Self-consistent equation-of-motion approach for polarons*

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The polaron problem is treated in a self-consistent manner. The treatment is based on Heisenberg's equations of motion starting from a trial expression for the electron position which includes only one real phonon but any number of virtual phonons. Numerical results for the polaron effective mass and the optical-absorption coefficient are given for electron-LO-phonon coupling strengths ranging from 0 to 4.5. The results are discussed and compared with those obtained by Feynman and by Lee, Low, and Pines.

INTRODUCTION

The static (effective mass, ground-state energy) and dynamical (optical absorption, mobility, response to a magnetic field) properties of free Fröhlich polarons have been studied using a relatively large number of theoretical methods. At weak electron–phonon coupling (small α) energy and mass were obtained in Refs. 1–4, while Refs. 5–9 treat mobility and other response properties. At strong coupling (large α) the static polaron properties were calculated in Refs. 10–12 and dynamical response was treated in Refs. 13 and 14. For a more complete list of references and a review on polarons see Refs. 15 and 16.

Until now only one method has been able to bridge the gap between weak and strong coupling in a natural way and to treat accurately intermediate coupling. It is the path-integral method as first introduced by Feynman[17] and Feynman and co-workers[18,19] and applied by others.[20–23] Although the path-integral formulation of the quadratic approximation is a powerful device in polaron theory, there are different reasons to set up alternative schemes to treat the polaron at all α:

(a) The path-integral formalism does not allow for a treatment of spin. This aspect might be important for the generalization of polaron theory to problems in both elementary-particle physics and solid-state physics (e.g., magnetism, Raman spin-flip, etc.).

(b) Furthermore some approximations have been made in the path-integral method (to study the polaron mobility problem for example) which might be easier to generalize in other formalisms.

(c) The path-integral formulation represents an alternative description of quantum mechanics which is less familiar to solid-state physicists than the Schrödinger or Heisenberg formalism.

For all these reasons it remains useful to formulate a polaron theory valid at all coupling strengths α in a more conventional language of quantum mechanics.[24] In this paper a self-consistent treatment for the polaron problem is presented which starts from the Heisenberg equations of motion and which is used here to derive the effective mass m^* and the optical properties of the polaron at all α (the weak- and strong-coupling limits are treated analytically, the intermediate coupling is treated numerically up to $\alpha \approx 4$). In a subsequent paper[25] the mobility is derived (with the Boltzmann equation) using the formalism explained here.

In Sec. I the equations of motion (with elimination of the phonon variables) for the polaron are written and the weak-coupling results are reviewed.

In Sec. II a trial form for the force (operator) $d\vec{\mathfrak{p}}(t)/dt$ acting on the electron is derived by iterating the equation for $d\vec{\mathfrak{p}}(t)/dt$ [Eq. (20)] with $d\vec{\mathfrak{p}}(t)/dt = 0$ as starting expression. This trial expression for $d\vec{\mathfrak{p}}(t)/dt$ contains a function $f(k)$, which depends on m^*, the polaron effective mass, and which can be considered as a generalization to all coupling strengths of the well-known f_k derived by Lee, Low, and Pines.[3]

In Sec. III this function $f(k)$ is determined by the condition that the polaron equation of motion be self-consistently satisfied. This leads to an integral equation for $f(k)$ which is solved formally in Secs. IV and V. A justification based on continued-fraction theory is given in favor of the approximation used in this solution.

In Sec. VI m^*, and the self-consistency parameters A and B are obtained analytically for $\alpha \to 0$ and $\alpha \to \infty$ and numerically for $0 < \alpha < 4.5$. The formalism developed in Secs. I–VI is valid at temperatures low enough so that the probability of two-phonon excitation remains negligible.

In Sec. VII the expression for the operator $\vec{\mathfrak{p}}(t)$ is used to calculate the optical-absorption coefficient Γ at zero temperature.

A number of appendixes are added to facilitate

the reading of the present paper. The results for the effective mass and the optical absorption are analyzed in the discussion. For $0<\alpha<4$ and $\alpha\to\infty$, our results are quite close to those obtained with the path-integral method.

I. EQUATIONS OF MOTION FOR THE POLARON

The equations-of-motion method was used to study the polaron in the classical case[26] and in the weak-coupling limit for the quantum case.[27,28] Let us reformulate the main features of this method. It is well known[15,16] that the polaron Hamiltonian can be written in the form

$$H_F = H_0 + H_1 , \tag{1}$$

where H_0 is the Hamiltonian for the electron and the longitudinal-optical phonons in the absence of interaction and H_1 is the electron–phonon coupling Hamiltonian.

Explicitly:

$$H_0 = \frac{p^2}{2m} + \sum_{\vec{k}} \hbar\omega\, a_{\vec{k}}^{\dagger} a_{\vec{k}} , \tag{2}$$

$$H_1 = \sum_{\vec{k}} (V_k a_{\vec{k}} e^{i\vec{k}\cdot\vec{r}} + V_k^* a_{\vec{k}}^{\dagger} e^{-i\vec{k}\cdot\vec{r}}) , \tag{3}$$

where

$$V_k = i(4\pi/V)^{1/2}\alpha^{1/2}\hbar\omega(\hbar/2m\omega)^{1/4}k^{-1} . \tag{4}$$

In these expressions, $\vec{r}$ and $\vec{p}$ denote the position and momentum of the particle, V is the volume of the crystal, ω is the longitudinal-optical phonon frequency, while $a_{\vec{k}}$ and $a_{\vec{k}}^{\dagger}$ are the phonon annihilation and creation operators whose commutation relation is

$$[a_{\vec{k}}, a_{\vec{k}'}^{\dagger}] = \delta_{\vec{k},\vec{k}'} . \tag{5}$$

The notation α is used for Fröhlich's coupling constant, defined as

$$\alpha = \frac{e^2}{\hbar c}\left(\frac{mc^2}{2\hbar\omega}\right)^{1/2}\left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0}\right) , \tag{6}$$

where e is the charge of the electron and ϵ_0 and ϵ_∞ are the static and high-frequency dielectric constants, respectively.

An adiabatic switching will be used from now on. This means that the electron–phonon coupling is applied adiabatically from $t=t_0$ ($t_0\to-\infty$) to $t=0$. Thus the Fröhlich Hamiltonian H_F is replaced by

$$H = H_0 + e^{-\epsilon|t|} H_1 , \tag{7}$$

where, after all calculations are performed, the positive quantity ϵ tends to zero. This new Hamiltonian depends explicitly on time.

The wave function $|t\rangle$ at time t can be related to the wave function $|t_0\rangle$ at time t_0 by means of the evolution operator $U(t, t_0)$. The Heisenberg operators

$$A(t) = e^{iH_0 t/\hbar}\, U^{\dagger}(t, t_0) A(t, t_0)\, e^{-iH_0 t/\hbar} \tag{8}$$

obey the equation of motion

$$\dot{A}(t) = (i/\hbar)[H(t), A(t)] , \tag{9}$$

where $H(t)$ is the Hamiltonian expressed in terms of the Heisenberg operators.

For the polaron system, one obtains

$$\frac{da_{\vec{k}}(t)}{dt} = -i\omega a_{\vec{k}}(t) - i\, e^{-\epsilon|t|}(V_k^*/\hbar)\, e^{-i\vec{k}\cdot\vec{r}(t)} , \tag{10a}$$

$$\frac{d\vec{r}(t)}{dt} = \vec{p}(t)/m , \tag{10b}$$

$$\frac{d\vec{p}(t)}{dt} = -i\, e^{-\epsilon|t|}\sum_{\vec{k}} \vec{k}(V_k a_{\vec{k}} e^{i\vec{k}\cdot\vec{r}(t)} - V_k^* a_{\vec{k}}^{\dagger} e^{-i\vec{k}\cdot\vec{r}(t)}) . \tag{10c}$$

A formal solution of Eqs. (10a) and (10b) can be obtained by means of the method of variation of the constant of integration. The procedure has been described in previous papers.[26,27] It leads to

$$a_{\vec{k}}(t) = a_{\vec{k}}(t_0)\, e^{-i\omega(t-t_0)} - i(V_k^*/\hbar)\, e^{-i\omega t} \times \int_{t_0}^{t} dt'\, e^{i\omega t'}\, e^{-\epsilon|t'|}\, e^{-i\vec{k}\cdot\vec{r}(t')} . \tag{11}$$

Now, at t_0 far in the past, the phonons are decoupled from the electron so that

$$a_{\vec{k}}(t_0) = a_+(\vec{k})\, e^{-i\omega t_0} , \tag{12}$$

where $a_+(\vec{k})$ is an annihilation operator for "incoming solutions" as introduced in scattering theory. This means that, if $|\Psi\rangle$ is an eigenstate of the Hamiltonian H_F with energy E and total momentum $\vec{P}$, the state $a_+^{\dagger}(\vec{k})|\Psi\rangle$ is also an eigenstate with energy $E+\hbar\omega$ and total momentum $\vec{P}+\hbar\vec{k}$. In particular, if $|\Psi\rangle$ is the polaron ground state, then $a_+^{\dagger}(\vec{k})|\Psi\rangle$ is a stationary solution describing the collision between a phonon $\vec{k}$ and the polaron.

