Linear energy-momentum relations for acoustic deformation-potential polarons

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Several variational theories for the energy-momentum (*EP*) relation of the acoustic deformation-potential (ADP) polaron are considered. We show that the trial states of these theories need not be translationally invariant to yield upper bounds to the actual *EP* relation. A theory which interpolates between weak- and strong-coupling theories is thus shown to give an asymptotically linear upper bound to the ADP polaron *EP* relation, as predicted by energy-crossing arguments.

I. INTRODUCTION

The nature of the energy-momentum relation for the acoustic deformation-potential (ADP) polaron has been the subject of several recent papers.[1-3] It has been pointed out that the onset of degeneracy between the ADP polaron and the acoustic phonon states occurs when the polaron velocity reaches the sound velocity.[3] Thus, on the basis of energy level splitting arguments, one might expect the energy-momentum relations of acoustic-mode polarons to approach linearity, as their velocity approaches the sound velocity. Such behavior is found in many different approaches to the piezoelectric polaron.[4-6]

In the ADP-polaron problem, the results obtained have not been as straightforward, due largely to the short-range character of the deformation potential interaction. We show in Sec. II that the classical theory does give the predicted energy resonance as the velocity approaches the sound velocity. The quantum theories are discussed in Sec. III, and substantial differences between the energy-momentum relations are found. Perturbation theories are complicated by the nature of the degeneracies encountered and produce questionable results. As a result, the approaches to the problem have been primarily variational, with the assumption that, at a given momentum, the state of lowest energy represents the polaron. A trial state which is an eigenstate of momentum $\vec{P}$ is varied to minimize the expected value of energy, which then represents an upper bound to the lowest energy at that momentum. As shown in Sec. III A, a trial state which is not a momentum eigenstate but has an expected value of momentum $\vec{P}$ is also an upper bound to the lowest energy at the momentum $\vec{P}$.

The variational theory of Lee, Low, and Pines,[7] appropriate to weak electron-lattice coupling, is not found to have a linear energy-momentum relation but only yields solutions for polaron velocities less than the sound velocity. The strong-coupling theory of Pekar[8] does yield a linear *EP* upper bound, although unphysically large lattice dilations and a high degree of localization seriously question the relevance of the results. A theory of Buimistrov and Pekar[9] which interpolates between the Lee-Low-Pines and Pekar states is shown to produce a linear *EP* upper bound and not to involve the questionable aspects of the Pekar state above.

II. CLASSICAL THEORY

The classical behavior of the polaron is given by the classical analog of the usual Fröhlich Hamiltonian[10]

$$H=\frac{\vec{p}^{\,2}}{2m}+\tfrac{1}{2}\sum_{\vec{q}}\left(P_{\vec{q}}P_{-\vec{q}}+\omega_{\vec{q}}^{2}Q_{\vec{q}}Q_{-\vec{q}}\right)+\sum_{\vec{q}}V_{\vec{q}}Q_{\vec{q}}e^{i\vec{q}\cdot\vec{r}}. \tag{1}$$

Here $\vec{r}$ and $\vec{p}$ are the electronic coordinate and momentum, respectively, $Q_{\vec{q}}$ and $P_{\vec{q}}$ are the lattice normal coordinate and conjugate momentum for the longitudinal acoustic (LA) modes, respectively, $\omega_{\vec{q}}$ is the LA dispersion relation, and $V_{\vec{q}}$ is the deformation-potential interaction:

$$V_{\vec{q}}=[(D^2/V\rho s)\omega_{\vec{q}}|\vec{q}|]^{1/2}, \tag{2}$$

where D is the deformation potential, ρ is the lattice mass density, V is the crystal volume, and s is the LA sound velocity.

Applying Hamilton's equations;

$$m\ddot{\vec{r}}=-i\sum_{\vec{q}}\vec{q}V_{\vec{q}}Q_{\vec{q}}e^{i\vec{q}\cdot\vec{r}},\quad \vec{p}=m\dot{\vec{r}},$$

$$\ddot{Q}_{\vec{q}}=-\omega_{\vec{q}}^{2}Q_{\vec{q}}-V_{-\vec{q}}e^{-i\vec{q}\cdot\vec{r}},\quad P_{\vec{q}}=\dot{Q}_{-\vec{q}}.$$

A uniformly translating solution is

$$\vec{r}=\vec{r}_0+\vec{v}t,\quad \vec{p}=m\vec{v},$$

$$Q_{\vec{q}}(t)=Q_{\vec{q}}(0)e^{i\vec{q}\cdot\vec{v}t},\quad P_{\vec{q}}(t)=-i\vec{q}\cdot\vec{v}\,Q_{-\vec{q}}(t),$$

for which

$$Q_{\vec{q}}(0)=-V_{-\vec{q}}[\omega_{\vec{q}}^{2}-(\vec{q}\cdot\vec{v})^2]^{-1}. \tag{3}$$

Using $\omega_{\vec{q}} \cong s|\vec{q}|$ gives

$$Q_{\vec{q}}(0) = -(8\pi\alpha/V)^{1/2} s^{1/2} |\vec{q}| [s^2 q^2 - (\vec{q}\cdot\vec{v})^2]^{-1} \quad (4)$$

and we notice that the amplitude of the lattice modes increases indefinitely as the velocity $|\vec{v}|$ approaches s.

