

## Theory of intraband magnetoabsorption in weakly polar semiconductors

J. P. Vigneron, R. Evrard, and E. Kartheuser

University of Liège, Sart Tilman, Liège 1, Belgium

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The intraband magnetoabsorption by optical polarons has been calculated to linear order in the carrier-phonon coupling constant for the case of a simple parabolic band. A finite lifetime has been introduced for the Landau levels to take into account scattering mechanisms other than that due to optical phonons. The pinning effect, line broadening, and phonon-assisted transitions are studied under these assumptions.

### I. INTRODUCTION

Great interest has been shown during the last several years in the study of the properties of polar semiconductors. It is well known that in these materials the polaron interaction deeply modifies the behavior of the charge carriers. Since the idea of Landau<sup>1</sup> and Pekar's works<sup>2</sup> (strong electron-phonon coupling), important progress has been achieved by Fröhlich,<sup>3</sup> when, in 1954, we wrote the polaron Hamiltonian. More recently, a certain number of experimental observations seem to confirm many of the theoretical conjectures. Most of the experimental material was found in the study of intraband and interband magnetoabsorption. Especially, in the very last years, the development of infrared lasers allowed for direct observation of the absorption spectrum as a function of the incident frequency.<sup>4</sup>

The purpose of this paper is to present a theory of the cyclotron resonance of polarons at zero temperature and for weak electron-phonon coupling, taking into account the broadening of the Landau levels due to scattering by acoustic phonons and impurity centers.

Some features of the absorption spectrum have already been obtained by several authors in considering a system for which the Landau levels are perfectly sharp. Much information about magnetoabsorption in polar materials can be found in the review papers by Levinson and Rashba,<sup>5</sup> Kaplan and Ngai,<sup>6</sup> and Harper *et al.*<sup>7</sup> The main results of the theoretical works on the cyclotron-resonance spectrum in such a system can be indicated by the following statements:

(i) The electron-phonon interaction is able to break the harmonic-oscillator selection rules by the allowance of the emission of optical phonons. This leads to the so-called "phonon-assisted transitions"<sup>8,9</sup> giving rise to an absorption band starting at the LO-phonon frequency ( $\omega_{LO}$ ).

(ii) The accidental degeneracy of the zero-phonon  $n=1$  Landau state and the one-phonon  $n=0$  Landau states is lifted. This leads to the doublet

structure of the main absorption peak observed in interband magnetoabsorption of InSb when the cyclotron frequency  $\omega_c$  is close to the LO-phonon frequency.<sup>10</sup>

(iii) The cyclotron-resonance line experiences a sudden increase in the broadening when the cyclotron frequency passes through the LO-phonon frequency.<sup>11-13</sup>

Despite the extensive literature on the subject, a number of problems have not yet been completely solved.

The singularities in the density of states of a charged particle in a uniform magnetic field induce corresponding divergences at incident radiation frequencies  $\omega = \omega_{LO} + n\omega_c$  ( $n=0, 1, 2, \dots$ ) in the one-phonon sideband for the geometrical configuration where the electric field of the incident radiation is perpendicular to the direction of the uniform magnetic field. The suggestion of Enck, Saleh, and Fan<sup>9</sup> to take into account the finite resolution of the experimental device to overcome this unphysical result is far from giving a definitive answer to this problem. The first goal of the present paper is to study the effect of a natural broadening of the Landau states on the shape of these phonon-assisted lines. The introduction of a finite lifetime erases the singularities of the density of states and, as will be shown in Sec. IV, leads to a line shape in qualitative agreement with experiment.

The next interesting question lies in the interpretation of the so-called "pinning effect" first observed by Johnson<sup>10</sup> in the interband magnetoabsorption of InSb. A self-consistent calculation of this effect has been developed by means of Wigner-Brillouin perturbation theory<sup>14</sup> and Green's-function techniques.<sup>15,16</sup> A concise review of both theoretical and experimental aspects of this problem can be found in a review paper by Larsen.<sup>14</sup>

In polar materials, the electron-phonon interaction does not allow for a level crossing between the  $n=1$  and  $n=0$ , one-phonon Landau levels. The consequence of this for the absorption curve is to

induce a doublet structure in the cyclotron resonance line for magnetic fields such that  $\omega_c \approx \omega_{LO}$ . This behavior of the absorption line has almost no chance to be observed in direct (spin-up or spin-down) cyclotron resonance, since  $\omega \approx \omega_c \approx \omega_{LO}$  is within the reststrahl region. Nevertheless this problem has been overcome by producing combined transitions that involve a spin flip and therefore occur at higher frequencies.<sup>17</sup> Larsen's theory of the pinning effect predicts a discrete line in the magneto-optical spectrum just below  $\omega_{LO}$ . The behavior of this line with increasing field has not yet been cleared up. As Larsen suggested<sup>18</sup> the existence of a natural broadening could drastically alter the pinning behavior. The study of this effect on the natural broadening of the Landau states is the second purpose of the present paper. We will show that, due to its finite lifetime, the pinned state loses its individuality and becomes just a part of a broad phonon band located about  $\omega_{LO}$ .

Some questions also arise from the consideration of the linewidth of the cyclotron resonance as a function of magnetic field strength. In 1967, Dickey *et al.*<sup>11</sup> observed a sudden broadening of the cyclotron line as  $\omega_c$  was swept through  $\omega_{LO}$ . A detailed experimental study of this effect in InSb has been performed<sup>13</sup> and an expression for this broadening has been obtained by Harper<sup>12</sup> in the limit of perfectly sharp Landau levels. This theory is derived on the assumption that the polaron states involved in the transition consist of a mixing of three unperturbed states: the  $n=0$  Landau state containing one or no phonon and the  $n=1$  zero-phonon Landau state.

The calculation presented in this paper shows that this assumption leads to the correct result in the limit of perfectly sharp Landau levels. With the introduction of a finite lifetime the singularity in the Lorentzian broadening<sup>12</sup> disappears. On the other hand, taking into account the effects of the higher-order Landau states introduce corrections which will be discussed in Sec. IV.

In Sec. II, our model is described and the approximations are made clear. In Sec. III this model is solved and the absorption coefficient is obtained in a form convenient for a precise numerical evaluation. Finally, Sec. IV is devoted to the presentation and discussion of the numerical results.

## II. DESCRIPTION OF THE MODEL AND APPROXIMATIONS

To describe the absorption spectrum the usual relation<sup>19</sup> between the absorption coefficient  $K(\omega)$  and the transition probability  $P(\omega)$  is used, i.e.,

$$K(\omega) = 16\pi m_e P(\omega) / n(\omega) \omega \alpha_0^2, \quad (1)$$

where  $n(\omega)$  is the refractive index of the material,  $n_e$  is the carrier concentration,  $\alpha_0^2 = |A_0|^2 / mc \omega_{LO}$  has no dimension,  $A_0$  is the amplitude of the vector potential of the incident radiation, and  $m$  the effective mass of the carrier. Units are such that  $\hbar = 1$ ,  $\omega_{LO} = 1$ ,  $2m = 1$ ,  $\Gamma(\omega)$  being given in units of  $(2m\omega_{LO}/\hbar)^{1/2}$ . In writing relation (1) one assumes a carrier concentration low enough to avoid collective oscillations (plasmons). This is generally satisfied in semiconductors at the very low temperatures required for observing cyclotron resonance.

In this paper all transitions are supposed to start from the true ground state of the system, with zero momentum along the magnetic field axis. This assumption ( $p_x = 0$ ) is not completely justified at low but finite temperature. Indeed, a low but nonzero temperature leads to a weak increase of energy (proportional to  $p_x^2$ ) which does not necessarily imply a small value for  $p_x$ . Moreover, even at zero temperature, most of the electrons in the conduction band have nonzero  $p_x$  because of the Pauli exclusion principle. Nevertheless it is reasonable to expect that these effects lead only to a slight smearing of the overall spectrum, especially as the most probable transitions are "vertical," i.e., they do not modify  $p_x$ .

In the evaluation of expression (1) a mean value has been used for the refractive index. This is not an essential assumption in the theory and a more detailed expression could be introduced. Data concerning the behavior of  $n(\omega)$  for III-V compounds can be found in Refs. 20, 21.

It is then assumed that the absorption probability  $P(\omega)$  is well described by Fermi's Golden Rule. In the dipolar approximation, this leads to

$$P(\omega) = \pi \alpha_r \alpha_0^2 \sum_{f \neq i} \left| \langle f | \left( \vec{p} + \frac{e|\vec{A}|}{c} \right) \cdot \vec{\eta} | i \rangle \right|^2 \times \delta(\epsilon_f - \epsilon_i - \omega). \quad (2)$$

$\alpha_r$  is the atomic fine structure constant,  $\eta$  is the polarization vector of the incident light, and  $\vec{A}$  is the vector potential due to the presence of the magnetic field  $\vec{B}$ . Here the transitions occur from the ground state  $|i\rangle$  to the intermediate states  $|f\rangle$  with corresponding energy levels  $\epsilon_i$  and  $\epsilon_f$ . In the following development the term  $f=0$  is added to the summation, making the set of intermediate states  $|f\rangle$  complete. As a consequence the contribution arising for  $\omega=0$  is here treated incorrectly. However, this part of the spectrum is of no interest in the purpose of the present work.