In terms of these operators, the formal solution (11) can be written as ($t_0\to-\infty$)

$$a_{\vec{k}}(t) = a_+(\vec{k})\, e^{-i\omega t} - i(V_k^*/\hbar)\, e^{-i\omega t} \times \int_{-\infty}^{t} dt'\, e^{i\omega t'}\, e^{-\epsilon|t'|}\, e^{-i\vec{k}\cdot\vec{r}(t')} . \tag{13}$$

Let us now replace $a_{\vec{k}}(t)$ and $a_{\vec{k}}^{\dagger}(t)$ in Eq. (10c) by their expressions deduced from Eq. (13). This leads to the following integro-differential equation:

$$\frac{d\vec{p}(t)}{dt} = -i\, e^{-\epsilon|t|}\sum_{\vec{k}} \vec{k} V_k\, e^{i[\vec{k}\cdot\vec{r}(t)-\omega t]} a_+(k) - e^{-\epsilon|t|}\sum_{\vec{k}} \vec{k}\,\frac{|V_k|^2}{\hbar}\, e^{i[\vec{k}\cdot\vec{r}(t)-\omega t]} \times \int_{-\infty}^{t} dt'\, e^{-\epsilon|t'|}\, e^{-i[\vec{k}\cdot\vec{r}(t')-\omega t']} + \text{H.c.} , \tag{14}$$

where H.c. denotes the Hermitian conjugate of the first two terms on the right-hand side (rhs) of Eq. (14). It is easy to show that this relation expresses the conservation of the total momentum. Indeed, it is also possible to obtain Eq. (14) by introducing the solu-

tion (13) for $a_{\vec{k}}(t)$ [and for $a_{\vec{k}}^{\dagger}(t)$] into the expression

$$\frac{d}{dt}\left(\vec{p}(t)+\sum_{\vec{k}}\hbar\vec{k}\,a_{\vec{k}}^{\dagger}(t)a_{\vec{k}}(t)\right)=0\ . \tag{15}$$

II. ITERATIVE SOLUTION

Equation (14) has been solved by iteration[27] starting from a free electron in translation, which leads to introduce

$$\vec{r}(t)=\vec{r}(t_0)+[\vec{p}(t_0)/m](t-t_0)$$

in the rhs of Eq. (14). The first iteration gives

$$\vec{p}(t)=\vec{p}(t_0)-\frac{e^{-2\epsilon|t|}}{\hbar}\sum_{\vec{k}}\vec{k}\,|V_k|^2(\gamma_{\vec{k}}^2+\epsilon^2)^{-1}$$
$$+\sum_{\vec{k}}[\vec{k}V_k(\gamma_{\vec{k}}+i\epsilon)^{-1}e^{-i\gamma_{\vec{k}}t}e^{i\vec{k}\cdot\vec{R}_0}a_+(\vec{k})+\text{H.c.}]\ , \tag{16}$$

where $\vec{R}_0=\vec{r}(t_0)-[\vec{p}(t_0)/m]\,t_0$ and H.c. denotes the Hermitian conjugate of the first term between brackets. The notation $\gamma_{\vec{k}}$ has been used for

$$\gamma_{\vec{k}}=\omega-\vec{k}\cdot\vec{p}(t_0)/m+\hbar k^2/2m\ . \tag{17}$$

Let us expand the second term of the rhs of Eq. (16) in a power series of $p(t_0)$. To first order, this gives

$$\vec{p}(t)=(m/m^*)\vec{p}(t_0)+\sum_{\vec{k}}[\vec{k}V_k(\gamma_{\vec{k}}+i\epsilon)^{-1}$$
$$\times e^{-\epsilon|t|}e^{-i\gamma_{\vec{k}}t}e^{i\vec{k}\cdot\vec{R}_0}a_+(\vec{k})+\text{H.c.}]\ , \tag{18}$$

where m^* is defined by

$$\frac{m^*}{m}=\left[1-\frac{2}{3}\sum_{\vec{k}}\frac{k^2|V_k|^2}{m\hbar}\left(\omega+\frac{\hbar k^2}{2m}\right)^{-3}\right]^{-1}\ . \tag{19}$$

The integration over $\vec{k}$ gives

$$m^*/m=(1-\tfrac{1}{6}\alpha)^{-1}\ , \tag{20}$$

which is the well-known perturbation result for the polaron effective mass.

As the total momentum

$$\vec{P}=\vec{p}(t)+\sum_{\vec{k}}\hbar\vec{k}\,a_{\vec{k}}^{\dagger}(t)a_{\vec{k}}(t) \tag{21}$$

is conserved, one can write

$$\vec{p}(t_0)=\vec{P}-\sum_{\vec{k}}\hbar\vec{k}a_+^{\dagger}(\vec{k})a_+(\vec{k})\ . \tag{22}$$

The second term of the rhs in Eq. (22) represents the momentum of the incoming real phonons. This contribution is subtracted from the total momentum. Therefore $\vec{p}(t_0)$ not only has the meaning of the electron momentum at $t=t_0$, but it is also the momentum of the polaron $\vec{P}^*$ in an incident plane wave (which is scattered by the real phonons) so that

$$\vec{p}(t)=\frac{m}{m^*}\vec{P}^*+\sum_{\vec{k}}[\vec{k}V_k(\gamma_{\vec{k}}+i\epsilon)^{-1}$$
$$\times e^{-i\gamma_{\vec{k}}t}e^{i\vec{k}\cdot\vec{R}_0}a_+(\vec{k})+\text{H.c.}]\ . \tag{23}$$

An interesting particular case occurs when there is no real phonon present in the system. In that case, the polaron momentum $\vec{P}^*$ is not changed by scattering with phonons. Equation (23) shows that the motion of the electron has two components. The first one is a translation and results from the motion of the polaron as a whole. The second component is an oscillation produced by the interaction with the real-phonon field.

In the remainder of this section we put $\epsilon=0$. The solution for the position operator of the electron is easily obtained from (23) by integration. It is

$$\vec{r}(t)=\vec{R}+\frac{\vec{P}^*}{m^*}t+\frac{i}{m}\sum_{\vec{k}}\vec{k}\left(V_k\frac{e^{-i\gamma_{\vec{k}}t}}{\gamma_{\vec{k}}^2}e^{i\vec{k}\cdot\vec{R}}a_+(\vec{k})\right.$$
$$\left.-V_k^*a_+^{\dagger}(\vec{k})\,e^{-i\vec{k}\cdot\vec{R}}\frac{e^{i\gamma_{\vec{k}}t}}{\gamma_{\vec{k}}^2}\right), \tag{24}$$

where $\vec{R}$ is the position of the polaron center at time $t=0$, as given by the first iteration

$$\vec{R}=\vec{r}(t_0)-(\vec{P}^*/m^*)\,t_0$$
$$-\frac{i}{m}\sum_{\vec{k}}\frac{\vec{k}}{\gamma_{\vec{k}}^2}[V_k\,e^{-i\gamma_{\vec{k}}t_0}e^{i\vec{k}\cdot\vec{R}_0}a_+(\vec{k})$$
$$-V_k^*e^{i\gamma_{\vec{k}}t_0}a_+^{\dagger}(\vec{k})\,e^{-i\vec{k}\cdot\vec{R}_0}]\ . \tag{25}$$

This variable $\vec{R}$ and the polaron momentum $\vec{P}^*$ are canonically conjugated (cf. Ref. 27) so that

$$i[P_j^*,R_l]=\hbar\delta_{jl}\quad(j,l=x,y,z)\ . \tag{26}$$

Equation (24) will be used in Sec. III to define a trial expression for $\vec{r}(t)$ which is the starting expression for a self-consistent calculation.

III. SELF-CONSISTENT APPROACH

The solution (24) obtained above by iteration is valid for small coupling strengths. If the coupling strength is increased, it is expected that one or more resonances show up in the oscillatory part of the expression of the electron position $\vec{r}(t)$. These resonances are related to the possibility of excitation of the electron in the potential well caused by the induced polarization.

These excitations have been analyzed by different authors.[11,29,30] Their effects on the absorption of electro-magnetic radiation at strong coupling has been studied in detail by the present authors.[13] As one of the authors (J.D.) and co-workers[16,23] have shown, a resonance involving lattice relaxation also appears in the optical spectrum of Feynman's model for polarons.

Although to treat these resonances one has to include a substantial number of virtual phonons, it seems sufficient to take into account one real phonon to describe them and to obtain the physical properties of polarons at all coupling. Thus, it

can be expected that a solution of the type (24), but in addition allowing for resonances, constitutes a valid approximation. A supplementary argument for a one real-phonon approximation can be found in the calculation of Feynman *et al.*[18] (hereafter referred to as FHIP) which is generally considered as an accurate polaron description. Indeed, in the FHIP approach, the real part of the polaron impedance z (Ω) shows maxima for energies equal to a multiple of the resonance energy plus one LO phonon only. This indicates that, in Feynman's model, polarons interact with one real phonon at most.

On the other hand, a calculation of the optical-absorption coefficient has been performed in Ref. 23, starting from $\mathrm{Re}(z^{-1})$. The results of this treatment confirm the dominant role played by the interaction with one real phonon in the optical spectrum.

These considerations lead us to propose the following approach. We start from a trial expression for the position operator of the electron of the type

$$\vec{r}(t)=\vec{R}+\frac{\vec{P}^*}{m^*}t+\frac{i}{m}\sum_{\vec{k}}\vec{k}\left(f_{\vec{k}}\frac{e^{-i\gamma_{\vec{k}}t}}{\gamma_{\vec{k}}}e^{i\vec{k}\cdot\vec{R}}a_+(\vec{k})-f_{\vec{k}}^*a_+^\dagger(\vec{k})\,e^{-i\vec{k}\cdot\vec{R}}\frac{e^{i\gamma_{\vec{k}}t}}{\gamma_{\vec{k}}}\right), \tag{27}$$

where $f_{\vec{k}}$, $\gamma_{\vec{k}}$, and the polaron effective mass m^* are c numbers to be determined by self-consistency.

The corresponding force acting on the electron is

$$\frac{d\vec{p}(t)}{dt}=m\frac{d^2\vec{r}}{dt^2}=-i\sum_{\vec{k}}\gamma_{\vec{k}}\vec{k}\left[f_{\vec{k}}e^{-i\gamma_{\vec{k}}t}e^{i\vec{k}\cdot\vec{R}}a_+(\vec{k})-f_{\vec{k}}^*e^{i\gamma_{\vec{k}}t}a_+^\dagger(\vec{k})\,e^{-i\vec{k}\cdot\vec{R}}\right]. \tag{28}$$

It is assumed now that the translation and oscillation components of the motion of the electron are independent and, therefore, that they commute. This enables us to write

$$e^{i\vec{k}\cdot\vec{r}(t)}=e^{i\vec{k}[\vec{R}+(\vec{P}^*/m^*)t]}\exp\left(-\sum_{\vec{k}'}\frac{\vec{k}\cdot\vec{k}'}{m}\times\left[f_{\vec{k}'}\gamma_{\vec{k}'}^{-1}e^{-i\gamma_{\vec{k}'}t}e^{i\vec{k}'\cdot\vec{R}}a_+(\vec{k}')-f_{\vec{k}'}^*a_+^\dagger(\vec{k}')\,e^{-i\vec{k}'\cdot\vec{R}}\gamma_{\vec{k}'}^{-1}e^{i\gamma_{\vec{k}'}t}\right]\right). \tag{29}$$

