

## Phonon-Correlation Effects in the Optical-Absorption Spectra of Free Polarons

W. Huybrechts\* and J. Devreese\*

*Physics Department, University of Antwerp, Antwerp, Belgium*

(Received 24 May 1973)

The influence of correlation between the optical phonons on the one- and two-phonon contribution to the free-polaron absorption spectrum in polar semiconductors ( $\alpha < 1$ ) is evaluated by extending the Lee-Low-Pines formalism. This correlation can be related to the concept of the relaxed excited state in the intermediate and strong electron-phonon coupling region ( $\alpha > 3$ ).

### I. INTRODUCTION

The infrared intraband absorption due to the interaction between a free charge carrier and the optical phonons of a polar material has been studied recently in the Feynman model of the free polaron for all values of the electron-LO-phonon coupling.<sup>1-3</sup> It has been shown<sup>2</sup> that even for "weak coupling" (say  $\alpha \approx 1$ , where  $\alpha$  is the dimensionless electron-phonon coupling constant) the obtained absorption spectrum deviates from the second-order perturbation result (which is of order  $\alpha$ ).

The purpose of the present work is to derive an expression for the free-polaron absorption coefficient, taking into account correlation between optical phonons. Furthermore, the relative importance of one- and two-phonon processes in the absorption process will be evaluated.

We start from the well-known relationship between the absorption coefficient  $\Gamma(\Omega)$  as a function of the frequency of light  $\Omega$  and the probability that a photon be absorbed by the polaron in its ground state:

$$\Gamma(\Omega) = \frac{4\pi N e^2}{m^2 n c} F(\Omega), \quad (1)$$

where  $n$  is the refraction index,  $N$  is the density of polarons, and  $m$  is the band mass of the electron. In (1) the independent particle approximation has been made. In the dipole approximation for the oscillating field, the function  $F(\Omega)$  is given by ( $\hbar = m = \omega = 1$ ;  $\omega$  is the LO-phonon frequency):

$$F(\Omega) = \frac{\pi}{\Omega} \sum_f |\langle i | \vec{u} \cdot \vec{p} | f \rangle|^2 \times \delta(E_f - E_i - \Omega), \quad (2)$$

where  $\vec{p}$  is the momentum of the electron,  $\vec{u}$  is the unit vector in the direction of the polarization of light,  $|i\rangle$  is the ground state of the polaron with energy  $E_i$ , and  $|f\rangle$  represents the possible final states to which the polaron can be excited.

Equation (2) will be evaluated in the canonical-transformation formalism by introducing, in addition to the two Lee-Low-Pines transformations,

a third unitary transformation, which takes account of correlation in processes involving two LO phonons.

As a result of this simple analytic calculation, an expression for the absorption coefficient is obtained which contains a term linear in  $\alpha$  (corresponding to the second-order perturbation result<sup>4</sup>) and a term quadratic in the coupling. This last contribution will demonstrate the importance of correlation between virtual phonons of the polaron on the absorption spectrum even at weak coupling. This correlation will be related to the concept of the relaxed excited state (as studied in Refs. 1 and 3) in the intermediate and strong coupling region.

A further consequence of the present calculation is that the emission of the two correlated free LO phonons by the polaron can be neglected at weak coupling. This means that in the region  $\alpha < 1$ , the absorption spectrum can be considered as due to emission of one optical phonon by the polaron. In fact this latter point is somewhat surprising, as one would expect a two-phonon threshold which becomes more pronounced when  $\alpha$  increases.

### II. CALCULATION OF THE ABSORPTION COEFFICIENT

The Gurevich-Lang-Firsov perturbation result<sup>4</sup> (Appendix A) gives the absorption coefficient in linear order of the electron-LO-phonon coupling constant  $\alpha$ . In this approximation the correlation between LO phonons as a result of their interaction with the charge carrier is neglected. This is only valid in the asymptotic limit  $\alpha \rightarrow 0$ . For larger values of the coupling constant  $\alpha$ , such correlation should be included. In the following calculation of the optical-absorption coefficient, correlation is taken into account to order  $\alpha^2$ . In the first step the matrix element in Eq. (2),

$$\langle f | \vec{u} \cdot \vec{p} | i \rangle \quad (3)$$

is rewritten by performing the two Lee-Low-Pines canonical transformations:

$$U_1 = \exp\left(-i \sum_{\vec{k}} \vec{k} \cdot \vec{r} a_{\vec{k}}^\dagger a_{\vec{k}}\right) \quad (4)$$

$$U_2 = \exp\left(\sum_{\vec{k}} [f(\vec{k})a_{\vec{k}}^\dagger - f^*(\vec{k})a_{\vec{k}}]\right). \quad (5)$$