The polaron energy is

$$E = \frac{p^2}{2m} + \tfrac{1}{2}\sum_{\vec{q}} V_{\vec{q}}^2 [\omega_{\vec{q}} - (\vec{q}\cdot\vec{v})]^{-2} - \sum_{\vec{q}} v_{\vec{q}}^2 [\omega_{\vec{q}}^2 - (\vec{q}\cdot\vec{v})^2]^{-1} \quad (5)$$

which gives

$$E = \frac{p^2}{2m} + \frac{2\alpha s}{3\pi}\kappa^3 \left((s^2 - v^2)^{-1} - (vs)^{-1} \ln\left|\frac{s+v}{s-v}\right| \right). \quad (6)$$

The κ is the wave-number cutoff which arises from the usual substitution,

$$\sum_{\vec{q}} \rightarrow (V/8\pi^3)\int_0^{\kappa} d^3q .$$

This solution of the classical equation of motion has the behavior predicted from the energy-crossing arguments, where the polaron energy is unbounded as the polaron velocity approaches the speed of sound in the lattice. This is due to the fact that a lattice displacement moving at the sound velocity is a stable configuration, even in the absence of an attractive interaction with an electron. The presence of such an interaction results in a resonance for electrons approaching the sound velocity s. Although the resonance will be affected by anharmonic lattice terms which limit the lattice dilation (proportional to $\ln|v-s|$), the lattice energy grows much more quickly [proportional to $(s^2-v^2)^{-1}$] and can become quite large before the anharmonic limit obtains. We show in Sec. III C that this classical electron-lattice resonance is just that seen in the quantum strong-coupling theory of Pekar.

III. QUANTUM THEORY

The ADP polaron problem is formulated in terms of the Fröhlich Hamiltonian, which in dimensionless units is

$$\hat{H} = \tfrac{1}{2}\hat{\vec{p}}^2 + \sum_{\vec{q}} \omega_{\vec{q}} \hat{a}_{\vec{q}}^{\dagger}\hat{a}_{\vec{q}} + \sum_{\vec{q}} Q_{\vec{q}} (\hat{a}_{-\vec{q}}^{\dagger} + \hat{a}_{\vec{q}}) e^{i\vec{q}\cdot\vec{r}} . \quad (7)$$

Here $\vec{r}$ and $\hat{\vec{p}} = -i\nabla$ are the electron coordinate and momentum, respectively, $\hat{a}_{\vec{q}}^{\dagger}$ and $\hat{a}_{\vec{q}}$ are the LA phonon creation and destruction operators, respectively, and $\omega_{\vec{q}}$ is the LA dispersion relation. The deformation-potential interaction term is

$$Q_{\vec{q}} = [(4\pi\alpha/V)|\vec{q}|]^{1/2} \quad (8)$$

and the dimensionless coupling constant α is

$$\alpha = D^2 m^2/8\pi\hbar^3\rho s . \quad (9)$$

The m is the band effective mass of the electron and the other parameters are as given in the classical theory. We use ms^2 as the unit of energy and $\hbar/ms$ as the unit of length (essentially taking $\hbar = m = s = 1$).

In using the Fröhlich-Hamiltonian formalism, we assume that all wave vectors are much less than the lattice cutoff κ in order to apply an effective-mass theory for the electron, and that the lattice dilation is much less than unity, so that anharmonic lattice effects are avoided.

In this theory, the Hamiltonian is translationally invariant, so the polaron states are chosen to be simultaneous eigenstates of total momentum

$$\hat{\vec{P}} = \hat{\vec{p}} + \sum_{\vec{q}} \vec{q}\,\hat{a}_{\vec{q}}^{\dagger}\hat{a}_{\vec{q}} . \quad (10)$$

We want to find the ground-state energy of $\hat{H}$ as a function of the eigenvalues of $\hat{\vec{P}}$. In keeping with the assumptions above, we shall only be concerned with momenta $\vec{P}$ which are much less than κ.

A. Upper-bound theorem

We consider several variational approaches to the ADP problem, assuming that for a given total momenta, the state of lowest energy will represent the polaron. This is commonly done by assuming a trial state for the polaron which is a momentum eigenstate and then varying the expected value of energy. The result represents an upper bound to the actual ground state at that momentum. This is the procedure followed by Lee, Low, and Pines for weak coupling and done here in Sec. III B. The same procedure may be followed for the adiabatic strong-coupling theory (Sec. III C) by using a modified trial state due to the work of Höhler.[11] However, the resulting analysis is invariably difficult,[12] and normally a simpler procedure is used. One assumes a trial state appropriate for the moving Pekar theory which is not a momentum eigenstate and then minimizes the expected value of energy with the constraint that the expected value of momentum be fixed. We thus find the minimal polaron energy as a function of expected momenta. It is not immediately clear that such an EP relation represents an upper bound to the actual values and this has been the basis for skepticism concerning the results of this procedure. We shall show that, under reasonable assumptions, such EP relations do represent upper bounds to the actual lowest energy of the Hamiltonian, evaluated at the expected momentum of the trial state.

