PHYSICAL REVIEW VOLUME 140, NUMBER 1A 4 OCTOBER 1965

Electric-Field-Induced Indirect Optical Absorption in Solids and the Fine Structure of the Stark Splitting*

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(Received 4 May 1965)

The problem of optical transitions in solids subject to a strong electric field is discussed using time-dependent wave functions. Expressions for the change in transition rate for direct and indirect transitions are obtained in terms of integrals of Bessel functions. Moreover, using approximations for the Bessel functions, it is shown that the change in transition rate for direct transitions, T_d, and for indirect transitions, T_i, are given by

$$T_d = E^{1/3} F_d(\epsilon_p - \epsilon_{g0})/E^{2/3}$$

and

$$T_i = E^{4/3} F_i(\epsilon_p - \epsilon_{g0})/E^{2/3},$$

where E is the applied electric field, ϵ_p the excitation energy, and ϵ_{g0} the minimum energy gap. These formulas are valid both below and above the absorption edge. As a special case a machine computation of the change in the transition rate for Si with field applied in the [100] direction is presented. The results show a series of decaying peaks. In addition, a fine structure is predicted of the same kind as that predicted by Callaway for the Stark splitting. The details of the fine structure are analyzed and are shown to depend on the particular structure of the bands both near and far above the minimum energy gap. Difficulties in the experimental observation of the fine structure arising from inhomogeneity of the electric field are discussed.

INTRODUCTION

THE effect of an electric field on the absorption of light in the vicinity of the absorption edge has been discussed by a few authors. The results of Franz[1] and Keldysh[2] which are based on time-dependent wave functions are restricted mainly to direct-gap materials and to photon energies below the absorption edge. Callaway,[3,4] using a stationary-states method and Tharmalingam,[5] using an effective-mass theorem, obtain more general results for the direct-gap case which are valid for wavelengths both below and above the absorption edge. Moreover, Callaway,[3,4] using the stationary-states method, predicts that Stark splitting will produce fine structure in the absorption of light.

In this paper we approach the problem using time-dependent wave functions. It is shown that expressions for the change in absorption may be obtained which are valid below, at, and above the absorption edge, for both direct and indirect transitions. Moreover, it is shown that the fine structure predicted by Callaway[3,4] is also predicted by the time-dependent wave-function approach.

We shall focus attention here on indirect transitions in the presence of an electric field, and on some details of the fine structure.

Expressions describing the change in indirect transitions in the presence of electric field with preliminary application to silicon, have been reported previously.[6] Penchina[7] has also found expressions equivalent to ours, and has also given a calculation for silicon with the field applied in the [100] direction. However, a special feature of our results is the inclusion of an approximate general form of the relation between changes in transition probability excitation energy and applied field which is valid continuously from below to above the absorption edge. In addition, we give the result of an accurate calculation for silicon when the field is applied in the [111] direction. This case is of interest since both Handler's[8] and our experiments were made for this direction of the applied field. Our calculation for this case goes to high excitation energies, where the required accuracy of the computation is very high and shows a set of secondary peaks for each process in addition to the one reported earlier.[6,7] It should be pointed out that the measurements are still quite sensitive at this range of excitation energies.

By analyzing the details of the fine structure beyond the results reported by Callaway,[3,4] it is shown that the fine structure might differ considerably in different materials, depending upon their particular band structure and upon the direction of the applied field. Finally, however, certain experimental difficulties in observing this fine structure are discussed.

Throughout the development, the only factors expressed explicitly are those which are needed in the analysis of experimental data in which the absorption and the change in the absorption with applied electric field are measured directly, and where the attention is focused on $\Delta\alpha/\alpha$.

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* Research supported in part by the Advanced Research Projects Agency under Contract No. SD-90. This paper is based on a thesis submitted to the Massachusetts Institute of Technology in partial fulfillment of the requirements for the degree of Doctor of Science.

