Breakdown of perturbation theory for the deformation potential polaron

George Whitfield and Peter B. Shaw
Department of Physics, The Pennsylvania State University, University Park, Pennsylvania 16802
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By comparing variational forms of weak and strong coupling we show that the crossover point is near $\alpha K \approx 1$, where α is the coupling constant and K is the largest allowed (dimensionless) wave vector. Analysis of the perturbation series gives a leading term of the form $K^2 \Sigma b_n \alpha^n$, which contains no indication that when $\alpha K > 1$ the ground state is better described by strong coupling. It is suggested that when $\alpha K > 1$ the perturbation series approximates a metastable state of the system.

I. INTRODUCTION

In its original formulation the polaron[1] was composed of an electron interacting with the longitudinal optical mode of an ionic crystal. One of the attractive features of this problem was that it involved only one parameter, α, the coupling constant. Although integrals on a wave vector should really be taken over only one Brillouin zone, a reasonable approximation was obtained by integrating to infinity. Nothing in the theory diverged and all results would then be expressed as functions of α.

As the ideas of this original work were applied to a wider range of electron-phonon interactions,[2-4] it became clear that in general two parameters were needed. Besides the coupling constant many of the results depended directly on the cutoff wave vector K, or an approximate wave vector for the first Brillouin zone. This was noted to be the case for the piezoelectric polaron first[5] in weak coupling and then in the corrections to strong coupling.[2] Cutoff dependence is most pronounced in the theory discussed here, namely the interaction with acoustic phonons via the deformation potential.[6] This was probably the first type of electron-phonon interaction to be analyzed. It applies to carriers in nonpolar semiconductors and metals, and forms the basis of the theory of conductivity for these systems as well as the basis of the theory of superconductivity. In spite of all this work very little has been done on the deformation potential acoustic polaron, and much of what has been done has been in the context of small-polaron theory. This is due largely to the strong cutoff dependence and closely related short-range character of the theory. The argument is that if the coupling is large enough for strong coupling to apply, then the radius of the polaron will be about a lattice constant, and the displacements of the lattice in that unit cell will be very large. Hence, it should be treated by small- rather than large-polaron theory. This argument is quite reasonable and is probably appropriate to many cases. However, it is conceivable that in some cases strong coupling and large-polaron theory would still apply. For instance, if one estimates the strain that would be present for a polaron in a unit cell, it is about one, which is much too large, and, at a radius of several lattice constants, anharmonic effects in the lattice could well arrest further collapse. Also in superconductors it is clear that perturbation theory breaks down, and we are interested in a detailed understanding of this process. Lastly, we are interested in this problem as a model of a cutoff-dependent particle-field theory.

In the next section we compare variational versions of the weak and strong coupling theories and find that the parameter that determines which gives the lower energy is αK, and that at $\alpha K \approx 1$ the energies cross. We therefore expect that the parameter that determines the convergence of perturbation theory should be αK, not α.

In Sec. III we look at perturbation corrections to weak coupling and find that, as far as we can see, higher orders contribute a series in α^n. Hence the perturbation theory gives the mistaken impression that it should be a reasonable approximation whenever $\alpha \ll 1$, when comparison with strong coupling has shown that the range of usefulness of perturbation theory should be $\alpha \ll 1/K \approx 1/100$.

II. WEAK AND STRONG COUPLING

A. Weak coupling

We use the Fröhlich Hamiltonian[3]

$$H = \tfrac{1}{2}p^2 + \sum_k a_k^\dagger a_k \omega(k) + \sum_k V(k)(a_{-k}^\dagger + a_k) e^{i\vec{k}\cdot\vec{r}}. \quad (1)$$

The electron's position is $\vec{r}$ and its momentum is $\vec{p} = -i\vec{\nabla}_r$. The operators a_k and $a_k^\dagger$ destroy and create longitudinal acoustic phonons with wave vector $\vec{k}$. For the deformation potential acoustic interaction,[7] $\omega(k) = |k|$ and

$$V(k) = (4\pi\alpha k/\upsilon)^{1/2}. \quad (2)$$

The volume is $\mathcal{V}$ and the coupling constant

$$\alpha = D^2 m^2/8\pi\rho\hbar^3 s \approx \tfrac{1}{100}\,, \tag{3}$$

where D is the deformation potential, m is the electron band mass, ρ is the density of the crystal, and s is the speed of sound. In the above, ms^2 is the unit of energy and $\hbar/(ms)$ is the unit of length.