Consider simultaneous eigenstates of $\hat{H}$ and $\hat{\vec{P}}$:

$$\hat{H}|\vec{P};\xi\rangle = E_{\xi}(\vec{P})|\vec{P};\xi\rangle$$

and

$$\hat{\vec{P}}\,|\vec{P};\zeta\rangle = \vec{P}\,|\vec{P};\zeta\rangle,$$

where the index ζ symbolizes any remaining quantum numbers in the system and gives bands of energy denoted by E_ζ. We will take $\zeta = 0$ to give the ground state of the polaron, $E_\zeta(\vec{P}) \geq E_0(\vec{P})$, and find an upper bound to $E_0(\vec{P})$.

Define $\hat{K}_{\vec{\lambda}} \equiv \hat{H} - \vec{\lambda}\cdot\hat{\vec{P}}$, for which

$$\hat{K}_{\vec{\lambda}}\,|\vec{P};\zeta\rangle = K_{\vec{\lambda};\zeta}(\vec{P})\,|\vec{P};\zeta\rangle$$

and

$$K_{\vec{\lambda};\zeta}(\vec{P}) = E_\zeta(\vec{P}) - \vec{\lambda}\cdot\vec{P}.$$

For some arbitrary $\vec{\lambda}$, we find the minimal value of $K_{\vec{\lambda};\zeta}(\vec{P})$ by

$$\nabla_{\vec{P}} K_{\vec{\lambda};\zeta}(\vec{P})\,|_{\vec{\lambda},\zeta} = \nabla_{\vec{P}} E_\zeta(\vec{P})\,|_\zeta - \vec{\lambda} = 0$$

and note that the quantity $\nabla_{\vec{P}} E_\zeta(\vec{P})\,|_\zeta \equiv \vec{v}_\zeta(\vec{P})$ is the polaron velocity. To find minima for reasonable $E_\zeta(P)$ we require

$$\nabla^2_{\vec{P}} K_{\vec{\lambda};\zeta}(\vec{P})\,|_{\vec{\lambda},\zeta} = \nabla_P\cdot\vec{v}_\zeta(\vec{P})\,|_\zeta \equiv 1/m_{eff} > 0.$$

Since the $\hat{\vec{P}}$ is Hermitian, we take the $|\vec{P};\zeta\rangle$ to form a complete orthonormal basis and can write an arbitrary trial state as

$$|\psi\rangle = \sum_{\vec{P},\zeta} c_\zeta(\vec{P})\,|\vec{P};\zeta\rangle, \tag{11}$$

with normalization

$$\langle\psi\,|\,\psi\rangle = \sum_{\vec{P},\zeta} |c_\zeta(\vec{P})|^2 = 1$$

and expected momentum

$$\langle\hat{\vec{P}}\rangle \equiv \sum_{\vec{P},\zeta} \vec{P}\,|c_\zeta(\vec{P})|^2.$$

Consider the quantity

$$\langle\psi\,|\,\hat{K}_{\vec{\lambda}}\,|\,\psi\rangle = \sum_{\vec{P},\zeta} K_{\vec{\lambda};\zeta}(\vec{P})\,|c_\zeta(\vec{P})|^2.$$

By the definition of $\zeta = 0$, $K_{\vec{\lambda};0} \leq K_{\vec{\lambda};\zeta}$, so that

$$\langle\psi\,|\,\hat{K}_{\vec{\lambda}}\,|\,\psi\rangle \geq \sum_{\vec{P},\zeta} K_{\vec{\lambda};0}(\vec{P})\,|c_\zeta(\vec{P})|^2.$$

The minimal value of $K_{\vec{\lambda};0}(\vec{P})$ is given by $\vec{P} = \vec{P}_0(\vec{\lambda})$, where $\vec{V}_0(\vec{P}_0(\vec{\lambda})) = \vec{\lambda}$:

$$\langle\psi\,|\,\hat{K}_{\vec{\lambda}}\,|\,\psi\rangle \geq K_{\vec{\lambda};0}(\vec{P}_0(\vec{\lambda})).$$

In the case that $\vec{P}_0(\vec{\lambda})$ is multiple valued, we must pick that value which gives the smallest $K_{\vec{\lambda};0}(\vec{P}_0(\vec{\lambda}))$ where $c_\zeta(\vec{P}_0(\vec{\lambda}))$ is not vanishing.

