron increases through the growth of the lattice deformation, rather than through the polaron velocity. Alternatively, we could say that the large lattice deformation traps the electron and prevents it from going faster than the speed of sound. At this point, however, we can see a flaw in the picture; namely, if there is a really large lattice deformation, we would expect it to react back on the electron and produce some structure in the electron wave function. Such an effect is not included in the intermediate coupling trail wave function. This type of electronic structure in response to a large lattice deformation is a characteristic of the strong coupling theory. In fact, the thought that, at large P, the polaron involves a large lattice deformation [which it must if it is to have a linear $E(P)$] suggests that in this range the strong coupling theory should be better than the intermediate coupling theory. (Although we call this theory "intermediate coupling", it is really an extension of weak coupling, not an interpolation theory.) This thought is the motivation for the present paper, in which we apply the strong coupling polaron theory to the moving piezoelectric polaron, and indeed find that we get not only a linear energy-momentum relation, but one that, in general, crosses below the intermediate coupling theory at high P. Unfortunately, although the intermediate coupling theory is an upper bound, the strong coupling theory at high P is not, so we have not proven that the strong coupling is better at high P.

As is true in the case of the optical polaron, the strong coupling theory applies for coupling constants much larger than those for common crystals. A crude way of determining where the two theories cross is by equating the perturbation-theory results to the Pekar strong coupling theory. In the optical-polaron problem, this crossover occurs at $\alpha \approx 3\pi$, and for the piezoelectric problem it is at about $\alpha = 25$ (about ten times the coupling constant in CdS). Even though the strong coupling is an academic theory, it is very helpful in clarifying some difficult points. The purpose of this paper (mentioned above) is one example. Another is associated with the question of whether or not the linear energy-momentum relation is caused by the infrared divergences in the piezoelectric polaron problem. Although we have previously shown that the anomaly survives when we cut off the interaction, the whole situation is much clearer in the strong coupling theory, where we will show that the wave-vector integral factors out from the divergent terms, and hence it is clear that no reliance is placed on the small-wave-vector part.

II. STRONG COUPLING

We start with the spherically symmetric piezoelectric polaron Hamiltonian[4,7,8]

$$H = \tfrac{1}{2}p^2 + \sum_q q a_q^\dagger a_q + \left(\frac{4\pi\alpha}{V}\right)^{1/2} \sum_q \frac{1}{\sqrt{q}} (a_q + a_{-q}^\dagger) e^{i\vec{q}\cdot\vec{r}} \ . \tag{1}$$

The coupling constant[4,7,8] is α, and V is the volume. The units of length and energy are $\hbar/ms$ and ms^2, where m is the electron band mass and s is an average[4,8] speed of sound. The electronic position and momentum are $\vec{r}$ and $\vec{p}$.

We will use a modification of a method devised by Pekar[9] for the optical polaron. The method is based on the trial state

$$|\psi\rangle = e^{i\vec{W}\cdot\vec{r}} \phi(r-R) e^{S(R)} |0\rangle \ , \tag{2}$$

where

$$S(R) = \sum_q d_q (a_q e^{i\vec{q}\cdot\vec{R}} - a_q^\dagger e^{-i\vec{q}\cdot\vec{R}}) \ . \tag{3}$$

The operator $e^{S(R)}$ produces a lattice distortion centered at the point R. The electron is then in a bound state $\phi(r-R)$ centered around the lattice distortion, and the factor $e^{i\vec{W}\cdot\vec{r}}$ sets the system in motion. Since H is invariant to translations of the electron and the lattice distortion together, the total momentum

$$\vec{\mathcal{P}} = \vec{p} + \sum_q \vec{q} a_q^\dagger a_q \tag{4}$$

is a constant of the motion. Hence the eigenstates of H can be chosen to be eigenstates of $\vec{\mathcal{P}}$. Unfortunately, $|\psi\rangle$ is not an eigenstate of $\mathcal{P}$, nor is it a simple combination of a few such eigenstates. It is possible to choose states similar to $|\psi\rangle$ that are eigenstates[10] of $\mathcal{P}$, but the subsequent variations then become very involved.

Instead, we will follow a procedure similar to, but slightly simpler than that described by Allcock.[11] We will find the $|\psi\rangle$ which minimizes $\langle\psi|H|\psi\rangle$ subject to the constraint that

$$\langle\psi|\vec{\mathcal{P}}|\psi\rangle = \vec{P} \ . \tag{5}$$

We do this with the help of the technique of Lagrange multipliers. The procedure is to minimize the functional

$$I = \langle\psi|H - \vec{v}\cdot(\vec{\mathcal{P}} - \vec{P})|\psi\rangle \ , \tag{6}$$

where v is an undetermined multiplier. The state $|\psi(P,v)\rangle$ that minimizes I is then substituted into the constraint equation (5) to determine v. Because of the way the functions are written [Eq. (2) and (12)], we are able to vary the functional so that it remains normalized; hence

$$\delta\langle\psi|\psi\rangle = 0 \ ,$$

and this need not be included as an additional constraint. Also we see that

$$\delta\langle\psi|\vec{P}\cdot\vec{v}|\psi\rangle = \vec{P}\cdot\vec{v}\,\delta\langle\psi|\psi\rangle = 0 \ ,$$

so this term can be dropped from Eq. (6).