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THEORY

The one-electron Hamiltonian for the problem of indirect optical transitions in the presence of an electric field is

$$H=H_0+H_1,$$

$$H_1=e\mathbf{E}\cdot\mathbf{R}+O_le^{-i\omega_lt}e^{st}+O_se^{-i\mathbf{k}_s\cdot\mathbf{R}}e^{\pm i\omega_st}e^{s't}. \quad (1)$$

H_0 is the unperturbed Hamiltonian; $\mathbf{E}$ the applied electric field; $\mathbf{R}$ the space coordinate of the electron; O_l the time-independent part of the electromagnetic-radiation operator; and $O_se^{-i\mathbf{k}_s\cdot\mathbf{R}}$ the time-independent part of the lattice-vibration operator corresponding to a phonon with wave number $\mathbf{k}_s$; ω_l and ω_s are the frequencies of the electromagnetic radiation and of the lattice vibrations correspondingly; s and s' are positive constants which at the end will be taken to the limit zero. The electron-hole interaction has been neglected.

We choose to solve the problem in terms of a time dependent wave function of the form

$$\psi=a(t)B_v(\mathbf{k};\mathbf{R};t)+a'(t)B_v(\mathbf{k}';\mathbf{R};t)$$
$$+b'(t)B_c(\mathbf{k};\mathbf{R};t)+b(t)B_c(\mathbf{k}';\mathbf{R};t) \quad (2)$$
$$\mathbf{k}'-\mathbf{k}=\mathbf{k}_s.$$

Here subscripts v and c denote valence and conduction bands correspondingly. B is the Houston wave function

$$B=\phi_{\mathbf{k}}(\mathbf{R})e^{-i\mathbf{k}\cdot\mathbf{R}}\exp\left[-\frac{i}{\hbar}\int^t\epsilon(\mathbf{k})d\tau\right],\ \mathbf{k}=\mathbf{k}_0+e\mathbf{E}t, \quad (3)$$

$\phi_{\mathbf{k}}(\mathbf{R})$ is the periodic part of the Block wave function, $\epsilon(\mathbf{k})$ is the corresponding energy for the unperturbed Hamiltonian.

We assume that at $t\rightarrow-\infty$ $a(t)=1$ and all other coefficients are zero. We then proceed to evaluate the other coefficients as functions of time. In the process we neglect the interaction between B_v and B_c produced by the operator $e\mathbf{E}\cdot\mathbf{R}$. This interaction has been estimated[6] and found to be completely negligible in comparison with the main effect of interest.

Using the usual methods, we obtain

$$i\hbar\frac{db'}{dt}=\langle\phi_c(\mathbf{k})|0_l|\phi_v(\mathbf{k})\rangle$$
$$\times\exp\left\{\frac{i}{\hbar}\int^t[\epsilon_c(\mathbf{k})-\epsilon_v(\mathbf{k})-\hbar\omega_l]d\tau\right\}e^{st}. \quad (4)$$

Both the matrix element and the exponent in Eq. (4) are time-dependent through the dependence of $\mathbf{k}$ on time. However, since the time variation of these elements is much slower than that of the exponential term, we can write approximately

$$i\hbar b'=\langle\phi_c(\mathbf{k})|0_l|\phi_v(\mathbf{k})\rangle\exp\left\{\frac{i}{\hbar}\int^t[\epsilon_c(\mathbf{k})-\epsilon_v(\mathbf{k})-\hbar\omega_l]d\tau\right\}e^{st}\Big/ i\left(\frac{\epsilon_c(\mathbf{k})-\epsilon_v(\mathbf{k})}{\hbar}\right)-i\omega_l+s. \quad (5)$$

We now obtain a' in the same way, and use the results for a' and b' to find

$$(i\hbar)^2\frac{db}{dt}=g_i\exp\left[-\frac{i}{\hbar}\int^t(\epsilon-\epsilon_p)d\tau\right]e^{(s+s')t}; \quad (6)$$
$$\epsilon=\epsilon_c(\mathbf{k}')-\epsilon_v(\mathbf{k});$$
$$\epsilon_p=\hbar\omega_l\pm\hbar\omega_s.$$

The factor g_i is independent of the electric field of $\epsilon-\epsilon_p$ and of temperature and thus will not be given explicitly.