Then the phonon operators are ordered in such a way that the creation operators appear at the left of the annihilation operators. Disentangling the translation term gives

$$e^{i\vec{k}\cdot[\vec{R}+(\vec{P}^*/m^*)t]}=e^{i(\vec{k}\cdot\vec{P}^*/m^*)t}e^{i\vec{k}\cdot\vec{R}}e^{-i(\hbar k^2/2m^*)t}. \tag{30}$$

One then obtains

$$e^{i\vec{k}\cdot\vec{r}(t)}=e^{i(\vec{k}\cdot\vec{P}^*/m^*)t}e^{i\vec{k}\cdot\vec{R}}e^{-i(\hbar k^2/2m^*)t}\exp\left[-\frac{1}{2}\sum_{\vec{k}'}\left(\frac{\vec{k}\cdot\vec{k}'}{m}\right)^2\frac{|f_{\vec{k}'}|^2}{\gamma_{\vec{k}'}^2}\right]\times\exp\left(\sum_{\vec{k}'}\frac{\vec{k}\cdot\vec{k}'}{m}f_{\vec{k}'}^*\gamma_{\vec{k}'}^{-1}a_+^\dagger(\vec{k}')\,e^{-i\vec{k}'\cdot\vec{R}}e^{i\gamma_{\vec{k}'}t}\right)\exp\left(-\sum_{\vec{k}'}\frac{\vec{k}\cdot\vec{k}'}{m}f_{\vec{k}'}\gamma_{\vec{k}'}^{-1}\,e^{i\vec{k}'\cdot\vec{R}}a_+(\vec{k}')\,e^{-i\gamma_{\vec{k}'}t}\right). \tag{31}$$

Another expression that appears in the equation of motion is a product of exponentials of the type studied above, but measured at different times. The factors are written so that the creation operators for real phonons appear at the left of the annihilation operators. For this purpose, the commutation rule

$$e^a\,e^b=e^b\,e^a\,e^{[a,b]} \tag{32}$$

is used again. Here

$$a=-\sum_{\vec{k}'}\frac{\vec{k}\cdot\vec{k}'}{m}f_{\vec{k}'}\gamma_{\vec{k}'}^{-1}\,e^{i\vec{k}'\cdot\vec{R}}a_+(\vec{k}')\,e^{-\gamma_{\vec{k}'}t}, \tag{33a}$$

$$b=-\sum_{\vec{k}'}\frac{\vec{k}\cdot\vec{k}'}{m}f_{\vec{k}'}^*\gamma_{\vec{k}'}^{-1}a_+^\dagger(\vec{k}')\,e^{-i\vec{k}'\cdot\vec{R}}\,e^{i\gamma_{\vec{k}'}t'}, \tag{33b}$$

and

$$[a,b]=\sum_{\vec{k}'}\left(\frac{\vec{k}\cdot\vec{k}'}{m}\right)^2|f_{\vec{k}'}|^2\gamma_{\vec{k}'}^{-1}e^{-i\gamma_{\vec{k}'}(t-t')}. \tag{33c}$$

After ordering the operators, and introducing the results of the disentangling Eqs. (31), (32) with (33), into Eq. (14), one obtains

$$\frac{d\vec{p}(t)}{dt}=-ie^{-\epsilon|t|}\sum_{\vec{k}}\vec{k}V_k\exp\left[-i\left(\omega-\frac{\vec{k}\cdot\vec{P}^*}{m^*}+\frac{\hbar k^2}{2m^*}\right)t\right]e^{-F(\vec{k},0)/2}\exp\left(\sum_{\vec{k}'}\frac{\vec{k}\cdot\vec{k}'}{m}f_{\vec{k}'}^*\gamma_{\vec{k}'}^{-1}a_+^\dagger(\vec{k}')\,e^{i\vec{k}'\cdot\vec{R}}e^{i\gamma_{\vec{k}'}t}\right)\times\exp\left(-\sum_{\vec{k}'}\frac{\vec{k}\cdot\vec{k}'}{m}f_{\vec{k}'}\gamma_{\vec{k}'}^{-1}e^{i\vec{k}'\cdot\vec{R}}a_+(\vec{k}')e^{-i\gamma_{\vec{k}'}t}\right)e^{i\vec{k}\cdot\vec{R}}a_+(\vec{k})-\sum_{\vec{k}}\vec{k}\,\frac{|V_k|^2}{\hbar}e^{i(\vec{k}\cdot\vec{P}^*/m^*)t}\times e^{-2\epsilon|t|}\int_{-\infty}^{0}d\tau\,e^{-\epsilon|\tau|}\exp\left(\sum_{\vec{k}'}\frac{\vec{k}\cdot\vec{k}'}{m}f_{\vec{k}'}^*\gamma_{\vec{k}'}^{-1}(1-e^{i\gamma_{\vec{k}'}\tau})a_+^\dagger(\vec{k}')\,e^{-i\vec{k}'\cdot\vec{R}}e^{i\gamma_{\vec{k}'}t}\right)$$

$$\times\exp\left(-\sum_{\vec{k}'}\frac{\vec{k}\cdot\vec{k}'}{m}-f_{\vec{k}'}\gamma_{\vec{k}'}^{-1}(1-e^{-i\gamma_{\vec{k}'}\tau})\,e^{i\vec{k}'\cdot\vec{R}}a_+(\vec{k}')\,e^{-i\gamma_{\vec{k}'}t}\right)$$

$$\times\exp\left[i\left(\omega-\frac{\vec{k}\cdot\vec{P}^*}{m^*}+\frac{\hbar k^2}{2m^*}\right)\tau\right]\exp[F(\vec{k},\tau)-F(\vec{k},0)]\,e^{-i(\vec{k}\cdot\vec{P}^*/m^*)t}+\text{H.c.}\,, \tag{34}$$

where $\tau=t'-t$ and

$$F(\vec{k},\tau)=\sum_{\vec{k}'}\left(\frac{\vec{k}\cdot\vec{k}'}{m}\right)^2|f_{\vec{k}'}|^2\gamma_{\vec{k}'}^{-2}\,e^{i\gamma_{\vec{k}'}\tau}\,. \tag{35}$$

In the derivation of Eq. (34) it has been assumed that $\vec{R}$ and $\vec{P}^*$ commute with the oscillation terms, i.e., the terms containing the field operators $a_+(\vec{k})$ and $a_+^\dagger(\vec{k})$. From the physical point of view, this approximation seems reasonable, since a measure of the polaron position or momentum does probably not modify its internal oscillations. However the property seems difficult to prove mathematically. Indeed, to our knowledge, the relation between $\vec{R}$ and the field operators is not known.

The principle of our approach is to require that the matrix elements of Eq. (34) between states with zero and one real phonon be identical with the matrix elements obtained from the initial expression (28) of $d\vec{p}(t)/dt$. For instance one can calculate the matrix element between the ground state $\langle 0|$ and a state $|1_{\vec{k}}\rangle$ containing one real phonon with wave vector $\vec{k}$.

Relations of the type,

$$\langle 0|\exp\left(\sum_{\vec{k}'}Q_{k'}^*a_+^\dagger(\vec{k}')\,e^{-i\vec{k}'\cdot\vec{R}}\right)=\langle 0|\,,$$

$$\exp\left(-\sum_{\vec{k}'}Q_{k'}\,e^{i\vec{k}'\cdot\vec{R}}a_+(\vec{k}')\right)|1_{\vec{k}}\rangle=-Q_k\,e^{i\vec{k}\cdot\vec{R}}|0\rangle+|1_{\vec{k}}\rangle \tag{36a}$$

are used. In these relations

$$Q_{k'}=\frac{\vec{k}''\cdot\vec{k}'}{m}f_{\vec{k}'}\gamma_{\vec{k}'}^{-1}\,e^{-i\gamma_{\vec{k}'}t} \tag{36b}$$

or

$$Q_{k'}=\frac{\vec{k}''\cdot\vec{k}'}{m}f_{\vec{k}'}\gamma_{\vec{k}'}^{-1}(1-e^{-i\gamma_{\vec{k}'}\tau})\,e^{-i\gamma_{\vec{k}'}t} \tag{36c}$$

depending on the term considered in Eq. (34).

Because it is assumed that the translations do not interfere with the oscillations, the factor $\exp[-i(\vec{k}\cdot\vec{P}^*/m^*)\tau]$ commutes with the oscillatory part of the equation. This property is used to order the factors in such a way that $\vec{P}^*$ be applied to the initial state which is an eigenstate of $\vec{P}^*$. Therefore this latter takes the corresponding eigenvalue and can be treated as a c number from here on. The matrix element of the left-hand side (lhs) of Eq. (34) is easily evaluated using Eq. (28).

With all these considerations, one obtains the following equation:

$$\vec{k}f_{\vec{k}}\gamma_{\vec{k}}e^{-i\gamma_{\vec{k}}t}=V_k\vec{k}\,e^{-F(\vec{k},0)/2}\exp\left[-i\left(\omega-\frac{\vec{k}\cdot\vec{P}^*}{m^*}+\frac{\hbar k^2}{2m^*}\right)t\right]-if_{\vec{k}}\gamma_{\vec{k}}^{-1}e^{-i\gamma_{\vec{k}}t}\sum_{\vec{k}'}\vec{k}'\,\frac{\vec{k}\cdot\vec{k}'}{m\hbar}|V_{k'}|^2\int_{-\infty}^{0}d\tau\,(1-e^{-i\gamma_{\vec{k}}\tau})$$

$$\times\left\{\exp\left[i\left(\omega-\frac{\vec{k}'\cdot\vec{P}^*}{m^*}+\frac{\hbar k'^2}{2m^*}\right)\tau\right]\exp[F(\vec{k}',\tau)-F(\vec{k}',0)]\right.$$

$$\left.-\exp\left[-i\left(\omega-\frac{\vec{k}'\cdot\vec{P}^*}{m^*}+\frac{\hbar k'^2}{2m^*}\right)\tau\right]\exp[F^*(\vec{k}',\tau)-F(\vec{k}',0)]\right\}. \tag{37}$$

From the time dependence in Eq. (37), it immediately follows that

$$\gamma_{\vec{k}}=\omega-\frac{\vec{k}\cdot\vec{P}^*}{m^*}+\frac{\hbar k^2}{2m^*}\,, \tag{38}$$

which is the first result of our requirement of self-consistency. In this result for the frequency $\gamma_{\vec{k}}$ the recoil is taken by the polaron ($\hbar^2k^2/2m^*$) contrary to the result of perturbation theory [Eq. (17)] where the bare electron takes the recoil ($\hbar^2k^2/2m$). Of course the main difference between the present self-consistent treatment and the iteration method will be reflected in the resonant denominator of $f_{\vec{k}}$.

