Optical Absorption of Polarons in the Feynman-Hellwarth-Iddings-Platzman Approximation

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The optical absorption of polarons at rest at zero temperature is calculated starting from the Feynman-Hellwarth-Iddings-Platzman (FHIP) theory of the impedance. The results are compared with the results of theories whose physical interpretation is clearer [weak-coupling theory of Gurevich, Lang, and Firsov (GLF) and product-ansatz strong-coupling theory of Kartheuser, Evrard, and Devreese (KED)] in order to obtain a better understanding of the FHIP approximation. We are particularly interested in the possible role of lattice relaxation [leading to relaxed excited states (RES)] in the optical absorption process. If the FHIP perturbation method were used to expand the *conductivity* (this would be the normal procedure), essentially Franck-Condon transitions would be found in the spectrum, and lattice relaxation would be absent. In this case the results do not fit with the product ansatz and provide merely the asymptotic limit $\alpha \to 0$, where α is the electron-phonon coupling constant. If, however, the *impedance function* rather than the conductivity is expanded (as preferred by FHIP for intuitive reasons, without further justification) more reliable results for the optical absorption appear. For $\alpha > 5$, intense absorption peaks now occur, which presumably correspond to transitions to RES, and the results are in qualitative agreement with the predictions of the product-ansatz treatment in this coupling region. Also in the limit $\alpha \to 0$ the correct behavior is found. For $3 \lesssim \alpha \lesssim 5$, the *interpretation* of the results is somewhat delicate but the possibility that RES contribute to the oscillator strength as soon as $\alpha > 3$ should be considered. The results so obtained for the optical absorption seem reliable at all α. This provides an indirect justification for the expansion of $Z(\Omega)$ rather than $1/Z(\Omega)$ in FHIP theory and a confirmation of the qualitative strong-coupling predictions of KED. The present study indicates that optical absorption peaks due to free polarons should be observable experimentally in crystals for which $\alpha > 1$.

INTRODUCTION

The optical absorption of Fröhlich polarons has been studied in the asymptotic limit $\alpha \to 0$ (α is the electron-phonon coupling constant) by Gurevich, Lang, and Firsov (GLF).[1] The result of this study at $T = 0$ to order α for a one-polaron approach gives a single peak which is due to absorption accompanied by emission of one phonon. The theory of Gurevich *et al.* is highly involved. A simple derivation of the optical absorption coefficient of polarons for $T = 0$, $\alpha \to 0$ is given in Ref. 2.

For large coupling, Kartheuser, Evrard, Devreese[3] (KED) have presented a theory of the optical absorption of polarons based on the product-ansatz strong-coupling theory. These authors have emphasized the role of the internal structure of the polaron. They find that for values of α, where the Born-Oppenheimer approximation is meaningful, the so-called relaxed excited states (RES) play a predominant role in the absorption process. (The authors of Ref. 3 claim that the Born-Oppenheimer approximation leads to *qualitatively* correct results if $\alpha > 4$; Larsen[4] has made reservations on this point.) The energy level of the first "RES" had been studied before (see Refs. 5 and 6). Let us recall that RES correspond to an excitation of the electron in the polaron potential for which the lattice is adapted to the new electron distribution. In Ref. 3 it is argued that absorption from the ground state into the RES gives the most pronounced peak as soon as $\alpha > 3$ and that the sideband structure of this RES peak gives a broader envelope with a maximum at the Franck-Condon transition frequency. The Franck-Condon transition occurs from the ground state of the polaron to an excited state without any lattice readaptation taking place.

The results of Ref. 3 are at most qualitatively valid for $\alpha < 8$. It is an open question how important the role of RES is in the absorption at intermediate α.

The purpose of the present investigation is to calculate the optical absorption of polarons at all coupling using the Feynman-Hellwarth-Iddings-Platzman (FHIP) approximation[7] and to discuss the validity of the results so obtained. This study also throws some light on the nature of the Feynman polaron.[8] The extreme accuracy of the Feynman model in describing the ground-state properties has been shown on an exactly soluble model.[9]

It should be realized that the quantity $\mathrm{Im}\chi(\Omega)$ (where Ω is the frequency of the incident light) plotted by FHIP is not the optical absorption coefficient. To obtain the optical absorption coefficient

we can start both from the expansion of the conductivity $[1/Z(\Omega)]$ or from the expansion of the impedance function [$Z(\Omega)$ in the FHIP approximation]. The former expansion is the only one which is really justified in the framework of the FHIP perturbation scheme; the latter is preferred by FHIP for intuitive reasons.

What we will do is to make use of the results obtained by FHIP for the frequency-dependent impedance function $Z = \mathrm{Re}Z + i\,\mathrm{Im}Z$ of the polaron. FHIP studied only $\mathrm{Re}Z(\Omega)$ in order to obtain an expression for the mobility. Although this is sufficient to obtain the optical absorption in the case of the expansion of the conductivity $1/Z(\Omega)$, we have to carry through a mathematical and numerical treatment starting from the quite involved analytical expressions obtained by path-integral formalism for $\mathrm{Re}Z$ and $\mathrm{Im}Z$ in Ref. 7, in order to obtain the optical absorption coefficient for all α in the case of the expansion of the impedance function $Z(\Omega)$.

Both treatments will then be compared with the existing results at weak and strong coupling, which are understood physically, and the intermediate coupling region will be investigated.

In Sec. I we recall some results of FHIP and discuss the approximations involved.

In Sec. II it is shown how the optical absorption coefficient can be calculated starting from FHIP's formalism. The necessary mathematical developments are presented.

In Secs. III and IV the analytic and numerical data obtained for the optical density in the FHIP approximation (both for the expansion of the conductivity and the impedance function) are presented and discussed.

I. GENERAL FORMULAS FOR THE IMPEDANCE OF THE POLARON AS OBTAINED BY FHIP

First a general expression for the frequency-dependent conductivity $1/Z(\Omega)$ of the polaron obtained by FHIP is recalled, together with the approximations made by these authors. This expression, which is the starting point of the present calculations, is as follows:

$$1/Z(\Omega) = \Omega(G_0 + G_1)\,, \tag{1}$$

where $-iG_0$ is the classical response function for the Feynman model [see Ref. 7, Eq. (39a)] and

$$G_1 = iG_0^2\left(\chi(\nu) + \frac{v^2 - w^2}{\Omega^2 - w^2}\,\Omega^2\right)\,, \tag{2}$$

where $\chi(\Omega)$ is defined as

$$\chi(\Omega) = \int_0^\infty (1 - e^{i\Omega u})\,\mathrm{Im}S(u)\,du\,, \tag{3}$$

and

$$S(u) = (2\alpha/3\sqrt{\pi})\,[D(u)]^{-3/2}\,[e^{iu} + 2P(\beta)\cos u]\,. \tag{4}$$

Here $P(\beta) = 1/(e^\beta - 1)$, $\beta = 1/kT$, and $\hbar = m = \omega_0 = 1$, where ω_0 is the frequency of the longitudinal-optical phonons at long wavelength. Furthermore, we have

$$D(u) = \frac{w^2}{v^2}\left(\frac{v^2 - w^2}{w^2 v}\,[1 - e^{ivu} + 4P(\beta v)\sin^2(\tfrac{1}{2}vu)] - iu + \frac{u^2}{\beta}\right), \tag{5}$$

where v and w are variational parameters, which have been introduced by Feynman to minimize the self-energy in the case of the harmonic approximation.

If one considers the "physical" model of two particles bound harmonically to each other[10] as corresponding to the Feynman polaron, v describes the vibration frequency of this system and $w = (v^2 - K)^{1/2}$, where K is the force constant of the model system.

In the strong-coupling limit, the vibration frequency v of the Feynman polaron behaves as $v = 4\alpha^2/9\pi$, which is the Franck-Condon transition frequency of the Fröhlich polaron as obtained from the product ansatz.

The approximations made by FHIP are twofold:

(a) The "double" path integral

$$g = \int\!\!\int e^{i\phi}\,D\chi\,D\chi'\,, \tag{6}$$

where ϕ is the exact action for the polaron in a relatively weak external oscillating field at any temperature is replaced by

$$g_{\text{approx}} = \int\!\!\int e^{i\phi_0}\,D\chi\,D\chi' + \int\!\!\int e^{i\phi_0}\,i(\phi - \phi_0)\,D\chi\,D\chi'\,, \tag{7}$$

where ϕ_0 is a "quadratic" approximation to ϕ. This is essentially Feynman's approximation of replacing the potential energy part in the action, which is Coulomb-like, by a *quadratic approximation.*

(b) FHIP calculate the expression $G(\Omega) = 1/\Omega Z(\Omega)$ in the form (1), where G_0 and G_1 correspond to the first and second terms in (7), respectively. To obtain a resonance structure for $\mathrm{Im}\chi$ (which they call dissipation), they expand the impedance function $Z(\Omega)$ as follows:

$$\Omega Z(\Omega) = \frac{1}{G_0(\Omega)} - \frac{1}{[G_0(\Omega)]^2}\,G_1(\Omega)\,. \tag{8}$$

FHIP present intuitive arguments for preferring this expansion of the impedance to the expansion of the conductivity:

$$1/\Omega Z(\Omega) = G_0(\Omega) + G_1(\Omega)\,. \tag{9}$$

Of course, this is a delicate point because the different conclusions resulting from (8) and (9) question the degree of accuracy of the approximation (7). It may be recalled that only the expansion (9) is justified in the FHIP perturbation scheme.