The energy difference ϵ is time-dependent through its dependence on $\mathbf{k}$. Moreover, since ϵ is a periodic function of $\mathbf{k}$, it is also a periodic function of time, with basic frequency ω_0, where ω_0 is given by

$$\omega_0=\frac{\lambda eE}{\hbar}, \quad (7)$$

where $2\pi/\lambda$ is the periodicity of the reciprocal lattice in the direction of the applied field. If $\bar{\epsilon}$ denotes the average of ϵ then, the exponential $\exp[i\int^t(\epsilon-\bar{\epsilon})d\tau]$ is also periodic with the same basic frequency. It is therefore possible to expand this exponential in Fourier series as follows:

$$\exp\left[\frac{i}{\hbar}\int^t(\epsilon-\bar{\epsilon})d\tau\right]=\sum_n C_ne^{in\omega_0t}. \quad (8)$$

Using this expansion, we find that the transition rate for given $k_\perp$ and $\mathbf{k}_s$ is given by

$$T(k_\perp;\mathbf{k}_s)=\int_0^\infty\lim_{(s+s')\rightarrow0}\frac{d}{dt}|b|^2I(\epsilon_l)\frac{d\epsilon_p}{\hbar}$$
$$=\int_0^\infty\left[\sum_{n=-\infty}^{\infty}|g_i|^2|c_n|^2\frac{2\pi}{\hbar^4}\delta\left(\frac{\bar{\epsilon}}{\hbar}+n\omega_0-\frac{\epsilon_p}{\hbar}\right)\right]$$
$$\times I(\epsilon_l)\frac{d\epsilon_p}{\hbar}. \quad (9)$$

Here $k_\perp$ is the component of $\mathbf{k}$ perpendicular to the applied electric field and $I(\epsilon_l)$ is the relative intensity of the electromagnetic radiation as function of photon energy. We discuss only the transition rate since $\Delta T/T=\Delta\alpha/\alpha$ for any photon energy.

The transition rate given in Eq. (9) is a sum of contributions of discrete values of ϵ_p and therefore for discrete values of photon energy. The separation between the absorption lines is $\hbar\omega_0$ which is exactly equal to the magnitude of the Stark splitting predicted by Callaway.[3,4] In fact, this result and the Stark splitting are essentially the same phenomenon observed from two different points of view.

We now evaluate the coefficients c_n. According to Eq. (8)

$$c_n=\frac{4\pi}{\omega_0}\int_0^{2\pi/\omega_0}\exp\left(i\int^t\frac{\epsilon}{\hbar}d\tau-i\frac{\bar{\epsilon}}{\hbar}t-in\omega_0 t\right)dt.$$

Using the relation $\bar{\epsilon}+n\hbar\omega_0-\epsilon_p=0$ implied by Eq. (9) we get

$$c_n=\frac{4\pi}{\omega_0}\int_0^{2\pi/\omega_0}\exp\left(i\int^t\frac{\epsilon}{\hbar}d\tau-i\frac{\epsilon_p}{\hbar}t\right)dt. \tag{10}$$

We now define a continuous function

$$P(k_\perp;\mathbf{k}_s;\epsilon_p)=\frac{4\pi}{\omega_0}\int_0^{2\pi/\omega_0}\exp\left(i\int^t\frac{\epsilon}{\hbar}d\tau-i\frac{\epsilon_p}{\hbar}t\right)dt. \tag{11}$$

Thus the transition rate for given $k_\perp$, $\mathbf{k}_s$ is given by

$$\bar{T}=\int_{-\infty}^{\infty}\bigg[|P(k_\perp;\mathbf{k}_s;\epsilon_p)|^2 \times|g_i|^2\frac{2\pi}{\hbar^4}\sum_{n=-\infty}^{\infty}\delta\left(\frac{\bar{\epsilon}}{\hbar}+n\omega_0-\frac{\epsilon_p}{\hbar}\right)\bigg]\frac{d\epsilon_p}{\hbar}. \tag{12}$$

We approximate $\epsilon(\mathbf{k})$ by a sinusoidal function which preserves the positions of the minima, the value of ϵ at the minimum and its curvature there; thus we obtain

$$\epsilon_{k_\perp;\mathbf{k}_s}(k_{\shortparallel})\cong\epsilon_g(k_\perp;\mathbf{k}_s)+\frac{\hbar^2}{\lambda^2\bar{m}}[\sin(\lambda k_{\shortparallel}+c)+1]. \tag{13}$$