Using the definition of $K_{\vec{\lambda}}$, we rewrite the inequality as

$$\langle\psi\,|\,\hat{H}\,|\,\psi\rangle \geq E_0(\vec{P}_0(\vec{\lambda})) - \vec{\lambda}\cdot[\vec{P}_0(\vec{\lambda}) - \langle\hat{\vec{P}}\rangle].$$

If $\langle\hat{\vec{P}}\rangle$ is in the range of $\vec{P}_0$ over its domain $\vec{\lambda}$, we may choose $\vec{\lambda}$ such that $\vec{P}_0(\vec{\lambda}) = \langle\hat{\vec{P}}\rangle$ and have

$$\langle\psi\,|\,\hat{H}\,|\,\psi\rangle \geq E_0(\langle\hat{\vec{P}}\rangle), \tag{12}$$

which is the desired upper bound.

We have found an upper bound to the actual ground-state energy for all momenta in the range of the function $\vec{P}_0(\vec{\lambda})$. Assume that $m^{-1}_{eff} \equiv \nabla^2_{\vec{P}} E_0(\vec{P}) > 0$ for all $\vec{P}$ where the $c_\zeta(\vec{P})$ are significant. Presumably $c_\zeta(\vec{P})$ will be peaked around $\langle\hat{\vec{P}}\rangle$, which is always much less than κ, so we want $m^{-1}_{eff} > 0$ for $|\vec{P}| \ll \kappa$. In this region we may invert $v_0(\vec{P})$ to find that $\vec{P}_0(\vec{\lambda})$ can take on those values of $|\vec{P}| \ll \kappa$. This assumption about the actual ground-state energy $E_0(\vec{P})$ is reasonable in that we consider $\vec{P}$ only near the band edge and have no reason to expect energy bendovers in this region.

B. Weak-coupling theory

In the regime of weak coupling, the polaron energy-momentum relation is usually computed by a suitable perturbation series or by a variational method due to Lee, Low, and Pines.[7,13] Use of ordinary perturbation theory is questionable, as it ignores the degeneracy due to phonon emission at polaron velocities greater than the lattice sound velocity. As discussed in relation to the piezopolaron,[4] degenerate perturbation methods (such as the Tamm-Dancoff theory) for acoustic polarons account for this degeneracy, but place its onset at the wrong energy. Owing to this problem, we shall do weak coupling by variational methods. Lee, Low, and Pines use a trial state similar to

$$|\psi_{\vec{P}}\rangle = e^{i\vec{P}\cdot\vec{r}} e^{S(\vec{r})}\,|0\rangle, \tag{13}$$

where

$$S(\vec{r}) = \sum_{\vec{q}} (f_{\vec{q}}\, e^{i\vec{q}\cdot\vec{r}}\hat{a}_{\vec{q}} - f^*_{\vec{q}}\, e^{-i\vec{q}\cdot\vec{r}}\hat{a}^\dagger_{\vec{q}})$$

generates a lattice deformation about the electron position and $|0\rangle$ represents the phonon vacuum. Minimizing the energy $\langle\psi_{\vec{P}}\,|\,\hat{H}\,|\,\psi_{\vec{P}}\rangle$ by varying $f_{\vec{q}}$ gives

$$f_{\vec{q}} = Q_{\vec{q}}\,(\tfrac{1}{2}q^2 + \omega_{\vec{q}} - \vec{q}\cdot\vec{v})^{-1},$$

where $\vec{v} = \vec{P} + \sum_{\vec{q}} \vec{q}\,|f_{\vec{q}}|^2$. The energy and momentum in terms of the velocity $\vec{v}$, are

$$\langle E\rangle = \vec{P}\cdot\vec{v} - \tfrac{1}{2}v^2 - \sum_{\vec{q}} Q^2_{\vec{q}}\,(\tfrac{1}{2}q^2 + \omega_{\vec{q}} - \vec{v}\cdot\vec{q})^{-1} \tag{14}$$

and

$$\vec{P} = \vec{v} + \sum_{\vec{q}} \vec{q}\,Q^2_{\vec{q}}\,(\tfrac{1}{2}q^2 + \omega_{\vec{q}} - \vec{v}\cdot\vec{q})^{-2}. \tag{15}$$

Using $\omega_{\vec{q}} = |\vec{q}|$ and $Q^2_{\vec{q}} = (4\pi\alpha/v)\,|\vec{q}|$, we find (for $v \leq 1$)

$$\langle E\rangle = \vec{P}\cdot\vec{v} - \tfrac{1}{2}v^2 + \frac{8\alpha}{3\pi v}\left((1+3v^2+\kappa^3/8)\ln\left|\frac{\kappa/2+1-v}{\kappa/2+1+v}\right| + (1+3v^2)\ln\left|\frac{1+v}{1-v}\right| - v(v^2+3)\ln\left|\frac{(\kappa/2+1)^2-v^2}{1-v^2}\right| - \kappa v(\kappa/4-2)\right) \tag{16}$$

and

$$\vec{P}=\vec{v}\left[1+\frac{\alpha}{3\pi^2 v^3}\left((1-3v^2+\kappa^3/8)\ln\left|\frac{\kappa/2+1-v}{\kappa/2+1+v}\right|+(1-3v^2)\ln\left|\frac{1+v}{1-v}\right|+2v^3\ln\left|\frac{(\kappa/2+1)^2-v^2}{1-v^2}\right|+\kappa v(\kappa/2-1)\right)\right]. \quad (17)$$