The standard[1,8] weak coupling polaron theory (which gives the same self-energy as perturbation theory) can be obtained from the trial wave function

$$|T_w\rangle = (1/\sqrt{\mathcal{V}})e^{S(r)}|0\rangle\,, \tag{4}$$

where

$$a_k|0\rangle = 0\,, \tag{5}$$

$$S(r) = \sum_k f_k(a_k e^{i\vec{k}\cdot\vec{r}} - a_k^\dagger a^{-i\vec{k}\cdot\vec{r}})\,. \tag{6}$$

The c number f_k will be chosen to minimize the energy with $f_k = f_{-k} = f_k^*$. The factor $1/\sqrt{\mathcal{V}}$ is an electron plane wave with zero momentum, and the factor $\exp S(r)|0\rangle$ describes a lattice distortion centered on the instantaneous position of the electron $\vec{r}$.

The expected value of H in this state is

$$E_w \equiv \langle T_w|H|T_w\rangle$$

$$= \sum_k f_k^2[\omega(k) + \tfrac{1}{2}k^2] - \sum_k 2V(k)f_k\,. \tag{7}$$

Minimizing E_w with respect to f_k gives

$$f_k = V(k)/[\omega(k) + \tfrac{1}{2}k^2] \tag{8}$$

and

$$E_w = -\sum_k V^2(k)/[\omega(k) + \tfrac{1}{2}k^2]\,. \tag{9}$$

It is easy to see that (9) is just the result of second-order Rayleigh-Schrödinger perturbation theory when the third term in (1) is treated as a perturbation. We evaluate the sum in (9) imposing a cutoff wave vector of $K \equiv (\pi/a)(\hbar/ms) \approx 100$. The lattice constant is a. We obtain

$$E_w = -\frac{2}{\pi}\alpha K^2 + \frac{8}{\pi}\alpha K - \frac{16}{\pi}\alpha \ln(1 + \tfrac{1}{2}K)\,. \tag{10}$$

The dominant term is $\approx \alpha K^2 \approx 100$. Since our unit of energy is $ms^2 \approx 10^{-4}$ eV, $E_w \approx 10^{-2}$ eV, which is much less than the band gap but about equal to the maximum phonon energy. This is quite large.

Since E_w is also the result of the lowest order of perturbation theory one wonders what the higher orders will look like. If they appear in powers of αK^2, the series is clearly useless, but if they appear in powers of α we would expect the series to be useful for almost all practical values of the parameters. In Sec. III we will examine higher orders of perturbation theory where we will find a series in powers of α, but K^2 is the highest power of K appearing. However, a much simpler and in some respects more reliable method is to compare this variational weak coupling theory to a variational strong coupling theory and see where they cross. We will do this next.

B. Strong coupling

The strong coupling theory of the polaron was done first by Pekar[9] and applied to the deformation potential coupling by Toyozawa.[4] In what follows we use a slightly modified version of Pekar's calculation. The procedure has been shown to yield the correct strong coupling limit for a variety of polarons.[10]

In previous versions of the polaron theory we found that although we had to cut off the integrals that appear in weak coupling it was not necessary in strong coupling because the electronic form factor provided a cutoff. We will start by ignoring the cutoff and show that without it the strong coupling is unstable in this instance.

We use the trial wave function

$$|T_s\rangle = \phi(\vec{r} - \vec{R})e^{S(R)}|0\rangle\,, \tag{11}$$

where

$$S(R) \equiv \sum_k d_k(a_k e^{i\vec{k}\cdot\vec{R}} - a_k^\dagger e^{-i\vec{k}\cdot\vec{R}})\,. \tag{12}$$

Here the c number $d_k = d_{-k} = d_k^*$. This is similar to the weak coupling trial state (4) except that the lattice distortion $\exp[S(R)]|0\rangle$ is centered about the arbitrary c number $\vec{R}$ rather than the electron position $\vec{r}$, and the electron is in a bound state $\phi(r - R)$ rather than a plane-wave state.

Again we calculate the expected value of H and we get this time

$$E_s \equiv \langle T_s|H|T_s\rangle$$

$$= \tfrac{1}{2}\int \phi p^2\phi\, d^3r + \sum_k d_k^2\,\omega(k) - 2\sum_k V(k)d_k\rho_k\,, \tag{13}$$

where

$$\rho_k \equiv \int d^3r\, e^{i\vec{k}\cdot(\vec{r}-\vec{R})}\,\phi^2(\vec{r} - \vec{R})\,. \tag{14}$$

We have chosen ϕ real.