In this paper the calculation is restricted to the case of zero polaron momentum, $\vec{P}^*=0$.

After symmetry considerations explained in Appendix A, one obtains from Eq. (37) the following integral equation, which determines the function f_k:

$$f_k=\frac{V_k\,e^{-F(k,0)/2}}{\gamma_k}\left(1-\frac{i}{\gamma_k^2}[M(0)-M^*(0)-M(-\gamma_k)+M^*(\gamma_k)]\right)^{-1}, \tag{39}$$

where

$$M(z)=\frac{1}{3}\sum_{\vec{k}'}\frac{k'^2|V_{k'}|^2}{m\hbar}\,g(k',\gamma_{k'}+z)\,; \tag{40a}$$

furthermore

$$g(k,z)=\int_0^\infty d\tau\, e^{-iz\tau}\, e^{-\epsilon|\tau|}\exp[F^*(\vec{k},\tau)-\vec{F}(\vec{k},0)]\,, \tag{40b}$$

with

$$F(\vec{k},\tau)=k^2C(\tau) \tag{40c}$$

and

$$C(\tau)=\sum_{\vec{k}'}\frac{k'^2}{3m^2}|f_{k'}|^2\gamma_{k'}^{-2}\,e^{i\gamma_{k'}t}\,. \tag{40d}$$

Equation (39) is an integral equation, since the expression of $M(z)$ contains a sum over the f_k via Eqs. (40) and (35). It does not seem possible to solve Eq. (39) exactly at all α. We rather prefer to look for approximate solutions. The solution proposed in this paper is based on an approximate way of calculating $M(z)$.

Before describing this approximation in Sec. IV, we give an equation for the mass, the third parameter which together with f_k and γ_k is to be determined self-consistently.

The motion of the electron has two components: a component of translation due to the translation of the polaron as a whole and a component of oscillation representing the motion of the electron around the polaron center. Obviously the mean value of the oscillation component of the electron momentum is zero and the mean value of $\vec{p}(t)$ is due to the polaron translation. In the present work, the polaron effective mass is defined in such a way that the polaron velocity is $\vec{P}^*/m^*$. Therefore one must have

$$\langle\vec{p}(t)\rangle=(m/m^*)\,\vec{P}^*\,, \tag{41}$$

where the brackets $\langle\,\rangle$ denote the expectation value taken between states containing no real phonons.

This relation can be used to define the polaron mass m^* in the following way. Let us go back to Eq. (34), giving the force acting on the electron. The diagonal matrix element between states with no real phonons gives

$$\frac{d\vec{p}(t)}{dt}=-\sum_k \vec{k}\,\frac{|V_k|^2}{\hbar}\,e^{-\epsilon|t|}\int_{-\infty}^0 d\tau\, e^{i\gamma_k\tau}\,e^{-\epsilon|\tau|}\times\exp[F(\vec{k},\tau)-F(\vec{k},0)]+\text{c.c.}\,, \tag{42}$$

where the adiabatic switching, necessary here, has been reintroduced. With our notation (40b), this relation becomes

$$\left\langle\frac{d\vec{p}(t)}{dt}\right\rangle=-2\sum_{\vec{k}}\vec{k}'\,\frac{|V_k|^2}{\hbar}\,e^{-2\epsilon|t|}\,\mathrm{Re}g(\vec{k},\gamma_k)\,. \tag{43}$$

Integrating Eq. (43) from a time t_0, when the interaction is not yet applied, to the time $t=0$ gives

$$\langle\vec{p}(0)\rangle=\vec{P}^*-\frac{1}{\epsilon}\sum_k\frac{|V_k|^2}{\hbar}\,\mathrm{Re}g(k,\gamma_k)\,. \tag{44}$$

The operator $\vec{p}(t_0)$ is a constant of the motion and can be identified with the polaron momentum. The second term of the rhs of Eq. (44) is subtracted from the polaron momentum to give the mean electron momentum. Therefore this term represents the contribution of the cloud of virtual phonons to the polaron momentum. Comparing Eq. (44) with (41) gives

$$\frac{m^*-m}{m^*}\,\vec{P}^*=\frac{1}{\epsilon}\sum_{\vec{k}}\vec{k}\,\frac{|V_k|^2}{\hbar}\,\mathrm{Re}g(\vec{k},\gamma_k)\,, \tag{45}$$

which is the equation needed to determine the polaron mass m^*.

IV. APPROXIMATE CALCULATION OF $M(z)$

In the expression (40d) let us replace the sum by an integral. Since $f_{k'}$ is isotropic the integration over the angles is readily performed. This gives

$$C(\tau)=\frac{V}{6\pi^2}\int_0^\infty dk'\,\frac{k'^4}{m^2}\,|f_{k'}|^2\gamma_{k'}^{-2}\,e^{i\gamma_{k'}\tau}\,. \tag{46}$$

Then the frequency $\gamma_{k'}$ is taken as the new integration variable, leading to

$$C(\tau)=\int_\omega^\infty d\gamma_{k'}\,e^{i\gamma_{k'}\tau}\,W(\gamma_{k'})\,. \tag{47}$$

This appears as a spectrum of harmonic contributions with different frequencies $\gamma_{k'}$. The weight $W(\gamma_{k'})$ of the mode $\gamma_{k'}$ is

$$W(\gamma_{k'})=\frac{Vm^*}{6\pi^2m^2\hbar}\,|f_{k'}|^2\gamma_{k'}^{-2}\,[2m^*/\hbar(\gamma_{k'}-\omega)]^{3/2}\,. \tag{48}$$

The basic approximation in our method of solving Eq. (39) consists of replacing the frequency distribution $W(\gamma_{k'})$ [Eq. (48)] by a single frequency. This means that $W(\gamma_{k'})$ is approximated by

$$W(\gamma_{k'})=W\delta(\gamma_{k'}-\zeta)\,. \tag{49}$$

The frequency ζ and its weight W are chosen so that the first two moments μ_0^a and μ_1^a of the approximate spectrum (49) be equal to those of the self-consistent spectrum (48). Obviously the moments of the approximate spectrum are

$$\mu_0^a=W\,, \tag{50a}$$

$$\mu_1^a=W\zeta\,. \tag{50b}$$

Comparison between the moments of the approximate and self-consistent spectra gives

$$W=\int_\omega^\infty d\gamma_{k'}\,W(\gamma_{k'})=C(0)\,, \tag{51a}$$

$$\zeta=\frac{1}{C(0)}\int_\omega^\infty d\gamma_{k'}\,W(\gamma_{k'})\,\gamma_{k'}\,. \tag{51b}$$

Introducing the approximate spectrum (49) into (46) gives the approximate time dependence of $C(\tau)$

$$C(\tau) = C(0)\, e^{i\zeta\tau} . \tag{52}$$

A further justification of this approximation can be found in Appendix B.

Using relations (40) and (52), one has

$$g(k, z) = \int_0^\infty d\tau\, e^{-izt}\, e^{-\epsilon|\tau|} \exp[k^2 C(0)\,(e^{-i\zeta\tau} - 1)] \tag{53}$$

and

$$M(z) = \sum_{\vec{k}} \frac{1}{3} \int_0^\infty d\tau\, e^{-iz\tau}\, e^{-\epsilon|\tau|} \times \sum_{\vec{k}} \frac{k^2 |V_k|^2}{m\hbar}\, e^{-i\gamma_k\tau} \exp[k^2 C(0)\,(e^{-i\zeta\tau} - 1)] . \tag{54}$$

After integration over $\vec{k}$ this becomes

$$M(z) = \omega^3 \frac{\alpha}{3\sqrt{\pi}} \left(\frac{m^*}{m}\right)^{3/2} \int_0^\infty d\tau\, e^{-\epsilon|\tau|}\, e^{-i(1+z/\omega)\omega\tau} \times [C'(1 - e^{-i\zeta\tau}) + i\omega\tau]^{-3/2} , \tag{55a}$$

with

$$C' = C(0) r_0^{-2} m^*/m \tag{55b}$$

and

$$r_0 = (\hbar/2m\omega)^{1/2} .$$

In the subsequent analytical and numerical work it is convenient to introduce the following notations:

$$A = \frac{3\pi^2}{V} \left(\frac{m}{m^*}\right)^{5/2} \hbar^2 r_0 \mu_0 = C' \frac{3\pi^2}{V} \hbar^2 r_0^3 \left(\frac{m}{m^*}\right)^{7/2} , \tag{56a}$$

$$B = (\mu_1 - \omega\mu_0) \frac{3\pi^2}{V} r_0 \frac{\hbar^2}{\omega} \left(\frac{m}{m^*}\right)^{5/2} , \tag{56b}$$

so that

$$\zeta = \omega(1 + BA^{-1}) . \tag{56c}$$

V. SIMPLIFICATION OF $f(x)$ AND THE EXPRESSIONS FOR A, B, m^* IN TERMS OF $f(x)$

At this point the self-consistent determination of f_k, γ_k, and m^* is reduced to the self-consistent determination of A, B, and m^*. Our next purpose is to write f_k in a form which is more compact and suitable for numerical treatment. Therefore the different terms in the denominator of f_k [Eq. (39)] will be combined. This denominator is

$$1 - (i/\gamma_k^2)[M(0) - M^*(0) + M^*(\gamma_k) - M(-\gamma_k)] . \tag{57}$$

With the notation

$$\frac{m}{m^*}[C'(1 - e^{-i\omega\zeta\tau}) + i\omega\tau] = D(\omega\tau) , \tag{58}$$

one has

$$M^*(\gamma_k) - M(-\gamma_k) = +\, i \frac{2\alpha}{3\sqrt{\pi}} \omega^3 \int_0^\infty d\tau\, e^{-\epsilon|\tau|}\, e^{i\gamma_k\tau} \times \mathrm{Im}\{e^{+i\omega\tau}[D^*(\omega\tau)]^{-3/2}\} . \tag{59}$$

It follows that

$$M(0) - M^*(0) + M^*(\gamma_k) - M(-\gamma_k) = -\, i \frac{2\alpha}{3\sqrt{\pi}} \omega^3 \int_0^\infty d\omega\tau\, e^{-\epsilon|\tau|}(1 - e^{i\gamma_k\tau}) \times \mathrm{Im}\{[D^*(\omega\tau)]^{-3/2}\, e^{i\omega\tau}\} . \tag{60}$$

At this point we remark that the integrand on the rhs of Eq. (60) has the same analytical structure as the integrand of the function $\chi(\nu)$ defined by Feynman *et al.*[18] [see Eqs. (35a) and (36) in this reference].