Examination of these solutions shows that as the polaron velocity approaches the sound velocity ($v\to 1$ in our units), the energy-momentum relation does not become linear but rather approaches a critical value (momentum P_c on Fig. 1), beyond which no solutions exist.[2] This is due to the fact that Eq. (15) does not have a Cauchy principal value when $v>1$ but has a finite value when $v\to 1$ for $v<1$. This is unlike perturbation theories where the onset of singular integrands usually allow one to generalize the energy off the real axis and find the principal value plus an imaginary part, corresponding to a quasiparticle with a lifetime. The meaning of the behavior here is not clear.

C. Strong-coupled theory

In the strong-coupling regime, one assumes that the electron responds much faster than the lattice (adiabatic approximation), so the lattice sees only the average position of the electron.[8] In this adiabatic limit, the quantum fluctuations of the electron and lattice are neglected and the resulting theory is similar to the classical theory. We assume a moving Pekar state:

$$|\psi_{\vec{v}}\rangle=\phi_{\vec{v}}(\vec{r},\vec{R})e^{\hat{S}(\vec{R})}|0\rangle, \quad (18)$$

where

$$\hat{S}(\vec{R})=\sum_{\vec{q}}(d_{\vec{q}}e^{i\vec{q}\cdot\vec{R}}\hat{a}_{\vec{q}}-d_{\vec{q}}^{*}e^{-i\vec{q}\cdot\vec{R}}\hat{a}_{\vec{q}}^{\dagger})$$

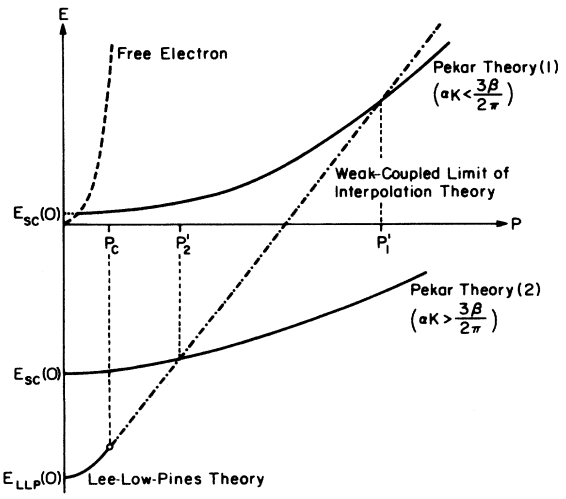

FIG. 1. Energy-momentum relations for variational theories of the ADP polaron (not to scale).

generates a lattice deformation about $\vec{R}$, the average position of the electron. The electron wave function is $\phi_{\vec{v}}(\vec{r},\vec{R})=e^{i\vec{v}\cdot\vec{r}}\psi(\vec{r}-\vec{R})$, which gives a nonvanishing expected momentum corresponding to a polaron velocity $\vec{v}$. We choose ψ so that $\psi^*=\psi$ and $\int d^3r|\psi|^2=1$. As discussed in Sec. III A, we minimize the expected value of energy with the constraint that the expected momentum is fixed. This is simply done by minimizing $\langle\psi_{\vec{v}}|K_{\vec{\lambda}}|\psi_{\vec{v}}\rangle$ for fixed $\vec{\lambda}$ while varying $d_{\vec{q}}$ and $\vec{v}$.
We find

$$d_{\vec{q}}=Q_{\vec{q}}\rho_{\vec{q}}(\omega_{\vec{q}}-\vec{\lambda}\cdot\vec{q})^{-1}$$

and

$$\vec{v}=\vec{\lambda},$$

where

$$\rho_{\vec{q}}=\int d^3r\, e^{i\vec{q}\cdot\vec{r}}|\psi(\vec{r})|^2.$$

In terms of the velocity $\vec{v}$, the results are

$$\langle E\rangle=\tau+\langle\vec{P}\rangle\cdot\vec{v}-\tfrac{1}{2}v^2-\sum_{\vec{q}}Q_{\vec{q}}^2\rho_{\vec{q}}^2(\omega_{\vec{q}}-\vec{v}\cdot\vec{q})^{-1} \quad (19)$$

and

$$\langle\vec{P}\rangle=\vec{v}+\sum_{\vec{q}}\vec{q}Q_{\vec{q}}^2\rho_{\vec{q}}^2(\omega_{\vec{q}}-\vec{v}\cdot\vec{q})^{-2}, \quad (20)$$

where

$$\tau=-\tfrac{1}{2}\int d^3r\psi^*\nabla^2\psi.$$

Although $|\psi_{\vec{v}}\rangle$ is not a momentum eigenstate, the upper-bound theorem in Sec. III A implies that $\langle E\rangle$ as a function of $\langle\vec{P}\rangle$ represents an upper bound to the exact solution $E_0(\vec{P})$ if it is well behaved (in the sense that $m_{\text{eff}}^{-1}>0$).

