Optical Absorption in an Electric Field

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A previous calculation of the effect of an external, uniform electric field on the optical absorption associated with a direct transition between bands is extended. The absorption is shown to consist of a series of steps whose width is determined by the separation of the discrete levels produced by the field.

IN a previous calculation[1] (which will be referred to as I), it was shown that the discrete levels which are formed when a uniform electric field is present in an insulating crystal[2] produce a periodic structure in the energy dependence of the absorption due to interband transitions. The expression for the absorption coefficient which was given in that paper is a slowly convergent Fourier series which cannot easily be interpreted. It is the object of the present work to transform this series in such a way that the energy dependence of the optical absorption is evident. I hope that this work will help to stimulate experimental investigation of the discrete levels.

An infinite crystal containing a uniform electric field $\mathcal{E}$, in the x direction is considered. From Eqs. (14) and (21) of I, the following expression is obtained for the absorption coefficient α, produced by a direct transition between bands n' and n (the notation conforms to I as far as possible):

$$\alpha=\frac{2\kappa^2}{F\pi\omega n\epsilon_0 c\mathcal{A}_0^2}\left[\int\frac{d\mathbf{k}_\perp}{(2\pi)^2}|M_{nn'}(x_0)|^2+2\sum_{l=1}^{\infty}\int\frac{d\mathbf{k}_\perp}{(2\pi)^2}|M_{nn'}(x_0)|^2\cos 2\pi l x_0\right], \quad (1)$$

in which κ is the width of the Brillouin zone in the x direction (which coincides with a reciprocal lattice vector); $\mathbf{k}_\perp$ represents components of the wave vector perpendicular to the field; $\mathcal{A}_0$ is the magnitude of the vector potential of the radiation field (which has circular frequency ω); n is the index of refraction, and ϵ_0 is the permittivity of free space. In addition, $F=e\mathcal{E}$, and

$$x_0=\frac{\kappa}{2\pi F}(\Delta_{nn'}-\hbar\omega), \quad (2a)$$

where

$$\Delta_{nn'}(\mathbf{k}_\perp)=\kappa^{-1}\int_{-\kappa/2}^{\kappa/2}[E_n(k_x,\mathbf{k}_\perp)-E_{n'}(k_x,\mathbf{k}_\perp)]dk_x. \quad (2b)$$

The matrix element $M_{nn'}$ is given by

$$M_{nn'}=\frac{2\pi e\mathcal{A}_0\beta^{1/3}}{m\kappa}(\boldsymbol{\epsilon}\cdot\mathbf{p}_{nn'})\,\mathrm{Ai}(\sigma\beta^{1/3}), \quad (3)$$

where $\mathbf{p}_{nn'}$ is the usual interband matrix element of the momentum and $\boldsymbol{\epsilon}$ is the polarization vector of the radiation. Both bands are treated in the effective mass approximation, so that the energy difference between bands is

$$E_n-E_{n'}=E_g+(\hbar^2k^2/2\mu). \quad (4)$$

In this equation, E_g is the energy gap at $k=0$, and μ is the reduced mass for the two bands: $\mu^{-1}=m_n^{-1}+m_{n'}^{-1}$. We also have

$$\beta=2\mu F/\hbar^2;\quad \sigma=F^{-1}(E_g-\hbar\omega+\hbar^2k_\perp^2/2\mu),$$

and Ai is the Airy integral, which is defined by

$$\mathrm{Ai}(z)=\frac{1}{\pi}\int_0^\infty\cos(sz+s^3/3)ds.$$

Other quantities which are not explicitly defined above have their usual significance.

In I, approximate expressions were worked out for the terms of the series, Eq. (1). It is, however, evident from Eqs. (1) and (2) without approximation that whenever the photon energy $\hbar\omega$ increases by $\Delta(\hbar\omega)$ with

$$\Delta(\hbar\omega)=2\pi F/\kappa, \quad (5)$$

all the cosines return to their original values. This energy change is just the separation between the discrete levels so that the absorption must contain some structure with this period. In order to investigate this structure, we proceed as follows. The sum of trigonometric functions is evaluated using the identity[3]

$$1+2\sum_{l=1}^{\infty}\cos ly=2\pi\sum_{m=-\infty}^{\infty}\delta(y-2m\pi). \quad (6)$$

Then with the use of (3)

$$\alpha=K\frac{2\mu}{\omega\beta^{1/3}}\int d\mathbf{k}_\perp\,\mathrm{Ai}^2(\sigma\beta^{1/3})\left[1+2\sum_{l=1}^{\infty}\cos 2\pi l x_0\right]$$

$$=(4\pi K\mu/\beta^{1/3}\omega)\sum_{j=-\infty}^{\infty}\int d\mathbf{k}_\perp\,\mathrm{Ai}^2(\sigma\beta^{1/3})\delta(2\pi x_0-2\pi j), \quad (7)$$

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[1] J. Callaway, Phys. Rev. **130**, 549 (1963). Other calculations of the effect of an electric field on optical absorption have been reported by: W. Franz, Z. Naturforsch. **13**, 484 (1958); L. V. Keldysh, Zh. Eksperim. i Teor. Fiz. **34**, 1138 (1958) [English transl.: Soviet Phys.—JETP **7**, 788 (1958)]; and K. Tharmalingam, Phys. Rev. **130**, 2204 (1963).