As with weak coupling we choose the lattice distortion that minimizes the energy, giving

$$d_k = V(k)\rho_k/\omega(k)\,, \tag{15}$$

and hence

$$E_s = \int \phi \tfrac{1}{2} p^2 \phi \, d^3r - \frac{4\pi\alpha}{v} \sum_k \rho_k^2, \tag{16}$$

or

$$E_s = \int \phi \tfrac{1}{2} p^2 \phi \, d^3r - 4\pi\alpha \int d^3r \, \phi^4. \tag{17}$$

However, unlike the weak coupling we must now choose ϕ to minimize E_s, subject to the constraint that $\int \phi^2 d^3r = 1$. This variation leads to the familiar "nonlinear Schrödinger equation"[11]

$$\tfrac{1}{2} p^2 \phi - 8\pi\alpha\phi^3 = \epsilon\phi. \tag{18}$$

This equation has a closed-form solution in one dimension which we have discussed extensively[3,12,13] in connection with the polaron. However, in three dimensions the lowest value of $\epsilon = -\infty$, unless we impose a cutoff. The easiest way to show this is to return to Eq. (17) and use a particular form for the bound state $\phi(\vec{r} - \vec{R})$. For the purposes of this argument the precise form of ϕ used is not very important (and no matter what form we use the result is still an upper bound), so for simplicity we choose

$$\phi(\vec{r} - \vec{R}) = (2/\pi\beta^2)^{3/4} e^{-|\vec{r}-\vec{R}|^2/\beta^2}, \tag{19}$$

and then (17) becomes

$$E_s = \langle T_s | H | T_s \rangle = \frac{3}{2}\frac{1}{\beta^2} - \frac{4}{\sqrt{\pi}}\alpha\frac{1}{\beta^3}. \tag{20}$$

We see that unless $\alpha/\beta > 3\sqrt{\pi}/8$ the energy is positive and then certainly higher than E_w. However, if $\alpha/\beta > 3\sqrt{\pi}/8$ then the smaller we make β the lower E_s will be (i.e., $\beta \to 0$ and $E_s \to -\infty$). Hence we see that the strong coupling theory is unstable.

We can introduce a cutoff into this theory simply by setting $\beta = 1/K$. Although this introduces a cutoff in a slightly different way than it was put into the weak coupling calculation, it is a considerable simplification and gives qualitatively the same results. In the Appendix we develop a systematic strong coupling theory, but for the present discussion we just set $\beta = 1/K$ which gives

$$E_s = \tfrac{5}{2} K^2 - (4/\sqrt{\pi})\alpha K^3. \tag{21}$$

We must compare this with

$$E_w \approx -(2/\pi)\alpha K^2 \tag{22}$$

which is the leading term in (10).

Although K varies from crystal to crystal it does not change much, so let us hold it fixed at $K = 100$ for this discussion. We will let α vary keeping in mind that we are interested in the range of values near $\alpha \approx 1/100$. In Fig. 1 we plot Eqs. (21) and (22) as a function of (αK). We see that when $\alpha K > 1$, E_s becomes lower than E_w, and it rapidly becomes much lower.

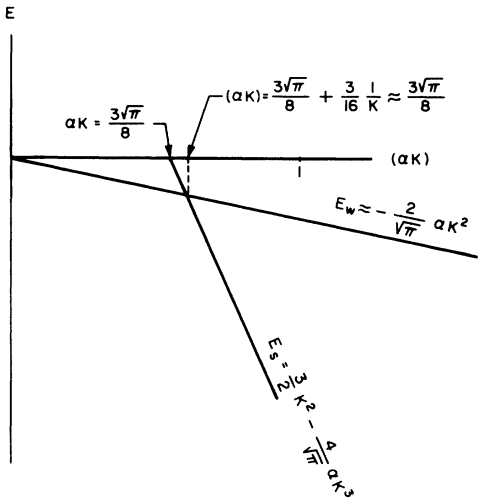

FIG. 1. Comparison of weak and strong coupling upper bounds. Assume that $K = 100$ and plot the energies as functions of αK.

It seems difficult to escape the conclusion that someplace near $\alpha K = 1$ the perturbation theory breaks down and the correct theory is some form of strong coupling. Therefore we would expect higher orders of perturbation theory to proceed in powers of (αK). We will show in the next section that this does not seem to be the case.

III. HIGHER ORDERS OF PERTURBATION THEORY

Our weak coupling result E_w given in Eq. (10) is the lowest order of Rayleigh-Schrödinger perturbation theory. This is the standard perturbation theory where one calculates the eigenvalues and eigenfunctions as a power series in the coupling constant. It is, however, extremely involved to calculate higher orders of this perturbation theory. If instead of trying to calculate the eigenfunction and eigenvalues we calculate the Green's function and expand it as a power series in the coupling constant, it is easier to generate higher-order corrections. The formalism for the calculation is well known from quantum electrodynamics and from electron-phonon interaction in metals. The present problem has some analogies to each of these. We follow a formulation originally[14] set up to deal with the optical polaron. First we rewrite the Hamiltonian (1) in second quantized form where it is

$$H = \sum_k \tfrac{1}{2} k^2 C_k^\dagger C_k + \sum_k a_k^\dagger a_k \omega(k) + \sum_k V(k)(a_{-k}^\dagger + a_k)\rho_{-k}, \tag{23}$$

where

$$\rho_k = \sum_q C^\dagger_{q-k} C_q . \tag{24}$$

The electron creation operator is $C^\dagger_k$, and we ignore spin and holes. The rest of the notation is the same as in Sec. II.