In the present study both the expansions (8) of $Z(\Omega)$ and (9) of $1/Z(\Omega)$ will be used.

II. OPTICAL ABSORPTION COEFFICIENT OF POLARONS IN THE QUADRATIC APPROXIMATION

A. Impedance and Absorption Coefficient of Polarons

The relation between the absorption coefficient $\Gamma(\Omega)$ and the impedance $Z(\Omega)$ of polarons is as follows:

$$\Gamma(\Omega) = (1/c\epsilon_0 n)\,\mathrm{Re}[1/Z(\Omega)] \,. \tag{10}$$

ϵ_0 is the dielectric constant of the vacuum, n is the index of refraction of the medium and c is the velocity of light.

One needs both the real and imaginary part of $Z(\Omega)$ or of $\chi(\Omega)$ to express the absorption coefficient at zero temperature if the expansion of $Z(\Omega)$ is used. We obtain the following formula in that case:

$$\Gamma_Z(\Omega) = \frac{1}{c\epsilon_0 n} \lim_{\beta\to\infty} \frac{\Omega\,\mathrm{Im}\chi(\Omega)}{\Omega^4 - 2\Omega^2\,\mathrm{Re}\chi(\Omega) + |\chi(\Omega)|^2} \,. \tag{11a}$$

If the expansion for $1/Z(\Omega)$ is used one finds

$$\Gamma_\sigma(\Omega) = [(\Omega^2 - w^2)^2/\Omega^3(\Omega^2 - v^2)^2]\,\mathrm{Im}\chi(\Omega) \,. \tag{11b}$$

B. Evaluation of Reχ

In the work of FHIP only $\mathrm{Im}\chi(\Omega)$ has been calculated (together with $\lim_{\Omega\to 0}[\mathrm{Im}\chi(\Omega)/\Omega] = 1/\mu$ resulting in the mobility μ). In FHIP's theory a closed expression is given for $\mathrm{Im}\chi(\Omega)$ valid at all T and α. This expression involves integrals which were evaluated numerically for large β ($\beta = 100$), after using an infinite-power-series expansion in terms of Bessel functions of imaginary arguments. FHIP also considered the limit $T \to 0$ ($\beta \to \infty$) for the first two terms in the series expansion.

In the present work we limit our attention to the case $T = 0$. It is important to take the limit $\beta \to \infty$ in a rigorous way, starting from the expressions obtained by FHIP. Therefore, we have demonstrated (Appendixes A and B) that the exact zero-temperature limit arises if the limit $\beta \to \infty$ is taken directly in the expression (3) for $\chi(\Omega)$. Doing so one obtains immediately

$$\mathrm{Im}\chi(\Omega) = \mathrm{Im}\int_0^\infty \frac{du\,\sin(\Omega u)\,e^{iu}}{[R(1 - e^{ivu}) - iu]^{3/2}} \qquad (\beta = \infty) \,. \tag{12}$$

$\lim_{\beta\to\infty}\mathrm{Re}\chi(\Omega)$ has not been calculated by FHIP. However, to study the optical absorption to the same approximation as FHIP's treatment of the impedance which corresponds to considering the expansion for $Z(\Omega)$, we have also to calculate $\lim_{\beta\to\infty}\mathrm{Re}\chi(\Omega)$ and use this result in (11a). This means that we must evaluate

$$\mathrm{Re}\chi(\Omega) = \mathrm{Im}\int_0^\infty \frac{du\,(1 - \cos\Omega u)e^{iu}}{[R(1 - e^{ivu}) - iu]^{3/2}} \qquad (\beta = \infty) \,. \tag{13}$$

In (12) and (13), $R = (v^2 - w^2)/w^2 v$.

Strictly speaking the *mathematical* contribution of the present work consists of evaluating the integral (13). Developing the denominator of both integrals on the right-hand side of (12) and (13) the calculations are reduced to the evaluation of a sum of integrals of the type

$$\mathrm{Im}\int_0^\infty \frac{du\,\sin\Omega u\,e^{i(1+nv)u}}{(R - iu)^{3/2+n}} \,, \tag{14a}$$

$$\mathrm{Im}\int_0^\infty \frac{du\,\cos\Omega u\,e^{i(1+nv)u}}{(R - iu)^{3/2+n}} \,. \tag{14b}$$

In Appendix B we show how such integrals are evaluated using a recurrence formula. For $\mathrm{Im}\chi(\Omega)$ at $\beta = \infty$ a very convenient result appears:

$$\mathrm{Im}\chi(\Omega) = \frac{2\alpha}{3}\,\frac{v^3}{w^3}\sum_{n=0}^{\infty} C^n_{-3/2}(-1)^n \frac{R^n 2^n}{(2n+1)\cdots 3\cdot 1} \times |\Omega - 1 - nv|^{n+1/2} e^{-|\Omega-1-nv|R}\,\frac{1 + \mathrm{sgn}(\Omega - 1 - nv)}{2} \,. \tag{15}$$

This expression is a finite sum and not an infinite series. FHIP gave the first two terms of (15) explicitly.

Using the same recurrence relation it is seen that (Appendix B) the analytical expression which we find for $\mathrm{Re}\chi(\Omega)$ ($\beta = \infty$) is far more complicated. Moreover, this expression is not efficient for the numerical work which has to be done. We have written it down in Appendix B nevertheless, because it might be of some use for analytical investigations. To circumvent the difficulty with the numerical treatment of $\mathrm{Re}\chi(\Omega)$ we have transformed the corresponding integrals in (14) to integrals with rapidly convergent integrands:

$$\mathrm{Im}\int_0^\infty \frac{du\,(1 - \cos\Omega u)\,e^{i(1+nv)u}}{(R - iu)^{n+3/2}} = -\frac{1}{\Gamma(n + \frac{3}{2})}\int_0^\infty dx[(n + \tfrac{1}{2})\,x^{n-1/2}\,e^{-Rx} - Rx^{n+1/2}\,e^{-Rx}] \times \ln\left|\left(\frac{(1 + nv + x)^2}{\Omega^2 - (1 + nv + x)^2}\right)\right|^{1/2} \,. \tag{16}$$

The integral on the right-hand side of (16) is adequate for computer calculations. These have been performed on the IBM 1130 computer of our Institute at the University of Antwerp, using the Gaussian-quadrature technique. In Appendix C some supplementary details of the computation of (16) are given.

We are now in a position to calculate the optical absorption coefficient both from Eq. (11a) and Eq. (11b).

III. NUMERICAL RESULTS

Starting from (15) and (16) we have obtained $\mathrm{Im}\chi(\Omega)$, $\mathrm{Re}\chi(\Omega)$, and the optical absorption coef-

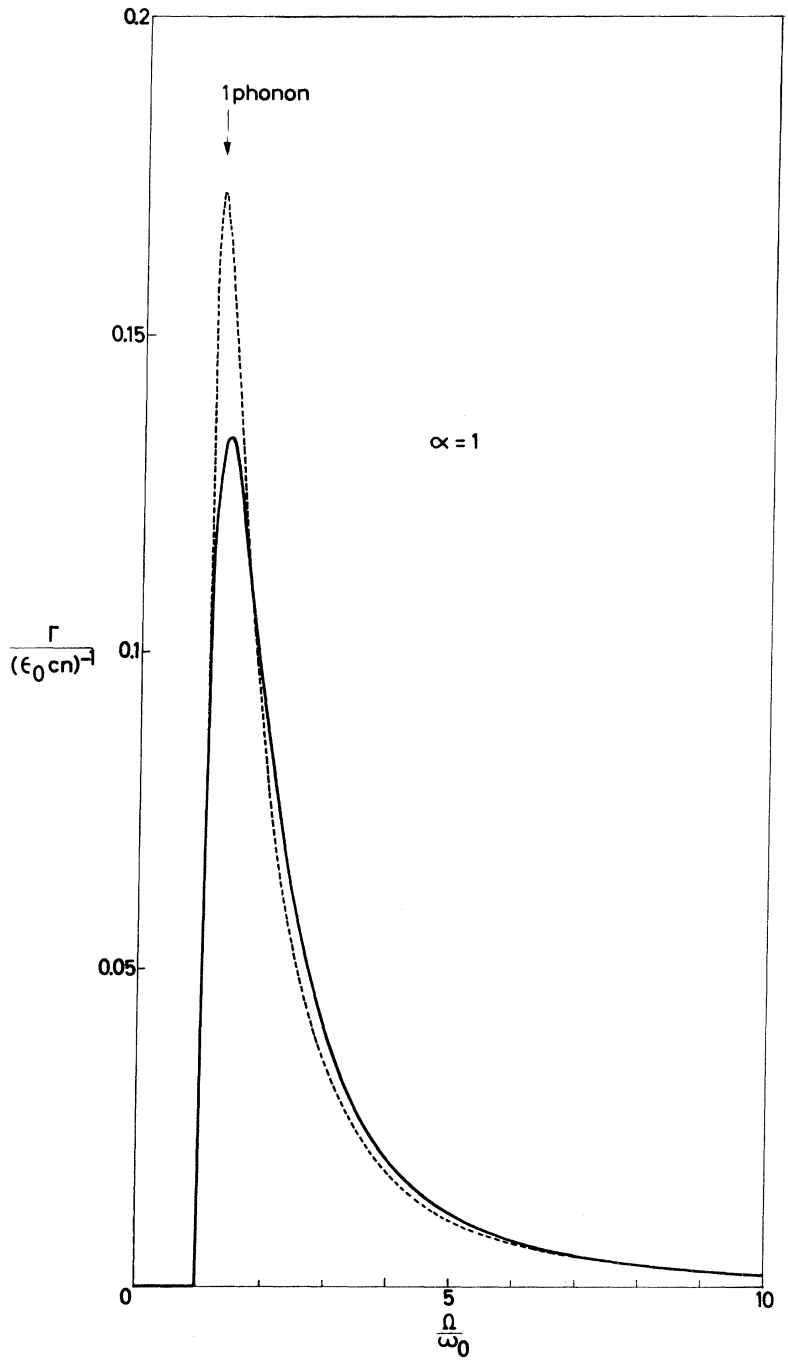

FIG. 1. Optical absorption of polarons for $\alpha = 1$. Full curve: present calculations; dotted curve: perturbation result of Gurevich *et al.*, Ref. 1. These peaks are one-phonon peaks.