Evaluation of Eqs. (19) and (20) gives

$$\langle E\rangle=\tfrac{1}{2}v^2+\beta R^{-2}+\frac{2\alpha}{3\pi}\kappa^3(1-\gamma\kappa^2R^2\cdots)\left((1-v^2)^{-1}-v^{-1}\ln\left|\frac{1+v}{1-v}\right|\right), \quad (21)$$

and

$$\langle\vec{P}\rangle=\vec{v}\left[1+\frac{\alpha}{3\pi v^2}\kappa^3(1-\gamma\kappa^2R^2\cdots)\times\left(2(1-v^2)^{-1}-v^{-1}\ln\left|\frac{1+v}{1-v}\right|\right)\right]. \quad (22)$$

Here β and γ are numerical constants[14] which depend on the exact form of $\psi(\vec{r})$, R is a measure

of the width of ψ, and κ is the wave-vector cutoff from before. When $\alpha\kappa \gtrsim 3\beta/2\pi$, the stationary self-energy [$E_{sc}(0)$ on Fig. 1] will be negative and a bound state exists for all $\vec{P}$. Even if the self-energy is positive (and no bound state exists) at zero momentum, at some critical velocity, the Pekar energy-momentum relation will pass below the free-electron curve and bound states will occur. The value of R which gives ground states of minimum energy approaches a value somewhat less than the lattice spacing π/κ and represents a highly localized state.

Comparison of the Pekar energy [Eq. (21)] and the classical energy [Eq. (7)], keeping in mind the dimensionless units of the quantum theory, shows very close correspondence, as mentioned earlier. The curve of $\langle E\rangle$ vs $\langle\vec{P}\rangle$ has the predicted linear behavior in the asymptotic region $v \to 1$:

$$\langle E\rangle \sim |\langle\vec{P}\rangle| + \frac{\alpha\kappa^3}{3\pi}\ln\left|\frac{\alpha\kappa^3}{3\pi}|\langle\vec{P}\rangle|^{-1}\right| + \cdots . \tag{23}$$

However, for reasonable values of the parameters ($\alpha \sim 10^{-2}$, $\kappa \sim 300$), this is a region of large $\langle\vec{P}\rangle$. The behavior of the Pekar theory for meaningful momenta is nearly quadratic, with an extremely large effective mass:

$$m_{eff} \cong 1 + (2\alpha/9\pi)\kappa^3. \tag{24}$$

We show in Sec. IIIE that the large mass is due to an unphysically large lattice dilation and related to the highly localized nature of the Pekar state for this polaron. This extreme localization is more appropriate to the theory of the small polaron and represents a violation of one of the basic assumptions involved in the Fröhlich formulation of the large polaron.

D. Interpolation theory

We consider a variational theory similar to Buiminstrov and Pekar[9] which combines features of the Lee, Low, and Pines trial state [Eq. (13)] and the Pekar trial state [Eq. (18)] in order to find an energy-momentum relation which interpolates smoothly from weak to strong coupling for increasing momentum. Our trial state is:

$$|\psi_{\vec{v}}\rangle = e^{i\vec{v}\cdot\vec{r}}\psi(\vec{r}-\vec{R})e^{\hat{S}_1(\vec{R})}e^{\hat{S}_2(\vec{r})}|0\rangle, \tag{25}$$

where

$$\hat{S}_1(\vec{R}) = \sum_{\vec{q}}\left(d_{\vec{q}}e^{i\vec{q}\cdot\vec{R}}\hat{a}_{\vec{q}} - d^*_{\vec{q}}e^{-i\vec{q}\cdot\vec{R}}\hat{a}^\dagger_{\vec{q}}\right),$$

$$\hat{S}_2(\vec{r}) = \sum_{\vec{q}}\left(f_{\vec{q}}e^{i\vec{q}\cdot\vec{r}}\hat{a}_{\vec{q}} - f^*_{\vec{q}}e^{-i\vec{q}\cdot\vec{r}}\hat{a}^\dagger_{\vec{q}}\right),$$

and $\vec{v}$ will be the polaron velocity. As in strong coupling, we minimize $\langle\psi_{\vec{v}}|\hat{K}_{\vec{\lambda}}|\psi_{\vec{v}}\rangle$ by varying $d_{\vec{q}}$, $f_{\vec{q}}$, and $\vec{v}$ and find