We define the electron Green's function at time t by

$$G(\vec{p}, t) = -i\langle 00 | T C_p(t) C^\dagger_p(0) | 00\rangle , \tag{25}$$

where $|00\rangle$ is the electron and phonon vacuum, and T is the time ordering symbol, and $C^\dagger_p(t)$ is the Heisenberg creation operator $e^{iHt} C_p e^{-iHt}$. We will make use of its Fourier transform $G(\vec{p}, \epsilon)$ defined by

$$G(\vec{p}, t) \equiv \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} G(\vec{p}, \epsilon) e^{-i\epsilon t} . \tag{26}$$

For $\alpha = 0$, $G(\vec{p}, \epsilon)$ becomes

$$G_0(\vec{p}, \epsilon) = \frac{1}{\epsilon - \frac{1}{2}p^2 + i\delta} . \tag{27}$$

The phonon Green's function is defined by

$$D(\vec{k}, t) \equiv -i\langle 00 | T[a_k(t) + a^\dagger_{-k}(t)][a_{-k}(0) + a^\dagger_k(0)] | 00\rangle , \tag{28}$$

and its Fourier transform through

$$D(\vec{k}, t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} D(\vec{k}, \omega) e^{-i\omega t} .$$

Since $|00\rangle$ is an electron vacuum and H commutes with the electron number operator,

$$D(\vec{k}, \omega) = D_0(\vec{k}, \omega) = \frac{1}{\omega - \omega(k) + i\delta} - \frac{1}{\omega + \omega(k) - i\delta} . \tag{29}$$

This greatly simplifies our problem in comparison to both quantum electrodynamics and electron-phonon interaction in metals. If we define the self-energy, $\Sigma'(p, \epsilon)$, by

$$G(\vec{p}, \epsilon) = \frac{1}{\epsilon - \frac{1}{2}p^2 - \Sigma'(\vec{p}, \epsilon) + i\delta} , \tag{30}$$

then we see that G has a pole at $\epsilon = E(p)$, where

$$E(p) = \tfrac{1}{2}p^2 + \Sigma'(\vec{p}, E(p)) , \tag{31}$$

which corresponds to the polaron energy. Above we have only considered $E(0)$ so we need only

$$E(0) = + \Sigma'(0, E(0)) . \tag{32}$$

Σ' can be expressed as a power series in the coupling constant, the terms of which can be obtained through an established set of rules for Feynman diagrams.[14] The first such term comes from the diagram in Fig. 2(a) which contributes to the self-energy

$$\Sigma^{(a)}(\vec{p}, \epsilon) = \sum_k \int \frac{d\omega}{(2\pi)} V(k) D_0(k, \omega) \times G_0(\vec{p} - \vec{k}, \epsilon - \omega) . \tag{33}$$

The integral on ω can be performed by closing the contour in the lower half plane giving

$$\Sigma^{(a)}(\vec{p}, \epsilon) = \sum_k \frac{V^2(k)}{\epsilon - \omega(k) - \frac{1}{2}(\vec{p} - \vec{k})^2 + i\delta} . \tag{34}$$

As above we will concern ourselves here only with $p = 0$. Also since we want $\Sigma(0, \epsilon)$ to use in (32) and since $E(0)$ is always negative, we need know $\Sigma(0, \epsilon)$ only for values of $\epsilon < 0$. Under these conditions

$$\Sigma^{(a)}(0, \epsilon) = \mathrm{Re}\Sigma^{(a)}(0, \epsilon) , \quad \epsilon < 0 \tag{35}$$

$$= \sum_k \frac{V^2(k)}{\epsilon - |k| - \frac{1}{2}k^2} , \quad \epsilon < 0 . \tag{36}$$

Note that if we neglect the ϵ in (36) then $\Sigma^{(a)}(0, 0) = E_w$, which is the lowest order of Rayleigh-Schrödinger perturbation theory. Moreover, we see that if we use (36) in (32) and compare the result with (9) then $E_w < E(0)$. Since E_w is an upper bound this means that E_w is better in the variational sense than $E(0)$ in the present approximation. This sort of thing has been seen before in the polaron problem.[15] With $\epsilon \approx -\alpha K^2 \ll -1$ the integral in (36) becomes