ficient $\Gamma_Z(\Omega)$ [corresponding to the expansion of $Z(\Omega)$] for a number of α values and typically for $0 < \Omega < 10$. $\mathrm{Im}\chi(\Omega)$ obtained here for $\beta = \infty$ coincides to the desired precision with FHIP's results for $\mathrm{Im}\chi(\beta)$ at $\beta = 100$. $\mathrm{Re}\chi(\Omega)$ and $\Gamma_Z(\Omega)$ are obtained here for the first time. Also $\Gamma_\sigma(\Omega)$ is calculated.

We have plotted here the optical absorption coefficient $\Gamma_Z(\Omega)$ for $\alpha = 1$ (Fig. 1), $\alpha = 3$ (Fig. 2), $\alpha = 5$ (Fig. 3), $\alpha = 6$ (Fig. 4), $\alpha = 7$ (Fig. 5) of polarons at $T = 0$. Furthermore, the frequencies where $\Gamma_\sigma(\Omega)$ diverges are indicated for $\alpha = 5$, $\alpha = 6$, and $\alpha = 7$.

IV. DISCUSSION

A. Weak-Coupling Limit

The development (7) should lead to the exact result for the absorption coefficient in order α for weak coupling. For $\alpha \to 0$ one has (see Appendix B)

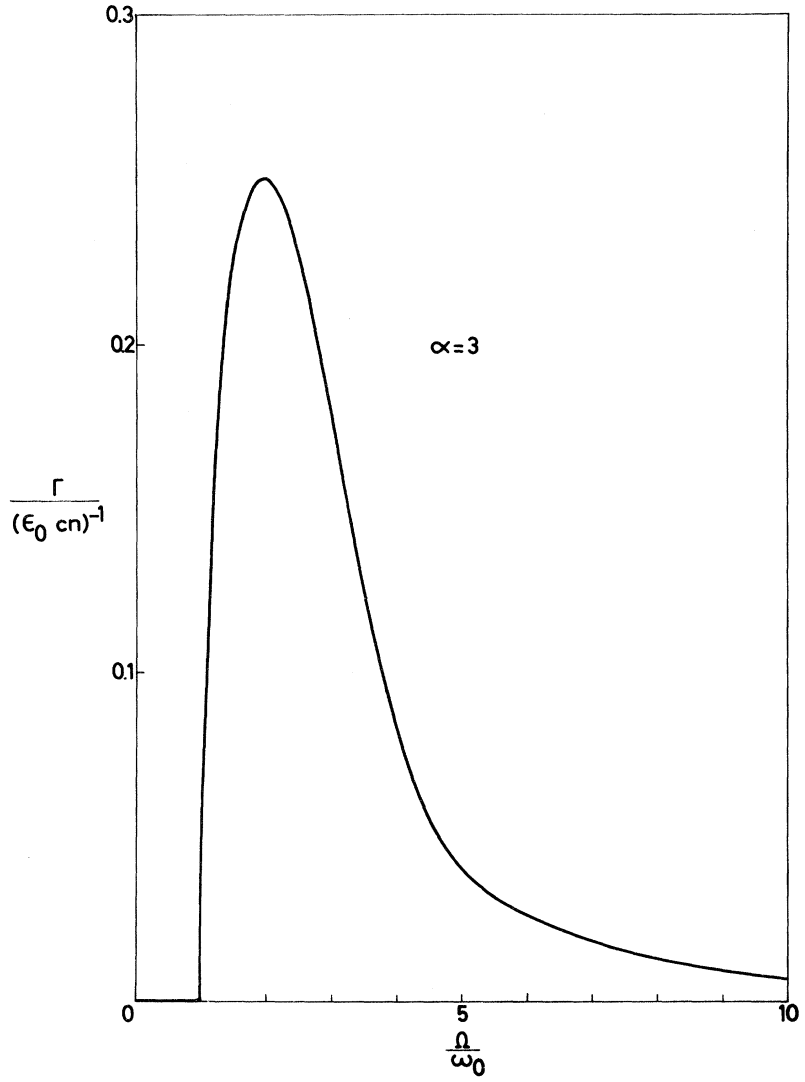

FIG. 2. Optical absorption Γ_z of polarons for $\alpha = 3$.

$$\lim_{\alpha \to 0} \mathrm{Re}\chi(\Omega) = 0 \quad \text{and} \quad \lim_{\alpha \to 0} |\chi(\Omega)|^2 = 0 \ .$$

From (15) and (11) it then follows immediately that

$$\lim_{\alpha \to 0} \Gamma_z(\Omega) = (1/c\epsilon_0 n)\, \tfrac{2}{3}\alpha\, [(\Omega - 1)^{1/2}/\Omega^3] \quad \text{for } \Omega > 1$$
$$= 0 \quad \text{for } \Omega < 1 \ . \qquad (17)$$

Γ_σ takes the same form to order α. This is exactly the result found in the Green's-function treatment of Gurevich *et al.* [1] (see also Ref. 2).

Equation (17) corresponds to a "one-phonon peak": The polaron (with the initial momentum equal to zero) emits a phonon during the absorption process and takes recoil energy from the incident light.

B. Behavior of the Optical Absorption in the FHIP Approximation as a Function of the Coupling Parameter α

1. *Case of the Expansion of the Conductivity* $1/Z(\Omega)$

The natural expansion to which the FHIP approximation scheme would lead is given in Eq. (1) and leads to Γ_σ [Eq. (11b)].

Equation (11b) gives the exact asymptotic limit for $\alpha \to 0$. The main characteristic of Γ_σ is that it becomes divergent for $\Omega = v$. The parameter v describes the vibrational frequency of the Feynman two-particle model of the polaron at all coupling and corresponds to a Franck-Condon transition of this Feynman polaron. For large α one has $v = 4\alpha^2/9\pi - (4\ln 2 - 1)$ following Feynman, which is close to the Franck-Condon (FC) frequency $\Omega_{FC} = 4\alpha^2/9\pi$ obtained with the product-ansatz strong-coupling theory for Fröhlich polarons.

The result for Γ_σ is not unexpected because it corresponds only to a perturbative correction to the Feynman two-particle polaron model. Such a model (corresponding to G_0 and Φ_0) would have an allowed transition at $\Omega = v$ only. The structure in Γ_σ due to the factor $\mathrm{Im}\chi(\Omega)$ (which is plotted in

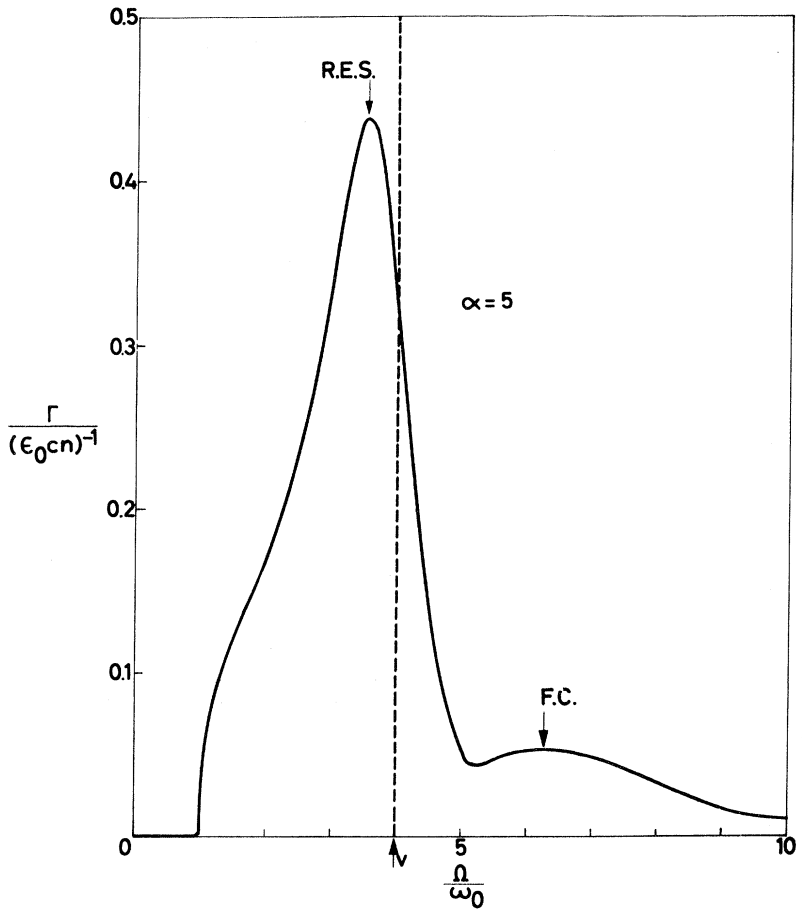

FIG. 3. Optical absorption Γ_z of polarons for $\alpha = 5$. The main peak at $\Omega = 3.51$ is interpreted here as due to transitions to relaxed excited states. A "shoulder" at the low-frequency side of the main peak is attributed to one-phonon transitions. The structure at about $\Omega = 6$ to 7 is attributed to a Franck-Condon band. The frequency $\Omega = v$ is indicated.

