Intermediate-Coupling Polaron Effective Mass

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We have calculated the polaron effective mass m^* by extending the variational ansatz of a previous paper, where, in the Lee-Low-Pines representation, pair correlations between wave vectors of virtually emitted phonons are taken into account. Our results suggest that (1) effective masses calculated by fourth-order perturbation theory lie below the true values but are quite accurate for $\alpha \lesssim 3$ and (2) the Feynman theory gives effective masses which are somewhat too high in the intermediate-coupling region. For $\alpha \gtrsim 4.5$ we propose the estimate $m^*/m = 1+\alpha/6+0.02363\alpha^2+0.0014\alpha^3$.

I. INTRODUCTION

THE measurement of cyclotron masses of conduction electrons in polar materials like the silver halides and alkali halides is very difficult. Only recently have accurate data become available.[1–3]

Because an electron placed in the conduction band of a polar insulator or semiconductor will distort the lattice and induce a local polarization charge, cyclotron-resonance experiments in such materials do not measure the rigid lattice conduction-band mass as calculated in conventional band theory, but rather the magnetic levels of a more complicated excitation (the polaron) consisting of the electron and its accompanying lattice distortion. Thus to determine the band mass from a cyclotron-resonance experiment, the shift in the cyclotron frequency due to electron-lattice interaction must be taken into account.

In view of the large amount of new data becoming available on the intermediate coupling polaron mass and the success of the variational ansatz of I[4] in producing a polaron ground-state energy lower than has previously been reported for $\alpha \leq 3.5$, we thought it would be useful and interesting to extend the ansatz of I for the purpose of calculating the polaron effective mass.

Calculations based on the Fröhlich[5] model for electron-lattice interaction indicate that in general the cyclotron frequency of the polaron $\omega_{\rm pol}$ is not linear in the magnetic field.[6,7] However, in the special case that the excitation energy of the final state remains sufficiently small compared to $\hbar\omega$, the energy of the long-wavelength LO phonons, we have

$$\omega_{\rm pol} = m\omega_c/m^* = eH/m^*c\,, \tag{1}$$

where m is the rigid lattice band mass for the conduction band (assumed parabolic), m^* is the polaron mass in the absence of magnetic fields, and $\omega_{\rm pol}$ is the observed (polaron) cyclotron frequency. (By "excitation energy" of a conduction-band state in a magnetic field we mean the energy difference between the given state and the conduction-band ground state in the magnetic field.)

Nonlinear behavior of $\omega_{\rm pol}$ with magnetic field has been observed in the weakly polar semiconductor InSb.[8] However, in electron-cyclotron resonance experiments reported to date in the silver and alkali halides $\omega_c/\omega < 0.1$; thus, if the resonating electrons are not moving too rapidly along the magnetic field, as seems to be the case in the reported experiments, (1) should be accurate.

Determining the relationship of m^* to m is one of the fundamental problems of polaron theory. Mathematically this amounts to calculating the coefficient of the P^2 term in the expansion of the polaron energy $E(P)$ (in the absence of magnetic fields) as a function of the polaron momentum P. Previous variational calculations of $E(P)$ for small P have been carried out by LLP,[9] Gurari,[10] Lee and Pines,[11] and Haga.[12] An important nonvariational calculation was made by Feynman[13] and calculations equivalent to fourth-order perturbation theory have been reported by Höhler and Müllenseifen,[14] LLP, and Röseler.[15] These calculations will be discussed in Sec. III.

II. VARIATIONAL CALCULATION

Starting from the Fröhlich Hamiltonian in dimensionless form,

$$H = p^2 + \sum n_{\mathbf{k}} + \sum \nu_k (e^{-i\mathbf{k}\cdot\mathbf{r}} b_{\mathbf{k}}^\dagger + \text{H.c.})\,, \tag{2}$$

where

$$\alpha = \tfrac{1}{2}(e^2/r_0)(1/\hbar\omega)(1/\epsilon_\infty - 1/\epsilon_0)\,,$$
$$n_{\mathbf{k}} = b_{\mathbf{k}}^\dagger b_{\mathbf{k}}\,, \quad \nu_k = (4\pi\alpha/(\Omega/r_0^3))^{1/2}(1/k)\,, \tag{3}$$