The description of the absorbing system involves two further approximations: first, the charge carriers are supposed to belong to a simple parabolic band. This is true only near a minimum (maximum) of the conduction (valence) band. This

assumption is questionable in the case of InSb, especially for transitions to higher excited Landau levels. Nevertheless, as will be discussed in Sec. III, this assumption is crucial to apply to the method proposed in this paper. On the other hand, the carrier-LO-phonon interaction will be described by the Fröhlich-interaction Hamiltonian. In itself, this involves a certain number of approximations.<sup>22</sup> The model is based on the assumption that the lattice can be described as a continuous polarizable medium and this medium is supposed to be quite isotropic. The model is further restricted to the cases where only one longitudinal optical phonon exists and the dispersion law of this mode is approximated by the simple relation  $\omega(\vec{k}) = \omega_{LO}$ .

The absorbing system will then be described by the following total Hamiltonian:

$$H = H_0 + H_1, \quad (3)$$

with

$$H_0 = \left( \vec{p} + \frac{|e|\vec{A}}{c} \right)^2 + \sum_{\vec{k}} b_{\vec{k}}^\dagger b_{\vec{k}}, \quad (4)$$

$$H_1 = \sum_{\vec{k}} \left( \frac{4\pi\alpha}{V} \right)^{1/2} k^{-1} (b_{\vec{k}}^\dagger e^{i\vec{k}\cdot\vec{r}} + b_{\vec{k}} e^{-i\vec{k}\cdot\vec{r}}) \quad (5)$$

The operators  $b_{\vec{k}}$  and  $b_{\vec{k}}^\dagger$  are annihilation and creation operators for phonons with wave vector  $\vec{k}$ , and  $\vec{p}$  and  $\vec{r}$  are, respectively, the electron momentum and position operators. The quantity  $V$  is the volume of the crystal and  $\alpha$  is the Fröhlich coupling constant defined as

$$\alpha = \frac{1}{2} e^2 (1/\epsilon_\infty - 1/\epsilon_0) \quad (6)$$

in terms of the static dielectric constant  $\epsilon_0$  and the high-frequency dielectric constant. In expression (6) the electric charge is expressed in units of  $(\hbar^3 \omega_{LO}/2m)^{1/4}$ .

For weakly polar crystals (III-V compounds) the basic parameter  $\alpha$  of Fröhlich's polaron theory is much below unity. The theory developed in Sec. III is valid in this weak-coupling limit since the solution obtained in the present paper for the absorption spectrum is exact in the framework of Fröhlich's polaron model up to linear order in  $\alpha$ . The configuration most used in experiments in the so-called Faraday configuration<sup>23</sup> for which the wave vector of the incident radiation is parallel to the direction of the uniform magnetic field  $\vec{B}$  and therefore its electric component is perpendicular to the same direction. This configuration is chosen for the present theoretical investigations. Moreover, we restrict ourselves to the case of a circularly polarized wave described by the polarization vector

$$\vec{\eta} = (1, i, 0)/\sqrt{2}, \quad (7)$$

the  $z$  axis being the direction of the static magnetic field. The case of a configuration such that the electric component of the incident radiation is parallel to the magnetic field has recently been treated by Van Royen *et al.*<sup>24</sup> in the case of perfectly sharp Landau levels.

For a circularly polarized radiation it is more convenient to use the symmetrical gauge in which the vector potential  $\vec{A}$  is given by  $\vec{A} = \frac{1}{2} B(-y, x, 0)$ . In this gauge  $H_0$  can be diagonalized exactly<sup>25,26</sup> if one defines the following operators:

$$A_\pm = (i/\lambda)(p_x \mp ip_y) + (\frac{1}{4}\lambda)(x \mp iy), \quad (8)$$

where following Larsen's<sup>18</sup> notations we put

$$\lambda^2 = 2|e|B/c.$$

These operators satisfy the usual boson commutation relations. The unperturbed Hamiltonian  $H_0$  is then written (in units of  $\hbar\omega_{LO}$ )

$$H_0 = p_z^2 + (A_+^\dagger A_+ + \frac{1}{2})\lambda^2 + \sum_{\vec{k}} b_{\vec{k}}^\dagger b_{\vec{k}} \quad (9)$$

and the term describing the interaction between the electron and the radiation field is

$$(\vec{p} + |e|\vec{A}/c) \cdot \vec{\eta} = (i\lambda/\sqrt{2}) A_+^\dagger. \quad (10)$$

The presence of a  $\delta$  function in Fermi's Golden Rule [Eq. (2)] indicates that the Landau levels are considered to be infinitely sharp in absence of electron-phonon interaction. If a finite lifetime is attributed to each energy level, the  $\delta$  function must be replaced by a Lorentzian defined as

$$\mathcal{L}(x; \gamma) = (\gamma/2\pi)(x^2 + \frac{1}{4}\gamma^2)^{-1}, \quad (11)$$

where  $\gamma$  describes the width of the state under consideration. The justification of this procedure is given in Appendix C. The main conclusions of Appendix C are the following: the parameter  $\gamma$  describes the broadening in the absence of interaction with the LO-phonons. As it is not the purpose of this paper to study the effects of the interaction with impurities or with acoustical phonons,  $\gamma$  is to be considered as a phenomenological parameter which can be deduced from comparison of the calculated curve and the actual measured absorption spectrum. In the region of interest (around  $\omega = \omega_{LO}$ ) these latter broadening mechanisms are not strongly frequency dependent (contrary to that due to electron LO-phonon interaction); so that, for the purpose of the present work,  $\gamma$  will be considered as frequency independent. As regarding the frequency dependent broadening or more generally the modification in the shape of the magneto-optical spectrum due to the interaction with the LO phonons, it is included in the formalism through the Fröhlich Hamiltonian. Therefore, these broaden and change in the ab-

sorption naturally appears in the results of our calculations.

Including these assumptions the Golden Rule is finally written

$$P(\omega) = 2\pi\lambda^2\alpha_r\alpha_0^2 \sum_f |\langle f | A_+^\dagger | i \rangle|^2 \mathcal{L}(\epsilon_f - \epsilon_i - \omega; \gamma). \quad (12)$$

The next step consists of expanding this transition probability  $P(\omega)$  in powers of the electron LO-phonon coupling constant  $\alpha$ . The expansion is restricted to linear order in  $\alpha$ . As it is shown in Sec. III, this linear contribution can be calculated exactly. In the compounds considered here,  $\alpha$  is small and the perturbationlike approximation described above is valid except for the description of the main cyclotron peak near the pinning point (i.e., when  $\omega_c \approx \omega_{LO}$ ). For this latter situation, the expansion in powers of  $\alpha$  is valid only when the broadening  $\gamma$  due to impurities and acoustic phonons is large compared to the shift of levels caused by the pinning effect. Indeed the change in the absorption produced by a shift is the difference between the shifted and unperturbed Lorentzians. This difference is small enough to be considered as a perturbation only when the shift is small as compared to the linewidth  $\gamma$  of the Lorentzian. It is experimentally known that this is not the case close to the pinning. The shift<sup>11</sup> as well as the broadening<sup>13</sup> due to the emission of LO-phonons when  $\omega_c$  is just beyond  $\omega_{LO}$  are in fact much larger than the natural linewidth  $\gamma$  measured at low magnetic fields. However our result for the transition probability [Eq. (26) in Sec. III] appears as the first terms of an expansion in  $\alpha$  of a modified (shifted and broadened) Lorentzian. This suggests to replace the expansion by this Lorentzian itself and to use the expression (49) of Sec. III in the numerical computations rather than the expansion (26) in powers of  $\alpha$ . The arguments in favor of this procedure are the following:

First, the linewidth is now the sum of  $\gamma$  and a frequency dependent contribution  $\alpha\Gamma(\omega)$  coming from the interaction with the LO phonons as expected from the physical point of view.

Second, with this form of the solution, there is no longer any difficulty at the pinning point, even if  $\gamma$  is given a low value in accordance with experiment. For instance, the absorption coefficient never becomes negative, contrary to the predictions obtained with an expansion in  $\alpha$  for too small values of  $\gamma$ .

Finally, for cyclotron frequencies larger than the LO-phonon frequency, the lines in the calculated spectrum have a natural width even when  $\gamma$  is taken equal to zero. This natural width is due,

as expected, to emission of LO phonons.