FHIP) seems difficult to interpret because of the unrealistic nature of Γ_σ: The total intensity (integrated over all frequencies) described by Γ_σ is divergent.

It may be noted however, that Γ_σ tends to zero for $\Omega \to \infty$ in the same way as Γ_Z. In practice the prominent structure shown by FHIP for $\mathrm{Im}\chi(\Omega)$ is smeared out in Γ_σ after the first maximum of $\mathrm{Im}\chi$ for all α.

We may conclude that the expansion of the conductivity in FHIP leads essentially to divergences for the optical absorption coefficient at $\Omega = v$ and to Franck-Condon-type transitions.

2. *Case of the Expansion of the Impedance Function $Z(\Omega)$*

FHIP present intuitive arguments to prefer an expansion of $Z(\Omega)$ rather than an expansion of $1/Z(\Omega)$. However, they did not study the optical absorption of polarons with these expansions.

Using Eq. (11a) we obtain results for the optical absorption based on the expansion of $Z(\Omega)$ which are plotted in Figs. 1–5.

First it should be remarked that these absorption curves do not present any divergences. The most striking feature of the curves, obtained from Eq. (11a), is the appearance of sharp absorption lines for $\alpha = 6$ at $\Omega = 4.3$ and for $\alpha = 7$ at $\Omega = 4.86$.

In Fig. 6 (curve a) a plot of the positions of these peaks as a function of α is shown, together with the peak position of the main peaks at $\alpha = 5$ and $\alpha = 11$. It seems *plausible* from this plot that the peak at $\Omega = 3.59$ for $\alpha = 5$ is of the same nature as those for $\alpha = 6$ and $\alpha = 7$. The problem of the physical interpretation of these peaks arises.

In Fig. 3 ($\alpha = 5$), Fig. 4 ($\alpha = 6$), and Fig. 5 ($\alpha = 7$) we have indicated the frequencies $\Omega = v$, which measure the FC transitions in the Feynman polaron. It is seen that the intense absorption peaks arising from the expansion of $Z(\Omega)$ are shifted substantially from the FC frequencies, at which the absorption diverges for the expansion of $1/Z(\Omega)$ towards lower frequencies. Such a shift would be typical for RES.

In Fig. 6, the transition frequencies for a transition from the ground state to the first relaxed excited state resulting from product-ansatz calculations have been plotted (curve b).[5,6] One can make the following remarks. (a) The peak positions of

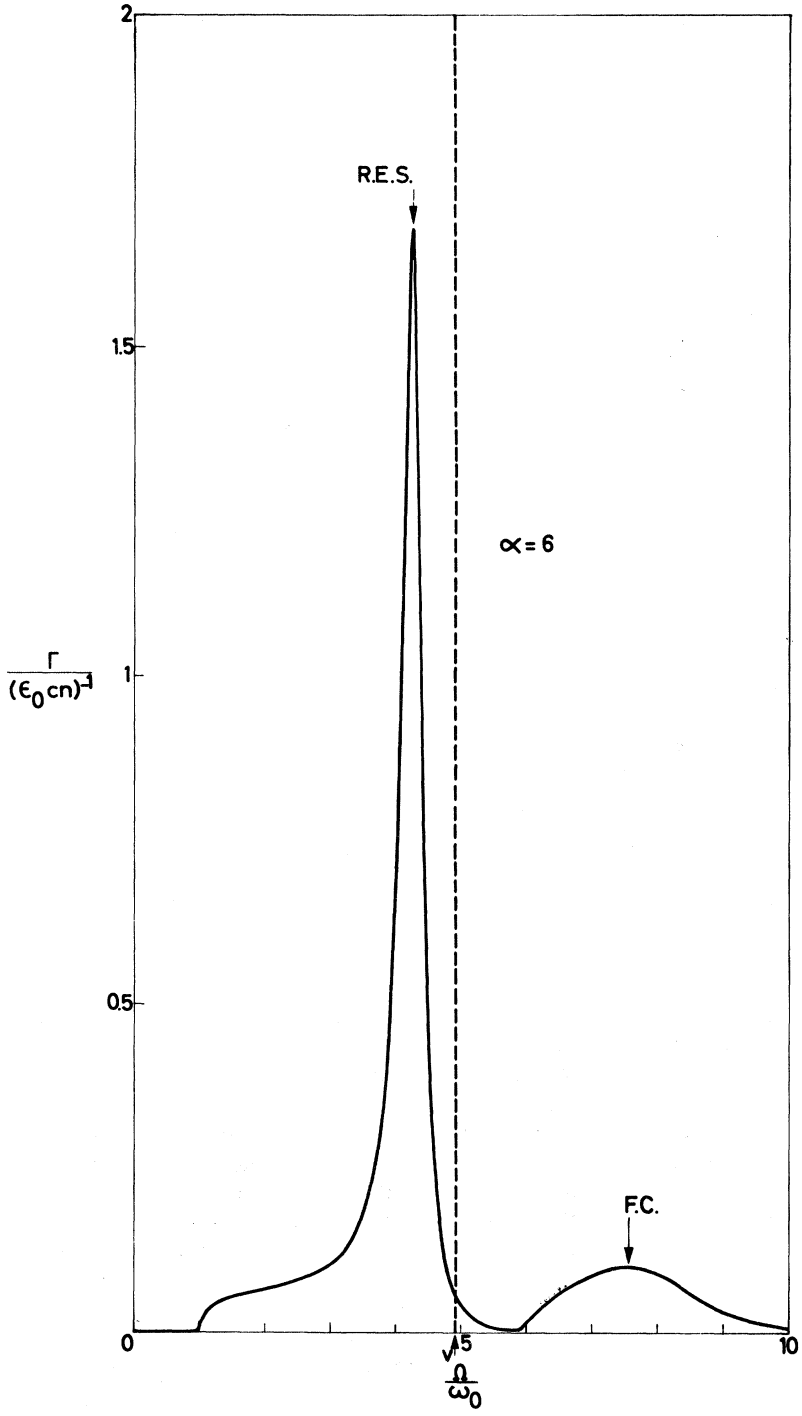

FIG. 4. Optical absorption Γ_z of polarons at $\alpha = 6$. The RES peak is very intense compared with the Franck-Condon peak. The frequency $\Omega = v$ is indicated.

the intense absorption peaks found here (curve a) are shifted towards lower frequencies with respect to the FC frequencies of the *Feynman polaron* (curve d) in qualitatively the same way as the relaxed excited states (curve b) are shifted from the FC states (curve c) for the Fröhlich polaron in the product ansatz. (b) Further arguments will be given to consider curve e (and *not* curve d) as corresponding to the FC transition frequencies in the FHIP approximation. In that case the shift from the FC-transition frequencies (curve e) to the frequencies of the intense absorption peaks (curve a) in the FHIP approximation is comparable to the shift between the product-ansatz approximation for