Here $\epsilon_g(k_\perp;\mathbf{k}_s)$ is the minimum value of $\epsilon_{k_\perp;\mathbf{k}_s}(k_{\shortparallel})$; $k_{\shortparallel}$ is the component of $\mathbf{k}$ parallel to the applied field; and $\bar{m}$ is an effective mass defined by

$$\text{(a)}\quad \frac{1}{\bar{m}}=\frac{1}{m_e^*}+\frac{1}{m_h^*},$$

$$\text{(b)}\quad \frac{1}{m_e^*}=\frac{1}{\hbar^2}\frac{\partial^2}{\partial k_{\shortparallel}^2}\epsilon_c(\mathbf{k}), \tag{14}$$

$$\text{(c)}\quad -\frac{1}{m_h^*}=\frac{1}{\hbar^2}\frac{\partial^2}{\partial k_{\shortparallel}^2}\epsilon_v(\mathbf{k}).$$

Note that Eqs. (14) are not restricted to any particular direction of the applied field. Using the approximation Eq. (13) for ϵ, it is easily seen that when ν is an integer

$$|P(k_\perp;\mathbf{k}_s;\epsilon_p)|=J_\nu(\chi)$$

$$\nu=\frac{\epsilon_g(k_\perp;\mathbf{k}_s)+(\hbar^2/\bar{m}\lambda^2)-\epsilon_p}{\hbar\omega_0};\quad \chi=\frac{\hbar}{\bar{m}\lambda^2\omega_0}. \tag{15}$$

Here $J_\nu(\chi)$ is the Bessel function of order ν. In most semiconductors and insulators and for electric fields of the order of 10^6 V/cm or less, it is found that ν and χ are large numbers. In such a case the Bessel function is a smoothly varying function and thus we can interpolate $P(k_\perp;\mathbf{k}_s;\epsilon_p)$ by $J_\nu(\chi)$ also for nonintegral values of ν. For excitation energies close to the minimum energy gap we find that $|\nu-\chi|\ll\nu;\chi$. In this case the following approximations are valid[9]:

$$J_\nu(\chi)\simeq\frac{\exp[-2^{3/2}(\nu-\chi)^{3/2}/3\chi^{1/2}]}{[2\pi\sqrt{2}(\nu-\chi)^{1/2}\chi^{1/2}]^{1/2}}\quad\text{for}\quad\chi<\nu,$$

$$J_\nu(\chi)\simeq\frac{\sin(\pi/3)\Gamma(1/3)}{3\pi(\chi/6)^{1/3}}+\frac{\sin(2\pi/3)\Gamma(2/3)}{3\pi(\chi/6)^{2/3}}(\chi-\nu)\quad\text{for}\quad\chi\simeq\nu, \tag{16}$$

$$J_\nu(\chi)=\left(\frac{2}{\pi 2^{1/2}\chi^{1/2}(\chi-\nu)^{1/2}}\right)^{1/2}\cos\left(-\frac{\pi}{4}+\frac{2^{3/2}(\chi-\nu)^{3/2}}{3\chi^{1/2}}\right)\quad\text{for}\quad\chi>\nu.$$

To obtain the transition rate for direct transitions for given $k_\perp$, the same steps are to be followed. It is readily seen that changes will enter only in the value of g_i and in the excitation energy ϵ_p, which will now be simply the photon energy alone. Thus for direct transitions we have

$$\bar{T}_d(k_\perp)=\int_0^{\infty}\left[|g_d|^2J_\nu^2(\chi)\frac{2\pi}{\hbar^4}\sum_{n=-\infty}^{\infty}\delta\left(\frac{\bar{\epsilon}}{\hbar}+n\omega_0-\frac{\epsilon_l}{\hbar}\right)\right]\times I(\epsilon_l)\frac{d\epsilon_l}{\hbar}, \tag{17}$$

and for indirect transitions we have

$$\bar{T}_i(k_\perp;\mathbf{k}_s)=\int_0^{\infty}\left[|g_i|^2J_\nu^2(\chi)\frac{2\pi}{\hbar^4}\sum_{n=-\infty}^{\infty}\delta\left(\frac{\bar{\epsilon}}{\hbar}+n\omega_0-\frac{\epsilon_p}{\hbar}\right)\right]\times I(\epsilon_l)\frac{d\epsilon_p}{\hbar}. \tag{18}$$

Two extreme cases can now be considered. If $I(\epsilon_l)$ is a square pulse of unit height and of a width $\Delta\epsilon_l$, then

case a: $\Delta\epsilon_l\gg\hbar\omega_0$,
case b: $\Delta\epsilon_l\ll\hbar\omega_0$.

It is clear that Stark splitting will show up only in case b.