It is important to note the following fact: As the contributions to the shift and broadening due to the interaction with LO phonons are strongly frequency dependent, the absorption curve obtained in the present work has a rather complicated structure, showing two peaks close to the pinning point as well as phonon assisted transitions at frequencies  $\omega = \omega_{LO} + n\omega_c$ . This is in agreement with preliminary results obtained with the perturbation expansion (26) and presented at two different conferences.<sup>27,28</sup> The main improvement with the present procedure lies in fact that the cyclotron resonance curve has a more realistic shape just beyond pinning (i.e., when  $\omega_c$  is slightly larger than  $\omega_{LO}$ ) at least for small values of  $\gamma$  as required by experiment.

### III. CALCULATION OF THE ABSORPTION COEFFICIENT

Using the completeness of the intermediate states, expression (12) for the transition probability can be written

$$P(\omega) = -2\lambda^2\alpha_r\alpha_0^2 \text{Im}G(\omega), \quad (13)$$

where

$$G(\omega) = \langle i | A_+ (\omega + \frac{1}{2}i\gamma - H + \epsilon_i)^{-1} A_+^\dagger | i \rangle. \quad (14)$$

Note that  $H$ ,  $|i\rangle$ ,  $\epsilon_i$  are the Hamiltonian, the ground state, and the ground-state energy level for a polaron, respectively. To calculate this expression to first order in the coupling constant  $\alpha$ , it is more convenient to use the following identity:

$$\frac{1}{\omega + \frac{1}{2}i\gamma - H + \epsilon_i} = \frac{1}{\omega + \frac{1}{2}i\gamma} \left( 1 + \frac{H - \epsilon_i}{\omega + \frac{1}{2}i\gamma - H + \epsilon_i} \right). \quad (15)$$

This leads to

$$\begin{aligned} (\omega + \frac{1}{2}i\gamma)G(\omega) &= \langle i | A_+ A_+^\dagger | i \rangle \\ &+ \langle i | [A_+, H] (\omega + \frac{1}{2}i\gamma - H + \epsilon_i)^{-1} A_+^\dagger | i \rangle. \end{aligned} \quad (16)$$

The commutator appearing in the right-hand side of Eq. (16) gives

$$[A_+, H] = A_+\lambda^2 + \sqrt{\alpha}P, \quad (17)$$

where

$$P = \frac{i}{\lambda} \left( \frac{4\pi}{V} \right)^{1/2} \sum_{\vec{k}} \left( \frac{k_x - ik_y}{k} \right) (b_{\vec{k}}^\dagger e^{i\vec{k}\cdot\vec{r}} - b_{\vec{k}} e^{-i\vec{k}\cdot\vec{r}}).$$

The very simple form of expression (17) arises from the hypothesis of parabolicity for the band, which is implicit in relation (9). For non parabolic bands, the following procedure can probably not be easily repeated.

The introduction of Eq. (17) into Eq. (16) makes the expression (14) for  $G(\omega)$  appear in the right-hand side of relation (16). This term can then be added to the left-hand side to give

$$(\omega + \frac{1}{2}i\gamma - \lambda^2)G(\omega) = \langle i | A_+ A_+^\dagger | i \rangle + \sqrt{\alpha} \langle i | P(\omega + \frac{1}{2}i\gamma - H + \epsilon_i)^{-1} A_+^\dagger | i \rangle. \quad (19)$$

The advantage of Eq. (19) over relation (14) is that the only term (the second term) in the right-hand side of Eq. (19) that depends on the frequency exhibits a factor  $\sqrt{\alpha}$ . Therefore, the calculation of Eq. (19) to first order in  $\alpha$  requires the evaluation of the second matrix elements only at order  $\sqrt{\alpha}$ .

One can apply to the frequency-dependent matrix element in Eq. (19) the same kind of reduction used to transform expression (14) into expression (19). This procedure leads to the final expression

$$G(\omega) = \frac{\langle i | A_+ A_+^\dagger | i \rangle}{\omega - \lambda^2 + \frac{1}{2}i\gamma} + \sqrt{\alpha} \frac{\langle i | P A_+^\dagger | i \rangle}{(\omega - \lambda^2 + \frac{1}{2}i\gamma)^2} + \alpha \frac{\langle i | P(H - \epsilon_i - \omega - \frac{1}{2}i\gamma)^{-1} P^\dagger | i \rangle}{(\omega - \lambda^2 + \frac{1}{2}i\gamma)^2}. \quad (20)$$

This relation is of course valid for any value of the coupling constant  $\alpha$ , but is especially well suited for a perturbation expansion. To obtain  $G(\omega)$  to first order in  $\alpha$ , it is sufficient to calculate the first, second, and third terms of the right-hand side of Eq. (20) up to order  $\alpha$ ,  $\alpha^{1/2}$ , and  $\alpha^{(0)}$ , respectively.

Let us now see how this transformation affects the transition probability  $P(\omega)$  [Eqs. (13) and (20)]. We first define  $\mu_0$  and  $\mu_1$ , the zeroth- and first-order moments of this probability, i.e.,

$$\mu_0 = \int_{-\infty}^{+\infty} P(\omega) d\omega, \quad (21)$$

$$\mu_1 = \int_{-\infty}^{+\infty} \omega P(\omega) d\omega. \quad (22)$$

Using the expression for  $P(\omega)$  [Eq. (2)], we obtain

$$\mu_0 = \frac{1}{2}\pi\lambda^2\alpha_r\alpha_0^2\langle i | A_+ A_+^\dagger | i \rangle \quad (23)$$

and

$$\mu_1 = \lambda^2\mu_0 + \frac{1}{2}\pi\lambda^2\alpha_r\alpha_0^2\sqrt{\alpha}\langle i | P A_+^\dagger | i \rangle. \quad (24)$$

With these notations the expression

$$G(\omega) = \frac{\mu_0}{\frac{1}{2}\pi\lambda^2\alpha_r\alpha_0^2(\omega - \lambda^2 + \frac{1}{2}i\gamma)} + \frac{\mu_1 - \lambda^2\mu_0}{\frac{1}{2}\pi\lambda^2\alpha_r\alpha_0^2(\omega - \lambda^2 + \frac{1}{2}i\gamma)^2} + \alpha \frac{\langle i | P(\omega + \frac{1}{2}i\gamma - H + \epsilon_i)^{-1} P^\dagger | i \rangle}{(\omega - \lambda^2 + \frac{1}{2}i\gamma)^2} \quad (25)$$

leads to an interesting form for the transition probability,

$$P(\omega) = \mu_0 \mathcal{L}(\omega - \lambda^2; \gamma) + [\lambda^2\mu_0 - \mu_1 + I(\omega)] \frac{\partial \mathcal{L}(\omega - \lambda^2; \gamma)}{\partial \omega} + R(\omega) \frac{\partial \mathcal{L}(\omega - \lambda^2; \gamma)}{\partial \gamma}, \quad (26)$$

with, successively,

$$I(\omega) = -\pi\lambda^2\alpha \operatorname{Re}[Q(\omega)], \quad (27)$$

$$R(\omega) = -2\pi\lambda^2\alpha \operatorname{Im}[Q(\omega)], \quad (28)$$

$$Q(\omega) = \langle i | P(\omega - H + \epsilon_i + \frac{1}{2}i\gamma)^{-1} P^\dagger | i \rangle. \quad (29)$$

Taking the zeroth- and first-order moments of this expression leads to the following sum rules:

$$\int_{-\infty}^{+\infty} \left( R(\omega) \frac{\partial \mathcal{L}(\omega - \lambda^2; \gamma)}{\partial \gamma} + I(\omega) \frac{\partial \mathcal{L}(\omega - \lambda^2; \gamma)}{\partial \omega} \right) d\omega = 0 \quad (30)$$

and

$$\int_{-\infty}^{+\infty} \left( R(\omega) \frac{\partial \mathcal{L}(\omega - \lambda^2; \gamma)}{\partial \gamma} + I(\omega) \frac{\partial \mathcal{L}(\omega - \lambda^2; \gamma)}{\partial \omega} \right) \omega d\omega = 0. \quad (31)$$

Note that these relations are not restricted to weak-coupling strengths. From Eq. (26) one can understand that the behavior of  $R(\omega)$  and  $I(\omega)$  close to  $\omega = \lambda^2$  governs the effect of carrier-phonon interaction on the shape (width and position, respectively) of the cyclotron resonance line. Especially an interesting quantity is  $R(\lambda^2)/\mu_0$ , which will be referred to as the "Lorentzian broadening." Its behavior as a function of the magnetic field strength will be considered in Sec. IV.