Therefore we define

$$\chi(\Omega) = \frac{2\alpha}{3\sqrt{\pi}} \int_0^\infty du\, e^{-\epsilon|u|}(1 - e^{i\Omega u})\mathrm{Im}\{[D^*(u)]^{-3/2}\, e^{iu}\} . \tag{61}$$

Using Eqs. (39), (57), and (60), it then follows that

$$f_k = \frac{V_k \gamma_k\, e^{-F(\vec{k},0)/2}}{\gamma_k^2 - \omega^2 \chi(\gamma_k/\omega)} . \tag{62}$$

One now easily obtains,

$$A = \int_0^\infty \frac{dx\, |f(x)|^2\, x^{3/2}}{(1+x)^2} , \tag{63a}$$

$$B = \int_0^\infty \frac{dx\, |f(x)|^2\, x^{5/2}}{(1+x)^2} , \tag{63b}$$

with

$$|f(x)|^2 = \frac{(4\pi\alpha/V)(\hbar\omega)^2(\hbar/2m\omega)^{3/2}(1+x)^2\, e^{-C'x}(m/m^*)}{\omega^2 x\{[(1+x)^2 - \mathrm{Re}\chi(1+x)]^2 + [\mathrm{Im}\chi(1+x)]^2\}} , \tag{64}$$

where $\gamma_k = (1+x)\omega$.

To determine $f(x)$ in a self-consistent manner, one needs not only A and B, but also the effective mass m^*. Using Eq. (45) one obtains the following expression for m^*:

$$\frac{m^*}{m} = \left[1 - \frac{\alpha}{3\sqrt{\pi}} \left(\frac{m^*}{m}\right)^{1/2} \int_0^\infty \frac{dx\, x^2\, e^{-x}}{[C'(1 - e^{-\zeta x/\omega}) + x]^{3/2}}\right]^{-1} . \tag{65}$$

The details of the derivation of Eq. (65) are given in Appendix C.

Equations (63a), (63b), and (65) allow one, in principle, to determine A, B, and m^*.

VI. NUMERICAL AND ANALYTICAL EVALUATION OF THE PARAMETERS A, B, m^*

It is relatively straightforward to determine A, B, and m^* analytically for weak and strong coupling. Let us first consider the case of weak coupling. If $\alpha \to 0$ then C', $\mathrm{Im}\chi(\Omega)$ and $\mathrm{Re}\chi(\Omega)$ tend to zero like α. This gives (to order α)

$$A = B = \alpha\, \tfrac{1}{4}\pi^2 (r_0^3 \hbar^2/V) . \tag{66}$$

To obtain the expression for the mass, it is sufficient to consider the term $n=0$ in Eq. (C5), leading immediately to

$$m/m^* = 1 - \tfrac{1}{6}\alpha , \tag{67}$$

consistent with the results of perturbation theory.

For strong coupling we need to examine the asymptotic behavior of $\mathrm{Re}\chi(\Omega)$, $\mathrm{Im}\chi(\Omega)$, etc. As shown in Appendix D one has

$$\mathrm{Re}\chi(\Omega)=\frac{2\alpha}{3\sqrt{\pi}}\left(\frac{m^*}{m}\right)^{3/2}\frac{1}{C'^{3/2}} \quad (\alpha\to\infty). \tag{68}$$

Furthermore, from Ref. 23,

$$\mathrm{Im}\chi(\Omega)=\frac{2\alpha}{3}\left(\frac{m^*}{m}\right)^{3/2}(\Omega-1)^{1/2}\,e^{-C'(\Omega-1)} \quad (\alpha\to\infty). \tag{69}$$

One needs also the asymptotic behavior of

$$\Delta=\int_0^\infty \frac{x^{1/2}\,e^{-C'x}\,dx}{[(1+x)^2-\mathrm{Re}\chi(1+x)]^2+[\mathrm{Im}\chi(1+x)]^2} \tag{70}$$

to evaluate A, B, and m^* at large α [the integral with $x^{3/2}$ follows immediately from Eq. (70)].

It is shown in Appendix D that

$$\Delta=\frac{3\pi}{4\alpha y_0(m^*/m)^{3/2}} \quad (\alpha\to\infty), \tag{71}$$

where y_0 is the solution of

$$y_0^2=\mathrm{Re}\chi(y_0) \quad (\alpha\to\infty). \tag{72}$$

With Eq. (71) and the expressions (51), (55b), and (56a), it follows that

$$C'=\frac{4\alpha}{3\pi}\left(\frac{m^*}{m}\right)^{5/2}\Delta \quad (\alpha\to\infty). \tag{73}$$

Elimination of Δ between Eqs. (71) and (73) leads to

$$y_0=(1/C')(m^*/m) \quad (\alpha\to\infty). \tag{74}$$

With Eqs. (68) and (72) it follows that

$$C'=(9\pi/4\alpha^2)(m^*/m) \quad (\alpha\to\infty). \tag{75}$$

From Eqs. (74) and (75) one also has

$$y_0=4\alpha^2/9\pi \quad (\alpha\to\infty). \tag{76}$$

Furthermore, from the definitions of A and B one sees

$$BA^{-1}=y_0=4\alpha^2/9\pi \quad (\alpha\to\infty). \tag{77}$$

Now the equation for the mass (65) becomes

$$\frac{m^*}{m}=\left[1-\frac{2\alpha}{3\sqrt{\pi}}\left(\frac{m^*}{m}\right)^{1/2}\frac{1}{C'^{3/2}}\right]^{-1} \quad (\alpha\to\infty). \tag{78}$$

Indeed in the limit $\alpha\to\infty$ the integral in the rhs of (65) tends to $2/C'^{3/2}$ (see Appendix D) (taking $C'\to\infty$ if $\alpha\to\infty$ as a solution). The only physically acceptable strong coupling solution occurs when the denominator of Eq. (78) tends to zero. Eliminating C' from Eqs. (75) and (78) one has for this solution

$$m^*/m=16\alpha^4/81\pi^2 \quad (\alpha\to\infty). \tag{79}$$

This is the same as the Pekar result[11] and the Feynman asymptotic value.[17]

The parameters A, B, and C' are given as follows for large α:

$$C'=4\alpha^2/9\pi \quad (\alpha\to\infty), \tag{80a}$$

$$A=3\pi^2\left(\frac{9\pi}{4\alpha^2}\right)^6\frac{r_0^3\hbar^2}{V} \quad (\alpha\to\infty), \tag{80b}$$

$$B=3\pi^2\left(\frac{9\pi}{4\alpha^2}\right)^5\frac{r_0^3\hbar^2}{V} \quad (\alpha\to\infty). \tag{80c}$$

For intermediate coupling the calculation of A, B, and m^* has been performed numerically by going to self-consistency between the three equations (63a), (63b), and (65). (Self-consistency can be achieved in different ways, e.g., as was done here,

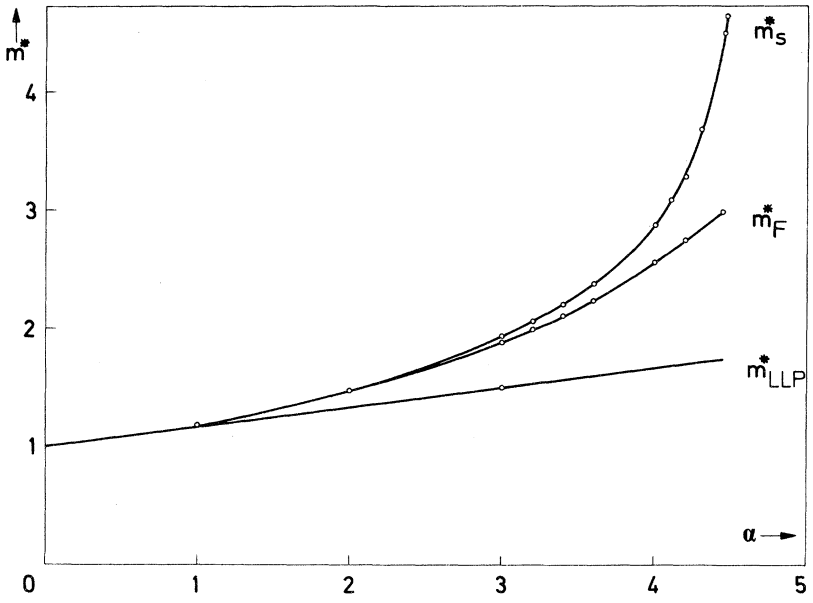

FIG. 1. Polaron effective mass as a function of the electron–phonon coupling strength α. m_S^*, present work; m_F^*, Feynman's result, m_{LLP}^*, Lee–Low–Pines result.