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where Ω is the crystal volume and $r_0=(\hbar/2m\omega)^{1/2}$, we take our trial functions in the form

$$e^{i\mathbf{r}\cdot(\mathbf{P}-\sum \mathbf{k}n_k)}U|\phi_P\rangle,$$
$$U=e^{\sum f_k(b_\mathbf{k}^\dagger-b_\mathbf{k})}. \tag{4}$$

The Schrödinger equation for $|\phi_P\rangle$ is

$$\begin{aligned}
&\mathcal{H}|\phi_P\rangle=E(P)|\phi_P\rangle,\\
&\mathcal{H}=\mathcal{H}_0+\mathcal{H}_1,\\
&\mathcal{H}_0=(\mathbf{P}-\textstyle\sum \mathbf{k}n_\mathbf{k})^2+\sum n_\mathbf{k}-\alpha,\\
&\mathcal{H}_1=-2\textstyle\sum(\mathbf{P}\cdot\mathbf{k})f_k(b_\mathbf{k}^\dagger+b_\mathbf{k})\\
&\qquad+2\textstyle\sum \mathbf{k}\cdot\mathbf{m}f_kf_mb_\mathbf{k}^\dagger b_\mathbf{m}\\
&\qquad+\textstyle\sum \mathbf{k}\cdot\mathbf{m}f_kf_m(b_\mathbf{k}^\dagger b_\mathbf{m}^\dagger+b_\mathbf{k}b_\mathbf{m})\\
&\qquad\qquad+2\textstyle\sum \mathbf{k}\cdot\mathbf{m}f_k(n_\mathbf{m}b_\mathbf{k}+b_\mathbf{k}^\dagger n_\mathbf{m}),
\end{aligned}\tag{5}$$

where $\mathbf{P}$, being the total momentum of the electron-phonon system, is conserved and is therefore a c vector. We have specialized our choice of f_k in (4) to

$$f_k=-\nu_k/(1+k^2), \tag{6}$$

which is used by LLP for the case $P=0$.

Imagine now a sequence of possible trial functions $|\phi_P\rangle$:

$$\tilde{d}|0\rangle+\textstyle\sum \tilde{d}_\mathbf{k}|\mathbf{k}\rangle, \tag{7a}$$
$$\tilde{d}|0\rangle+\textstyle\sum \tilde{d}_\mathbf{k}|\mathbf{k}\rangle+\sum \tilde{d}_{\mathbf{k}\mathbf{l}}|\mathbf{k}\mathbf{l}\rangle, \tag{7b}$$
$$\tilde{d}|0\rangle+\textstyle\sum \tilde{d}_\mathbf{k}|\mathbf{k}\rangle+\sum \tilde{d}_{\mathbf{k}\mathbf{l}}|\mathbf{k}\mathbf{l}\rangle+\sum \tilde{d}_{\mathbf{k}\mathbf{l}\mathbf{m}}|\mathbf{k}\mathbf{l}\mathbf{m}\rangle, \tag{7c}$$

etc., where $|0\rangle$ is the phonon vacuum, $|\mathbf{k}\rangle=b_\mathbf{k}^\dagger|0\rangle$, $|\mathbf{k}\mathbf{l}\rangle=b_\mathbf{k}^\dagger b_\mathbf{l}^\dagger|0\rangle$, etc., and the indexed $\tilde{d}$ coefficients are freely varied to produce in each case, (7a)–(7c), the lowest possible energy at given $\mathbf{P}$ (where P is sufficiently small).

Optimization of (7a), first attempted by Haga,[12] amounts to diagonalizing $\mathcal{H}$ in the subspace S_1 spanned by $|0\rangle$ and the one-phonon states $|\mathbf{k}\rangle$. For small P this can be done analytically yielding the simple result

$$E_H(P)=-\alpha+(1-\tfrac{1}{12}\alpha)/(1+\tfrac{1}{12}\alpha)P^2, \tag{8}$$

hence an effective mass

$$m^*/m=(1+\tfrac{1}{12}\alpha)/(1-\tfrac{1}{12}\alpha). \tag{9}$$

The variational problem posed by (7b), equivalent to diagonalizing $\mathcal{H}$ in S_2, the subspace spanned by the zero-, one-, and two-phonon states, requires the services of a computer, although the problem can be reduced, as will be described, to a set of coupled one-dimensional integral equations. The effective mass is deduced from the energy spectrum $E_v(P)$ by

$$(m^*/m)_v=P^2/[E_v(P)-E_v(0)], \tag{10}$$

where $E_v(P)$ is the energy found variationally to order P^2. Optimizing the energy for (7c) would be a major undertaking because of the multidimensional integrals involved. In fact, even the perturbation calculation of $E(P)$ to order α^3 has not yet been carried out, and this perturbed energy is the weak-coupling limit of the energy deduced from (7c).