As in the case of the free polaron,<sup>29</sup> the first term in Eq. (26) can be related to the ground-state energy. This can be seen by first noting that only  $H_0$  depends on  $\lambda^2$ ,

$$\frac{\partial H}{\partial \lambda^2} = A_+^\dagger A_+ + \frac{1}{2} = A_+ A_+^\dagger - \frac{1}{2}. \quad (32)$$

Invoking the Feynman-Hellmann theorem, one obtains

$$\frac{\partial \epsilon_i}{\partial \lambda^2} = \langle i | \frac{\partial H}{\partial \lambda^2} | i \rangle = \langle i | A_+ A_+^\dagger | i \rangle - \frac{1}{2}. \quad (33)$$

The ground-state energy correct to first order in the coupling constant has been obtained by Larsen<sup>18</sup> for arbitrary magnetic field strength. Larsen's result can be written in the form

$$\epsilon_i = \frac{\lambda^2}{2} - \frac{\alpha}{\lambda\sqrt{\pi}} \int_0^\infty \frac{f(u)}{\sqrt{u}} e^{-u/\lambda^2} du, \quad (34)$$

where

$$f(u) = \tanh^{-1} s(u) / s(u) \quad (35a)$$

and

$$s(u) = [(u - 1 + e^{-u})/\mu]^{1/2}. \quad (35b)$$

This leads to

$$\mu_0 = \frac{1}{2}\pi\lambda^2\alpha_r a_0^2 \langle i | A_+ A_+^\dagger | i \rangle = \frac{1}{2}\pi\lambda^2\alpha_r a_0^2 \left( 1 + \frac{\alpha}{\lambda^2\sqrt{\pi}} \int_{-\infty}^{+\infty} e^{-x^2} f(\lambda^2 x^2) (\frac{1}{2} - x^2) dx \right), \quad (36)$$

which can be computed numerically without any difficulty.

The second term in Eq. (26) has to be evaluated at first order in  $\alpha$ . Since we have

$$\sqrt{\alpha} \langle i | P A_+^\dagger | i \rangle = I_1 + I_2, \quad (38)$$

$$I_1 = \frac{4\pi\alpha}{V} \sum_k \frac{\exp(-k_x^2/\lambda^2)}{k^2} \sum_{n=0}^{\infty} \frac{(k_x^2/\lambda^2)^{n+1}}{n!} \left( \frac{1}{n\lambda^2 + k_z^2 + 1} \right), \quad (39)$$

$$I_2 = \frac{4\pi\alpha}{V} \sum_k \frac{\exp(-k_x^2/\lambda^2)}{k^2} \sum_{n=0}^{\infty} \frac{(k_x^2/\lambda^2)^{n+1}}{n!} \left( \frac{\lambda^2}{(n\lambda^2 + k_z^2 + 1)[(n+1)\lambda^2 + k_z^2 + 1]} \right), \quad (40)$$

with

$$k_{\perp} = (k_x^2 + k_y^2)^{1/2}. \quad (41)$$

The first term  $I_1$  in this expression turns out to be divergent, but as will be shown below, it is cancelled by a similar divergent term appearing in the calculation of the third matrix element in the right-hand side of Eq. (26). According to the procedure described in Appendix A,  $I_2$  in Eq. (38) can be reduced to the one-dimensional integral

$$I_2 = \frac{2\alpha[(1+\lambda^2)^{1/2} - 1]}{3\lambda^2} + \frac{\alpha}{2\lambda\sqrt{\pi}} \int_0^{\infty} e^{-s/\lambda^2} (1 - e^{-s}) \times [g(s) - \frac{2}{3}s^{-3/2}] ds, \quad (42)$$

where

$$g(s) = \frac{\sqrt{s}}{\delta_1 \delta_2} - \frac{1}{\delta_2^{3/2}} \ln \left[ \left( \frac{\delta_2}{\delta_1} \right)^{1/2} + \left( 1 + \frac{\delta_2}{\delta_1} \right)^{1/2} \right] \quad (43)$$

and

$$\delta_1 = 1 - e^{-s}, \quad (44a)$$

$$\delta_2 = s - \delta_1. \quad (44b)$$

The numerical evaluation of (42) is straightforward. The last term in Eq. (26) involves a matrix element that depends on the incident radiation frequency. Indeed we get

$$R(\omega) = -2\pi\lambda^2\alpha \operatorname{Im}Q(\omega), \quad (45)$$

with

$$\alpha Q(\omega) = \alpha \langle 0 | P(\omega - H_0 + E_0 + \frac{1}{2}i\gamma)^{-1} P^\dagger | 0 \rangle, \quad (46)$$

$$\lambda^2\mu_0 - \mu_1 = -\frac{1}{2}\pi\lambda^2\alpha_r a_0^2 \sqrt{\alpha} \langle i | P A_+^\dagger | i \rangle, \quad (37)$$

the wave function  $|i\rangle$  to be used must be correct up to first order of perturbation. This leads to

$$\alpha Q(\omega) = \frac{4\pi\alpha}{V} \sum_k \frac{e^{-k_x^2/\lambda^2}}{k^2} \sum_{n=0}^{\infty} \frac{(k_x^2/\lambda^2)^{n+1}}{n!} \times \frac{1}{\omega - n\lambda^2 - k_z^2 - 1 + \frac{1}{2}i\gamma}, \quad (47)$$

where  $|0\rangle$  and  $E_0$  are the ground state and energy of the unperturbed system. This expression turns out to diverge. For large value of  $n$ , the  $n$ th term of the series behaves like  $n^{-1/2}$ . Nevertheless, one can extract the divergence by writing the denominator in Eq. (47) in the following form, which holds for any positive integer  $N$ :

$$\frac{1}{n\lambda^2 + k_z^2 + 1 - \omega - \frac{1}{2}i\gamma} = \sum_{m=0}^N \frac{(\omega + \frac{1}{2}i\gamma)^m}{(n\lambda^2 + k_z^2 + 1)^{m+1}} + \frac{[(\omega + \frac{1}{2}i\gamma)/(n\lambda^2 + k_z^2 + 1)]^{N+1}}{n\lambda^2 + k_z^2 + 1 - \omega - \frac{1}{2}i\gamma}. \quad (48)$$

Introducing (48) into (47) one can see that the term corresponding to  $m=0$  is the divergent part of (47) and that all terms corresponding to  $m=1, 2, \dots, N$  are convergent and can be reduced to a one-dimensional integral according to the procedure described in Appendix A. This procedure cannot be directly applied to the last term of Eq. (48), because the sign of the denominator changes when  $k_z$  varies from 0 to  $\infty$ . However due to the high-order exponent appearing in this expression, the series corresponding to that term converges much faster than the original one (see Appendix B). On the other hand, one may easily see that the divergent term ( $m=0$ ) exactly cancels the divergent part of the second term in Eq. (20).

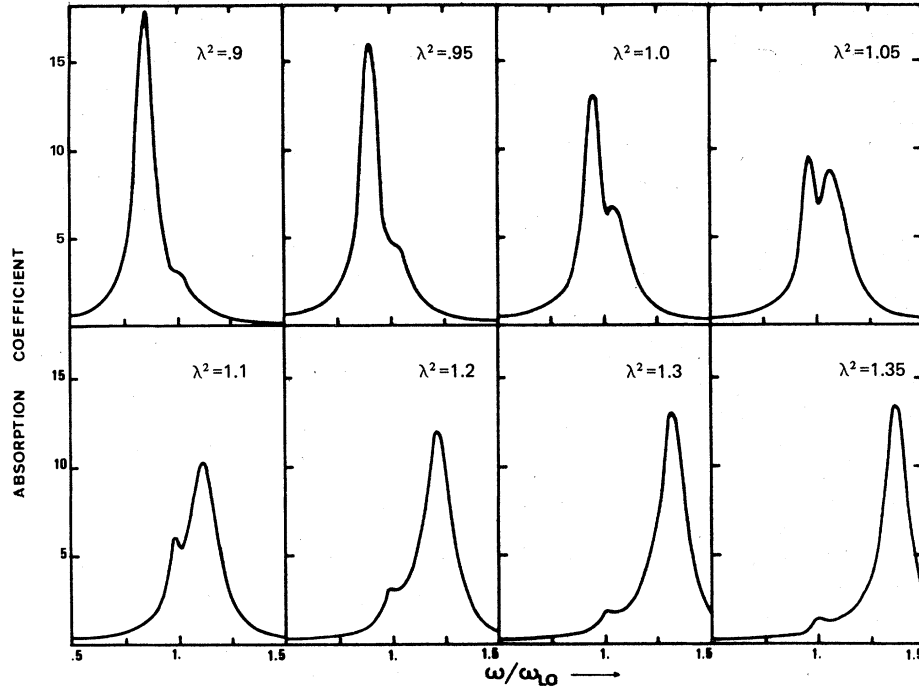


FIG. 1. Shape of the absorption coefficient obtained in the present paper as a function of  $\omega/\omega_{LO}$  in the region  $\omega_r \sim \omega_c \sim \omega_{LO}$ . Ordinates are in units of  $16\pi n_e \alpha_r / nr_o$ , where  $n_e$  is the carrier density,  $\alpha_r = 1/137$ ,  $n$  is the refractive index,  $r_o = (\hbar/2m\omega_{LO})^{1/2}$  is the polaron radius,  $\alpha = 0.02$  is the electron-phonon coupling constant, and  $\gamma = 0.2$ .