[2] G. Wannier, Phys. Rev. **117**, 432 (1960).

[3] G. Goertzel and N. Tralli, *Some Mathematical Methods of Physics* (McGraw-Hill Book Company, Inc., New York, 1960), p. 123.

in which

$$K=2e^2|\boldsymbol{\varepsilon}\cdot\mathbf{p}_{nn'}|^2/\pi\hbar^2m^2n\epsilon_0c. \tag{8}$$

From Eqs. (2) and (4), we obtain for x_0

$$2\pi x_0=\kappa(\sigma+\chi\beta^{-1/3}), \tag{9}$$

in which

$$\chi=\kappa^2/12\beta^{2/3}.$$

Then we change the variable of integration in (7) to $y=\sigma\beta^{1/3}$ and obtain

$$\alpha=\frac{4\pi^2K\mu\beta^{2/3}}{\omega\kappa}\sum_{j=-\infty}^{\infty}\int dy\,\mathrm{Ai}^2(y)\times\delta[y+\chi-(2\pi j\beta^{1/3}/\kappa)]. \tag{10}$$

If we allow $k_\perp$ to vary between 0 and ∞, then y varies from a lower value

$$y_0=R^2\beta^{-2/3};\quad R^2=(2\mu/\hbar^2)(E_g-\hbar\omega) \tag{11}$$

to ∞. Note that R^2 may be either positive or negative. We then find that the absorption coefficient is

$$\alpha=\frac{4\pi^2K\mu\beta^{2/3}}{\omega\kappa}\sum_{j=j_0}^{\infty}\mathrm{Ai}^2\left(\frac{2\pi j\beta^{1/3}}{\kappa}-\chi\right), \tag{12}$$

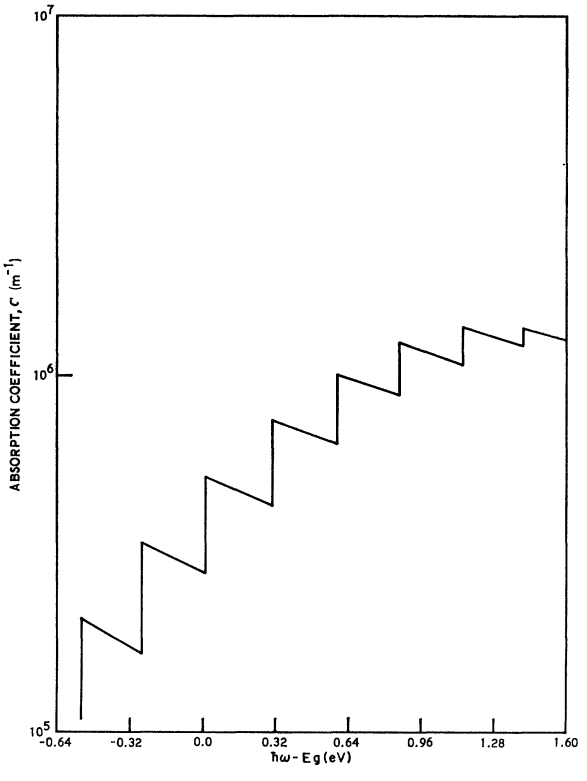

FIG. 1. The optical absorption coefficient α is shown as a function of $\hbar\omega-E_g$ as calculated from Eq. (12) using numbers appropriate to the transition between the heavy-hole band and the conduction band in gallium arsenide. For the purpose of illustration, a very large field, 10^9 eV/m has been chosen.

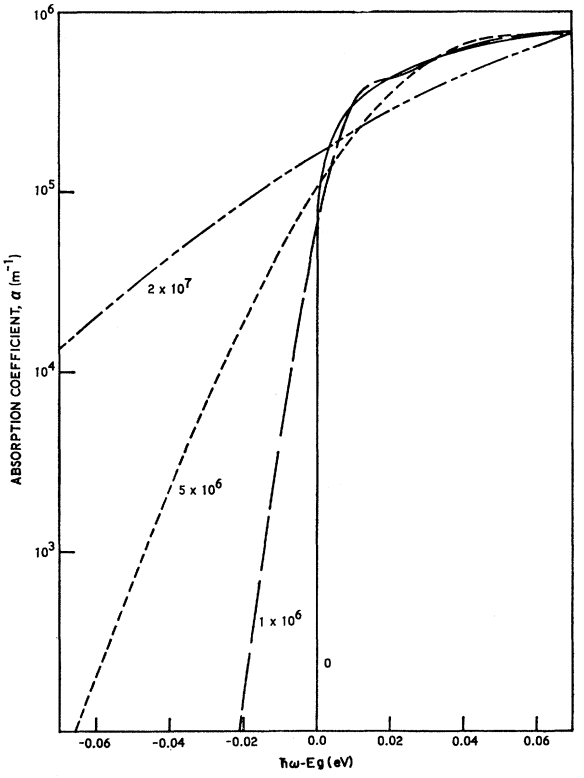

FIG. 2. The optical absorption coefficient is shown as a function of energy in the weak-field limit, Eq. (18), neglecting the discrete levels. Numbers appropriate to gallium arsenide, including both light and heavy holes, have been considered. Each curve is marked by the value of the field F in electron volts per meter which has been used in the calculation. The field is oriented along the [100] crystal axis.