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[9] This approximation is not valid in the direction of the applied field in which minima of values very close to ϵ_g are present within the basic period of ϵ.

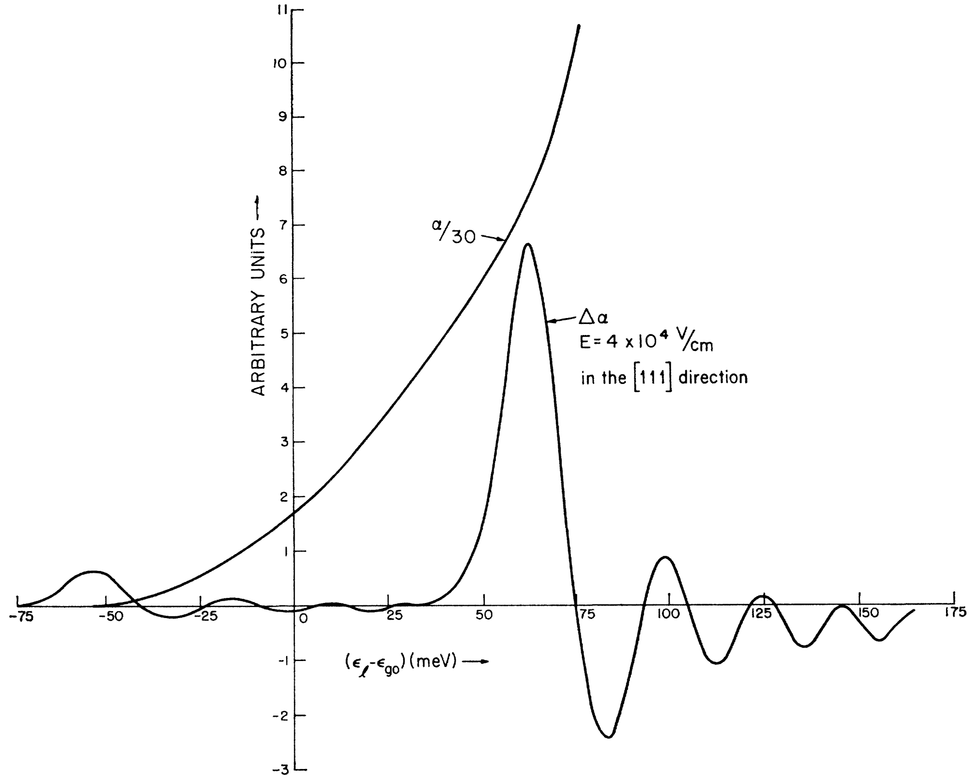

FIG. 1. Transition rate and change in transition rate for Si for an applied field of 40 kV/cm in the [111] direction.

Case a: From Eqs. (17) and (18) we obtain

$$\bar{T}_d = |g_d|^2 J_\nu^2(\chi) \frac{2\pi}{\hbar^4} \frac{\Delta\epsilon_l}{\hbar\omega_0}, \tag{19}$$

$$\bar{T}_i = |g_i|^2 J_\nu^2(\chi) \frac{2\pi}{\hbar^4} \frac{\Delta\epsilon_l}{\hbar\omega_0}. \tag{20}$$

The over-all transition rate in the direct transition case is given by

$$T_d = \textstyle\sum_{k_\perp} \bar{T}_d(k_\perp). \tag{21}$$

Since the dependence on $k_\perp$ enters only through $\epsilon_g(k_\perp)$ the transition rate can be expressed in the form

$$T_d = \int_{\epsilon_{g0}}^{\infty} \bar{T}_d(\epsilon_g) D_d(\epsilon_g) d\epsilon_g, \tag{22}$$

where $D_d(\epsilon_g)$ is an appropriate density of states function which can be shown to be given by

$$D_d(\epsilon_g) = C_d. \tag{23}$$

Here C_d is a number independent of ϵ_g and ϵ_{g0} is the minimum energy gap.

In the case of indirect transitions

$$T_i = \sum_{k_\perp;\, \mathbf{k}_s} N(\epsilon_s) \bar{T}_i(k_\perp; \mathbf{k}_s),$$

where $N(\epsilon_s) = (e^{\hbar\omega_s/k\theta} - 1)^{-1}$ for phonon absorption and $N(\epsilon_s) = (e^{\hbar\omega_s/k\theta} - 1)^{-1} + 1$ for phonon emission. Here θ is the absolute temperature. Since in the case of excitations close to the minimum energy gap the range of variation of ϵ_s is relatively small thus we can replace $N(\epsilon_s)$ by $N(\epsilon_{s0})$ where ϵ_{s0} is the energy of the phonon whose $\mathbf{k}_s$ vector is the difference between the k values of the extrema of the bands.