Turning now to the determination of $E_v(P)$ from (7b), which is the subject of this paper, we obtain from the diagonalization of $\mathcal{H}$ in S_2 the equations

$$\begin{aligned}
&E_v(P)+\alpha-P^2=-2\mathbf{P}\cdot\textstyle\sum \mathbf{k}f_kd_\mathbf{k}\\
&\qquad+\textstyle\sum \mathbf{k}\cdot\mathbf{m}f_kf_m(d_{\mathbf{k}\mathbf{m}}+d_{\mathbf{m}\mathbf{k}}),\\
&D_1(\mathbf{k})d_\mathbf{k}=-2\mathbf{P}\cdot\mathbf{k}f_k+2f_k\mathbf{k}\cdot\textstyle\sum \mathbf{m}f_md_\mathbf{m}\\
&\qquad+2(\mathbf{k}-\mathbf{P})\cdot\textstyle\sum \mathbf{l}f_l(d_{\mathbf{k}\mathbf{l}}+d_{\mathbf{l}\mathbf{k}}),\\
&D_2(\mathbf{k},\mathbf{l})d_{\mathbf{k}\mathbf{l}}=2\mathbf{k}\cdot\mathbf{l}f_kf_l\\
&\qquad+2(\mathbf{k}-\mathbf{P})\cdot\mathbf{l}f_ld_\mathbf{k}+2(\mathbf{l}-\mathbf{P})\cdot\mathbf{k}f_kd_\mathbf{l}\\
&\qquad+2\textstyle\sum \mathbf{k}\cdot\mathbf{m}f_kf_m(d_{\mathbf{l}\mathbf{m}}+d_{\mathbf{m}\mathbf{l}})\\
&\qquad\qquad+2\textstyle\sum \mathbf{l}\cdot\mathbf{m}f_lf_m(d_{\mathbf{k}\mathbf{m}}+d_{\mathbf{m}\mathbf{k}}),\\
&D_1(k)=E_v(P)+\alpha-(\mathbf{P}-\mathbf{k})^2-1,\\
&D_2(\mathbf{k},\mathbf{l})=E_v(P)+\alpha-(\mathbf{P}-\mathbf{k}-\mathbf{l})^2-2,\\
&d_\mathbf{k}=\tilde{d}_\mathbf{k}/d,\qquad d_{\mathbf{k}\mathbf{l}}=\tilde{d}_{\mathbf{k}\mathbf{l}}/d.
\end{aligned}\tag{11}$$

We have used the convention $d_{\mathbf{k}\mathbf{l}}=0$ for $k>l$.

In determining $E_v(P)$ to order P^2 it is convenient to introduce the quantities

$$\mathbf{P}\cdot\textstyle\sum \mathbf{m}f_md_\mathbf{m}=P^2\xi(P), \tag{12a}$$
$$\textstyle\sum \mathbf{l}f_l(d_{\mathbf{k}\mathbf{l}}+d_{\mathbf{l}\mathbf{k}})=\mathbf{k}f_k\zeta_1(\mathbf{P},\mathbf{k})+\mathbf{P}_{k\mathbf{l}}f_k\zeta_2(\mathbf{P},\mathbf{k}), \tag{12b}$$

where $\mathbf{P}_{k\mathbf{l}}=\mathbf{P}-(\mathbf{P}\cdot\mathbf{k})\mathbf{k}/k^2$, and therefore is a vector perpendicular to $\mathbf{k}$ in the plane of $\mathbf{P}$ and $\mathbf{k}$. We remark that (12a) and (12b) are more than definitions of ξ, ζ_1, and ζ_2. Actually these equations contain assumptions about the nature of the solutions of (11).