In the new representation  $\vec{p}$  is a  $c$  number, which represents the total polaron momentum and which can be taken equal to zero at zero temperatures. As a result of the evaluation of the two transformations  $U_1$  and  $U_2$ , the matrix element (3) is transformed into

$$\langle f' | \sum_{\vec{k}} (\vec{u} \cdot \vec{k}) [a_{\vec{k}}^\dagger + f^*(\vec{k})] [a_{\vec{k}} + f(\vec{k})] | i' \rangle, \quad (6)$$

where  $|i'\rangle = (U_1 U_2)^{-1} |i\rangle$  and  $|f'\rangle = (U_1 U_2)^{-1} |f\rangle$ .

In analogy with the wave function for the ground state of the polaron, as used by Röseler,<sup>5</sup> a third unitary transformation  $U_3$  is proposed<sup>6</sup>:

$$U_3 = \exp\left(\frac{1}{2} \sum_{\vec{k}_1, \vec{k}_2} g(\vec{k}_1, \vec{k}_2) (a_{\vec{k}_1}^\dagger a_{\vec{k}_2}^\dagger - a_{\vec{k}_1} a_{\vec{k}_2})\right). \quad (7)$$

One has

$$g(\vec{k}_1, \vec{k}_2) = g(\vec{k}_2, \vec{k}_1) = g^*(\vec{k}_1, \vec{k}_2). \quad (8)$$

The functions  $f(\vec{k})$  and  $g(\vec{k}, \vec{k}')$  can be evaluated by means of a variational calculation of the ground-state energy  $E_1$ .<sup>5</sup> This results in

$$f(\vec{k}) = f_1(\vec{k}) + f_2(\vec{k}), \quad (9a)$$

$$f_1(\vec{k}) = V_n / \omega_{\vec{k}}, \quad (9b)$$

$$f_2(\vec{k}) = \sum_{\vec{k}'} \frac{(\vec{k} \cdot \vec{k}')^2 f(\vec{k}) |f(\vec{k}')|^2}{\omega_{\vec{k}}(\omega_{\vec{k}} + \omega_{\vec{k}'} + \vec{k} \cdot \vec{k}')}, \quad (9c)$$

$$g(\vec{k}, \vec{k}') = \frac{(\vec{k} \cdot \vec{k}') f(\vec{k}) f(\vec{k}')}{(\omega_{\vec{k}} + \omega_{\vec{k}'} + \vec{k} \cdot \vec{k}')}, \quad (10)$$

$$\omega_{\vec{k}} = 1 + \frac{1}{2} k^2. \quad (11)$$

Applying the unitary transformation  $U_3$  of Eq. (7) to  $[a_{\vec{k}}^\dagger + f^*(\vec{k})]$  and  $[a_{\vec{k}} + f(\vec{k})]$  in Eq. (6), and retaining for both operators terms up to order  $\alpha$ , one obtains

$$\begin{aligned} \langle f'' | \sum_{\vec{k}} (\vec{u} \cdot \vec{k}) \left[ a_{\vec{k}}^\dagger + \sum_{\vec{k}'} g(\vec{k}, \vec{k}') a_{\vec{k}'}^\dagger + f^*(\vec{k}) \right] \\ \times \left[ a_{\vec{k}} + \sum_{\vec{k}'} g(\vec{k}, \vec{k}') a_{\vec{k}'} + f(\vec{k}) \right] | i'' \rangle, \end{aligned} \quad (12)$$

with

$$|f''\rangle = U_3^{-1} |f'\rangle$$

and

$$|i''\rangle = U_3^{-1} |i'\rangle. \quad (13)$$

In the new representation the ground state of the polaron is represented by  $|i''\rangle = |0\rangle$  with  $a_{\vec{k}} |0\rangle = 0$ ; Eq. (12) to order linear in  $\alpha$  is given by

$$\langle f'' | A | 0 \rangle, \quad (14a)$$

with

$$A = \sum_{\vec{k}} (\vec{u} \cdot \vec{k}) \left( \sum_{\vec{k}'} g(\vec{k}, \vec{k}') a_{\vec{k}}^\dagger a_{\vec{k}'}^\dagger + f(\vec{k}) a_{\vec{k}}^\dagger \right)$$

$$+ \sum_{\vec{k}'} g(\vec{k}, \vec{k}') f^*(\vec{k}) a_{\vec{k}'}^\dagger. \quad (14b)$$