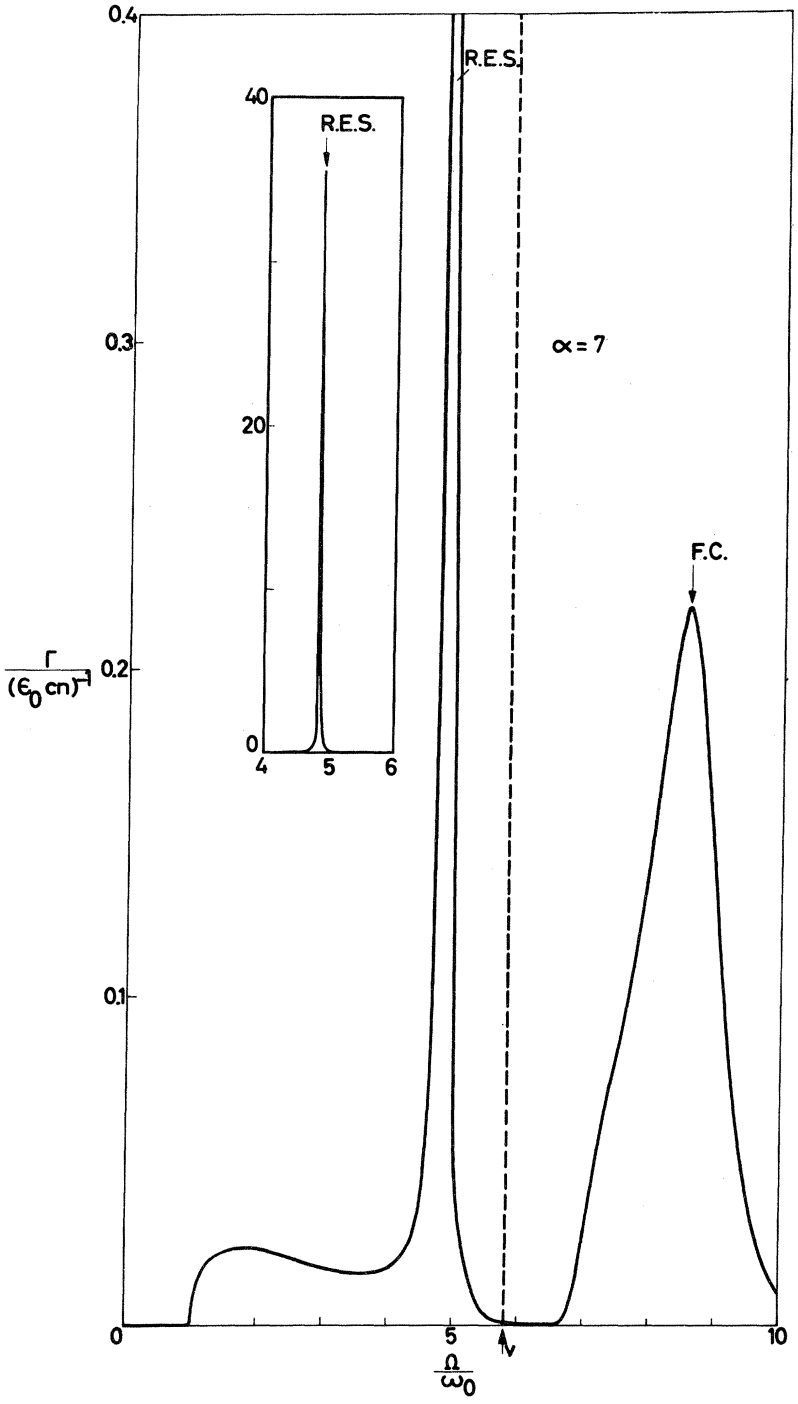

FIG. 5. Optical absorption Γ_z of polarons at $\alpha = 7$. An extremely intense RES transition occurs. The frequency $\Omega = v$ is indicated.

FC- and RES-transition frequencies (curves c and b) for $5 < \alpha < 10$. This again would indicate that curve a describes transition frequencies towards relaxed excited states in the FHIP approximation. There is a frequency difference between curve a and curve b (Fig. 6) of about $2\hbar\omega_0$ for $5 < \alpha < 10$. However, the precision of the product-ansatz calculation of the RES energy is questionable to some extent, because it does not lead to an upper bound for the energy contrary to the product-ansatz treatment of the ground state.

The intense absorption peaks at $\alpha = 5$, $\alpha = 6$, and $\alpha = 7$ should be investigated further. Nevertheless, in our opinion, the arguments presented above indicate that the peaks at $\Omega = 3.59$ for $\alpha = 5$, at $\Omega = 4.3$ for $\alpha = 6$, and at $\Omega = 4.86$ for $\alpha = 7$ describe transi-

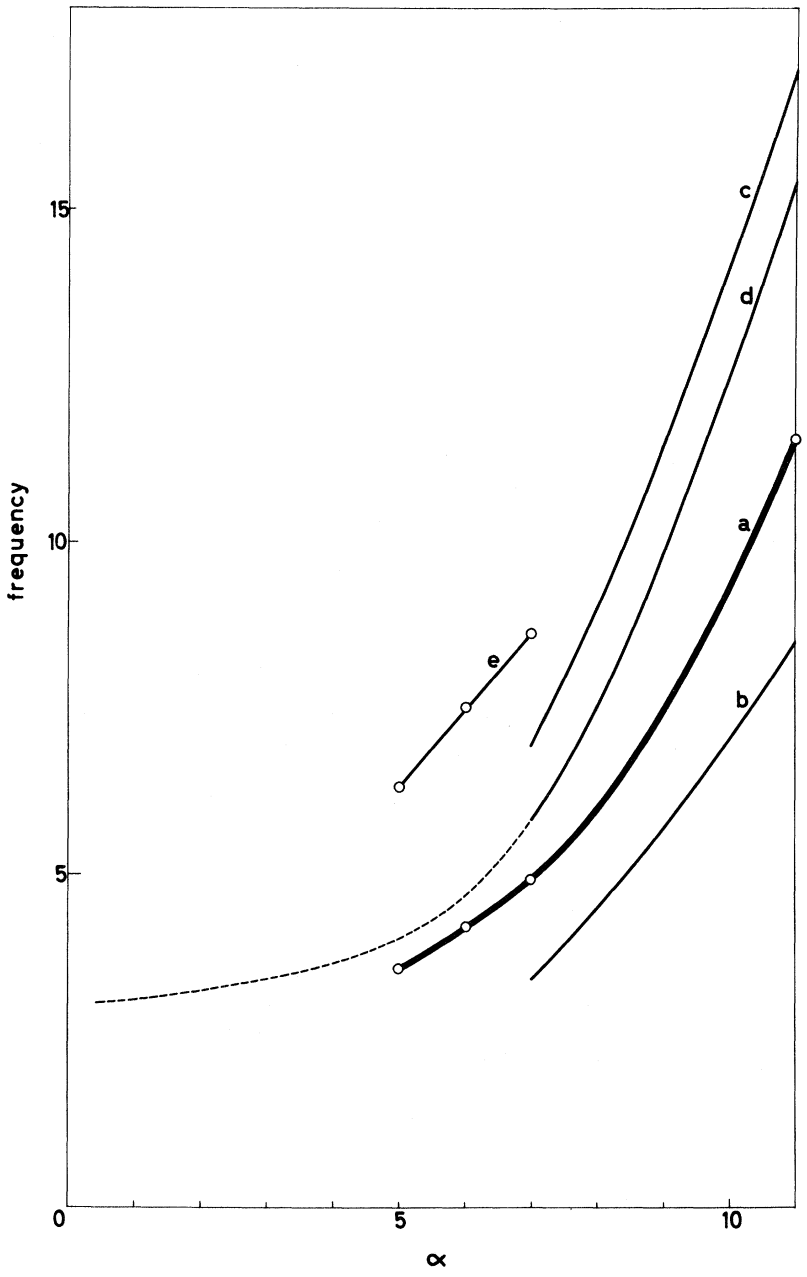

FIG. 6. Transition frequencies (in units ω_0) as a function of the coupling parameter α: curve a: frequencies of the RES transitions from the present calculations; curve b: frequencies of the RES transitions in the product-ansatz approximation; curve c: Franck-Condon transition frequency in the product ansatz; curve d: Franck-Condon transition frequency v of the two-particle Feynman polaron; curve e: Franck-Condon transition frequency from the present calculation.

tions from the ground state to the RES of polarons. Therefore, these peaks are indicated with the designation RES in Figs. 3–5.

Before discussing the broader, less intense peaks to the high-frequency side of the RES peaks, let us briefly recall the main predictions of KED from their *product-ansatz strong-coupling* treatment, based on the Fermi Golden Rule: (a) There is an intense absorption peak (zero-phonon line) corresponding to a transition from the ground state to the first RES (at Ω_{RES}). (b) At $\Omega_{\mathrm{RES}}+1$ a phonon sideband structure appears. (A one-phonon sideband at $\Omega_{\mathrm{RES}}+1$, a two-phonon sideband at $\Omega_{\mathrm{RES}}+2$, etc.) KED *expect* that this structure has a maximum at Ω_{FC}, the FC-transition frequency.

The treatment of KED is a strong-coupling treatment and shows the following limitations: (a) KED calculate only the one-phonon sideband. This treatment is incomplete for strong coupling where many-phonon processes enter the picture. The present authors[11] have calculated the two-phonon sideband. They showed that, in the product ansatz, this sideband is more important than the one-phonon sideband if $\alpha > 6$. The treatment of three-phonon side-

bands seems unrealistic (because of the mathematical complexity) in the KED treatment. (b) The lifetime of the RES has been calculated only in the one-phonon approximation.

In conclusion, the treatment of KED is incomplete as soon as three-phonon processes are important (say for $\alpha > 6$) and at most *qualitatively* valid if mainly two-phonon processes are important ($4 < \alpha < 6$).

Nevertheless, the KED results are very useful for a comparison with the present results because they are obtained in an entirely different manner than the results obtained here and because they are physically well understood and similar to the zero- and multiphonon transitions in color centers.

It is immediately clear that our results for $\alpha = 6$ and $\alpha = 7$, and even for $\alpha = 5$, obtained with the FHIP approximation by expanding the impedance function, have the qualitative behavior predicted by KED: an intense zero-phonon (RES) line with a broader sideband at the high-frequency side. This is very reassuring.