As discussed in Sec. II, we now replace the expansion (26) in powers of  $\alpha$  by the following closed expression:

$$P(\omega) = \mu_0 \mathcal{L}(\omega - \lambda^2 - \alpha \Delta(\omega); \gamma + \alpha \Gamma(\omega)), \quad (49)$$

where  $\alpha \Delta(\omega)$  and  $\alpha \Gamma(\omega)$  are, respectively, the frequency dependent shift and broadening due to the interaction with the LO phonons. These latter quantities are easily obtained by comparison of the expansion of the Lorentzian in (49) in powers of  $\alpha$  restricted to first order, i.e.,

$$P(\omega) = \mu_0 \mathcal{L}(\omega - \lambda^2; \gamma) - \alpha \mu_0 \frac{\partial \mathcal{L}(\omega - \lambda^2; \gamma)}{\partial \omega} \Delta(\omega) + \alpha \mu_0 \frac{\partial \mathcal{L}(\omega - \lambda^2; \gamma)}{\partial \gamma} \Gamma(\omega), \quad (50)$$

with relation (26). Then, the developments described at the end of Sec. II allow the computation of  $\Delta(\omega)$  and  $\Gamma(\omega)$  and therefore of the transition probability (49). Finally, using definition (1) of the absorption coefficient  $K(\omega)$  we obtain the results shown in Figs. 1 and 2 for the absorption curve as a function of the incident radiation frequency and at Fig. 5 versus magnetic field.

#### IV. RESULTS AND DISCUSSION

An overall look at the spectrum (cf. Figs. 1 and 2) obtained by the method described in Sec. III

shows, as expected, the presence of peaks for  $\omega = 1 + n\lambda^2$ , besides the cyclotron line at  $\omega \approx \lambda^2$  ( $\lambda^2$  being the dimensionless cyclotron frequency).

The peak corresponding to  $n=0$  and therefore located at frequencies close to  $\omega_{LO}$  cannot be observed in pure cyclotron resonance. Indeed it lies in a region of strong absorption by the crystal lattice, the so-called reststrahl region. In principle, this band could be observed in interband magnetooptics or in combined resonance. However, the spectrum is usually obtained in

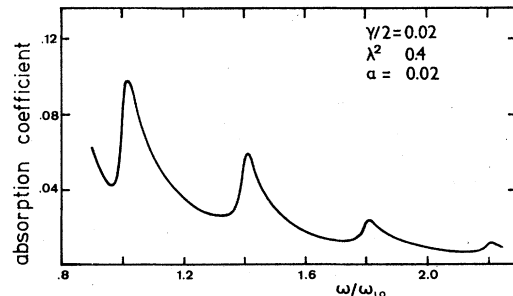


FIG. 2. Phonon sideband as a function of incident frequency. The magnetic field is such that  $\omega_c = 0.4\omega_{LO}$  and  $\gamma = 0.04\omega_{LO}$ . The Fröhlich coupling constant is taken as  $\alpha = 0.02$ . Ordinates are in units of  $16\pi n_e \alpha_r / nr_o$ , where  $n_e$  is the carrier density,  $\alpha_r = 1/137$ ,  $n$  is the refractive index, and  $r_o = (\hbar/2m\omega_{LO})^{1/2}$  is the polaron radius.

varying the magnetic field rather than the frequency of the incident radiation. In this case, it is obvious that the contribution of the  $n=0$  phonon assisted transition is a weak constant background that cannot be revealed experimentally. However, we see no reason why this peak should not appear in the interband or combined resonance spectrum taken versus frequency at constant magnetic field.

No peak appears at frequencies near  $\omega = n\lambda^2$  ( $n=1, 2, \dots$ ). Therefore, the harmonics observed in InSb (Ref. 4) are most probably not due to the effects of the coupling with the LO phonons.

The pinning effect, described by Larsen on basis of Wigner-Brillouin perturbation theory, as well as the broadening of the cyclotron line when  $\omega_c$  passes through  $\omega_{LO}$  show up in the calculated spectrum.

Let us first consider the manifestation of the pinning effect. The spectrum shown in Fig. 1 for various cyclotron frequencies exhibits a characteristic doublet structure with a line close to  $\omega = \lambda^2$  and a broader band starting near  $\omega = 1$ . For weak magnetic fields the cyclotron line is almost perfectly lorentzian with the initial linewidth  $\gamma$ . No appreciable broadening appears due to the electron LO-phonon interaction. For these magnetic fields, the peak at  $\omega \approx 1$  has the line shape exhibited in Fig. 2. The asymmetric line shape is characteristic of a transition involving emission of a phonon. The high energy tail arises from the possibility of recoil of the electron during creation of the LO phonon. The amplitude of this band is small compared to the cyclotron line (1% for  $\alpha = 0.02$  and  $\gamma = 0.2$ ). With increasing magnetic field the oscillator strength of the phonon assisted transition increases, the zero-phonon  $n=1$  Landau state being more strongly admixed to the one-phonon state responsible for the absorption at  $\omega \approx 1$ . At the same time, the oscillator strength of the cyclotron line decreases. For a value of  $\lambda^2$  slightly above 1, the doublet looks almost symmetric (for  $\alpha = 0.02$ ,  $\lambda^2 = 1.05$ , and  $\gamma = 0.2$ ). For higher values of  $\lambda^2$  the peak of smaller energy remains in the vicinity of  $\omega = 1$  whereas the peak of higher energy is located near  $\omega = \lambda^2$ . This is characteristic of the absorption by an oscillator of variable frequency  $\lambda^2$  coupled to another oscillator of fixed frequency 1.

As demonstrated in Fig. 2, a weak absorption at a frequency near  $\omega = 1$  is present even for weak magnetic field. This is in contradiction with the result obtained by Larsen<sup>18</sup> [Wigner-Brillouin perturbation theory (WBPT)] and by Korovin and Pavlov<sup>15</sup> (Poles of the Green's function), who did not find any solution for the upper branch  $E^+$  of the polaron first excited state for a value of the cyclotron frequency smaller than a fixed frequency  $\omega_{th}$

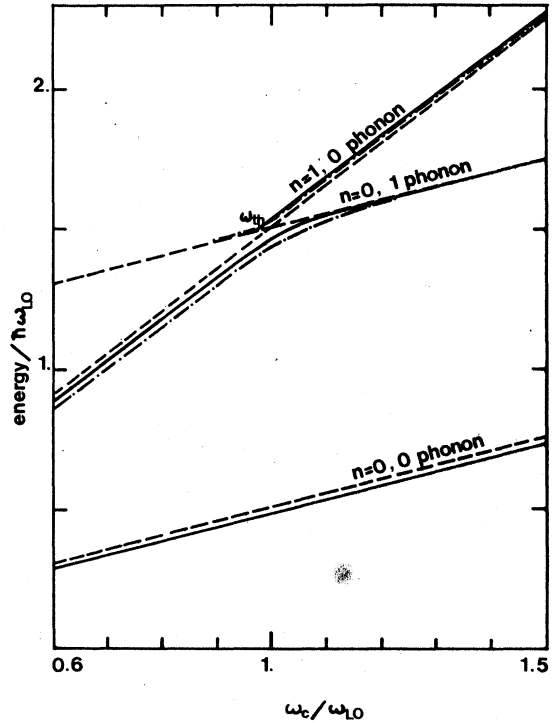


FIG. 3. Comparison of Larsen's results (Ref. 18) for the energy spectrum of polarons in a magnetic field with the maxima of the absorption curve obtained in the present theory. The reference level is the polaron ground state as calculated by Larsen. Here  $\gamma = 0.2$  and  $\alpha = 0.02$ . -----, unperturbed Landau energy levels; ———, results obtained by Larsen using Wigner-Brillouin perturbation theory; ·····, results obtained by using the position of the maxima of the absorption curve derived in this paper, to predict the difference between energies of excited and ground polaron state.

slightly below unity.

However, our results are confirmed by previous work on optical absorption by polarons for zero magnetic fields.<sup>30,31</sup>

Figure 3 shows a comparison between the results obtained by WBPT and the position of the maxima of the spectrum obtained from the present work. The reference level is the polaron ground state calculated by Larsen.<sup>18</sup> As expected good agreement is obtained in the regions where WBPT produces a solution. The shift and broadening of the cyclotron line become important when  $\lambda^2 = 1$ .