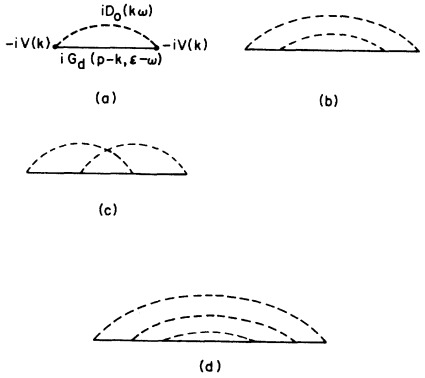

FIG. 2. Feynman diagrams for the self-energy of the polaron. (a) Second-order correction, (b) and (c) are the fourth-order, and (d) is one of the sixth-order terms.

$$\Sigma^{(a)}(0,\epsilon) = -\frac{2\alpha}{\pi}\left[K^2 - 4K + 4(1+\tfrac{1}{2}\epsilon)\ln\frac{-\epsilon+K+\frac{1}{2}K^2}{-\epsilon} - \frac{\beta(1+\frac{3}{2}\epsilon)}{(-2\epsilon-1)^{1/2}}\left(\arctan\frac{1+K}{(-2\epsilon-1)^{1/2}} - \arctan\frac{1}{(-2\epsilon-1)^{1/2}}\right)\right]. \tag{37}$$

The leading terms $-(2\alpha/\pi)K^2+(8/\pi)\alpha K$ are the same as in Eq. (10) for E_w. Now although this perturbation series is different from the Rayleigh-Schrödinger series, we can analyze the higher terms to determine if higher powers of K appear. In particular we want to know if there is a series in $(\alpha K)^n$ which would suggest when strong coupling obtains (i.e., $\alpha K \gg 1$).

The next terms in the series for Σ' are given by the diagrams in Figs. 2 (b) and 2 (c) and lead to the integrals:

$$\Sigma^{(b)}(\vec{p},\epsilon) = \sum_{k,k'}\int\frac{d\omega}{2\pi}\frac{d\omega'}{2\pi}V^2(k)V^2(k')D_0(\vec{k}\omega)D_0(\vec{k}'\omega')G_0^2(\epsilon-\omega,\vec{p}-\vec{k})G_0(\epsilon-\omega-\omega',\vec{p}-\vec{k}-\vec{k}'), \tag{38}$$

$$\Sigma^{(c)}(\vec{p},\omega) = \sum_{kk'}\int\frac{d\omega}{2\pi}\frac{d\omega'}{2\pi}V^2(k)V^2(k')D_0(\vec{k}\omega)D_0(\vec{k}'\omega')G_0(\epsilon-\omega,\vec{p}-\vec{k}) \times G_0(\epsilon-\omega-\omega',\vec{p}-\vec{k}-\vec{k}')G_0(\epsilon-\omega',\vec{p}-\vec{k}'). \tag{39}$$

The ω integration can be done by closing the contour in the lower half plane and we are left with

$$\Sigma^{(c)}(0,\epsilon) = \sum_{k,k'}\frac{V^2(k)V^2(k')}{(\epsilon-|k|-\frac{1}{2}k^2)[\epsilon-|k|-|k'|-\frac{1}{2}(\vec{k}+\vec{k}')^2](\epsilon-|k'|-\frac{1}{2}k'^2)}, \tag{40}$$

and a similar integral for $\Sigma^{(b)}(0,\epsilon)$. Since we are interested only in the region $\epsilon<0$ there are no infrared divergences in (40).

We are interested here in seeing how $\Sigma^{(c)}$ depends on K, so we concern ourselves only with the region of integration near the upper limit. We replace $\epsilon-|k|-\frac{1}{2}k^2$ by $\frac{1}{2}k^2$, but for $\epsilon-|\vec{k}|-|\vec{k}'|-\frac{1}{2}(\vec{k}+\vec{k}')^2$ we write $-|k|-|k'|-\frac{1}{2}(\vec{k}+\vec{k}')^2$ because there is a region when $(\vec{k}+\vec{k}')^2<|k|+|k'|$ and our main concern in these integrals will be to see if this region is large enough to change the K dependence of the result. We get the K dependence of (40) from the integral

$$-\frac{(4\pi\alpha)^2}{(2\pi)^6}\,4\int\frac{k^3dk\,k'^3dk'\,d(\cos\phi)(4\pi)(2\pi)}{k^2(k+k'+\frac{1}{2}k^2+k'^2+2kk'\cos\phi)k'^2}. \tag{41}$$

If we introduce the scale change $q=k/K$ and $q'=k'/K$ we have

$$-\frac{(8\pi\alpha)^2K^2}{(2\pi)^6}\int_0^1 dq\int_0^1 dq'\int_{-1}^1 dx\,\frac{qq'(4\pi)(2\pi)}{(q+q')/K+\frac{1}{2}(q^2+q'^2+2qq'x)}. \tag{42}$$

If we expand the integrand in powers of $(q+q')/K$ all the coefficients are finite and independent of K and the leading term is K^2 as in second-order perturbation theory. The integral for $\Sigma^{(b)}$ behaves in the same way.