The interpretation of the broad peaks centered at $\Omega = 6.3$ for $\alpha = 5$, at $\Omega = 7.5$ for $\alpha = 6$, and at $\Omega = 8.62$ for $\alpha = 7$ as phonon sidebands is now natural in view of the prediction of such sidebands by KED. However, the absorption sidebands in Figs. 4 and 5 fail at least in one respect: The *exact* sidebands should start at $\Omega_{RES} + 1$, while the "sidebands" shown in Figs. 3–5 start at about $\Omega_{RES} + 2$. The maximum of these "sidebands" are indicated in Fig. 6 (curve e). The maxima indicate the positions of the FC levels in the FHIP approximation if the interpretation is based on KED. It may be recalled that the frequency differences between curve e (FC position in FHIP approximation) and curve a (RES position in FHIP approximation) on the one hand, and the frequency differences between curve c (FC position with the product ansatz) and curve b (RES position with the product ansatz) on the other hand, are close to each other for $5 < \alpha < 10$.

It thus seems that although the FHIP approximation [with expansion of $Z(\Omega)$] gives the gross features of the phonon sidebands, the results are not quantitatively exact. Because the lineshape of the sidebands is related to the lifetime of the RES this would also imply that the shapes of the RES peaks are approximate.

The next point to discuss concerns the absorption for $\alpha < 5$. For $\alpha = 1$ there is no doubt that the peak found from Eq. (11a) has the meaning of a "one-phonon" peak. From Fig. 1 it is seen that the peak of the absorption coefficient obtained here from FHIP approximation for $\alpha = 1$ is less intense than that obtained from perturbation theory. [1,2]

For $\alpha = 3$ the absorption peak is relatively broad and has a maximum for $\Omega = 2.2$. This peak has a shape which connects the absorption curve at $\alpha = 1$ with that at $\alpha = 5$ in a smooth manner.

The interpretation of the absorption peak for $\alpha = 3$ is an unsolved problem. It might correspond to a one-phonon peak at this larger coupling, but it is also possible that part of the oscillator strength in the absorption for $\alpha = 3$ is due to *lattice relaxation* in the final states, be it to a smaller extent than for $\alpha = 5$. As noted by KED, arguments on conservation of energy and momentum indicate that RES should play a role in the optical absorption for $\alpha > 3$. Considerations following Larsen's calculations, however, [4] tend to restrict the role of RES to $\alpha > 6$. So this point is not clear and the *physical interpretation* of the optical absorption curves obtained here for $\alpha \approx 3$ needs further investigation.

Despite the uncertainties and difficulties in the understanding of our results for $\alpha \approx 3$ we find it worthwhile to *propose* the following interpretation of these results: For $\alpha = 1$ the optical absorption spectrum consists of a one-phonon peak. For increasing α more and more oscillator strength is added due to transitions towards final states for which lattice adaptation to the (excited) electronic configuration has occured. For futher increase of the coupling the one-phonon line and the RES separate off and this is visible for $\alpha = 5$ where the "shoulder" at the low-energy side of the main peak can be attributed to a one-phonon line and the main peak at $\Omega = 3.59$ to a RES transition. For $\alpha = 6$ and $\alpha = 7$ this splitting-off between the one-phonon line and RES is completed. From $\alpha = 5$ on, a sideband structure leading to a broad FC peak, which is much less pronounced than the RES peak, appears.

Although this interpretation is probably not a unique one, it has the feature of joining the weak-coupling behavior (one-phonon peak) in a natural way to the strong-coupling behavior (essentially RES absorption).

It should be remarked indeed that the FHIP approximation with expansion of the impedance leads to much more realistic results for the optical absorption than the FHIP approximation with expansion of the conductivity. This then provides an indirect but convincing argument in favor of the expansion of the impedance rather than the conductivity. Especially the appearance of the well-known results for the optical absorption at weak and strong coupling gives evidence for the reliability of the method. The results on Fig. 3 ($\alpha = 5$) illustrate the power of the FHIP approximation which allowed us to obtain a kind of envelope function for the optical absorption as a function of frequency. This envelope function accounts at the same time for both the lifetime of the RES (which is unstable for $\alpha = 5$) and the shape of the FC band (which is a superposition of multiphonon sidebands and which is almost untractable in the product ansatz if more than two phonons are included).

It would be possible to improve the accuracy of the present results by using the recent developments (similar to the FHIP treatment) by Thornber and Feynman[12] and by Thornber.[13] However, although Ref. 12 contributes to the justification of the expansion of $Z(\Omega)$, neither in Ref. 12 nor in Ref. 13 has it been found that an expansion of $Z(\Omega)$ leads to sharp resonances in the optical absorption of polarons.

It is reasonable to expect that the main absorption peaks calculated here can be observed experimentally certainly for $\alpha > 1$. Indeed, the peak for $\alpha = 5$ at $\Omega = 3.59$ is about three times more intense than the one-phonon peak at $\alpha = 1$. The one-phonon peak now has been observed experimentally by Finkenrath *et al.*[14] in CdO, where $\alpha \approx 0.75$, as a function of temperature. If a sufficient density of polarons can be obtained for $\alpha \gtrsim 3$ this should then lead to the first observation of optical absorption due to free polarons for highly ionic crystals.

It should still be remarked that for $\alpha > 7$ the results which are obtained numerically using the FHIP treatment with the expansion of $Z(\Omega)$ are unreliable. Indeed, the linewidth of the FC peak becomes then much smaller than ω_0, the LO phonon frequency. However, as pointed out by KED, a FC state has a lifetime of the order of $1/\omega_0$, as follows from the uncertainty relations, because this is the time necessary for lattice readaptation. This is presumably the well-known failure of the Fröhlich model at strong coupling (due to the contradiction between the *continuum* polaron concept and the small polaron radius at large α) and not a failure of the FHIP approximation.

Nevertheless, we have assumed that for $\alpha \approx 11$ the *position* of the RES peak is meaningful for comparison with other strong-coupling treatments.

It may still be noted that Fig. 4 ($\alpha = 6$) is a quantitative picture for the qualitative results shown in Fig. 11 by Devreese *et al.*[15]

CONCLUSIONS

We have calculated the optical absorption coefficient of polarons at rest at zero temperature using the FHIP theory.

Essentially different results are found if the optical absorption is calculated starting from an expansion of the conductivity rather than of the impedance function. As the results are unquestionably more realistic in the latter case, we have given an indirect justification for the expansion of $Z(\Omega)$ rather than $1/Z(\Omega)$.

The results which we obtain for the optical absorption coefficient in the FHIP approximation [expansion of $Z(\Omega)$] give the exact weak-coupling behavior as first derived by Gurevich, Lang, and Firsov and join qualitatively the product-ansatz strong-coupling treatment of the optical absorption of KED if $\alpha > 5$.

The somewhat unexpected result appears that the FHIP approximation [with expansion of $Z(\Omega)$] takes into account the possibility of *lattice relaxation* to the electronic configuration of internal excited-polaron states. This is seen from the analysis of intense peaks which the present study reveals for $\alpha > 5$ in the optical absorption spectrum obtained here with the FHIP approximation.

Therefore, although FHIP thought that their trial action could not "give such detailed results (excitation spectrum) correctly" our study of the optical absorption of polarons with their approximation shows that the polaron excitation spectrum, including the fundamental possibility of lattice relaxation, should be described quite accurately by the FHIP approximation at all α.

It may still be emphasized that the function $\mathrm{Im}\chi(\Omega)$ studied by FHIP is not simply related to the optical absorption coefficient.

The physical interpretation of our results presents some difficulties for $2 < \alpha < 5$. This does not affect our conclusion that the main absorption peaks, which are calculated here, should be detectable experimentally certainly for $\alpha > 1$. This last conclusion follows from comparison with the case of CdO ($\alpha \approx 0.75$) where the absorption calculated here has been observed for *finite* temperatures.

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APPENDIX A

The expression for $\mathrm{Im}\chi(\Omega)$ and its zero temperature limit will be calculated starting from FHIP's formula

$$\mathrm{Im}\chi(\Omega) = \lim_{\beta\to\infty} \frac{2\alpha}{3\sqrt{\pi}} \left(\frac{v}{w}\right)^3 \beta^{3/2} \frac{\sinh(\frac{1}{2}\beta\Omega)}{\sinh(\frac{1}{2}\beta)} \times \int_0^\infty \frac{du \cos\Omega u \cos u}{(u^2+a^2-b\cos vu)^{3/2}} \,. \quad \text{(A1)}$$

a, b and R are defined as

$$a^2 = \tfrac{1}{4}\beta^2 + R\beta\coth(\tfrac{1}{2}\beta v) \,, \qquad b = \frac{R\beta}{\sinh(\frac{1}{2}\beta v)} \,, \qquad R = \frac{v^2-w^2}{w^2 v} \,. \quad \text{(A2)}$$

Because

$$|b\cos vu/(u^2+a^2)| < 1 \quad \text{if} \quad v > 0 \quad \text{and} \quad \beta > 0$$

$\mathrm{Im}\chi(\Omega)$ can be written as

$$\mathrm{Im}\chi(\Omega) = \lim_{\beta\to\infty} \frac{2\alpha}{3\sqrt{\pi}} \left(\frac{v}{w}\right)^3 \beta^{3/2} \frac{\sinh(\frac{1}{2}\beta\Omega)}{\sin(\frac{1}{2}\beta)}$$

$$\times \int_0^\infty \frac{du \cos\Omega u \cos u}{(u^2+a^2)^{3/2}} \times \sum_{n=0}^{\infty} C_n^{-3/2} \frac{(-1)^n b^n \cos^n vu}{(u^2+a^2)^n} \quad . \tag{A3}$$