We then have

$$\langle\psi|H - \vec{v}\cdot\vec{\mathcal{P}}|\psi\rangle = \tfrac{1}{2}W^2 + \vec{W}\cdot\int \phi^* \vec{p}\phi\, d^3r + \int \phi^* \tfrac{1}{2}p^2\phi\, d^3r - (\vec{v}\circ\vec{W}) - \vec{v}\cdot\int \phi^*\vec{p}\phi\, d^3r - \vec{v}\cdot\sum_q \vec{q} d_q^2 + \sum_q d_q^2 q + \sum_q \left(\frac{4\pi\alpha}{Vq}\right)^{1/2} d_q(\rho_q + \rho_{-q}) \ . \tag{7}$$

For the variation of the lattice part of the trial wave function we set

$$\frac{\partial I}{\partial d_q} = 0 \rightarrow d_q = -\frac{(4\pi\alpha/Vq)^{1/2}(\rho_q + \rho_{-q})}{2(q - \vec{v}\circ\vec{q})} \ , \tag{8}$$

where

$$\rho_q = \int \phi^*(r)\phi(r) e^{i\vec{q}\cdot\vec{r}} d^3r \ .$$

to minimize I with respect to $\vec{W}$ we set

$$\frac{\partial I}{\partial W_\alpha} = 0 \ ,$$

which gives

$$W_\alpha = v_\alpha - \int \phi^* P_\alpha \phi\, d^3r \ . \tag{9}$$

Here the subscripts refer to Cartesian components of the corresponding vectors.

We will leave the determination of ϕ until later. We then have

$$E(\phi, P) = \langle\psi|H|\psi\rangle = \tfrac{1}{2}p^2 - \tfrac{1}{2}(\vec{p} - \vec{v})^2 + \tfrac{1}{2}\int \phi^* p^2 \phi\, d^3r - \sum_q \frac{4\pi\alpha}{Vq} \frac{(\mathrm{Re}\rho_q)^2}{(q - \vec{v}\cdot\vec{q})} \ . \tag{10}$$

The equation of constraint which determines the Lagrange multiplier $\vec{v}(\vec{P})$, is

$$v = P - \sum_q \left(\frac{4\pi\alpha}{Vq}\right) \frac{\vec{q}(\mathrm{Re}\rho_q)^2}{[q - \vec{v}(p)\cdot\vec{q}]} \ . \tag{11}$$

By differentiating Eq. (10) with respect to the momentum P_α we can show that

$$\frac{d}{dP_\alpha}\langle\psi|H|\psi\rangle=\frac{\partial}{\partial P_\alpha}\langle\psi|H|\psi\rangle+\frac{\partial}{\partial v_\beta}\langle\psi|H|\psi\rangle\frac{\partial v_\beta}{\partial P_\alpha}=v_\alpha \quad ,$$

since, with the help of Eq. (11), we can show that $\partial\langle\psi|H|\psi\rangle/\partial v_\beta=0$. In these calculations we have taken ϕ to the independent of P. We will show in Sec. II A, when we discuss an anisotropic ϕ, which does depend on P, that v is still the polaron velocity.

Note that Eqs. (10) and (11) are very similar to the equations of the intermediate coupling theory [see Eqs. (4.1) and (4.2) of Ref. 1]. In particular, the singularity in the integral in Eq. (11) is very similar to that in the intermediate coupling theory. Both diverge as $v\to 1$ and do not exist for $v>1$. (Note that for $v>1$ these integrals do not even have a principal value, which is quite unusual for integrals encountered in electron-phonon interaction.) However, there is one interesting difference. Note that, when we change the sum on q to an integral in Eq. (11), we get

$$\frac{\alpha}{\pi}\int_0^\infty(\mathrm{Re}\rho_q)^2\,dq\int_{-1}^{1}\frac{x\,dx}{(1-vx)^2} \quad .$$

The integral on q is well behaved, and the singularity which comes in the angular integral has no special sensitivity to $q\approx 0$. Hence it is clear that the anomalous $E(p)$ in this theory has nothing to do with infrared divergences. This point was not clear in the intermediate coupling theory.