The one-phonon peak in the infrared absorption spectrum of free polarons results from the emission of one optical phonon by the polaron in its ground state, caused by the absorption of a photon with energy  $\Omega$ . In this case the possible final states  $|f''\rangle$  are given by

$$|f''\rangle = a_{\vec{k}}^\dagger |0\rangle \quad (15)$$

and the corresponding energy is

$$E_f = E_1 + (1 + \frac{1}{2} k^2). \quad (16)$$

The one-phonon contribution to the absorption spectrum can be written as

$$F_1(\Omega) = \frac{\pi}{\Omega} \sum_{\vec{k}} \delta((1 + \frac{1}{2} k^2) - \Omega) |\langle 0 | a_{\vec{k}} A | 0 \rangle|^2. \quad (17)$$

Working out Eq. (17), using (14), we find a term linear in  $\alpha$ :

$$\frac{\pi}{\Omega} \sum_{\vec{k}} \delta((1 + \frac{1}{2} k^2) - \Omega) (\vec{u} \cdot \vec{k})^2 |f_1(\vec{k})|^2; \quad (18a)$$

and a correction term quadratic in  $\alpha$ :

$$\begin{aligned} \frac{2\pi}{\Omega} \sum_{\vec{k}} \delta((1 + \frac{1}{2} k^2) - \Omega) \left( (\vec{u} \cdot \vec{k})^2 f_1(\vec{k}) f_2^*(\vec{k}) \right. \\ \left. + \sum_{\vec{k}'} (\vec{u} \cdot \vec{k}) (\vec{u} \cdot \vec{k}') f_1^*(\vec{k}) f_1(\vec{k}') g(\vec{k}, \vec{k}') \right). \end{aligned} \quad (18b)$$

In order to evaluate  $F_1(\Omega)$ , the summation over  $\vec{k}$  and  $\vec{k}'$  is converted into an integration over  $\vec{k}$  and  $\vec{k}'$ . As a result of the integration over  $\vec{k}$  in Eq. (18a) the Gurevich-Lang-Firsov perturbation result is obtained.

The two terms in Eq. (18b) are of order  $\alpha^2$  and can be taken together to give:

$$\frac{2}{3} \frac{\alpha^2}{\pi} \frac{[2(\Omega - 1)]^{1/2}}{\Omega^4} (\Omega - 2) I(k_0), \quad (19a)$$

where

$$I(k_0) = \int_0^\infty dk' \int_{-1}^1 dt \frac{k'^2 t^2}{(1 + \frac{1}{2} k'^2)^2 (2 + \frac{1}{2} k_0^2 + \frac{1}{2} k'^2 + k_0 k' t)}, \quad (19b)$$

with  $k_0 = [2(\Omega - 1)]^{1/2}$  and  $t = \cos(\vec{k}, \vec{k}')$ . Performing the integration over  $t$  in Eq. (19b),  $I(k_0)$  is reduced to

$$\begin{aligned} I(k_0) = \frac{-4}{k_0^2} \int_0^\infty dk' \frac{4 + k_0^2 + k'^2}{(2 + k'^2)^2} \\ + \frac{1}{k_0^3} \int_0^\infty \frac{dk'}{k'} \left( \frac{4 + k_0^2 + k'^2}{2 + k'^2} \right)^2 \ln \frac{4 + (k_0 + k')^2}{4 + (k_0 - k')^2}, \end{aligned} \quad (20)$$

where the second integral can be calculated using the formulas given in Appendix B.