To calculate this limit the integral

$$\int_0^\infty du \ \frac{\cos\Omega u \cos u \cos^n vu}{(u^2+a^2)^{n+3/2}} \tag{A4}$$

has to be worked out. Therefore, the identity

$$\cos^n vu \cos u \cos\Omega u = \frac{1}{2^{n+2}} \sum_{k=0}^{n} C_n^k \{\cos[u(\Omega+1+v(2k-n))] + \cos[u(\Omega-1+v(2k-n))] + \cos[u(v(2k-n)-\Omega+1)] + \cos u[(v(2k-n)-\Omega-1)]\} \tag{A5}$$

is used. Making use of

$$\int_0^\infty \frac{du \cos xu}{(x^2+z^2)^{n+1/2}} = \frac{K_n(xz)x^n\sqrt{\pi}}{\Gamma(n+\frac{1}{2})(2z)^n} = \frac{K_n(xz)x^n}{(2n-1)\cdots 3\cdot 1\, z^n} \ , \tag{A6}$$

there comes (K_n being the normal notation for Bessel functions with imaginary argument)

$$\int_0^\infty du \ \frac{\cos^n vu \cos\Omega u \cos u}{(u^2+a^2)^{n+3/2}} = \frac{1}{2^{n+2}} \sum_{k=0}^{n} C_n^k \left[K_{n+1}(|\Omega+1+v(2k-n)|a)|\Omega+1+v(2k-n)|^{n+1} + K_{n+1}(|\Omega-1+v(2k-n)|a)|\Omega-1+v(2k-n)|^{n+1} + K_{n+1}(|v(2k-n)-\Omega+1|a)|v(2k-n)-\Omega+1|^{n+1} + K_{n+1}(|v(2k-n)-\Omega-1|a)|v(2k-n)-\Omega-1|^{n+1}\right] \frac{a^{-n-1}}{(2n+1)\cdots 3\cdot 1} \quad . \tag{A7}$$

Because of the uniform convergence of the series obtained for $\mathrm{Im}\chi(\Omega)$ the limit sign and the summation sign may be interchanged so that the calculation reduces to evaluating the following limit:

$$\lim_{\beta\to\infty} \beta^{3/2} \frac{\sinh(\frac{1}{2}\beta\Omega)}{\sinh(\frac{1}{2}\beta)} \frac{b^n}{2^{n+2}} \sum_{k=0}^{n} C_n^k \left[K_{n+1}(|\Omega+1+v(2k-n)|a)|\Omega+1+v(2k-n)|^{n+1} + K_{n+1}(|\Omega-1+v(2k-n)|a)|\Omega-1+v(2k-n)|^{n+1} + K_{n+1}(|v(2k-n)-\Omega+1|a)|v(2k-n)-\Omega+1|^{n+1} + K_{n+1}(|v(2k-n)-\Omega-1|a)|v(2k-n)-\Omega-1|^{n+1}\right] \frac{a^{-n-1}}{(2n+1)\cdots 3\cdot 1} \quad . \tag{A8}$$

For β sufficiently large one has

$$K_n(a|x|) \cong \left(\frac{\pi}{2a|x|}\right)^{1/2} e^{-\beta|x|} \sum_{m=0}^{p-1} C_{n-1/2}^m \frac{1}{(2a|x|)^m} \frac{\Gamma(n+m+\frac{1}{2})}{\Gamma(n+\frac{1}{2})}$$

and

$$b \cong 2R\beta e^{-\beta v/2}, \quad a^2 = \tfrac{1}{4}\beta^2 + R\beta\coth(\tfrac{1}{2}\beta v), \quad a \cong \tfrac{1}{2}\beta + R\coth\tfrac{1}{2}\beta v, \quad \sinh(\tfrac{1}{2}\beta\Omega) \cong \tfrac{1}{2}e^{\beta\Omega/2}, \quad \sinh(\tfrac{1}{2}\beta) \cong \tfrac{1}{2}e^{\beta/2} . \tag{A10}$$

This limit is equal to

$$\lim_{\beta\to\infty} \frac{2^{n-1}R^n\sqrt{\pi}}{(2n+1)\cdots 5\cdot 3\cdot 1} e^{(\beta/2)(\Omega-1-nv)} \sum_{k=0}^{n} C_n^k \left[|\Omega+1+v(2k-n)|^{n+1/2} e^{-(\beta/2)|\Omega+1+v(2k-n)|-R|\Omega+1+v(2k-n)|} + |\Omega-1+v(2k-n)|^{n+1/2} e^{-(\beta/2)|\Omega-1+v(2k-n)|-R|\Omega-1+v(2k-n)|} + |v(2k-n)-\Omega+1|^{n+1/2} e^{-(\beta/2)|v(2k-n)-\Omega+1|-R|-\Omega+1+v(2k-n)|} + |v(2k-n)-\Omega-1|^{n+1/2} e^{-(\beta/2)|v(2k-n)-\Omega-1|-R|v(2k-n)-\Omega-1|}\right] , \tag{A11}$$

which results in

$$\frac{2^n R^n\sqrt{\pi}}{(2n+1)\cdots 5\cdot 3\cdot 1} |\Omega-1-nv|^{n+1/2} \times e^{-R|\Omega-1-nv|} \frac{1+\mathrm{sgn}(\Omega-1-nv)}{2} \quad . \tag{A12}$$

So that there comes as a final result for $\mathrm{Im}\chi(\Omega)$

$$\mathrm{Im}\chi(\Omega) = \tfrac{2}{3}\alpha\left(\frac{v}{w}\right)^3 \sum_{n=0}^{\infty} C_n^{-3/2} \frac{(-1)^n 2^n R^n}{(2n+1)\cdots 3\cdot 1} \times |\Omega-1-nv|^{n+1/2} e^{-R|\Omega-1-nv|} \times \tfrac{1}{2}[1+\mathrm{sgn}(\Omega-1-nv)] \quad . \tag{A13}$$

The first two terms of this expansion are also given in FHIP's work.

APPENDIX B

The calculation of

$$\mathrm{Im}\int_0^\infty \frac{du\,\sin\Omega u\, e^{iu}}{[R(1-e^{ivu})-iu]^{3/2}} \tag{B1}$$

and

$$\mathrm{Im}\int_0^\infty \frac{du(1-\cos\Omega u)\, e^{iu}}{[R(1-e^{ivu})-iu]^{3/2}} \tag{B2}$$

is performed. In the complex plane it is easily seen that

$$\left|\frac{Re^{ivu}}{R-iu}\right| \leq 1 . \tag{B3}$$

The equality occurs if and only if $u=0$, so that

$$\frac{1}{[R(1-e^{ivu})-iu]^{3/2}} = \frac{1}{[R-iu]^{3/2}} \sum_{n=0}^{\infty} C^n_{-3/2}(-1)^n \frac{R^n e^{invu}}{[R-iu]^n} . \tag{B4}$$

Therefore, the following equalities hold

$$\mathrm{Im}\int_0^\infty \frac{du\,\sin\Omega u\, e^{iu}}{[R(1-e^{ivu})-iu]^{3/2}} = \sum_{n=0}^{\infty} C^n_{-3/2}(-1)^n R^n \times \mathrm{Im}\int_0^\infty \frac{du\,\sin\Omega u\, e^{i(1+nv)u}}{[R-iu]^{3/2+n}} \tag{B5}$$

and

$$\mathrm{Im}\int_0^\infty \frac{du(1-\cos\Omega u)\, e^{iu}}{[R(1-e^{ivu})-iu]^{3/2}} = \sum_{n=0} C^n_{-3/2}(-1)^n R^n \times \mathrm{Im}\int_0^\infty \frac{du\,(1-\cos\Omega u)\, e^{i(1+nv)u}}{(R-iu)^{3/2+n}} . \tag{B6}$$

Making use of the following notations:

$$\varphi(R,\Omega,n,v) = \mathrm{Im}\int_0^\infty \frac{du\,\sin\Omega u\, e^{i(1+nv)u}}{(R-iu)^{3/2+n}} , \tag{B7}$$

$$\psi(R,\Omega,n,v) = \mathrm{Im}\int_0^\infty \frac{du\,(1-\cos\Omega u)\, e^{i(1+nv)}}{(R-iu)^{3/2+n}} , \tag{B8}$$

it is easily seen that φ and ψ satisfy the following relation:

$$-\frac{1}{n+3/2}\,\frac{\partial\phi(R,\Omega,n,n+1/n)\,v}{\partial R} = \phi(R,\Omega,n+1,v) , \tag{B9}$$

so that, to calculate all φ and ψ it is sufficient to evaluate

$$E(a,R) = \int_0^\infty \frac{du\, e^{-iua}}{(R-iu)^{5/2}} \quad \text{for real } a \text{ and } R . \tag{B10}$$

This integral can be evaluated in different manners and it equals

$$E(a,R) = \tfrac{2}{3}\, iR^{-3/2} + \tfrac{4}{3}\, a^2 e^{-aR}\{(1+i)(\pi/|a|)^{1/2} \times [1-i\,\mathrm{sgn}(a)] - i2R^{1/2}\int_0^1 dt\, e^{aRt^2}\} , \tag{B11}$$

from which it follows that

$$\varphi(R,\Omega,1,v) = \tfrac{1}{2}\,\mathrm{Im}\{-iE[-(\Omega+1+v),R] + iE[\Omega-1-v,R]\} = \tfrac{1}{3}\,|\Omega-1-v|^{3/2}\pi^{1/2}e^{-(\Omega-1-v)R}[1+\mathrm{sgn}(\Omega-1-v)] ; \tag{B12}$$

an analogous calculation gives

$$\varphi(R,\Omega,0,0) = |\Omega-1|^{1/2} e^{-(\Omega-1)R}\,\pi^{1/2}\,\tfrac{1}{2}[1+\mathrm{sgn}(\Omega-1)] . \tag{B13}$$