$$d_{\vec{q}} = Q_{\vec{q}}(s_{\vec{q}} - w_{\vec{q}})\rho_{\vec{q}}[s_{\vec{q}}(s_{\vec{q}}|\rho_{\vec{q}}|^2 - w_{\vec{q}})]^{-1},$$

$$f_{\vec{q}} = Q_{\vec{q}}(1 - |\rho_{\vec{q}}|^2)(w_{\vec{q}} - s_{\vec{q}}|\rho_{\vec{q}}|^2)^{-1},$$

and

$$\vec{v} = \vec{\lambda},$$

where $s_{\vec{q}} = \omega_{\vec{q}} - \vec{\lambda}\cdot\vec{q}$, $w_{\vec{q}} = s_{\vec{q}} + \frac{1}{2}q^2$, and $\rho_{\vec{q}} = \int d^3r\, e^{i\vec{q}\cdot\vec{r}}|\psi|^2$. Note that where $\rho_{\vec{q}}$ is small, the lattice distortion approaches the weak-coupling (Lee-Low-Pines) form, and when $\rho_{\vec{q}} \to 1$, the strong-coupling (Pekar) form is obtained. This implies that the electron tends to couple adiabatically to the long-wavelength components of the lattice deformation and more rigidly to the short-wavelength (high-frequency) components, so this theory should be an improvement on the simple adiabatic approximation of the Pekar theory or the rigid coupling of the Lee, Low, and Pines method.

The expected energy and momentum are

$$\langle E\rangle = \tau + \langle\vec{P}\rangle\cdot\vec{v} - \tfrac{1}{2}v^2 - \sum_{\vec{q}} Q_{\vec{q}}(f_{\vec{q}} + \rho^*_{\vec{q}}b_{\vec{q}}) + \sum_{\vec{q}}\omega_{\vec{q}}(b_{\vec{q}}f^*_{\vec{q}}\rho^*_{\vec{q}} + b^*_{\vec{q}}f_{\vec{q}}\rho_{\vec{q}}) \tag{26}$$

and

$$\langle\vec{P}\rangle = \vec{v} + \sum_{\vec{q}}\vec{q}(|f_{\vec{q}}|^2 + |b_{\vec{q}}|^2), \tag{27}$$

where τ is the electron kinetic energy

$$\tau = -\tfrac{1}{2}\int d^3r\,\psi^*\nabla^2\psi .$$

As in the Pekar theory, $\langle E\rangle$ and $\langle\vec{P}\rangle$ represent an upper bound to the exact solutions if the exact solutions are well behaved. If we approximate $\rho_{\vec{q}}$ by a sharp cutoff at $\vec{q} = 2\mu$, representing an electronic localization of radius π/μ, we find

$$\langle E\rangle \cong \beta\mu^2 + \langle\vec{P}\rangle\cdot\vec{v} - \tfrac{1}{2}v^2 + \frac{8\alpha}{3\pi v}\left[(1+3v^2+\kappa^3/8)\ln\left(\frac{(1+\kappa/2)-v}{(1+\kappa/2)+v}\right) - (1+3v^2+\mu^3)\ln\left(\frac{(1+\mu)-v}{(1+\mu)+v}\right)\right.$$

$$\left. - v(v^2+3)\ln\left(\frac{(1+\kappa/2)^2-v^2}{(1+\mu)^2-v^2}\right) + \frac{\kappa v}{2}(4-\kappa/2) - \mu v(4-\mu) - \mu^3\ln\left(\frac{1+v}{1-v}\right)\right] \tag{28}$$

and

$$\langle\vec{P}\rangle \cong \vec{v}\left\{1+\frac{8\alpha}{3\pi v^3}\left[(1-3v^2+\kappa^3/8)\ln\left(\frac{1+\kappa/2-v}{1+\kappa/2+v}\right)-(1-3v^2+\mu^3)\ln\left(\frac{1+\mu-v}{1+\mu+v}\right)\right.\right.$$
$$\left.\left.+2v^3\ln\left(\frac{(1+\kappa/2)^2-v^2}{(1+\mu)^2-v^2}\right)+\kappa v(\kappa/2-1)-2\mu v(\mu-1)+\mu^3 v\left(2v(1-v^2)^{-1}+\ln\left|\frac{1-v}{1+v}\right|\right)\right]\right\}, \quad (29)$$

where β is a numerical factor. It may be easily verified that the weak-coupling theory is given by $\mu \to 0$ and strong-coupling theory is given by $\mu \to \kappa/2$. In principle, we should choose the μ for each value of $\langle\vec{P}\rangle$ that gives the lowest $\langle E\rangle$. Even with μ fixed, however, we still have an upper bound with an asymptotically linear energy-momentum relation. Examination of the behavior of Eqs. (28) and (29) reveals that the lowest energy-momentum curve is given by $\mu \to 0$ for $\langle\vec{P}\rangle < \vec{P}'$, and $\mu = \kappa/2$ thereafter (see Fig. 1). Thus the theory does not smoothly interpolate between weak and strong coupling, giving an abrupt transition where the coupling extremes cross. However, the theory does give a suggestive extension to the Lee-Low-Pines theory beyond the critical momentum $\vec{P}_c$ due to an infinitesimal, but nonzero, amount of strong coupling as $v \to 1$ and $\mu \to 0$, and represents an upper bound in this region.