The assumption embodied in (12a) is almost self-evidently correct. From the equation for $d_\mathbf{k}$ it is clear that $d_\mathbf{k}$ contains only one preferred direction, namely, the direction of $\mathbf{P}$. Hence $\sum \mathbf{m}f_md_\mathbf{m}$ must be a vector lying along $\mathbf{P}$, as is implicit in (12a). Less easy to predict *a priori* is the direction of $\mathbf{V}\equiv\sum \mathbf{l}f_l(d_{\mathbf{k}\mathbf{l}}+d_{\mathbf{l}\mathbf{k}})$, since the two vectors, $\mathbf{k}$ and $\mathbf{P}$, determine $\mathbf{V}$. It turns out to be self-consistent to assume merely that $\mathbf{V}$ lies in the plane of $\mathbf{k}$ and $\mathbf{P}$. Within this restriction, (12b) gives the most general expression possible for $\mathbf{V}$.

Introducing (12a) and (12b) into (11) produces the equations

$$E_v(P)=-\alpha+P^2-2P^2\xi(P)+\textstyle\sum k^2f_k^2\zeta_1(\mathbf{P},\mathbf{k}), \tag{13a}$$

$$\tfrac{1}{2}P^2\xi=-\sum\frac{(\mathbf{P}\cdot\mathbf{k})^2}{D_1(k)}f_k^2(1-\xi+\zeta_1(\mathbf{P},\mathbf{k}))+\sum\frac{(\mathbf{P}\cdot\mathbf{k})}{D_1(k)}f_k^2k^2\zeta_1(\mathbf{P},\mathbf{k}), \tag{13b}$$

TABLE I. Comparison of the ratio of the polaron mass to the band mass as calculated from Eqs. (13) and (10) $[(m^*/m)_v]$, (16) $[(m^*/m)_L]$, and (15) $[(m^*/m)_{\text{we4}}]$ as a function of Fröhlich's coupling constant α, given in (3).

α	0.5	1.0	1.5	2.0	2.5	3.0	3.5	4.0	5.0
$(m^*/m)_v$	1.08940	1.19146	1.30656	1.4344	1.5738	1.7229	1.8788	2.0383	2.3543
$(m^*/m)_L$	1.08968	1.19417	1.31749	1.4653	1.6458	1.8711	2.1605	2.5467	3.8940
$(m^*/m)_{\text{we4}}$	1.08924	1.19029	1.30316	1.4278	1.5643	1.7126	1.8728	2.0447	2.4240

$$\tfrac{1}{2}l^2\zeta_1(\mathbf{P},\mathbf{l})=\sum\frac{[(\mathbf{k}\cdot\mathbf{l})-(\mathbf{P}\cdot\mathbf{k})]\mathbf{k}\cdot\mathbf{l}}{D_2(\mathbf{k},\mathbf{l})}f_k^2(C(\mathbf{l})+C(\mathbf{k}))$$
$$+\sum\frac{(\mathbf{k}\cdot\mathbf{l})^2f_k^2}{D_2(\mathbf{k},\mathbf{l})}(1+\zeta_1(\mathbf{P},\mathbf{l})+\zeta_1(\mathbf{P},\mathbf{k}))$$
$$+\sum\frac{\mathbf{k}\cdot\mathbf{l}}{D_2(\mathbf{k},\mathbf{l})}f_k^2(\mathbf{P}_{kl}\cdot\mathbf{l}\zeta_2(\mathbf{P},\mathbf{k})$$
$$+\mathbf{P}_{ll}\cdot\mathbf{k}\zeta_2(\mathbf{P},\mathbf{l})), \quad (13c)$$

$$\tfrac{1}{2}P_{ll}^2\zeta_2(\mathbf{P},\mathbf{l})=\sum\frac{[(\mathbf{k}\cdot\mathbf{l})-(\mathbf{P}\cdot\mathbf{k})]\mathbf{P}_{ll}\cdot\mathbf{k}}{D_2(\mathbf{k},\mathbf{l})}f_k^2(C(\mathbf{l})+C(\mathbf{k}))$$
$$+\sum\frac{(\mathbf{k}\cdot\mathbf{l})(\mathbf{P}_{ll}\cdot\mathbf{k})}{D_2(\mathbf{k},\mathbf{l})}f_k^2(1+\zeta_1(\mathbf{P},\mathbf{l})+\zeta_1(\mathbf{P},\mathbf{k}))$$
$$+\sum\frac{\mathbf{P}_{ll}\cdot\mathbf{k}}{D_2(\mathbf{k},\mathbf{l})}f_k^2(\mathbf{P}_{kl}\cdot\mathbf{l}\zeta_2(\mathbf{P},\mathbf{k})$$
$$+\mathbf{P}_{ll}\cdot\mathbf{k}\zeta_2(\mathbf{P},\mathbf{l})), \quad (13d)$$

where

$$C(\mathbf{k})=-[2\mathbf{P}\cdot\mathbf{k}/D_1(\mathbf{k})][1-\xi+\zeta_1(\mathbf{P},\mathbf{k})]$$
$$+[2/D_1(\mathbf{k})][k^2\zeta_1(\mathbf{P},\mathbf{k})-P_{kl}^2\zeta_2(\mathbf{P},\mathbf{k})].$$