A. Spherically symmetric ϕ

First we choose ϕ to be of the form

$$\phi(r-R)=[(1/\beta)(2/\sqrt{\pi})]^{3/2}e^{-|r-R|^2/\beta^2} \quad . \tag{12}$$

Using this simple form for ϕ, Eqs. (10) and (11) become

$$E(\beta,P)=-\frac{v^2}{2}+vP+\frac{3}{2\beta^2}-\frac{\alpha}{\sqrt{\pi}\beta v}\ln\left(\frac{1+v}{1-v}\right),\quad |v|<1 \tag{13}$$

and

$$v=P-\frac{\alpha}{\sqrt{\pi}}\frac{1}{\beta v^2}\left[\ln\left(\frac{1-v}{1+v}\right)+\frac{2v}{1-v^2}\right],\quad |v|<1. \tag{14}$$

The value of β that minimizes E for each value of P is given by

$$\beta_m(v)=\frac{3\sqrt{\pi}v}{\alpha\ln[(1+v)/(1-v)]} \quad ,\quad v<1 \quad . \tag{15}$$

Note that

$$\beta_m(0)=3\sqrt{\pi}/2\alpha \tag{16}$$

is the Pekar value (in our units). We note that as $v\to 1$, $\beta\to 0$, and hence the electron wave function becomes very narrow.

Substituting the $\beta_m(v)$ into Eqs. (13) and (14) we have for the minimum energy

$$E(\beta_m(v),P)=-\frac{v^2}{2}+vP-\frac{1}{6}\frac{\alpha^2}{\pi v^2}\ln^2\left(\frac{1+v}{1-v}\right),\quad v<1 \tag{17}$$

and

$$v=P-\frac{\alpha^2}{3\pi v^3}\ln\left(\frac{1+v}{1-v}\right)\left[\ln\left(\frac{1-v}{1+v}\right)+\frac{2v}{1-v^2}\right],\quad v<1 \quad . \tag{18}$$

We again note that $dE/dP=v$; so even when ϕ is P dependent, v is the polaron velocity.

It is clear from Eq. (18) that as $v\to 1$, the momentum $P\to\infty$; so we get the same qualitative behavior as we did from the intermediate coupling theory.

For small P we get

$$E(\beta_m,P)\simeq-\frac{2\alpha^2}{3\pi}+\frac{p^2}{2[1+\frac{8}{9}(\alpha^2/\pi)]} \quad . \tag{19}$$

As we expect, the self-energy is the same value (in our units) that Pekar obtained for the optical polaron. This is because the optical and piezoelectric problems are identical[12] to the lowest order in $1/\alpha$. The effective mass

$$m^*=1+\tfrac{8}{9}(\alpha^2/\pi) \tag{20}$$

is proportional to α^2 rather than to α^4, which is appropriate for the optical problem. The numerical factors in m^* differ from those obtained by different strong coupling theories.[9,12]

In Fig. 1 we plot $E(\beta_m,P)$. We see that this curve crosses below the intermediate coupling results $E_I(P)$, thus confirming the heuristic argument in Sec. I which concluded that for large P the strong coupling theory would tend to be favored.

B. Asymmetric ϕ

Since it seems reasonable that, as $v\to 1$, the electron wave function may behave differently in the direction of motion than it does in the direction perpendicular to the motion, we have tried a wave function which leads to

$$\rho_k=e^{-(k_z/2q)-(k_\perp/2p)} \quad . \tag{21}$$

We define an asymmetric parameter ϵ by

$$p^2=q^2(1+\epsilon) \quad ; \tag{22}$$

then we get

$$E=\frac{P^2}{2}-\frac{(P-v)^2}{2}+\frac{q^2}{4}+\frac{q^2(1+\epsilon)}{2}-\frac{\alpha q(1+\epsilon)^{1/2}}{(2\pi)^{1/2}(\epsilon+v^2)^{1/2}}\ln A \tag{23}$$

and

$$v=P+\frac{\alpha q(1+\epsilon)^{1/2}}{(2q)^{1/2}(\epsilon+v^2)}\left(\frac{v}{(\epsilon+v^2)^{1/2}}\ln A-\frac{2v(1+\epsilon)^{1/2}}{1-v^2}\right) \quad . \tag{24}$$