The one-phonon peak in the absorption spectrum up to order  $\alpha^2$  ( $\Omega \geq 1$ ) is given by

$$F_1(\Omega) = \frac{2\alpha}{3} \left[ \frac{(\Omega-1)^{1/2}}{\Omega^3} \left( 1 + \alpha \frac{\Omega-2}{\Omega} B(\Omega) \right) \right], \quad (21a)$$

where  $B(\Omega)$  is a slowly varying function which is positive for all frequencies  $\Omega > 1$ :

$$B(\Omega) = \frac{1}{\Omega-1} \left[ \frac{(\Omega+1)^2}{(\Omega-1)^{1/2}} \arctan\left(\frac{\Omega-1}{2}\right)^{1/2} - \frac{\Omega(\Omega+2)}{(\Omega-1)^{1/2}} \arctan\frac{(\Omega-1)^{1/2}}{1+\sqrt{2}} - \frac{\Omega^2 + (2+\sqrt{2})\Omega + 2 + 2\sqrt{2}}{2+2\sqrt{2}+\Omega} \right]. \quad (21b)$$

If the absorption process leads to the emission of two real phonons, the possible final states  $|f''\rangle$  and their energy  $E_f$  are

$$|f''\rangle = a_{\vec{k}}^\dagger a_{\vec{k}'}^\dagger |0\rangle \quad (22)$$

and

$$E_f = E_i + [2 + \frac{1}{2}(\vec{k} + \vec{k}')^2]. \quad (23)$$

To order  $\alpha^2$  the two-phonon contribution to the optical-absorption coefficient takes the form

$$F_2(\Omega) = \frac{\pi}{\Omega} \frac{1}{2} \sum_{\vec{k}, \vec{k}'} \delta(2 + \frac{1}{2}(\vec{k} + \vec{k}')^2 - \Omega) \times |\langle 0 | a_{\vec{k}} a_{\vec{k}'} A | 0 \rangle|^2, \quad (24)$$

with  $A$  given by (14b). It follows that

$$F_2(\Omega) = \frac{\pi}{2\Omega} \sum_{\vec{k}, \vec{k}'} \delta(2 + \frac{1}{2}(\vec{k} + \vec{k}')^2 - \Omega) \times [\vec{u} \cdot (\vec{k} + \vec{k}')]^2 g^2(\vec{k}, \vec{k}'). \quad (25)$$

After converting the summation over  $\vec{k}$  and  $\vec{k}'$  into an integral, the change of variables,

$$\vec{k} + \vec{k}' = \vec{v}, \quad \vec{k} = \vec{k}, \quad (26)$$

is made. So one obtains

$$F_2(\Omega) = \frac{\alpha^2}{16\pi^3 \Omega} \int d\vec{v} \int d\vec{k} \delta(\Omega - (2 + \frac{1}{2}v^2)) \frac{(\vec{u} \cdot \vec{v})^2 (\vec{v} \cdot \vec{k} - k^2)^2}{k^2 (1 + \frac{1}{2}k^2)^2 (v^2 + k^2 - 2\vec{v} \cdot \vec{k}) (1 + \frac{1}{2}v^2 + \frac{1}{2}k^2 - \vec{v} \cdot \vec{k})^2 (2 + \frac{1}{2}v^2)^2}. \quad (27)$$

The integration over  $\vec{v}$  reduces  $F_2(\Omega)$  to

$$F_2(\Omega) = \frac{\sqrt{2}\alpha^2}{3\pi} \frac{(\Omega-2)^{3/2}}{\Omega^3} I(\Omega), \quad (28a)$$

with

$$I(\Omega) = \int_0^\infty dk \int_{-1}^1 dt \frac{(v_0 kt - k^2)^2}{(1 + \frac{1}{2}k^2)^2 (v_0^2 + k^2 - 2v_0 kt) (1 + \frac{1}{2}v_0^2 + \frac{1}{2}k^2 - v_0 kt)^2} \quad (28b)$$

and

$$v_0 = [2(\Omega-2)]^{1/2} \quad (\Omega \geq 2), \\ t = \cos(\vec{k}, \vec{v}).$$

After the integration over  $t$ , the remaining integral over  $k$  can be transformed to an expression involving integrals of the type given in Appendix B.