Since $\bar{T}_i$ given in Eq. (20) depends on $k_\perp$ and $\mathbf{k}_s$ only through $\epsilon_g(k_\perp; \mathbf{k}_s)$ we may write

$$T_i = N(\epsilon_{s0}) \int_{\epsilon_{g0}}^{\infty} \bar{T}_i(\epsilon_g) D_i(\epsilon_g) d\epsilon_g, \tag{24}$$

$$D_i(\epsilon_g) = C_i(\epsilon_g - \epsilon_{g0})^{3/2}. \tag{25}$$

By taking the case $E \to 0$ we obtain the transition rate for the case $E = 0$.

$$\begin{aligned} T_d &= M_d \int_{\epsilon_{g0}}^{\infty} J_\nu^2(\chi) \frac{\Delta\epsilon_l}{\hbar\omega_0} d\epsilon_g, \\ T_d(E=0) &= M_d \frac{2^{1/2}(\epsilon_l - \epsilon_{g0})^{1/2}}{\pi(\hbar^2/\bar{m}\lambda^2)^{1/2}} \Delta\epsilon_l. \end{aligned} \tag{26}$$

$$\begin{aligned} T_i &= M_i \int_{\epsilon_{g0}}^{\infty} J_\nu^2(\chi) \frac{\Delta\epsilon_l}{\hbar\omega_0} (\epsilon_g - \epsilon_{g0})^{3/2} d\epsilon_g, \\ T_i(E=0) &= M_i \frac{\Gamma(5/2)\Gamma(1/2)/\Gamma(3)}{\pi 2^{1/2}(\hbar^2/\bar{m}\lambda^2)^{1/2}} \Delta\epsilon_l (\epsilon_p - \epsilon_{g0})^2. \end{aligned} \tag{27}$$

M_i and M_d are factors independent of the electric field,

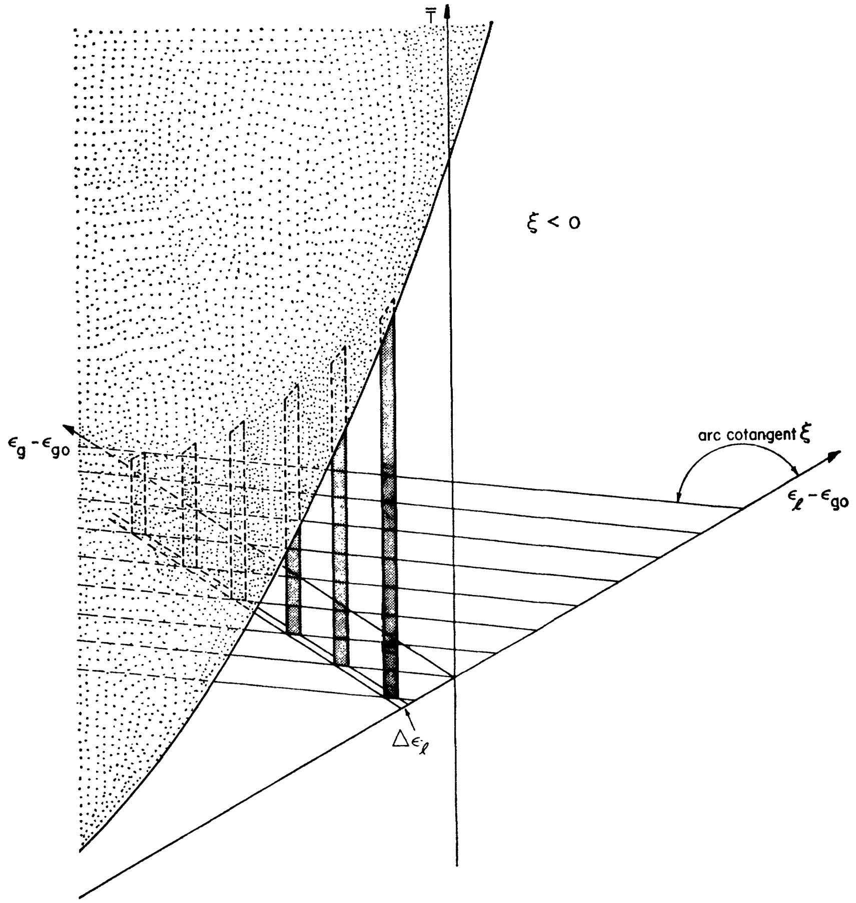

FIG. 2. Evaluation of the transition-rate fine structure for $\xi < 0$.