The effect of carrier-phonon interaction on the position of the peak located at  $\omega = \lambda^2$  is governed by the coefficient  $\lambda^2 - \mu_1/\mu_0 + I(\omega)/\mu_0$  in Eq. (26). The effect on the linewidth is contained in the behavior of the coefficient  $R(\omega)/\mu_0$ . Relation (28) can be rewritten in the form



$$\frac{R(\omega)}{\mu_0} = \frac{8\pi^3\lambda^2\alpha}{\mu_0V} \sum_{\mathbf{k}} \frac{e^{-k_1^2/\lambda^2}}{k^2} \sum_{n=0}^{\infty} \frac{(k_1^2/\lambda^2)^{n+1}}{n!} \times \mathcal{L}(\omega - n\lambda^2 - k_z^2 - 1; \gamma), \quad (51)$$

where the energies in the argument of the Lorentzian and the states in the matrix element are unperturbed quantities. It is easy to show from the explicit expression for the operator  $P$  [Eq. (18)] that the final state  $|n\rangle$  must contain a phonon to give rise to a nonvanishing matrix element. In the limit  $\gamma = 0$  the Lorentzian function has to be replaced by a  $\delta$  function. In a region of frequencies close to  $\omega = \lambda^2$ , and if the shift of the cyclotron line is neglected, the resonance is Lorentzian with a width  $R(\lambda^2)/\mu_0$ . For  $\lambda^2 < 1$ , no final energy level containing one phonon can satisfy the condition  $\epsilon_n - \epsilon_0 - \lambda^2 = 0$  so that  $R(\lambda^2)/\mu_0 = 0$ . In the contrary, for  $\lambda^2 > 1$ , the state  $n = 0$  with one phonon gives a nonvanishing contribution to  $R(\lambda^2)$ . In this particular case ( $\gamma = 0$ ), the function  $R(\lambda^2)/\mu_0$  can be calculated and the following analytic expression is obtained:

$$\frac{R(\lambda^2)}{\mu_0} = -\left(\frac{\alpha\gamma\lambda^2}{\mu_0}\right) \sum_{n=0}^{\infty} \int_0^{\infty} dk_1 k_1 \left(\frac{(k_1^2/\lambda^2)^{n+1}}{n!}\right) \exp\left(\frac{-k_1^2}{\lambda^2}\right) \int_{-\infty}^{\infty} dk_z \frac{1}{(k_z^2 + k_1^2) \{[(n-1)\lambda^2 + 1 + k_z^2]^2 + \frac{1}{4}\gamma^2\}}. \quad (55)$$

This expression leads to the numerical results presented for several values of  $\gamma$  in Fig. 4. It may be noted that the values obtained for  $R(\lambda^2)$  near the point  $\omega_c = \omega_{LO}$  lose their significance, since, in that region, the cyclotron line is far from being Lorentzian.

Let us now consider how the phonon-assisted absorption band shown in Fig. 2 arises from our calculation. Because of the energy denominator in  $Q(\omega)$  [Eq. (24)] the coefficients  $I(\omega)$  and  $R(\omega)$  exhibit an oscillatory behavior for  $\omega > \omega_{LO}$ . In this region of frequency the derivatives of the Lorentzian function appearing in (26) decrease monotonically. This leads to the phonon assisted transition peaks. The line shape of these peaks is highly asymmetric. The tail in high energy reflects the shape of the density of states of a charged particle in a uniform magnetic field. Most experimental results give the absorption versus the magnetic field. For this reason, we have plotted at Fig. 5 the absorption coefficient in the region of the phonon-assisted transitions as a function of  $\lambda^2$  for a fixed value of the incident frequency. This corresponds to Fig. 1 of the paper of Van Royen *et al.*,<sup>23</sup> although the line shape is slightly different due to the difference in the configuration used for the cyclotron resonance. However, for  $\gamma \rightarrow 0$ , both

$$R(\lambda^2)/\mu_0 = 0 \text{ for } \lambda^2 < 1, \quad (52)$$

$$\frac{R(\lambda^2)}{\mu_0} = \left(\frac{\alpha\lambda^2\pi}{\mu_0(\lambda^2 - 1)^{1/2}}\right) [1 - \beta e^{\beta} E_1(\beta)], \quad (53)$$

$$\beta = (\lambda^2 - 1)/\lambda^2 \text{ for } \lambda^2 > 1,$$

where the function  $E_1(x)$  is the exponential integral defined by

$$E_1(x) = \int_x^{\infty} \frac{e^{-t}}{t} dt. \quad (54)$$

This is exactly the result obtained by Harper.<sup>12</sup> This perfect agreement arises from the fact that, as shown above, no one-phonon Landau state other than  $n = 0$  contributes to  $R(\lambda^2)$ . This is not true for the shift term  $I(\lambda^2)$  appearing in Eq. (26) nor for both coefficients when  $\gamma \neq 0$ .

In the case  $\gamma \neq 0$  a small deviation from strict energy conservation law is allowed, so that phonon creation takes place even for  $\lambda^2 < 1$ . This gives rise to a nonzero value for  $R(\lambda^2)$  in this region. On the other hand the singularity for  $\lambda^2 = 1$  has disappeared.

For weak electron-phonon coupling and  $\gamma \neq 0$  we obtain, from Eq. (28),

spectra present absorption edges at magnetic fields such that  $\omega = \omega_{LO} + n\omega_c$ , with a maximum just before this edge and a minimum beyond it.

Recently, the development of spin-flip Raman lasers have made possible the measurement of the absorption coefficient as a function of incident frequency.<sup>4</sup> With a polaron radius of 12.5 Å and a coupling constant<sup>32</sup>  $\alpha = 0.02$ , the use of Fröhlich-interaction Hamiltonian and the choice of perturbation theory is certainly well justified in InSb. Unfortunately, this material is well known to exhibit a highly nonparabolic conduction band.<sup>33-35</sup> This makes difficult the comparison of our results with the existing experimental data. Nevertheless it can be seen from comparison between Fig. 3 of Ref. 4 and Fig. 2 of the present paper that a qualitative agreement is found concerning the line shape of the phonon-assisted transitions.

Our work shows that the harmonics demonstrated by the experimental results of Ref. 4 at  $\omega = 2\lambda^2$  or  $3\lambda^2$  do not appear at first order in the phonon coupling constant  $\alpha$ . As this coupling is very weak for InSb, it is quite probable that contributions of higher orders are negligible. This indicates that these harmonics are not due to the coupling of the electrons with the LO phonons.

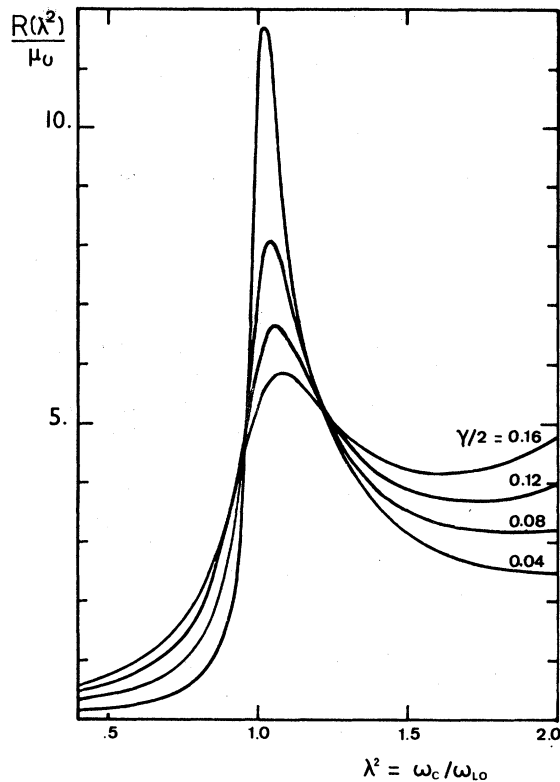


FIG. 4. Lorentzian broadening  $R(\lambda^2)$  of the main absorption line for different values of the initial linewidth  $\gamma$ .

V. CONCLUSIONS

In this paper, the cyclotron resonance by polarons has been calculated to linear order in the electron-phonon coupling constant  $\alpha$  and for a simple

parabolic band. The main contribution of our work is to allow the treatment of Landau states with a finite lifetime. The linewidth of the Landau states describes the effect of all the possible scattering processes other than the particular one involving LO phonons.

The introduction of a finite lifetime for the Landau states leads to predict an intraband magneto-absorption spectrum different from what could be deduced by simple considerations based on the pinning effect predicted by Larsen.<sup>10</sup> Indeed, the pinning effect suggests that, for  $\omega_c > \omega_{LO}$ , the spectrum has an extra line pinned just below  $\omega_{LO}$ . Our results show that this extra line does not show up when the finite lifetime of the Landau states is taken into account. The threshold of the phonon band is no longer sharply defined and the "pinned" state is just a small contribution to the low-energy tail of this band and can no longer be distinguished from it when  $\omega_c$  becomes appreciably larger than  $\omega_{LO}$ . The situation is similar to the cases of two interacting harmonic oscillators, the first one with variable frequency  $\omega_c$  and the second with a fixed frequency  $\omega_{LO}$ . The polaron states responsible for the doublet structure appearing in the spectrum when  $\omega_c \approx \omega_{LO}$  play the same role as the normal modes of these two oscillators.