Higher orders. Higher-order integrals always lead to an expression like (42) after all the wave vectors are scaled, i.e., K^2 multiplied by an integral where all wave vectors range from zero to one and where there are factors like

$$\frac{1}{[(q+q')/K+\frac{1}{2}(q^2+q'^2+2qq'x)]^a}.$$

If $a=1$ or 2, we can expand the integrand in powers of $(q+q')/K$ and the highest power of K that occurs is K^2. But if $a>2$, higher powers of K will appear.

In noncrossing diagrams like that shown in Fig. 2(d) terms with $a=2$ appear but in no diagram that we have seen do we ever get $a=3$ or higher. So as far as we can see even in higher orders of perturbation theory the highest power of K that occurs is K^2. This suggests strongly that there is no sequence of terms in $(\alpha K)^n$ and hence the perturbation theory does not indicate the critical region $\alpha K\approx 1$ where strong coupling should be used. This analysis is not sufficiently detailed to include terms like $(\alpha \ln K)^n$.

IV. DISCUSSION

It is quite clear that when $\alpha K\gg 1$ the strong coupling theory gives a better approximation to the ground state than the weak coupling does. It would have been reassuring to see that the perturbation series contained a series of terms like $\sum_n(\alpha K)^n b_n$, but as far as we have been able to see no such term exists. We can see a possible resolution to

this dilemma.

It has recently been suggested[16] that the system that we are discussing here has a large radius metastable state, and the perturbation series might be approximating this state. The easiest way to see that such a state exists is to return to Eq. (20) which gives the energy in the adiabatic approximation

$$E_s=\frac{3}{2}\frac{1}{\beta^2}-\frac{4}{\sqrt{\pi}}\alpha\frac{1}{\beta^3} \tag{20}$$

as a function of the radius of the electronic state β. In Fig. 3 E_s is plotted as a function of $1/\beta$. As we mentioned above the lowest energy is clearly obtained for $1/\beta\to\infty$. But we note that there is a stable minimum at $1/\beta=0$ suggesting that a large radius metastable state exists. This analysis and an alternative are both discussed by Toyozawa and Shinozuka.[16] The strange behavior of perturbation theory that we have pointed out lends support to the existence of such a metastable state.

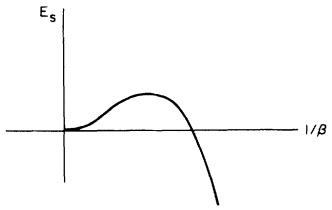

FIG. 3. The strong coupling energy vs the inverse of the radius of the electronic wave function β.

APPENDIX

In this Appendix we give a more systematic treatment of strong coupling. In the variational theory of Sec. II the electronic wave function for the polaron was found to obey the nonlinear Schrödinger equation given in Eq. (18). As noted there, the polaron described by this equation has the unphysical feature of being collapsed to a point with infinite binding energy. This collapse is avoided by taking into account the discrete nature of the lattice through the introduction of a maximum phonon wave vector K. In the variational argument it was assumed for simplicity that this cutoff gives the electronic wave function of a radius K^{-1}. A more consistent approach is to introduce the cutoff on phonon wave vectors in the Hamiltonian, which was essentially the method of cutting off the weak coupling theory. It is convenient to make the scale change

$$\begin{aligned}\vec{k}&\to K\vec{k},\\ \vec{r}&\to K^{-1}\vec{r},\end{aligned} \tag{A1}$$

under which the Hamiltonian becomes

$$H=K^2\Big(-\tfrac{1}{2}\nabla^2+\frac{1}{K}\sum_{\vec{k}}{}'a_k^\dagger a_k|\vec{k}| + \sum_{\vec{k}}{}'V(\vec{k})(a_{-k}^\dagger+a_k)e^{i\vec{k}\cdot\vec{r}}\Big). \tag{A2}$$