Using the recurrence relation φ can be written as

$$\varphi(R,\Omega,n,v) = \frac{\pi^{1/2}2^n|\Omega-1-nv|^{n+1/2}}{(2n+1)\cdots 3\cdot 1}\, e^{-|\Omega-1-nv|R} \times \frac{1+\mathrm{sgn}(\Omega-1-nv)}{2} . \tag{B14}$$

Also writing $\cos a$ as $\frac{1}{2}(e^{ia}+e^{-ia})$ there comes, after a few elementary calculations,

$$\psi(R,\Omega,1,v) = -\tfrac{2}{3}(\Omega+1+v)^2 e^{(\Omega+1+v)R}[(\pi/|\Omega+1+v|)^{1/2} - 2R^{1/2}\int_0^1 dt\, e^{(\Omega-1-v)Rt^2}] - \tfrac{2}{3}(\Omega-1-v)^2 e^{-(\Omega-1-v)R} \times\{\tfrac{1}{2}(\pi/|\Omega-1-v|)^{1/2}[1-\mathrm{sgn}(\Omega-1-v)] - 2R^{1/2}\int_0^1 dt\, e^{(\Omega-1-v)Rt^2}\} + \tfrac{4}{3}(1+v)^2 e^{(1+v)R}[(\pi/|1+v|)^{1/2} - 2R^{1/2}\int_0^1 dt\, e^{-(1+v)Rt^2}] . \tag{B15}$$

The notation

$$A(a,b,R) = \tfrac{2}{3}\,a^2 e^{-aR}(b - 2R^{1/2}\int_0^1 dt\, e^{aRt^2})$$

is introduced. This gives the expression

$$\psi(R,\Omega,1,v) = 2A\left(-(1+v),\ \frac{\pi^{1/2}}{(|1+v|)^{1/2}},R\right) - A\left(-(\Omega+1+v),\ \frac{\pi^{1/2}}{(|\Omega+1+v|)^{1/2}},R\right) - A\left(\Omega-1-v,\ \frac{\pi^{1/2}}{(|\Omega-1-v|)^{1/2}},\ \frac{1-\mathrm{sgn}(\Omega-1-v)}{2},\ R\right) . \tag{B16}$$

For every n, ψ takes the form

$$\psi(R,\Omega,n,v)=2B_n\left(-(1+nv),\ \frac{\pi^{1/2}}{(|1+nv|)^{1/2}},\ R\right)-B_n\left(-(\Omega+1+nv),\ \frac{\pi^{1/2}}{(|\Omega+1+nv|)^{1/2}},\ R\right)$$
$$-B_n\left(\Omega-1-nv,\ \frac{\pi^{1/2}}{(|\Omega-1-nv|)^{1/2}},\ \frac{1-\mathrm{sgn}(\Omega-1-nv)}{2},\ R\right), \quad \text{(B17)}$$

with B_n defined as

$$B_n(a,b,R)=\frac{(-1)^{n-1}2^n}{(2n+1)\cdots5\cdot3\cdot1}\frac{\partial^{n-1}}{\partial R^{n-1}}A(a,b,R). \quad \text{(B18)}$$

The derivation is easily carried out and results in

$$B_n(a,b,R)=\frac{2^n}{(2n+1)\cdots3\cdot1}\left(a^{n+1}e^{-aRb}+a^2\sum_{i=0}^{n-2}{}'a^{n-2-i}\tfrac{1}{2}\cdot\tfrac{3}{2}\cdots\tfrac{1}{2}(2i-1)R^{-(2i+1)/2}-2a^{n+1}e^{-aR}\int_0^{R^{1/2}}dv\,e^{av^2}\right) \quad \text{(B19)}$$

[$\sum'$ means that for $i=0$ one has $\frac{1}{2}\cdot\frac{3}{2}\cdots\frac{1}{2}(2i-1)=1$] valid for every $n\geq1$. In the case $n=0$, $B(a,b,R)$ becomes

$$B_0(a,b,R)=R^{-1/2}+ae^{-aR}(b-2R^{1/2}\int_0^1 dt\,e^{aRt^2}). \quad \text{(B20)}$$

For $n=0$ the function ψ takes the same form as in the case $n\geq1$.

APPENDIX C

To study the integral

$$I=\mathrm{Im}\int_0^\infty[du\,(1-\cos\Omega u)\,e^{i(1+nv)u}/(R-iu)^{n+3/2}] \quad \text{(C1)}$$

numerically it is necessary to transform it to a rapidly converging expression. Using the integral transformation

$$\frac{1}{(R-iu)^{3/2+n}}=\frac{1}{\Gamma(n+\frac{3}{2})}\int_0^\infty dx\,e^{-(R-iu)x}\,x^{n+1/2} \quad (R>0), \quad \text{(C2)}$$

I can be written as

$$I=\lim_{\epsilon\to0}\mathrm{Im}\int_0^\infty du\,(1-\cos\Omega u)\,e^{i(1+nv)u}[\Gamma(n+\tfrac{3}{2})]^{-1}\times\int_0^\infty dx\,e^{-Rx+iux-\epsilon u}x^{n+1/2}; \quad \text{(C3)}$$

or, interchanging the order of integration,

$$I=\lim_{\epsilon\to0}\frac{1}{\Gamma(n+\frac{3}{2})}\mathrm{Im}\int_0^\infty dx\,x^{n+1/2}e^{-Rx}\times\int_0^\infty du\,(1-\cos\Omega u)\,e^{i(1+nv+x)u-\epsilon u}. \quad \text{(C4)}$$

The integration over the variable u is elementary and gives

$$I=\lim_{\epsilon\to0}\frac{1}{\Gamma(n+\frac{3}{2})}\mathrm{Im}\int_0^\infty dx\,x^{n+1/2}e^{-Rx}\times\frac{\epsilon-i(1+nv+x)}{[\epsilon-i(1+nv+x)]^2+\Omega'^2}\Bigg|_{\Omega'=\Omega}^{\Omega'=0}. \quad \text{(C5)}$$

After a partial integration one obtains

$$I=\lim_{\epsilon\to0}\frac{1}{2\Gamma(n+\frac{3}{2})}\int_0^\infty dx\left(\frac{d}{dx}x^{n+1/2}e^{-Rx}\right)\times\ln|[\epsilon-i(1+nv+x)]^2+\Omega'^2|\Big|_{\Omega'=\Omega}^{\Omega'=0} \quad \text{(C6)}$$

or

$$I=-\frac{1}{\Gamma(n+\frac{3}{2})}\int_0^\infty dx\,[(n+\tfrac{1}{2})x^{n-1/2}e^{-Rx}-Rx^{n+1/2}e^{-Rx}]\times\ln\left|\frac{(1+nv+x)^2}{\Omega^2-(1+nv+x)^2}\right|^2. \quad \text{(C7)}$$

These integrals ($n=1,\ 2,\ \ldots$) can be performed numerically using Gaussian quadrature formulas.

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[1]V. L. Gurevich, I. E. Lang, and Yu. A. Firsov, Fiz. Tverd. Tela 4, 1252 (1962) [Sov. Phys. Solid State 4, 918 (1962)].

[2]J. Devreese, W. Huybrechts, and L. Lemmens, Phys. Status Solidi 48b, 77 (1971).

[3]E. Kartheuser, R. Evrard, and J. Devreese, Phys. Rev. Letters 22, 94 (1969).

[4]D. Larsen, Phys. Rev. 187, 1147 (1969).

[5]T. D. Schultz, MIT Solid State and Molecular Theory Group Technical Report No. 9, 1956 (unpublished).

[6]J. Devreese and R. Evrard, Phys. Letters 11, 278 (1966); R. Evrard, *ibid.* 14, 295 (1956).

[7]R. Feynman, R. Hellwarth, C. Iddings, and P. Platzman, Phys. Rev. 127, 1004 (1962).

[8]R. P. Feynman, Phys. Rev. 97, 660 (1955).

[9]J. Devreese and R. Evrard, Phys. Letters 23, 196 (1966).

[10]T. D. Schultz, in *Polarons and Excitons*, edited by C. G. Kuper and G. D. Whitfield (Oliver and Boyd, London, 1962), p. 71.

[11]M. Goovaerts, J. De Sitter, and J. Devreese, Studie-

Centrum Kernenergie Centre d'Etudes Nucléaires Report No. BLG439 (unpublished).

[12]K. Thornber and R. Feynman, Phys. Rev. B 1, 4099 (1970).

[13]K. Thornber, Phys. Rev. B 3, 1929 (1971).

[14]H. Finkenrath, N. Uhle, and W. Waidelich, Solid State Commun. 7, 11 (1969).

[15]J. Devreese, L. Lemmens, R. Evrard, and E. Kartheuser, Mem. Soc. Roy. Sci. Liège 20, 249 (1970). (1970).