To obtain $E_v(P)$ to order P^2 it turns out to be sufficient to expand ξ, ζ_1, and ζ_2:

$$\zeta_1(\mathbf{P},\mathbf{l})=\eta(l)+\mathbf{P}\cdot\mathbf{l}\eta_1(l)+(\mathbf{P}\cdot\mathbf{l})^2\eta_2(l)+P_{ll}^2\eta_3(l), \quad (14a)$$

$$\zeta_2(\mathbf{P},\mathbf{l})=\gamma(l)+\mathbf{P}\cdot\mathbf{l}\gamma_1(l), \quad (14b)$$

$$\xi(P)=\xi(0), \quad (14c)$$

where the η and γ functions in (14a) and (14b) are spherically symmetric and independent of $\mathbf{P}$.

Inserting (14a) and (14b) into (13b)–(13d) and equating terms of the same order in $\mathbf{P}\cdot\mathbf{l}$, P_{ll}^2, and $(\mathbf{P}\cdot\mathbf{l})P_{ll}^2$ gives a set of coupled integral equations for the etas and the gammas. These equations are quite complicated and will not be reproduced here. An important point is that all 14 nontrivial distinct angular integrals which appear can be done analytically, and the resulting coupled system of linear one-dimensional integral equations can be solved iteratively by machine.

The iteration proceeds in steps. First the equation for η is solved and $E_v(0)$ found as discussed in I. Then the two coupled equations for η_1 and γ are solved by iteration using an initial guess for ξ; ξ is computed from (13b) after each solution for η_1 and γ_1 is found. When ξ becomes stable the three coupled equations for η_2, η_3, and γ_1 are iterated using the already established values for ξ, η, η_1, and γ and a guessed value for $[E_v(P)-E_v(0)]/P^2$. Computation of a current value for $[E_v(P)-E_v(0)]/P^2$ is alternated with iteration of the equations for η_2, η_3, and γ_1 until $[E_v(P)-E_v(0)]/P^2$ becomes stable. The final result $(m^*/m)_v$ is then computed from (10).

III. RESULTS

In Table I we compare the effective mass deduced from (11) with the fourth-order perturbation result[15]

$$(m^*/m)_{\text{we4}}=1+\tfrac{1}{6}\alpha+\alpha^2$$
$$\times[7/36-\tfrac{5}{8}\sqrt{2}+\tfrac{4}{3}\ln(1+\sqrt{2})-\tfrac{2}{3}\ln 2]$$
$$\cong 1+\tfrac{1}{6}\alpha+0.0236276\alpha^2 \quad (15)$$

and Langreth's[16] expression

$$(m^*/m)_L=(1-0.0008\alpha^2)/(1-\tfrac{1}{6}\alpha+0.0034\alpha^2). \quad (16)$$

Equation (16) was devised in order to provide a convenient and accurate interpolation between (15) (for $\alpha\ll 1$) and the values of m^*/m given by Feynman's theory[13] for $\alpha\leq 5$. [Actually Feynman's theory comes remarkably close to (15) in the weak coupling limit.]

Inspection of Table I shows that over the range of α for which we expect our variational calculation to be quite accurate (say, $\alpha<3.5$—see I), our effective masses lie slightly higher than (15) and lower than (16). From one point of view we can say that our variational results certify the accuracy of fourth-order perturbation theory for the effective mass for $\alpha\leq 3$. The validity of the variational calculation, in turn, is supported by the calculation in I of the mean number $\bar{N}$ of virtual phonons in the field. It is shown in I that $\bar{N}$, which increases monotonically with α, becomes equal to 2 for $\alpha\sim 3.3$.