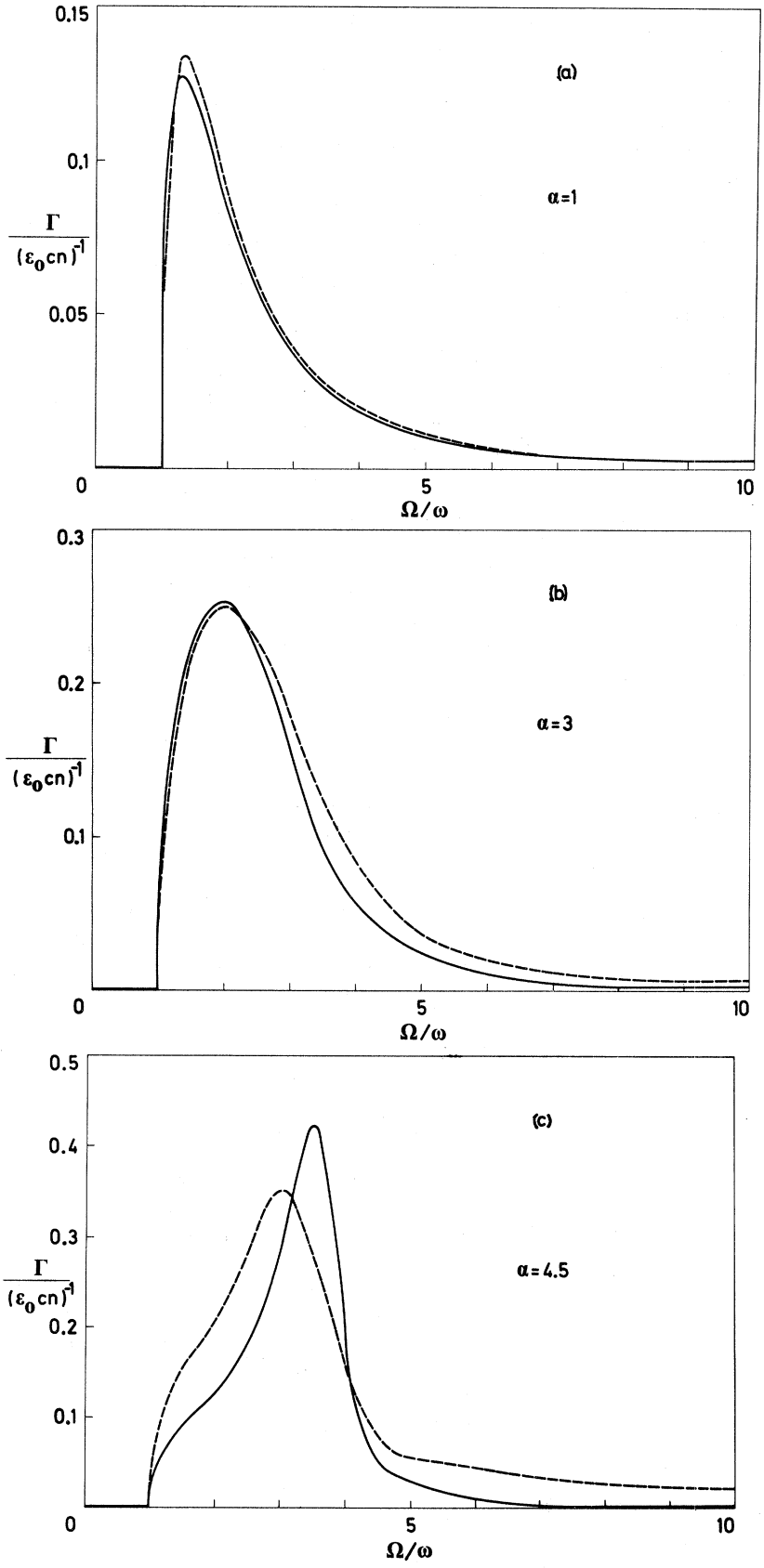

FIG. 2. Reduced optical absorption $\Gamma(\Omega)$ as a function of the frequency of the incident light measured in units ω. Full line, present work; dashed line, results of Ref. 23. The results are given for $\alpha=1$, $\alpha=3$, $\alpha=4.5$.

by starting from the weak-coupling value of A, B, and m^* and extrapolating these to larger α after self-consistency is achieved for smaller α.)

In Fig. 1 the mass m^* as obtained from the present theory is plotted and compared with the results obtained by Feynman.[17] It is seen that for α ranging from 0 to about 3.5 the effective mass m^* obtained here is very close to the value obtained by Feynman. Furthermore for large α our result has the correct asymptotic behavior.

Once the parameters A, B, and m^* are obtained it is possible to derive other physical quantities like the self-energy of the polaron and the transition probabilities determining optical absorption or mobility. In Sec. VII the optical absorption will be discussed.

VII. OPTICAL ABSORPTION

The formalism developed here is suitable to calculate the optical absorption coefficient K of polarons in the independent particle approximation, which can easily be expressed once $p(t)$, the electron momentum operator, which follows easily from Eq. (78), is known.[9,27]

$$K(\Omega)=\frac{4\pi N}{\sqrt{\epsilon_\infty}}\frac{e^2}{\hbar c}\frac{\hbar}{m\omega}\Gamma(\Omega)\,, \tag{81a}$$

with

$$\Gamma(\Omega)=\frac{\omega}{m\hbar\Omega}\,\mathrm{Re}\int_{-\infty}^{0}dt\,e^{-i(\Omega+i\epsilon)t}\langle P^*|\,p_z(0)p_z(t)\,|P^*\rangle\,, \tag{81b}$$

where $|P^*\rangle$ stands for the ground state of the polaron with total momentum P^* at $t=t_0$.

In what follows $\vec{P}^*=0$ is chosen. Equation (81b) is identified as the $T=0$ Kubo formula and can easily be related to the Fermi golden rule.

From (27) it follows

$$\vec{p}(t)=\sum_{\vec{k}}\vec{k}f_k\,e^{-i\gamma_k(t-t_0)}\,e^{-\epsilon|t|}\,e^{i\vec{k}\cdot\vec{r}}\,a_k+\mathrm{H.c.} \tag{82}$$

With Eq. (82) in (81b), one obtains

$$\begin{aligned}\Gamma(\Omega)&=\frac{\omega}{m\hbar\Omega}\,\mathrm{Re}\int_{-\infty}^{0}dt\,e^{-i(\Omega+i\epsilon)t}\\&\quad\times\langle 0|\sum_{\vec{k}}k_z f_k\,e^{i\gamma_k t_0}\,e^{i\vec{k}\cdot\vec{r}}\,a_k\\&\quad\times\sum_{k'}k'_z f^*_{k'}\,e^{i\gamma_{k'}(t-t_0)}\,e^{-i\vec{k}'\cdot\vec{r}}\,a^\dagger_{k'}\,|0\rangle\\&=\frac{\omega}{m\hbar\Omega}\,\mathrm{Re}\,\frac{1}{\pi}\sum_{\vec{k}}\int_{-\infty}^{0}dt\,e^{-i(\Omega+i\epsilon-\gamma_k)t}\,k_z^2|\,f_k\,|^2\\&=\pi\frac{\omega}{m\hbar\Omega}\sum_{\vec{k}}k_z^2\,|\,f_k\,|^2\delta(\Omega-\gamma_k)\\&=\frac{\omega}{m\hbar\Omega}\,\frac{V}{12\pi}\,(k^3\,|\,f_k\,|^2)_{\Omega=\gamma_k}\,,\end{aligned} \tag{83}$$

with

$$\gamma_k=\omega+\hbar k^2/2m\,,$$

therefore it is easy to express the optical-absorption coefficient of a free polaron in terms of the f_k calculated here.

With the expression (64) for $|\,f_k\,|^2$ in (83) one is then led to

$$\Gamma(\Omega)=\frac{\frac{2}{3}\alpha(\Omega/\omega)\sqrt{\Omega/\omega-1}\;e^{-C'(\Omega/\omega-1)}}{[(\Omega/\omega)^2-\mathrm{Re}\chi(\Omega/\omega)]^2+[\mathrm{Im}\chi(\Omega/\omega)]^2}\,. \tag{84}$$

This expression is very similar to that obtained by one of us (J.D.) and co-workers[16,23] using the FHIP impedance function. The analytical structure of the denominator is even identical to that found in Ref. 23, Eq. (11a). The numerator of Eq. (84) has only the leading term of that of Eq. (11a), Ref. 23. This is due to the one-real-phonon approximation in the present work but it does not alter the absorption curves considerably from those obtained in Ref. 23 in the α region of physical interest ($0<\alpha<4.5$) or for large α.

It should be remarked that the resonant structure introduced by the denominator of Eq. (84) is obtained here without any *ad hoc* approximation.

The results for the optical absorption $\Gamma(\Omega)$ obtained by numerical computation are shown in Fig. 2 for $\alpha=1$, 3, and 4.5. As can be seen from these curves, our results for $\alpha\lesssim 3.5$ are very close to those obtained in Ref. 23 with the FHIP approach. The main difference is the more rapid decrease of the absorption at high frequency in the case of the present treatment. This could be due to the fact that interactions with two or more real phonons are neglected here. For larger values of α, the disagreement between the two methods becomes considerable. However, the asymptotic limit $\alpha\to\infty$ results and those of Ref. 23 coincide again.

CONCLUSION

The method due to Lee, Low, and Pines is known[3] to give satisfactory results at intermediate coupling strengths ($\alpha\lesssim 3$) for the static properties of the polaron ground state, like the energy. The results are far less satisfactory for dynamical properties like the effective mass or the optical-absorption constant.

On the contrary, Feynman's method leads to apparently good results for these latter quantities.[17,23] However, the path-integral formalism used in this method is rather involved and not so universally known as the Schrödinger or Heisenberg formalisms.

There are probably two reasons for the success of Feynman's approach to the polaron problem. First, he is able to eliminate the phonon variables. Second, his model which is related to a harmonic oscillator, allows for internal excitations.

In this paper a different method is proposed which shares with Feynman's approach the two interesting features cited above. This method is based on the study of Heisenberg's equations, the concepts of which are probably familiar to more physicists than the path-integral formalism.

The results for the polaron effective mass and the optical absorption by free polarons as obtained here, are very close to those obtained with Feynman's method for values of the coupling constant up to α of the order of 4. Practically all the ionic crystals fall in this range of values of α.

For $\alpha \geq 4.5$ the equation giving the polaron effective mass has no longer a physically meaningful solution. This is probably due to the fact that our approach takes account of interactions with one real phonon only (but with any number of virtual phonons).

At the limit $\alpha \to \infty$ an asymptotic solution is found. This seems to be accidental and is due to a change in the meaning of the annihilation and creation operators $a_+(\vec{k})$ and $a_+^\dagger(\vec{k})$. At very strong coupling, these operators become associated with a coherent mode which now represents an internal oscillation of the electron in the potential well caused by the induced static polarization.

FHIP[18] and Thornber,[32] working in the path-integral formalism, have formulated a self-consistent scheme to calculate the response of polarons to external fields. As stated by Thornber,[32] the self-consistency in their work arises from expressing the response of the polaron system as a function of the effective electron–phonon interaction (via an "admittancelike" relation) and by simultaneously expressing the electron–phonon interaction as a function of the response (via an impedance-like expression). At different intermediate stages mathematical expressions occur in the present paper which are similar to those obtained by FHIP, Thornber, and in Ref. 23. It would certainly require an extensive study to obtain the precise relation between the present approximation and the Feynman-type self-consistency.

It seems to be relatively easy to generalize our method to study other properties of the polarons such as the mobility which will be treated in a forthcoming paper.