Finally, the two-phonon contribution to the infrared absorption spectrum in polar semiconductors is given by ( $\Omega \geq 2$ ):

$$F_2(\Omega) = \frac{\alpha^2}{3} \frac{(\Omega-2)^{3/2}}{\Omega^3} C(\Omega), \quad (29a)$$

with

$$C(\Omega) = \frac{\Omega^2 - 4\Omega + 2}{(\Omega-2)^{1/2}} \arctan \frac{(\Omega-2)^{1/2}}{2} - \frac{2(\Omega^2 - 4\Omega + 3)}{(\Omega-2)^{1/2}} \arctan(\Omega-2)^{1/2} + \frac{\pi(\Omega-2)^{3/2}}{2} - \frac{2(\Omega^2 + 2\Omega - 2)}{(\Omega+2)^2}. \quad (29b)$$

### III. DISCUSSION

As the influence of polaron effects on the refraction index  $n$  can be neglected, the structure of the absorption coefficient of free polarons is given by

$$F(\Omega) = F_1(\Omega)H(\Omega-1) + F_2(\Omega)H(\Omega-2), \quad (30)$$

where  $H(x)$  is the Heaviside unit function and where the one-phonon peak  $F_1(\Omega)$  and the two-phonon contribution  $F_2(\Omega)$ , respectively, are given by Eq. (21) and Eq. (29).

The structure of the absorption coefficient (30) to order  $\alpha^2$  is compared with the Gurevich-Lang-Firsov (GLF) perturbation result in Fig. 1. As a consequence of the correlation between the optical phonons, the intensity of the absorption peak is decreased with respect to the GLF result. At higher frequencies ( $\Omega > 2$ ) the absorption curve lies slightly higher than the perturbation result. This increase is a consequence of the broadening of the one-phonon peak and of the emission of two correlated phonons by the polaron.

The influence of the correlation between the op-

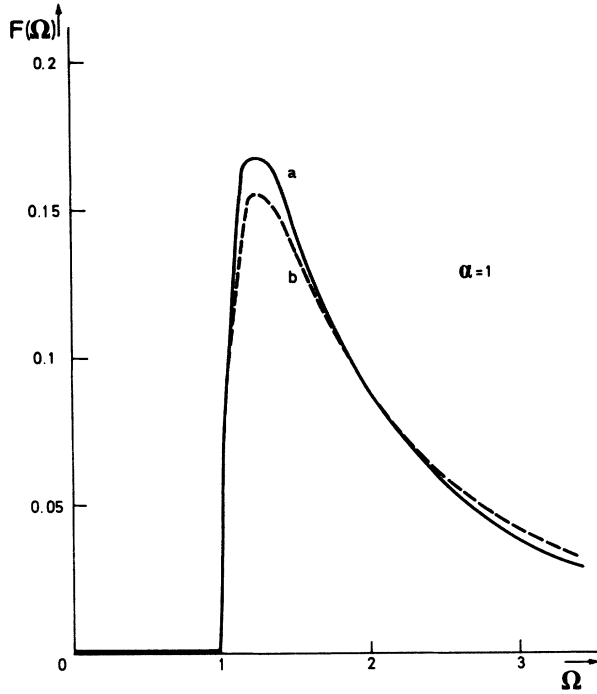


FIG. 1. The absorption coefficient of free polarons, taking into account correlation between optical phonons (30) (curve b), is compared with the second-order perturbation result linear in the electron-phonon coupling (Refs. 2 and 4) (curve a), ( $\alpha = 1$ ).

tical phonons on the absorption spectrum presumably provides the explanation for the difference which exists at weak coupling between the calculation of the absorption coefficient in the quadratic approximation of the polaron where phonon correlation is included,<sup>1</sup> and the GLF result.<sup>4</sup>

As a last point we can remark that a sum rule, relating the first moment of the absorption coefficient to the electronic part of the polaron self-energy has been discussed in Ref. 7:

$$\int_0^{\infty} \Omega F(\Omega) d\Omega = \pi \langle i | (\vec{u} \cdot \vec{p})^2 | i \rangle$$

or

$$\int_0^{\infty} \Omega F(\Omega) d\Omega = \frac{2}{3} \pi E_{e1}, \quad (31)$$

where  $E_{e1}$  can be calculated analytically by means of canonical transformations<sup>5</sup>:

$$E_{e1} = \frac{1}{2} \alpha + [2 \ln(1 + \sqrt{2}) - \frac{3}{2} \ln 2 - \frac{1}{2} \sqrt{2}] \alpha^2$$

The numerical evaluation of the integral in (31) with  $F(\Omega)$  given by (30) confirms this result.

#### IV. CONCLUSION

We have calculated the optical absorption for free polarons in the "weak" coupling regime, taking into account correlation in processes involving two phonons.