E. Lattice dilations for adiabatic theories

As discussed in the description of the Fröhlich Hamiltonian, we assume that all wave vectors and momenta are near the center of the Brillouin zone ($|\vec{P}| \ll \kappa$) and that the lattice dilation remains in the harmonic limit [$\vec{\nabla}\cdot\vec{u}(\vec{r}) \ll 1$ for lattice displacement $\vec{u}(\vec{r})$].[15] We must examine the behavior of the Pekar and interpolation theories to insure that these conditions are satisfied.

To examine the lattice dilation we use the fact that the deformation potential interaction is

$$\hat{H}_{\rm int} = D\vec{\nabla}\cdot\hat{\vec{u}}(\vec{r}) = \sum_{\vec{q}} Q_{\vec{q}}(\hat{a}_{\vec{q}} + \hat{a}^{\dagger}_{-\vec{q}})e^{i\vec{q}\cdot\vec{r}}. \quad (30)$$

The average dilation is given by

$$\Delta = -D^{-1}\langle\psi_{\vec{v}}|\hat{H}_{\rm int}|\psi_{\vec{v}}\rangle, \quad (31)$$

where $|\psi_{\vec{v}}\rangle$ is the trial state for the theory of interest. For the stationary Pekar theory ($\vec{v}=0$), we have

$$\Delta_{\rm sc} = \frac{2\alpha\kappa^3}{3\pi D} = \frac{\pi D}{12M}, \quad (32)$$

where M is the lattice mass. For typical physical systems, $\Delta_{\rm sc} \sim 0.3$, which is well into the anharmonic region for lattice displacements. This large dilation is a result of the localization of the electron and indicates that the extreme effective mass in the moving Pekar theory is due to neglect of anharmonic terms in the lattice Hamiltonian.

The weak-coupling limit of the interpolation theory ($\mu \to 0$) gives

$$\Delta_{\rm int} = \Delta_{\rm LLP} + \frac{16\alpha\mu^3}{3\pi D v}\ln\left|\frac{1+v}{1-v}\right|, \quad (33)$$

where $\Delta_{\rm LLP}$ is the dilation of the Lee-Low-Pines theory. In the linear EP region ($P \gg P_c$) we eliminate the velocity to find

$$\Delta_{\rm int} \cong \Delta_{\rm LLP}\big|_{v=1} + \frac{16\alpha\mu^3}{3\pi D}\left(1+\frac{16\alpha\mu^3}{3\pi(P-P_c)}\cdots\right)$$
$$\times\ln\left|\frac{3\pi(P-P_c)}{8\alpha\mu^3}\right|. \quad (34)$$

Clearly for a suitably small choice of μ, we can insure that $\Delta_{\rm int} \ll 1$ for all momenta $|\vec{P}| \ll \kappa$. This behavior is due to the fact that the linear EP region is the result of rapid growth of the lattice energy and its associated field momentum and not the lattice dilation, which grows at a much smaller rate. Thus we can have a self-consistent linear EP upper bound for the ADP polaron.

IV. CONCLUSIONS

On the basis of energy-crossing arguments,[4] one expects to find an asymptotically linear energy-momentum relation for the ADP polaron. Classical analysis of the problem does show this type of energy resonance for velocities approaching the sound velocity. Quantum theories, however, have yielded mixed results. The weak-coupling Lee-Low-Pines theory gives an EP relation which halts beyond a critical momentum, corresponding to a polaron velocity equal to s. The strong-coupling Pekar theory gives linear EP behavior in principle, but for the range of reasonable momenta, it exhibits an anomalously large effective mass. This is seen to be related to the extreme localization and the resulting large lattice dilation of the theory, inappropriate for a large polaron. We consider an interpolation theory which also gives a linear EP relation, due to an adiabatic strong-coupling component in the trial state. This theory seems to provide a linear upper-bound extension to the Lee-Low-Pines theory and does not involve the anomalous localization and lattice dilation of the pure Pekar theory.

By limiting the electron localization in adiabatic theories of the ADP polaron, we find that an

asymptotically linear EP upper bound seems to exist, consistent with the assumptions of the large-polaron formalism. The linear EP region arises from the growth of the lattice energy and its associated field momentum, not from the size of the lattice dilation and the electron localization. This suggests that the inclusion of anharmonic terms in the lattice Hamiltonian could naturally limit the dilation and localization of adiabatic theories, while retaining linear EP behavior.

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[15]We should actually ensure that the expected value of the strain tensor $\langle \partial u_i/\partial x_j \rangle$ is small. It may be shown that the requirement that its trace (the dilation) can be small is sufficient for the Pekar-type states.