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APPENDIX A

In this appendix the symmetry considerations leading to Eq. (39) are given. When $\vec{P}^* = 0$, the second term of the rhs of the integral Eq. (35) has the form

$$\vec{I} = \sum_{\vec{k}'} \vec{k}'(\vec{k} \cdot \vec{k}') Y(k^2, k'^2) , \tag{A1}$$

where $Y(k^2, k'^2)$ is a rather complicated expression which has no angular dependence. Using components for the vectors $\vec{k}$ and $\vec{k}'$ gives for the components of $\vec{I}$

$$I_j = \sum_{l=1}^{3} \sum_{\vec{k}'} k'_j k'_l k_l Y(k^2, k'^2) . \tag{A2}$$

Since $Y(k^2, k'^2)$ is isotropic in k' space, the terms with $l \neq j$ in the l sum are zero. Indeed, in the summation over $\vec{k}'$, it is always possible to associate the contributions of two vectors $\vec{k}'$ having opposite values (k'_j and $-k'_j$) for their j component so that the sum is zero.

These considerations lead to

$$I_j = k_j \sum_{\vec{k}'} k'^2_j Y(k^2, k'^2) \tag{A3}$$

but, because of symmetry

$$\vec{I} = \tfrac{1}{3} \vec{k} \sum_{\vec{k}'} k'^2 Y(k^2, k'^2) \tag{A4}$$

and the product $\vec{k}'(\vec{k} \cdot \vec{k}')$ in Eq. (37) is simply replaced by $\frac{1}{3} \vec{k} k'^2$, leading to Eq. (39) with (40).

A similar reasoning can be applied to obtain

$$F(\vec{k}, \tau) = \frac{k^2}{3m^2} \sum_{k'} k'^2 \left| f_{k'} \right|^2 \gamma_{k'}^{-2} e^{i\gamma_{k'}\tau} . \tag{A5}$$

APPENDIX B

In this appendix it is shown that the approximation used in the evaluation of $g(k, z)$ [cf. relation (45)] is equivalent to an expansion in a continued fraction limited to the first step. Moreover, it is proved that the choice of the coefficients of the continued fraction can be justified by a variational principle, at least when z is real and positive. These results give a further justification of our approximation.

Let us recall that [cf. Eq. (40)]

$$g(k, z) = \int_{-\infty}^{0} d\tau \, e^{iz\tau} \exp[-k^2 C(0)] \exp[k^2 C(\tau)] , \tag{B1}$$

with

$$C(\tau) = \frac{1}{3} \sum_{k'} \frac{k'^2}{m^2} \left| f_{k'} \right|^2 \frac{e^{i\gamma_{k'}\tau}}{\gamma_{k'}^2} . \tag{B2}$$

Expanding the last exponential of Eq. (B1) in a power series leads to

$$g(k,z)=e^{-k^2C(0)}\int_{-\infty}^{0}d\tau\,e^{iz\tau}\sum_{n=0}^{\infty}\frac{1}{n!}\left(\frac{1}{3m^2}\right)^{-n}\sum_{\vec{k}_1,\ldots,\vec{k}_n}\frac{k_1^2k_2^2\cdots k_n^2\,|f_{k_1}|^2\,|f_{k_2}|^2\cdots|f_{k_n}|^2}{\gamma_{k_1}^2\gamma_{k_2}^2\cdots\gamma_{k_n}^2}\,e^{i(\gamma_{k_1}+\gamma_{k_2}+\cdots+\gamma_{k_n})\tau}$$

$$=-ie^{-k^2C(0)}\sum_{n=0}^{\infty}\frac{(3m^2)^{-n}}{n!}\sum_{k_1,\ldots,k_n}\frac{k_1^2k_2^2\cdots k_n^2\,|f_{k_1}|^2\,|f_{k_2}|^2\cdots|f_{k_n}|^2}{\gamma_{k_1}^2\gamma_{k_2}^2\cdots\gamma_{k_n}^2}\,\frac{1}{\gamma_{k_1}+\gamma_{k_2}+\cdots+\gamma_{k_n}}\,. \tag{B3}$$

The multiple sum over the k's is in fact an integral with $3n$ variables. It is possible to change the variables in order that one of the new variables is

$$x_n=\gamma_{k_1}+\gamma_{k_2}+\cdots+\gamma_{k_n}\,. \tag{B4}$$

Then the multiple sum which appears in the last term of Eq. (B3) is of the following type:

$$J(z)=\int_{n\omega}^{\infty}\frac{L(x_n)}{x_n+z}\,dx_n\,, \tag{B5}$$

where

$$L(x_n)\geq 0$$

is the result of the integration over the $n-1$ other variables. An expansion of integrals of the type (B5) into Stieltjes continued fractions is known to give good results when z is real and not located on the cut of $J(z)$, i.e., when

$$z>-n\omega\,. \tag{B6}$$

The first nontrivial step in the continued fraction expansion is

$$J(z)=a_0/(a_1+z)\,, \tag{B7}$$

with

$$a_0=\int_{n\omega}^{\infty}L(x_n)\,dx_n\,, \tag{B8}$$

$$a_1=\frac{\int_{n\omega}^{\infty}x_nL(x_n)\,dx_n}{\int_{n\omega}^{\infty}L(x_n)\,dx_n}\,. \tag{B9}$$

A variational principle can be established, which gives a rather strong argument in favor of the approximation (B7). Let us introduce a variational parameter $\bar{x}$ writing

$$J(z)=\int_{n\omega}^{\infty}\frac{L(x_n)\,dx_n}{(x_n-\bar{x})+(z+\bar{x})}\,. \tag{B10}$$

Performing two steps of the division, this relation becomes

$$J(z)=\frac{1}{z+\bar{x}}\int_{n\omega}^{\infty}L(x_n)\,dx_n-\frac{1}{(z+\bar{x})^2}\int_{n\omega}^{\infty}(x_n-\bar{x})L(x_n)\,dx_n+K(z,\bar{x})\,, \tag{B11}$$

with

$$K(z,\bar{x})=\frac{1}{(z+\bar{x})^2}\int_{n\omega}^{\infty}\frac{(x_n-\bar{x})^2L(x_n)\,dx_n}{z+x_n}\,. \tag{B12}$$

The approximation consists of neglecting the term $K(z,\bar{x})$ in Eq. (B11). As this term is positive [cf. (B6)] the best approximation is obtained when it is minimum. Therefore let us use the freedom in the choice of $\bar{x}$ to minimize its expression (B12),

$$\frac{\partial K(z,\bar{x})}{\partial\bar{x}}=-2\,\frac{K(z,\bar{x})}{z+\bar{x}}-2\,\frac{1}{(z+\bar{x})^2}\int_{n\omega}^{\infty}\frac{(x_n-\bar{x})L(x_n)\,dx_n}{z+x_n}=0\,, \tag{B13}$$

which gives

$$\frac{-2}{(z+\bar{x})^2}\int_{n\omega}^{\infty}\left(\frac{x_n-\bar{x}}{z+\bar{x}}+1\right)\frac{x_n-\bar{x}}{z+x_n}L(x_n)\,dx_n=0 \tag{B14}$$

or

$$\frac{-2}{(z+\bar{x})^3}\int_{n\omega}^{\infty}(x_n-\bar{x})L(x_n)\,dx_n=0\,. \tag{B15}$$

This provides us with the best value of the variational parameter;

$$\bar{x}=\frac{\int_{n\omega}^{\infty}x_nL(x_n)\,dx_n}{\int_{n\omega}^{\infty}L(x_n)\,dx_n}\,, \tag{B16}$$

which is a_1 [cf. Eq. (B9)].

With this value of $\bar{x}$ and neglecting $K(z,\bar{x})$ the expression (B11) of the calculated quantity $J(z)$ becomes

$$J(z)=\frac{\int_{n\omega}^{\infty}L(x_n)\,dx_n}{z+\bar{x}}=\frac{a_0}{a_1+z}\,, \tag{B17}$$

which is the first step (B7) of a Stieltjes continued fraction.

To prove that this value of $\bar{x}$ gives a minimum of $K(z,\bar{x})$ let us calculate the second derivative

$$\frac{\partial^2K(z,\bar{x})}{\partial\bar{x}^2}=\frac{6}{(z+\bar{x})^4}\int_{n\omega}^{\infty}(x_n-\bar{x})L(x_n)\,dx_n+\frac{2}{(z+\bar{x})^3}\int_{n\omega}^{\infty}L(x_n)\,dx_n\,. \tag{B18}$$

Now the parameter $\bar{x}$ is replaced by its expression (B16). The relation (B18) becomes

$$\frac{\partial^2K(z,\bar{x})}{\partial\bar{x}^2}=\frac{2}{(z+a_1)^3}\int_{n\omega}^{\infty}L(x_n)\,dx_n\,, \tag{B18'}$$

which is positive if $z\geq-n\omega$, since it follows from relation (B16) that $a_1>n\omega$.

Our approximation is related to that used by Feynman[17] which is based on the following inequality:

$$\langle e^{-sx}\rangle\geq e^{-s\langle x\rangle}\,, \tag{B19}$$

where the brackets denote the expectation value of the random variable x. For instance

$$\langle e^{-sx}\rangle = \frac{\int_a^\infty L(x)\, e^{-sx}\, dx}{\int_a^\infty L(x)\, dx} , \tag{B20}$$

where $L(x)$ is the non-normalized probability density of x. The Laplace transform of Eq. (B19) gives

$$\int_0^\infty e^{-sz}\langle e^{-sx}\rangle\, ds \geq \int_0^\infty e^{-sz}\, e^{-s\langle x\rangle}\, ds , \tag{B21}$$

which after integration becomes

$$\int_a^\infty \frac{L(x)\, dx}{x+z} \geq \frac{\int_a^\infty L(x)\, dx}{\langle x\rangle + z} = \frac{a_0}{a_1+z} . \tag{B22}$$

This last inequality shows the relation with our procedure.