in which the lower limit of the summation j_0 is determined by the condition that j_0 be the next integer larger than q_0, where

$$(2\pi q_0\beta^{1/3}/\kappa)-\chi=R^2/\beta^{2/3}. \tag{13}$$

Let us interpret this result. Observe that the argument of the Airy functions in (12) is independent of energy. The energy dependence of the absorption coefficient is contained in the external factor ω^{-1} and implicitly in the lower limit of the summation. As the photon energy $\hbar\omega$ increases, R^2 decreases, and consequently y_0 and j_0 decrease. For each decrease in j_0, a new term appears in the sum in Eq. (12). Consequently, a graph of the absorption coefficient α as a function of energy resembles a staircase with slanted steps. The width of each step is the energy required to produce a unit change in j_0. This is easily seen to be given by Eq. (5). Hence, the width of a step is just the separation of the discrete levels. The height of a step is proportional to $\mathrm{Ai}^2(R^2\beta^{-2/3})$, evaluated at the energy at which the jump occurs. The downward slant of each step is determined by the factor ω^{-1}.

For large positive values of $R^2\beta^{-2/3}$, the change in α,

$\Delta\alpha$, produced by bringing in one more term (the height of the step), is easily obtained by using the asymptotic form of the Airy function:

$$\mathrm{Ai}(z)=(1/2\pi^{1/2}z^{1/4})\exp(-\tfrac{2}{3}z^{3/2}). \tag{14}$$

Hence, in this region

$$\Delta\alpha\approx(\pi K\beta/\omega\kappa R)\exp[-4R^3/3\beta]$$
$$=\frac{\pi KF}{\kappa\omega}\left[\frac{2\mu}{\hbar^2(E_g-\hbar\omega)}\right]^{1/2}$$
$$\times\exp\left[-\frac{4}{3}\left(\frac{2\mu}{\hbar^2}\right)^{1/2}\frac{(E_g-\hbar\omega)^{3/2}}{F}\right]. \tag{15}$$

Thus, the height of the steps decreases rapidly with increasing $E_g-\hbar\omega$ and decreasing field. For large negative values of $R^2\beta^{-2/3}$ we use

$$\mathrm{Ai}(-z)=(1/\pi^{1/2}z^{1/4})\sin(\tfrac{2}{3}z^{3/2}+\pi/4) \tag{16}$$

and find

$$\Delta\alpha\approx\frac{4\pi KF}{\kappa\omega}\left[\frac{2\mu}{\hbar^2(\hbar\omega-E_g)}\right]^{1/2}$$
$$\times\sin^2\left[\frac{2}{3}\left(\frac{2\mu}{\hbar^2}\right)^{1/2}\frac{(\hbar\omega-E_g)^{3/2}}{F}+\frac{\pi}{4}\right]. \tag{17}$$

These results are illustrated in Fig. 1.

The mathematical argument may be concluded by observing that the weak-field limit of the absorption is obtained by replacing the sum in (12) by an integral. Put

$$z=2\pi j\beta^{1/3}/\kappa-\chi,$$

and let z be continuous. Then we obtain in place of (12)

$$\alpha=2\pi\omega^{-1}K\mu\beta^{1/3}\int_{y_0}^{\infty}\mathrm{Ai}^2(z)dz$$
$$=2\pi\omega^{-1}K\mu\beta^{1/3}\left[\left(\frac{d\,\mathrm{Ai}(y)}{dy}\right)_{y_0}^2-y_0\,\mathrm{Ai}^2(y_0)\right], \tag{18}$$

where y_0 is given in Eq. (11). This is equivalent to an expression given by Tharmalingam,[1] and may also be obtained by discarding all but the constant term in Eq. (1). Expressions valid in the limit of large positive or negative values of y_0 may be obtained by inserting the asymptotic expansions of the Airy functions Eqs. (14) and (16) into (18). The results of this can be obtained in an obvious way from Eqs. (23), (29), and (31) of I, and also from Tharmalingam's paper, so they will not be repeated here. However, since the weak-field absorption coefficient has not been exhibited in the region near the band gap in which asymptotic forms cannot be used, we present in Fig. 2 the absorption coefficient as calculated in this approximation using numbers appropriate to gallium arsenide.

Finally, we should note that although the detailed results of this calculation depend on the use of the effective-mass approximation, Eq. (4), the existence of structure in the absorption constant with the period of the separation between the discrete levels is rigorous. In addition, one expects the effective-mass approximation to be valid in the region near a band gap which is accessible to experimental observation. The quantity χ, which cannot be calculated reliably in this approximation since it involves an integral over the entire band structure, acts only as a phase angle in (12). Therefore, uncertainty as to the value of χ does not affect these results in any essential way.

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