The prime on the summations indicates that they terminate at $|\vec{k}|_{\max}=1$. The function $V(\vec{k})$ is given by Eq. (2)

$$V(\vec{k})=(4\pi\alpha\,|\vec{k}|/V)^{1/2}, \tag{A3}$$

in which the volume V has been scaled

$$V\to K^{-3}V \tag{A4}$$

so that

$$\frac{1}{V}\sum_{\vec{k}}{}'(\,)\to\frac{1}{(2\pi)^3}\int_0^1 dk\,k^2\int\!\!\int d\Omega_k(\,). \tag{A5}$$

We point out that the electron-phonon interaction term in Eq. (A2) gives (in perturbation theory) a correction to the energy of order α. Since K^{-1} is itself a small number ($\sim10^{-2}$) the lattice energy serves as a rival perturbation in the Hamiltonian. In the case $\alpha\gg K^{-1}$ the electron-phonon interaction energy is large compared to the lattice deformation energy, and the sum of the first and third terms in Eq. (A2) should be diagonalized with the lattice energy then treated as a perturbation. This case of $\alpha K\gg1$ is the strong coupling regime and is the concern of this Appendix. The diagonalization of the Hamiltonian in strong coupling can be carried out systematically along the lines of Ref. 17. We will not fully develop that method here but note that it leads to a ground-state energy of the form

$$E_0=f(\alpha K)+K^{-1}g(\alpha K). \tag{A6}$$

We construct below a self-consistent method of determing $f(\alpha K)$ as a *descending* series in $(\alpha K)^{1/2}$.

As usual, we first made the canonical transformation

$$a_k\to a_k-d_k, \tag{A7}$$

where d_k is a real, even function of wave vector k. In terms of the new phonon operators the Hamiltonian becomes

$$H=K^2\Big(-\tfrac{1}{2}\nabla^2+\frac{1}{K}\sum_{\vec{k}}{}'a_k^\dagger a_k|\vec{k}|+\sum_{\vec{k}}{}'V(\vec{k})(a_{-k}^\dagger+a_k)e^{i\vec{k}\cdot\vec{r}} +\frac{1}{K}\sum_{\vec{k}}{}'d_k^2|\vec{k}|-\frac{1}{K}\sum_{\vec{k}}{}'(a_k+a_k^\dagger)d_k|\vec{k}|-2\sum_{\vec{k}}{}'V(\vec{k})d_k e^{i\vec{k}\cdot\vec{r}}\Big). \tag{A8}$$

We choose d_k by minimizing the expected value of H in the trial state

$$|\phi\rangle = \phi(\vec{r})|0\rangle\ , \tag{A9}$$

where $|0\rangle$ is the vacuum for the new phonon operator and the electronic wave function $\phi(\vec{r})$ is normalized to unity. The minimization is carried out with respect to both d_k and the functional form of $\phi(\vec{r})$, and leads to the results

$$d_k = V(\vec{k})\rho_k/|\vec{k}| \tag{A10}$$

and

$$-\tfrac{1}{2}\nabla^2\phi(\vec{r}) - \sum_k{}' V(\vec{k})d_k e^{i\vec{k}\cdot\vec{r}}\phi(\vec{r}) = \epsilon\,\phi(\vec{r})\,, \tag{A11}$$

where

$$\rho_k = \iiint d^3r\,\phi^2(\vec{r})e^{i\vec{k}\cdot\vec{r}}\,. \tag{A12}$$

In this Appendix ϵ is the expected value of H/K^2 with the energy of the deformed lattice subtracted out. We assume that $\phi(\vec{r})$ is a real spherically symmetric function. By combining Eqs. (A10), (A11), and (A12) we obtain the nonlinear Schrödinger equation

$$-\tfrac{1}{2}\nabla^2\phi(\vec{r}) - 8\pi\alpha K\iiint d^3r'\,\Delta(\vec{r}-\vec{r}')\phi^2(\vec{r}')\phi(\vec{r}) = \epsilon\,\phi(\vec{r})\,, \tag{A13}$$

where

$$\Delta(\vec{r}-\vec{r}') = \iint\frac{d\Omega_k}{(2\pi)^3}\int_0^1 dk\,k^2 e^{i\vec{k}\cdot(\vec{r}-\vec{r}')}\,. \tag{A14}$$

If the cutoff K had not been introduced in the Hamiltonian, the upper limit on the k integration in Eq. (A14) would extend to infinity. The function $\Delta(\vec{r}-\vec{r}')$ would then become a Dirac delta function, and with a scale change Eq. (A13) would reduce to Eq. (18). Because $\phi(\vec{r})$ is assumed to be spherically symmetric the angular part of the $\vec{r}'$ and $\vec{k}$ integrations can be done explicitly, and the Schrödinger equation reduces to

$$-\frac{1}{2}\frac{1}{r^2}\frac{d}{dr}r^2\frac{d}{dr}\phi(r) + U(r)\phi(r) = \epsilon\,\phi(r)\,, \tag{A15}$$

where the potential $U(r)$ is given by

$$U(r) = -\frac{4\alpha K}{\pi}\int_0^1 dk\iiint d^3r'\,\frac{\sin kr\sin kr'}{rr'}\phi^2(r')\,. \tag{A16}$$