This was performed by extending the Lee-Low-Pines canonical transformations with a third unitary transformation inspired by a wave function proposed by Röseler.<sup>5</sup> We find for the optical-absorption coefficient a term linear in  $\alpha^{2,4}$  and a correction term quadratic in the coupling:  $K(\Omega)\alpha^2$ . The coefficient  $K(\Omega)$  presumably is not exact because, e.g., in the argument of the  $\delta$  function in Eq. (2), which reflects the conservation of energy, the exact dispersion relation for polarons has not been used. Application of the sum rule (31), however, shows that this coefficient is very accurate.

Comparing our results with the optical-absorption coefficient calculated in Refs. 1 and 2, we find the same decrease of the absorption peak for increasing  $\alpha$ . Our result is perturbationlike and our correction is an estimate for the term in  $\alpha^2$ .

From the formula that we introduced for the absorption coefficient in Refs. 1 and 2,

$$F(\Omega) = \frac{\Omega \text{Im} \chi(\Omega)}{\Omega^4 - 2\Omega^2 \text{Re} \chi(\Omega) + |\chi(\Omega)|^2},$$

with

$$-i\Omega z(\Omega) = \Omega^2 - \chi(\Omega),$$

where  $z(\Omega)$  is the dynamical impedance of the polaron, it is clear that such a series expansion valid for  $\Omega > 1$  and for relatively small  $\alpha$  exists.

In conclusion we find a decrease of the weak coupling optical absorption as a result of correlation between the virtual phonons which build up the self-induced polarization cloud of the polaron. In fact this correlation is related to the concept of a relaxed excited state<sup>1,3</sup> (RES), which we propose to describe as follows: Starting from weak coupling and increasing the coupling constant  $\alpha$ , the initial and final states of the polaron are influenced more and more by the correlation between the phonons of the polarization cloud to which the electron distribution is readapted self-consistently. If the correlation is strong enough to enable the electron to be excited into a higher level in the polarization well ( $\alpha > 3$ ), the latter is re-adapted to the new electronic distribution. For strong electron-phonon coupling this tends to a quite stable RES.

#### APPENDIX A

In Eq. (2) the GLF perturbation result linear in  $\alpha$ ,

$$F(\Omega) = \frac{2\alpha}{3} \frac{(\Omega - 1)^{1/2}}{\Omega^3} \quad (\Omega \geq 1) \quad (A1)$$

is derived using the two canonical transformations of Lee-Low-Pines. A simple and elegant derivation of this result is obtained by rewriting the matrix element in Eq. (3) as

$$\frac{E_f - E_i}{\Omega} \langle i | \vec{u} \cdot \vec{p} | f \rangle = \frac{1}{\Omega} \langle i | \vec{u} \cdot [\vec{p}, H] | f \rangle, \quad (A2)$$

where  $H$  is the polaron Hamiltonian and

$$[\vec{p}, H] = \sum_{\vec{k}} \vec{k} (V_{\vec{k}} a_{\vec{k}} e^{i\vec{k}\cdot\vec{r}} - V_{\vec{k}}^* a_{\vec{k}}^\dagger e^{-i\vec{k}\cdot\vec{r}}). \quad (\text{A3})$$

After performing the first Lee-Low-Pines transformation  $U_1$  of Eq. (4), Eq. (A2) reduces to

$$\frac{1}{\Omega} \langle i_1 | \sum_{\vec{k}} (\vec{u} \cdot \vec{k}) (V_{\vec{k}} a_{\vec{k}} - V_{\vec{k}}^* a_{\vec{k}}^\dagger) | f_1 \rangle, \quad (\text{A4})$$

with

$$|i_1\rangle = U_1^{-1} |i\rangle \quad \text{and} \quad |f_1\rangle = U_1^{-1} |f\rangle. \quad (\text{A5})$$