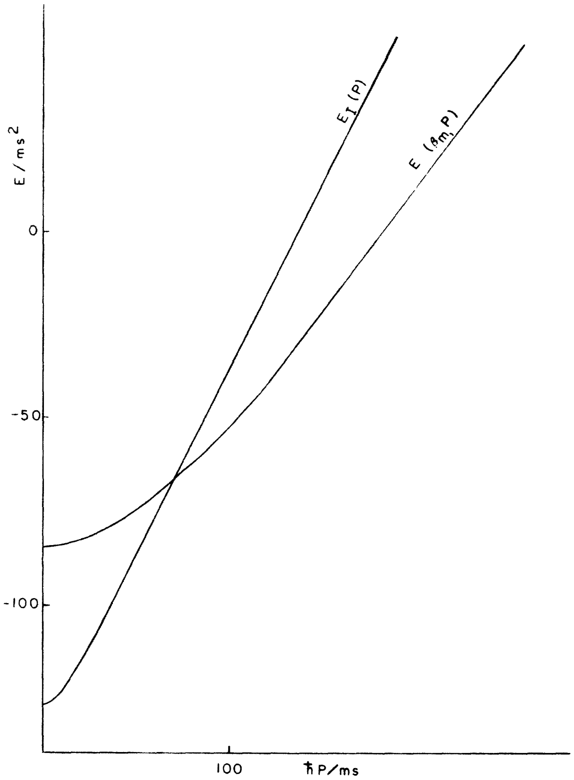

FIG. 1. Intermediate-coupling-theory $E_I(P)$ compared to strong-coupling $E(\beta_m, P)$. At higher P, $e(\beta_m, P)$ also approached a slope equal to the speed of sound. (m is the electron band mass and s is an average speed of sound.)

In Eqs. (23) and (24)

$$A = \frac{1+v}{1-v} \frac{v+\epsilon+[(1+\epsilon)(v^2+\epsilon)]^{1/2}}{v-\epsilon+[(1+\epsilon)(v^2+\epsilon)]^{1/2}} \quad . \tag{25}$$

We note in Eq. (24) that as $v \to 1$ the term with $(1-v^2)^{-1}$ dominates, and hence $P - v \to \infty$ like $(1-v^2)^{-1}$. Hence in Eq. (23), as $v \to 1$, the $(P-v)^2$ also dominates. We also note that as $v \to 1$ this dominant term in $(P-v)$ becomes independent of ϵ. Hence, since we expect the asymmetry in the wave function to occur only when $v \to 1$, the fact that in this region the dominant term in the energy is independent of ϵ indicates that the asymmetric wave function will give very little improvement. This conclusion was borne out by direct numerical calculations, when we found only a negligible reduction of the energy.

III. CONCLUSIONS

The main conclusion of this paper is that the strong coupling theory gives an energy-momentum relation in which the slope approaches the speed of sound asymptotically, thus lending support to the earlier suggestion that this is a real physical effect. It should be noted that the present version of the strong coupling theory gives a linear energy-momentum relation for the optical polaron as well. We feel that this theory is clearly inappropriate for the fast optical polaron because it treats the degeneracy in the theory in a completely wrong way. The same situation was pointed out for the intermediate coupling theory.[1]

It is also interesting to note that there is a range of α in which the strong coupling theory is higher than the intermediate coupling theory for small P, but crosses below it at high P. This supports the notion that as $v \to 1$ a large lattice deformation around the electron results, which tends to make the strong coupling theory more appropriate. However, we should note that although the intermediate coupling theory is an upper bound for each value of P, the present strong coupling theory is not. However, the fact that it is lower than the upper bound indicates that it could be a better theory. Moreover, the strong coupling theory still gives some argument against an $E(P)$ that increases as fast as P^2 at high P. For instance, assume that the correct ground state of H is $|\psi_c(P)\rangle$, which we suppose to have

$$H|\psi_c(P)\rangle = [E(0) + P^2/2m]|\psi_c(P)\rangle \tag{26}$$

and

$$\mathcal{P}|\psi_c(P)\rangle = P|\psi_c(P)\rangle \quad . \tag{27}$$

Let us further assume that our trial state can be written

$$|\psi\rangle = \sum_p A_p |\psi_c(P)\rangle \quad , \tag{28}$$

where we assume that we need not include polaron excited states of free-phonon states. We have shown that

$$\langle\psi|H|\psi\rangle = (\text{const}) + P \tag{29}$$

for large P. But

$$\langle\psi|H|\psi\rangle = \sum_k A_k^2 [E(0) + k^2/2m] \tag{30}$$

and

$$\langle\psi|\mathcal{P}|\psi\rangle = P = \sum_k A_k^2 k \quad , \tag{31}$$

and the fact that $|\psi\rangle$ is normalized gives

$$\sum_k A_k^2 = 1 \quad . \tag{32}$$

If we note that

$$0 < \sum_k A_k^2 (k-P)^2 \quad , \tag{33}$$

this implies that

$$\sum A_k^2 k^2 > P^2 \ , \quad (34)$$

which contradicts Eq. (29) for large P. This means that either Eq. (28) is incorrect, or the ground-state energy cannot be proportional to P^2 at high P. Although the assumption of Eq. (28) is not proven, we think it is very plausible.

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