Next we make a Taylor-series expansion of the sine functions in Eq. (A16) and make a scale change

$$r = (4\pi\alpha K)^{-1/4}x,\quad \chi(x) = (4\pi\alpha K)^{-3/8}\phi(x(4\pi\alpha K)^{-1/4})\,. \tag{A17}$$

The resulting Schrödinger equation is

$$-\frac{1}{2}\frac{1}{x^2}\frac{d}{dx}x^2\frac{d}{dx}\chi(x) + \sum_{n=1}^{\infty}\sum_{m=0}^{\infty}a_{n\,m}(4\pi\alpha K)^{(1-n-m)/2}x^{2n}\chi(x) = \bar{\epsilon}\chi(x)\,, \tag{A18}$$

where

$$a_{nm} = \frac{(-1)^{n+m-1}}{(2n+1)!(2m+1)!(2n+2m+3)}\iiint\frac{d^3y}{\pi^2}y^{2m}\chi^2(y)\,. \tag{A19}$$

The polaron self-energy ϵ is given in terms of the eigenvalue $\bar{\epsilon}$ of Eq. (A18) by

$$\epsilon = (4\pi\alpha K)^{1/2}\bar{\epsilon} - 4\pi\alpha K\sum_{m=0}^{\infty}\frac{(-1)^m}{(2m+1)!(2m+3)}\frac{1}{(4\pi\alpha K)^{m/2}}\iiint\frac{d^3y}{\pi^2}y^{2m}\chi^2(y)\,. \tag{A20}$$

In the strong coupling regime $(4\pi\alpha K)^{-1/2}$ is a small number, and Eqs. (A18), (A19), and (A20) serve as a coupled set of equations by which the polaron self-energy can be determined as an expansion in powers of $(4\pi\alpha K)^{-1/2}$. As an illustration, the leading contribution to the potential energy in Eq. (A18) comes from $n=1$ and $m=0$ (this term is the only one that survives in the potential energy of Eq. (A18) for $\alpha K\to\infty$). In this approximation we obtain

$$\chi(x) = (\Omega/\pi)^{3/4}e^{-\Omega x^2/2}\,,\qquad \bar{\epsilon} = \tfrac{3}{2}\Omega,\quad \Omega = 1/(15\pi)^{1/2}\,, \tag{A21}$$

and hence

$$\epsilon = \sum_{m=-2}^{\infty}\epsilon_m^0(4\pi\alpha K)^{-m/2}\,, \tag{A22}$$

where

$$\epsilon_{-2}^0 = -1/3\pi^2\,,\qquad \epsilon_{-1}^0 = 3\sqrt{15}/20\pi\,, \tag{A23}$$

and

$$\epsilon_m^0 = (-1)^{m+1}\frac{15(2m+5)!!(15\pi^2)^{m/2}}{(2m+5)!(2m+7)2^{m+2}},\quad m\geq 0\,.$$

The potential energy in Eq. (A18) is an *explicit* power series in $(4\pi\alpha K)^{-1/2}$. Ordinary perturbation theory can be used to calculate corrections to the zero-order eigenvalues $\bar{\epsilon}$ and wave function χ of Eq. (A21). Unfortunately, the coefficients a_{nm} which enter the potential energy power series also depend *implicitly* on $(4\pi\alpha K)^{-1/2}$ through the wave function χ as expressed in Eq. (A19). It is possible, although somewhat involved, to sort out all the dependence on αK and construct an expansion

$$\epsilon = \sum_{m=-2}^{\infty} \epsilon_m (4\pi\alpha K)^{-m/2}, \tag{A24}$$

where the coefficients ϵ_m are determined by a perturbative-iterative scheme involving Eqs. (A18), (A19), and (A20). The leading term of the expansion is given by $\epsilon_{-2} = \epsilon^0_{-2}$ as defined in Eq. (A23) and thus

$$\epsilon = -\frac{4}{3\pi}\alpha K + \cdots . \tag{A25}$$

We do not compute other terms in the expansion here. When the overall energy scale factor of K^2 in Eq. (A8) is reinserted, we obtain $-(4/3\pi)\alpha K^3$ for the leading strong coupling polaron self-energy, which should be compared with the lead term of Eq. (21).

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