and of the difference between excitation energy and the minimum energy gap; thus they will not be given explicitly.

The expression for T_d is equivalent to the result obtained by Callaway[4] and Tharmalingam[5] though they look different in form. The difference arises from the difference in the approximation for the shape of the bands. Similarly, Penchina's results[7] for the indirect case are equivalent to those presented above. However, Eqs. (26), (27), and (16) allow us to obtain some simple results which might be of use for experimental considerations.

From Eq. (16) it is evident that for all three ranges

$$\nu < \chi; \quad \nu \simeq \chi; \quad \text{and} \quad \nu > \chi$$

$$\frac{1}{\omega_0} J_\nu^2(\chi) = E^{-1/3} F\left(\frac{\epsilon_g - \epsilon_p}{E^{2/3}}\right).$$

Using this result in Eqs. (26) and (27), we obtain

$$T_d = E^{1/3} F_d\left(\frac{\epsilon_{g0} - \epsilon_l}{E^{2/3}}\right), \tag{28}$$

$$T_i = E^{4/3} F_i\left(\frac{\epsilon_{g0} - \epsilon_p}{E^{2/3}}\right). \tag{29}$$

These expressions are valid continuously from below to above the absorption edge, within the limits of accuracy of the approximation in Eq. (16). From Eqs. (26) and (27) for $E = 0$, we see that

$$T_d(E=0) \propto (\epsilon_l - \epsilon_{g0})^{1/2} = E^{1/3}(\epsilon_l - \epsilon_{g0}/E^{2/3})^{1/2},$$

$$T_i(E=0) \propto (\epsilon_p - \epsilon_{g0})^2 = E^{4/3}(\epsilon_p - \epsilon_{g0}/E^{2/3})^2.$$

Correspondingly, we obtain

$$\Delta T_d = E^{1/3} \bar{F}_d(\epsilon_{g0} - \epsilon_l/E^{2/3}), \tag{30}$$

$$\Delta T_i = E^{4/3} \bar{F}_i(\epsilon_{g0} - \epsilon_p/E^{2/3}). \tag{31}$$

A special case of particular interest is silicon with an applied field in the [111] direction, because much experimental work has been done on this material under this condition.[6,8] Moreover, the experiments actually show great sensitivity at excitation energies considerably above the absorption edge. Therefore we present here a calculation which is valid considerably above the absorption edge, and for field applied in the [111] direction.

The results shown in Fig. 1 are for transitions from the heavy hole valence band to the conduction band, with an applied field of 40 kV/cm and for the transverse optical phonon. It should be noted that the calculation