The results we have obtained for the magnetic field dependance of the line broadening are in perfect agreement with Harper's results<sup>12</sup> when  $\omega_c > \omega_{LO}$ , except when  $\omega_c$  is close to  $\omega_{LO}$  where Harper obtained a divergent linewidth. On the contrary, by following Harper's procedure to obtain the linewidth of the  $n = 1$  Landau state, we obtain a finite result when  $\omega_c \approx \omega_{LO}$ . However, the spectrum has then the shape of a doublet and this procedure be-

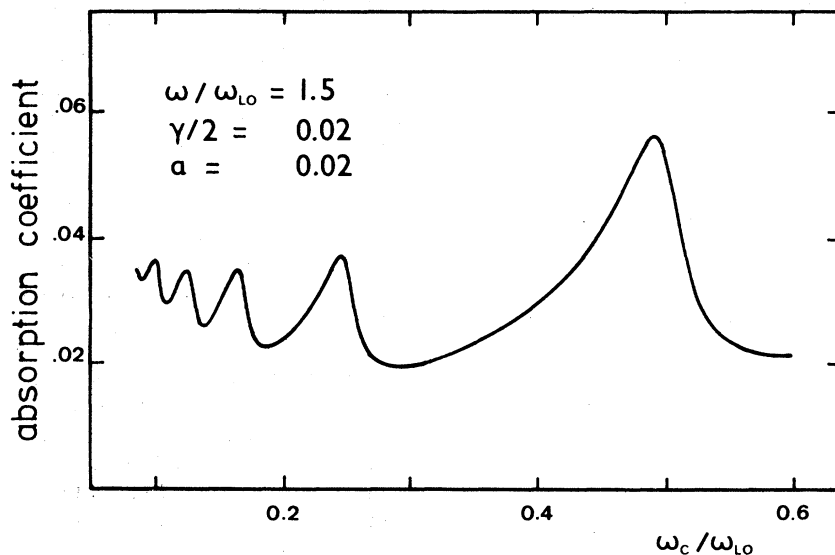


FIG. 5. Phonon sideband as a function of  $\omega_c / \omega_{LO}$  for a fixed incident frequency  $\omega = 1.5\omega_{LO}$ . Peaks appear for magnetic field strength such that:  $\omega_c = (\omega - \omega_{LO})/n$ ;  $n = 1, 2, 3, \dots$

comes questionable.

The region  $\omega \approx \omega_c \approx \omega_{LO}$  is masked by the strong free-lattice absorption. This is why the study of the phonon-assisted resonances, which fall outside the reststrahl region is of particular interest. Here also the introduction of a finite lifetime has a drastic effect on the shape of the spectrum. Indeed, one can predict<sup>5</sup> that, in the absence of natural broadening, the phonon-assisted spectrum consists of bands presenting a divergence at their low-energy threshold  $\omega = \omega_{LO} + n\omega_c$  ( $n=0, 1, \dots$ ). On the contrary, our results are divergence-free. Unfortunately, to our knowledge, the only case where phonon-assisted transitions have been observed is indium antimonide, for which nonparabolicity prevents comparison with the results of our calculations valid for a simple parabolic band. However, one can conclude that there is a good qualitative agreement on the shape of the phonon-assisted bands.

#### ACKNOWLEDGMENT

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#### APPENDIX A

Here it is shown how the function

$$\phi_m(a) = \sum_k \nu_k^2 e^{-(k_1^2/\lambda^2)} \sum_{n=0}^{\infty} \frac{(k_1^2/\lambda^2)^n}{n!} \times (\nu\lambda^2 + a + k_2^2 + 1)^{-m-1} \quad (\text{A1})$$

can be expressed as a one-dimensional integral when the variable  $a$  is real and positive. By using polar coordinates  $(k, \theta, \phi)$  and by making  $x = \cos\theta$  and  $u = k/\lambda$ , one may write Eq. (A1) under the following form:

$$\phi_m(a) = \frac{\alpha}{\pi\lambda^{2m+1}} \int_0^{\infty} du \int_{-1}^{+1} \exp[u^2(x^2 - 1)] \times \sum_{n=0}^{\infty} \frac{[u^2(1-x^2)]^{n+1}}{n!} \times (\nu + b + u^2x^2)^{-m-1}, \quad (\text{A2})$$

where we have put  $b = (a+1)/\lambda^2$ . One may then take advantage of the following identity, which holds for any positive value of  $b$ ,

$$(\nu + b + u^2x^2)^{-m-1} = \frac{1}{m!} \int_0^{\infty} t^m \exp[-(\nu + b + u^2x^2)t] dt. \quad (\text{A3})$$

When this expression is introduced into Eq. (A2), one is able to perform the summation over  $n$ . This leaves the next expression

$$\phi_m(a) = \frac{\alpha}{\pi\lambda^{2m+1} m!} \int_0^{\infty} dt t^m e^{-bt} \times \int_{-1}^{+1} dx (1-x^2) \times \int_0^{\infty} du u^2 \exp(\delta_1 + x^2\delta_2), \quad (\text{A4})$$

where we have

$$\delta_1 = 1 - e^{-t} \quad (\text{A5})$$

and

$$\delta_2 = t - \delta_1. \quad (\text{A6})$$

By performing consecutively integrations over  $u$  and  $x$ , one obtain the simple result

$$\phi_m(a) = \frac{\alpha}{2m! \sqrt{\pi} \lambda^{2m+1}} \int_0^{\infty} t^m g(t) \exp\left(\frac{-(a+1)t}{\lambda^2}\right) dt, \quad (\text{A7})$$

where

$$g(t) = (\sqrt{t}/\delta_1\delta_2) - \delta_2^{-3/2} \ln\{(\delta_2/\delta_1)^{1/2} + [1 + (\delta_2/\delta_1)]^{1/2}\}. \quad (\text{A8})$$

#### APPENDIX B

This Appendix is devoted to the calculation of the integral arising from the last term of Eq. (48) and to the consideration of the asymptotic behavior for  $n \rightarrow \infty$  of the general term of the corresponding series.

By introducing development (48) into expression (47) for  $\alpha Q(\omega)$ , one obtains for  $N=2$ ,

$$\alpha Q(\omega) = M_1 + M_2 + J_2, \quad (\text{B1})$$

where  $M_1$  is precisely equal to  $-I_1$  [Eq. (39)], which causes cancellation of these divergent terms in the general expression for  $G(\omega)$ .  $M_2$  turns out to be represented by

$$M_2 = - \sum_{m=1}^N \left(\omega + \frac{i\gamma}{2}\right)^m \phi_m(0), \quad (\text{B2})$$

where  $\phi_m(a)$  is calculated in Appendix A. Finally,  $J_2$  can be written in the form of a series

$$J_2 = \sum_{n=0}^{\infty} j_n, \quad (\text{B3})$$

where

$$j_n = \frac{-\alpha\beta^3}{\pi} \int_0^\infty k_1 dk_1 \frac{(k_1^2/\lambda^2)^{n+1}}{n!} e^{-k_1^2/\lambda^2} \times \int_{-\infty}^{+\infty} \frac{dk_z}{(b_n^2 + k_z^2)(\alpha_n^2 + k_z^2)^3(k^2 + k_z^2)}, \quad (\text{B4})$$

where

$$\begin{aligned} \beta &= +\frac{1}{2}i\gamma, \quad \alpha_n^2 = 1 + n\lambda^2, \\ b_n^2 &= n\lambda^2 + 1 - \omega - \frac{1}{2}i\gamma; \end{aligned} \quad (\text{B5})$$

these terms can be easily computed as

$$j_0 = \frac{-\alpha\beta^3}{2\pi\lambda^7} \int_0^\infty \sqrt{x} e^{-x} S(\alpha_0^2, \beta_0^2, x) dx, \quad (\text{B6})$$

$$j_n = \frac{-\alpha\beta^3}{2\pi\lambda^7 n^{5/2}} \int_0^\infty \sqrt{x} f_n(x) S(\alpha_n^2, \beta_n^2, x) dx, \quad (\text{B7})$$

where

$$\begin{aligned} \alpha_0^2 &= 1/\lambda^2, \quad \beta_0^2 = \frac{1-\omega}{\lambda^2} - i\frac{\gamma}{2\lambda^2}, \\ \alpha_n^2 &= \frac{\alpha_n^2}{n\lambda^2}, \quad \beta_n^2 = \frac{\beta_n^2}{n\lambda^2}, \\ f_n(x) &= \frac{(nx)^n e^{-nx}}{n!}. \end{aligned} \quad (\text{B8})$$

The function  $S(\alpha_n^2, \beta_n^2, x)$  is obtained by integrating exactly the factors depending on  $k_z$  in Eq. (B4).