APPENDIX C

This appendix is devoted to the evaluation of Eq. (65) determining m^*. The starting point is Eq. (45). It is supposed that the terms describing the internal oscillations of the electron do not contribute to the effective mass. Therefore it is sufficient to calculate the z component of the rhs of Eq. (45)

$$\frac{1}{\epsilon}\mathrm{Re}\sum_{\vec{k}} \frac{|V_k|^2}{\hbar} k_z g(k,\gamma_k) = \frac{\alpha}{\pi} r_0 \hbar\omega^2 \frac{1}{\epsilon}\mathrm{Re}\int_0^\pi d\theta \sin\theta\cos\theta \int_0^\infty dk\, k g(k,\gamma_k)$$

$$= \frac{\alpha}{\pi} r_0 \hbar\omega^2 \frac{1}{\epsilon}\mathrm{Re}\int_0^\pi d\theta \sin\theta\cos\theta \int_0^\infty dk\, k \int_0^\infty d\tau\, e^{-i\gamma_k\tau} e^{-\epsilon|\tau|} e^{-C(0)k^2(1-e^{-i\zeta\tau})}$$

$$= \frac{\alpha}{\pi} r_0 \hbar\omega^2 \frac{1}{\epsilon}\int_0^\pi d\theta \sin\theta\cos\theta \int_0^\infty dk\, k\, e^{-C(0)k^2} \mathrm{Re}\left(\sum_{n=0}^\infty \frac{C^n(0)}{n!} k^{2n} \int_0^\infty d\tau \exp[-i(\gamma_k + n\zeta - i\epsilon)\tau]\right) . \tag{C1}$$

Let us first consider

$$\Xi = \mathrm{Re}\sum_{n=0}^\infty \frac{C^n(0)}{n!} k^{2n} \int_0^\infty d\tau \exp[-i(\gamma_k + n\zeta - i\epsilon)\tau]$$

$$= \mathrm{Re}\sum_{n=0}^\infty \frac{C^n(0)k^{2n}}{n!} \frac{(-1)}{(-i)(\gamma_k + n\omega\zeta - i\epsilon)} . \tag{C2}$$

Developing this expression in powers of P^* and retaining the linear terms,

$$\Xi = 2\sum_{n=0}^\infty \frac{C^n(0)k^{2n}}{n!} \frac{\epsilon k P^* \cos\theta}{m^*(\gamma_k + n\omega\zeta)^3} . \tag{C3}$$

Going back to Eqs. (45) and (C1), it follows that

$$\frac{m}{m^*} = 1 - \frac{8\alpha}{3\pi}\left(\frac{m^*}{m}\right)^{1/2} \sum_{n=0}^\infty \left(\frac{2m^*\omega}{\hbar}\right)^n \times \frac{C(0)^n}{n!} \int_0^\infty \frac{du\, u^{2(n+1)}\, e^{-C'u^2}}{(1+u^2+n\zeta/\omega)^3} . \tag{C4}$$

Now consider

$$J = \sum_{n=0}^\infty \frac{(C')^n}{n!} \int_0^\infty \frac{du\, u^{2(n+1)}\, e^{-C'u^2}}{(1+n^2+n\zeta/\omega)^3} , \tag{C5}$$

where the notation C' is used; with

$$\frac{1}{a^n} = \frac{1}{\Gamma(n)} \int_0^\infty x^{n-1} e^{-ax}\, dx \tag{C6}$$

one obtains

$$J = \frac{1}{\Gamma(3)} \int_0^\infty du\, u^2 e^{-C'u^2} \int_0^\infty dx\, x^2 \times e^{-(1+u^2)x} \sum_{n=0}^\infty \frac{(C')^n}{n!} u^{2n} e^{-n\zeta x/\omega} . \tag{C7}$$

Now

$$\sum_{n=0}^\infty \frac{(C')^n}{n!} u^{2n} e^{-n\zeta x/\omega} = \exp(C'u^2 e^{-\zeta x/\omega}) . \tag{C8}$$

Subsequently

$$J = \frac{1}{\Gamma(3)} \int_0^\infty dx\, x^2 e^{-x} \int_0^\infty du\, u^2 \times \exp\{-u^2[x + C'(1-e^{-\zeta x/\omega})]\} . \tag{C9}$$

The integral over u, which is Gaussian, is easily performed now:

$$J = \frac{\sqrt{\pi}}{4\Gamma(3)} \int_0^\infty \frac{x^2 e^{-x}\, dx}{[C'(1-e^{-\zeta x/\omega}) + x]^{3/2}} , \tag{C10}$$

so that

$$\frac{m^*}{m} = \left[1 - \frac{\alpha}{3\sqrt{\pi}}\left(\frac{m^*}{m}\right)^{1/2} \int_0^\infty \frac{x^2 e^{-x}\, dx}{[C'(1-e^{-\zeta x/\omega}) + x]^{3/2}}\right]^{-1} . \tag{C11}$$

APPENDIX D

In this appendix a number of relatively straightforward analytical calculations are presented which facilitate the reading of the material in Sec. VI. First, the asymptotic behavior of $\mathrm{Re}(\chi)$ for $\alpha \to \infty$ is given. One has

$$\mathrm{Re}\chi(\Omega) = \frac{2\alpha}{3\sqrt{\pi}}\left(\frac{m^*}{m}\right)^{3/2} \int_0^\infty du$$

$$\times(1-\cos\Omega u)\,\mathrm{Im}\left(\frac{e^{iu}}{[C'(1-e^{i\xi u/\omega})-iu]^{3/2}}\right). \tag{D1}$$

Using a series expansion for the denominator, it follows that

$$\mathrm{Re}\chi(\Omega)=\frac{2\alpha}{3\sqrt{\pi}}\left(\frac{m^*}{m}\right)^{3/2}\sum_{n=0}^{\infty} C^n_{-3/2}\,C'^n(-)^n \times\mathrm{Im}\int_0^\infty \frac{du\,(1-\cos\Omega u)\,e^{i(1+\xi n/\omega)u}}{(C'-iu)^{n+3/2}}\,. \tag{D2}$$

Putting $x=\Omega u$ and with a further series expansion, one obtains

$$\mathrm{Re}\chi(\Omega)=\frac{2\alpha}{3\sqrt{\pi}}\left(\frac{m^*}{m}\right)^{3/2}\sum_{n=0}^{\infty} C^n_{-3/2}\,\frac{(-)^n}{\Omega(C')^{3/2}} \times\mathrm{Im}\int_0^\infty dx\,(1-\cos x)\,e^{i(1+n\xi/\omega)x/\Omega} \times\left(1+i(n+\tfrac{3}{2})\,\frac{x}{C'\Omega}+\cdots\right)\,. \tag{D3}$$

For large α, the dominant contribution to the imaginary part of the integral is (introducing a convergence factor $e^{-i\epsilon x}$) equal to Ω. Therefore

$$\mathrm{Re}\chi(\Omega)=\frac{2\alpha}{3\sqrt{\pi}}\left(\frac{m^*}{m}\right)^{3/2}\frac{1}{C'^{3/2}}\quad(\Omega\to\infty)\,. \tag{D4}$$

Now consider the imaginary part of $\chi(\Omega)$ [cf. Eq. (61)]. The asymptotic expansion for large α follows immediately from Ref. 23, Eq. (A13),

$$\mathrm{Im}\chi(\Omega)=\frac{2\alpha}{3}\left(\frac{m^*}{m}\right)^{3/2}(\Omega-1)^{1/2}\,e^{-C'(\Omega-1)}\quad(\alpha\to\infty)\,. \tag{D5}$$

Furthermore one needs the asymptotic behavior for large α of

$$\Delta=\int_0^\infty \frac{dx\,x^{1/2}\,e^{-C'x}}{[(1+x)^2-\mathrm{Re}\chi(1+x)]^2+[\mathrm{Im}\chi(1+x)]^2}\,. \tag{D6}$$

Let y_0 be a solution of $y_0^2=\mathrm{Re}\chi(y_0)$ (an equation which has solutions only if α is sufficiently large $\alpha\geq 6$). (D6) becomes

$$\Delta=\int_1^\infty \frac{dy\,(y-1)^{1/2}\,e^{-C'(y-1)}}{[(y-y_0)(y+y_0)]^2+[\mathrm{Im}\chi(y)]^2}\quad(\alpha\to\infty)\,, \tag{D7}$$

where the denominator is expanded around $y=y_0$. For strong coupling, the Lorentzian tends to a δ function and

$$\Delta=\pi\int_1^\infty \frac{dy\,\delta(y-y_0)(y-1)^{1/2}\,e^{-C'(y-1)}}{(y+y_0)\,\mathrm{Im}\chi(y)}\quad(\alpha\to\infty) \tag{D8}$$

or

$$\Delta=\frac{\pi(y_0-1)^{1/2}\,e^{-C'(y_0-1)}}{2y_0\,\mathrm{Im}\chi(y_0)}\quad(\alpha\to\infty)\,. \tag{D9}$$

Then using Eq. (D9), one finds

$$\Delta=\frac{3\pi}{4\alpha y_0(m^*/m)^{3/2}}\quad(\alpha\to\infty)\,, \tag{D10}$$

which is Eq. (71).

In the derivations of m^*, A, and B [from Eqs. (63) and (65)] one still needs the asymptotic behavior for large α of

$$J_1=\int_0^\infty \frac{dx\,x^2\,e^{-x}}{[C'(1-e^{-\xi x/\omega})+x]^{3/2}}\,. \tag{D11}$$

Using a series expansion, one has

$$J_1=\int_0^\infty \frac{dx\,x^2\,e^{-x}}{(C'+x)^{3/2}}\left(1-\frac{3}{2}\,\frac{C'\,e^{-\xi x/\omega}}{C'+x}+\cdots\right)\,. \tag{D12}$$

The second term in the brackets of (D12) is of lower order in $1/\alpha$ compared with the first term. At this point one can employ Eq. (8), Ref. 31,

$$\int_0^\infty dx\,x^{\nu-1}(x+\beta)^{-\rho}\,e^{-\mu x} =\beta^{(\nu-\rho-1)/2}\,\mu^{(\rho-\nu-1)/2}\,e^{\beta\mu/2}\,\Gamma(\nu)\,W_{(1-\nu-\rho)/2,(\nu-\rho)/2}(\beta\mu)\,, \tag{D13}$$

where $W_{\mu,\nu}$ is a Whittaker function. In Eq. (D12) one has $\nu=3$, $\mu=1$, $\beta=C'$, $\rho=\frac{3}{2}$, and therefore

$$J_1=2(C')^{1/4}(e^{C'})^{1/2}\,W_{-7/4,3/4}(C') \tag{D14}$$

or asymptotically

$$J_1=2(C')^{1/4}(e^{C'})^{1/2}(e^{-C'})^{1/2}(C')^{-7/4}=2(C')^{-3/2}\,. \tag{D15}$$

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