The ground state of the polaron in the new representation is given by  $|i_1\rangle = |0\rangle$  with  $a_{\vec{k}} |0\rangle = 0$  and the final state, corresponding to the emission of one optical phonon is

$$|f_1\rangle = a_{\vec{k}}^\dagger |0\rangle, \quad (\text{A6})$$

with energy

$$E_f = E_i + (1 + \frac{1}{2} k^2). \quad (\text{A7})$$

The evaluation of Eq. (A4) gives

$$\begin{aligned} \langle i | \vec{u} \cdot \vec{p} | f \rangle &= \frac{1}{\Omega} \langle 0 | \sum_{\vec{k}} (\vec{u} \cdot \vec{k}') \\ &\quad \times (V_{\vec{k}'} a_{\vec{k}'} - V_{\vec{k}'}^* a_{\vec{k}'}^\dagger) a_{\vec{k}}^\dagger | 0 \rangle \\ &= \frac{1}{\Omega} (\vec{u} \cdot \vec{k}) V_{\vec{k}}. \end{aligned} \quad (\text{A8})$$

Using Eq. (A8),  $F(\Omega)$  of Eq. (2) is:

$$F(\Omega) = \frac{\pi}{\Omega^3} \sum_{\vec{k}} (\vec{u} \cdot \vec{k})^2 |V_{\vec{k}}|^2 \delta(1 + \frac{1}{2} k^2 - \Omega) \quad (\text{A9})$$

Converting the summation over  $\vec{k}$  into an integral, the GLF result (A1) is obtained. In cgs units one has

$$\begin{aligned} \Gamma(\Omega) &= 0 & (\Omega < \omega) \\ \Gamma(\Omega) &= \frac{4\pi N e^2}{mnc\omega} \frac{2\alpha}{3} \frac{(\Omega/\omega - 1)^{1/2}}{(\Omega/\omega)^3} & (\Omega \geq \omega). \end{aligned} \quad (\text{A10})$$

In this way the perturbation result in order  $\alpha$  is obtained directly from the dynamical properties of the polaron without a variational calculation of the ground-state energy, as was necessary in Ref. 2.

#### APPENDIX B

In order to calculate the remaining integrals, (20) and (28b), the following types of integrals<sup>8</sup> are used ( $a > 0$  and  $b > 0$ ):

$$\begin{aligned} \int_0^\infty \frac{dx}{x} \frac{1}{a^2 + x^2} \ln \frac{b^2 + (x+c)^2}{b^2 + (x-c)^2} \\ = \frac{2\pi}{a^2} \left( \arctan \frac{c}{b} - \arctan \frac{c}{a+b} \right), \end{aligned} \quad (\text{B1})$$

$$\begin{aligned} \int_0^\infty \frac{dx}{x} \frac{1}{(a^2 + x^2)^2} \ln \frac{b^2 + (x+c)^2}{b^2 + (x-c)^2} \\ = \frac{2\pi}{a^4} \left( \arctan \frac{c}{b} - \arctan \frac{c}{a+b} \right) \\ - \frac{\pi}{a^3} \frac{c}{(a+b)^2 + c^2}, \end{aligned} \quad (\text{B2})$$

$$\int_0^\infty \frac{dx}{x} \ln \frac{b^2 + (x+c)^2}{b^2 + (x-c)^2} = 2\pi \arctan \frac{c}{b}. \quad (\text{B3})$$

Also at the Institute of Applied Mathematics Rijksuniversitair Centrum, Antwerp, Belgium.

<sup>1</sup>J. Devreese, J. De Sitter, and M. Goovaerts, *Solid State Commun.* **9**, 1383 (1971); *Phys. Rev. B* **5**, 2367 (1972).

<sup>2</sup>J. Devreese, W. Huybrechts, and L. Lemmens, *Phys. Status Solidi B* **48**, 77 (1971).

<sup>3</sup>*Polarons in Ionic Crystals and Polar Semiconductors*, edited by J. Devreese (North-Holland, Amsterdam, 1972).

<sup>4</sup>V. Gurevich, I. Lang, and Yu. Firsov, *Fiz. Tverd. Tela*

**4**, 1252 (1962) [*Sov. Phys.-Solid State* **4**, 918 (1962)].

<sup>5</sup>J. Röseler, *Phys. Status Solidi* **25**, 311 (1968).

<sup>6</sup>W. Huybrechts and J. Devreese, Technical Report No. BLG 458 (1971) (S. C. K.-C. E. N., Mol-Belgium) (unpublished).

<sup>7</sup>L. Lemmens, J. De Sitter, and J. Devreese, *Phys. Rev. B* **8**, 2717 (1973).

<sup>8</sup>C. C. Grosjean, *Simon Stevien Wis.-Natuurkd. Tijdschr.* **40-II**, 49 (1966).