One has

$$S(\alpha_n^2, \beta_n^2, x) = \pi M_5(x)/D_5(x),$$

where, successively,

$$M_5(x) = \frac{g_3^2 - g_1(g_5 - g_1g_4 + g_2g_3)}{\alpha_n^3/\beta_n},$$

$$D_5(x) = g_5[g_5 - g_2g_3 + g_1(g_2^2 - 2g_4)] + g_4[g_3^2 + g_1(g_1g_4 - g_2g_3)],$$

$$g_1 = -i(\sqrt{x} + A_3),$$

$$g_2 = -(A_3\sqrt{x} + 3\alpha_n A_3),$$

$$g_3 = i\alpha_n(3A_3\sqrt{x} + \alpha_n B_3),$$

$$g_4 = \alpha_n^2(B_3\sqrt{x} + \alpha_n\beta_n),$$

$$g_5 = -i\beta_n\alpha_n^3\sqrt{x},$$

$$A_3 = \beta_n + 3\alpha_n, \quad B_3 = \alpha_n + 3\beta_n, \quad A_b = \alpha_n + \beta_n.$$

In these expressions,  $\beta_n$  has a positive real part.

For large  $n$ , the function  $f_n(x)$ , which has a maximum for  $x=1$  [ $f_n(1) \sim (2\pi n)^{-1/2}$ ] can be approximated by  $n^{-1} \delta(x-1)$ . The function  $S(\alpha_n^2, \beta_n^2, x)$  behaves like a constant, so that  $j_n$  tends to zero like  $n^{-7/2}$ . Its convergency may then be considered as fairly fast.

#### APPENDIX C

This Appendix deals with the problem of calculating the absorption (or emission) spectrum of

radiation by a system in interaction with two types of perturbations. Due to the strength of the first type of perturbation (type I) as well as to its excitation spectrum, it is expected that it leads to a modification of the absorption spectrum with a complicated structure. Therefore the effects of this perturbation cannot be simply described by a frequency independent energy shift and broadening of the absorption lines, so that type-I perturbation must be treated separately. In the present paper, this perturbation is the interaction with the LO phonons.

The strength of the second type of perturbation (type II) is weak. Moreover, the perturbing system has a continuous spectrum of low-lying energy levels. Therefore, it is expected that the type-II perturbation gives rise only to a structureless change in the absorption spectrum that can be described by a frequency independent shift and broadening of the lines. In the case considered here, type-II perturbation is the interaction with the long wavelength acoustic phonons, the elastic collisions on impurities or defects of the crystal as well as the interaction with the radiation field itself.

The Hamiltonian for the system under consideration is

$$\mathcal{H} = \mathcal{H}_0 + H_1 + H_2, \quad (\text{C1})$$

where  $H_1$  and  $H_2$  are, respectively, the coupling terms for type-I and type-II perturbations.  $\mathcal{H}_0$  is the unperturbed Hamiltonian including the energy of the photon field. Since type II has a continuous spectrum of states, the time-dependent perturbation formalism as described by Louisell<sup>36</sup> can be applied. It gives the following result:

$$C_i(t) = \exp(-\frac{1}{2}\gamma t - i\Delta\omega t), \quad (\text{C2})$$

where  $C_i(t)$  is, in the interaction picture, the projection of the wave function at time  $t$  on the initial state  $|i\rangle$ . The frequency shift and the broadening in units such that  $\hbar=1$ , are given by

$$\Delta\omega = -P \sum_f \frac{|\langle f|H_2|i\rangle|^2}{E_f - E_i}, \quad (\text{C3})$$

$$\gamma = 2\pi \sum_f |\langle f|H_2|i\rangle|^2 \delta(E_f - E_i), \quad (\text{C4})$$

where  $|i\rangle$  and  $|f\rangle$  are, respectively, the initial and final states during transitions produced by the perturbation of type II. These states are eigenfunctions of

$$H = \mathcal{H}_0 + H_1, \quad (\text{C5})$$

with eigenvalues  $E_i$  and  $E_f$ . In the case discussed here,  $H$  is the Hamiltonian for the polaron in a constant and uniform magnetic field, including a

term describing the energy of the photon field. In the interaction picture, the projections  $C_f(t)$  of the wavefunction on the final states are given by relation (5.89) of the reference given above,<sup>36</sup> i.e.,

$$C_f(t) = -\frac{\langle f|H_2|i\rangle}{E_f - E_i - \Delta + \frac{1}{2}i\gamma} \{ \exp[i(E_f - E_i - \Delta\omega)t + \frac{1}{2}\gamma t] - 1 \}. \quad (C6)$$

The way of reasoning that we now follow is slightly different from Louisell's treatment. Taking the square of the modulus of (C6) gives the population of the states  $|f\rangle$  at time  $t$ . It is

$$|C_f(t)|^2 = |\langle f|H_2|i\rangle|^2 \times \frac{1 + e^{-\gamma t} - 2e^{-\gamma t/2} \cos(E_f - E_i - \Delta\omega)t}{(E_f - E_i - \Delta\omega)^2 + \frac{1}{4}\gamma^2}. \quad (C7)$$

As we are calculating the absorption spectrum, we are interested only in the probability of transitions to states  $|f\rangle$  containing one less photon than the initial state (one more for emission). Therefore, in (C7) we only keep the matrix elements of that part of  $H_2$  which describes the interaction with the radiation field, i.e., for the case of a polaron in a magnetic field.

$$H_r = -i\sqrt{2}\lambda\alpha_r\alpha_0^2(\hat{p} + |e|\vec{A}/c) \cdot \vec{n}, \quad (C8)$$

where the notations have been defined in Sec. II. Moreover, for an absorption process, we have  $E_i = \epsilon_i + \omega$  and  $E_f = \epsilon_f$ , where  $\epsilon_i$  and  $\epsilon_f$  are the polaron energies in the initial and final state and  $\omega_1$  is the frequency of the absorbed photon. The total population of states after absorption of a photon is

$$\sum_{f \neq i} |C_f(t)|^2 = \sum_{f \neq i} \frac{|\langle f|H_r|i\rangle|^2}{(\epsilon_f - \epsilon_i - \omega - \Delta\omega)^2 + \frac{1}{4}\gamma^2} \times \xi(\epsilon_f - \epsilon_i - \omega - \Delta\omega; t), \quad (C9)$$

where

$$\xi(a; t) = 1 - e^{-\gamma t} - e^{-\gamma t/2} e^{iat} - e^{-\gamma t/2} e^{-iat}. \quad (C10)$$

As the radiation field has a continuous spectrum of energy eigenvalues, the sum in (C9) includes an integration over  $\epsilon_f$ . Since  $\frac{1}{2}\gamma$  is small, one expects that the main contribution to the integral

comes from  $\epsilon_f \sim \epsilon_i + \omega + \Delta\omega$ . Therefore, extending the domain of integration from  $-\infty$  to  $+\infty$  does not appreciably change the result. The integration can then be performed in the complex plane, closing the integration contour by a circle in the upper-half complex plane for the term that contains the factor  $\exp i(\epsilon_f - \epsilon_i - \omega - \Delta\omega)t$  and in the lower-half plane for its complex conjugate. The denominator has two first order poles at

$$\epsilon_f = \epsilon_i + \omega + \Delta\omega \pm \frac{1}{2}i\gamma. \quad (C10)$$

Therefore, in the calculation of the residue of the numerator, the exponentials written above become

$$\exp[i(\epsilon_f - \epsilon_i - \omega - \Delta\omega)t] = \exp(-\frac{1}{2}\gamma t) \quad (C11)$$

and

$$\exp[-i(\epsilon_f - \epsilon_i - \omega - \Delta\omega)t] = \exp(-\frac{1}{2}\gamma t). \quad (C12)$$

As  $|\langle f|H_r|i\rangle|^2$  is expected to be a smooth function with no singularity, (C9) can be written in the equivalent form

$$\sum_{f \neq i} |C_f(t)|^2 = \sum_{f \neq i} \frac{|\langle f|H_r|i\rangle|^2 [1 - \exp(-\gamma t)]}{(\epsilon_f - \epsilon_i - \omega - \Delta\omega)^2 + \frac{1}{4}\gamma^2}, \quad (C13)$$

obtained by using (C11) and (C12). The transition probability per unit time is the total population (C13) measured before the decay of the initial state [i.e.,  $|C_i(t)|^2 \sim 1$  or  $\gamma t \ll t$ ] and divided by  $t$ . Expanding  $\exp(-\gamma t)$  in powers of  $\gamma t$  gives the following form of the absorption probability per unit time:

$$P(\omega) = 2\pi \sum_{f \neq i} |\langle f|H_r|i\rangle|^2 \frac{\gamma/2\pi}{(\epsilon_f - \epsilon_i - \omega - \Delta\omega)^2 + \frac{1}{4}\gamma^2}. \quad (C14)$$

Equation (12), i.e., the basic relation used in the present paper, is obtained from this expression by neglecting the constant energy shift  $\Delta\omega$ . This is not a crucial approximation since it only leads to a slight shift of the absorption curve along the frequency axis.

The present approach shows how to take the finite lifetime of the initial state into account. It should be possible to treat in the same way the lifetime of the final states.

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