It is very interesting to investigate the accuracy of (9), which, being derived from ansatz (7a), approaches correctly the exact effective mass only to order α and not to order α^2 as $\alpha\to 0$. We would hope that, for small α, Eq. (9), being a variational result, would pick up part of the correction to order α^2 in (15) even though (7a) neglects two-phonon correlations. Expanding (9) to order α^2 we obtain the approximation to E_{we4}: $m^*/m=1+\frac{1}{6}\alpha+0.01389\alpha^2$. Comparison with (15) shows

[16] D. C. Langreth, Phys. Rev. **159**, 717 (1967). Owing to a typographical error, (16) is misquoted in Ref. 1.

that Haga's ansatz, (7a), produces almost 60% of the exact α^2 term.

This result suggests that if we wish to estimate the coefficient of the α^3 term in the perturbation expansion of m^*/m in powers of α, we could reasonably use the corresponding coefficient obtained from the expansion of $(m^*/m)_v$ in powers of α even though we have neglected three-phonon correlations. To evaluate this coefficient we compute C for $0.1<\alpha<0.5$:

$$C=\{E_v(P)-E_v(0)-[E(P)-E(0)]_{\text{wc4}}\}/\alpha^3P^2, \quad (17)$$

where $[E(P)-E(0)]_{\text{wc4}}$ is the polaron-excitation energy correct to order α^2 in the weak-coupling limit.[15] We then expand $P^2/\{[E(P)-E(0)]_{\text{wc4}}+C\alpha^3P^2\}$ to order α^3 and obtain our estimate:

$$(m^*/m)_{\text{est}}=1+\tfrac{1}{6}\alpha+0.0236276\alpha^2+0.0014\alpha^3. \quad (18)$$

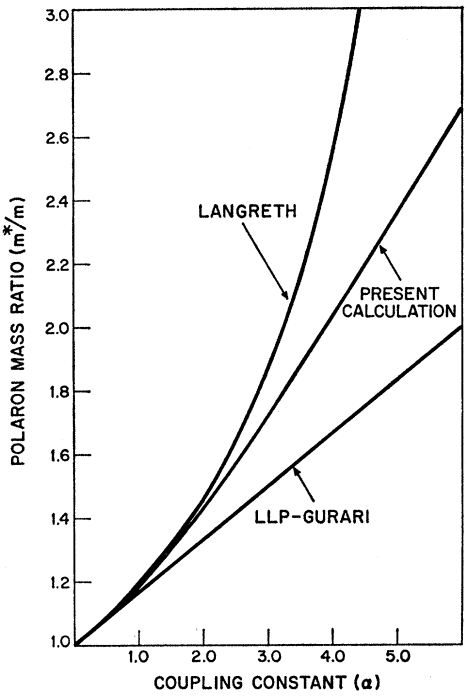

FIG. 1. Comparison of the ratio of the polaron mass to the band mass m^*/m, as calculated from Eqs. (16) (Langreth), (13) and (10) (present calculation), and (19) (LLP-Gurari).

The high accuracy of fourth-order perturbation theory for $\bar{N}<2$ suggests that sixth-order perturbation theory (order α^3) should have good accuracy for $\bar{N}<3$ or $\alpha\lesssim 4.5$. In the absence of knowledge of m^*/m to order α^3, we believe that (18) should provide a satisfactory estimate of m^*/m for $\alpha<4.5$.

We find that $(m^*/m)_{\text{est}}$ lies considerably closer to $(m^*/m)_v$ than to $(m^*/m)_L$ for $0<\alpha<5$.

In Fig. 1 we give a graphical comparison of $(m^*/m)_v$, $(m^*/m)_L$, and the lowest-order weak-coupling or LLP-Gurari result:

$$(m^*/m)_{\text{LLP-G}}=1+\tfrac{1}{6}\alpha. \quad (19)$$

In a more ambitious calculation one might have attempted to assume f_k is P-dependent and written a variational equation for $f_k(P)$ instead of assuming (6). For the ansatz (7a) we find that this procedure leads to the LLP-Gurari theory, which gives a less accurate energy spectrum than the Haga theory. We do not know what the results of a more general choice for f_k would be for ansatz (7b) but we doubt that significant improvement could be obtained.

Finally we remark that Haga has studied ansatz (7b) for the exactly soluble one-dimensional Gross model.[12] At a coupling constant corresponding to[17] $\alpha=2$ he finds extremely close agreement between the exact and variational solutions for small P.

ACKNOWLEDGMENT

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[17] J. Devreese and R. Evrard, Phys. Status Solidi 3, 2133 (1963).