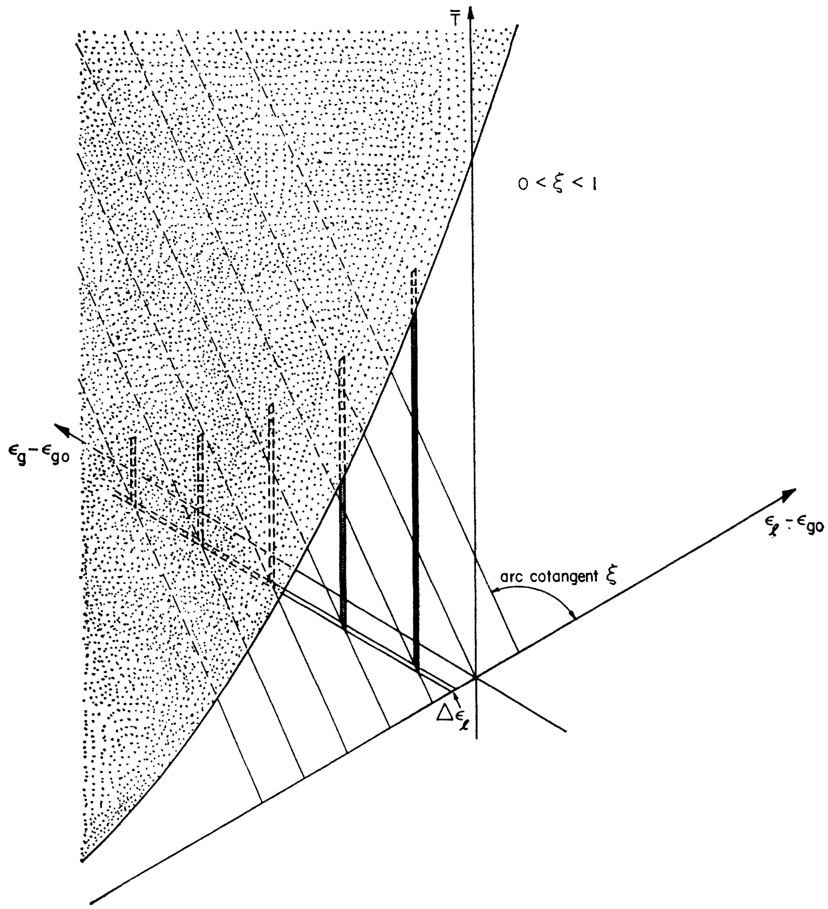

FIG. 3. Evaluation of the transition-rate fine structure for $0<\xi<1$.

of ΔT_i considerably above the absorption edge has to be very accurate, because $\Delta T_i/T_i$ gets to be about 5%. The calculation of T_i presented here is accurate to $\frac{1}{3}$%. The computation was done by machine and thus other cases may easily be computed.

Several points should be emphasized: As in the case of direct transitions (as shown by Callaway[4]) the indirect transitions also lead to several smaller peaks in addition to the large first one. The position of these peaks shifts as seen from Eq. (31) according to the rule $(\epsilon_l-\epsilon_{g0})\propto E^{2/3}$. The height of all these peaks varies as $E^{4/3}$ which is confirmed by the $E^{4/3}$ dependence of the first peak given by Penchina.[7] The width of all the peaks, however, is shown here to increase as $E^{2/3}$ meaning that smaller electric field will allow better resolution between different processes. It should be pointed out that the presence of internal electric fields like the ones calculated by Redfield[10] will tend to destroy the $E^{4/3}$ dependence at low applied fields and will also tend to smooth out the secondary peaks since their position depends on the electric field

Case b: Here $I(\epsilon_l/\hbar)$ in Eq. (17) is of unit height for a bandwidth $\Delta\epsilon_l$ such that $\Delta\epsilon_l\ll\hbar\omega_0$. In this section we discuss the fine structure of the transition rate; since this structure is more pronounced for direct transitions than for indirect ones we shall concentrate mainly on the direct transitions.

From Eq. (17) we see that contributions to the transition rate are made only when $\bar{\epsilon}/\hbar+n\omega_0-\epsilon_l/\hbar=0$, where

$$\bar{\epsilon}=\frac{\lambda}{2\pi}\int_0^{2\pi/\lambda}(\epsilon_c-\epsilon_v)dk_{\parallel}\,, \tag{32}$$

and ϵ_c and ϵ_v are the true, not the approximated functions of $k_{\parallel}$. The dependence of $\bar{\epsilon}$ on $k_\perp$ is not directly related to the dependence of ϵ_g on $k_\perp$. In fact, in some cases it does not have to have an extremum for the same $k_\perp$ for which ϵ_g has its minimum. However, for simplicity, we shall restrict ourselves here to the case where $\bar{\epsilon}$ has an extremum, i.e., $\partial\bar{\epsilon}/\partial k_\perp=0$ for any direction of $k_\perp$ at the same point that ϵ_g has its minimum.

In this case it is seen that $\bar{\epsilon}$ can be expressed around the extremum in the form

$$\bar{\epsilon}=\bar{\epsilon}_0+\xi(\varphi)(\epsilon_g-\epsilon_{g0})\,, \tag{33}$$

in which φ is the angle of $k_\perp$ measured with respect to an arbitrary reference on a plane perpendicular to the direction of the applied field.

In principle, ξ can be positive or negative, and its absolute value may be larger or smaller than 1 depend-

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[10] D. Redfield, Phys. Rev. **130**, 916 (1963).

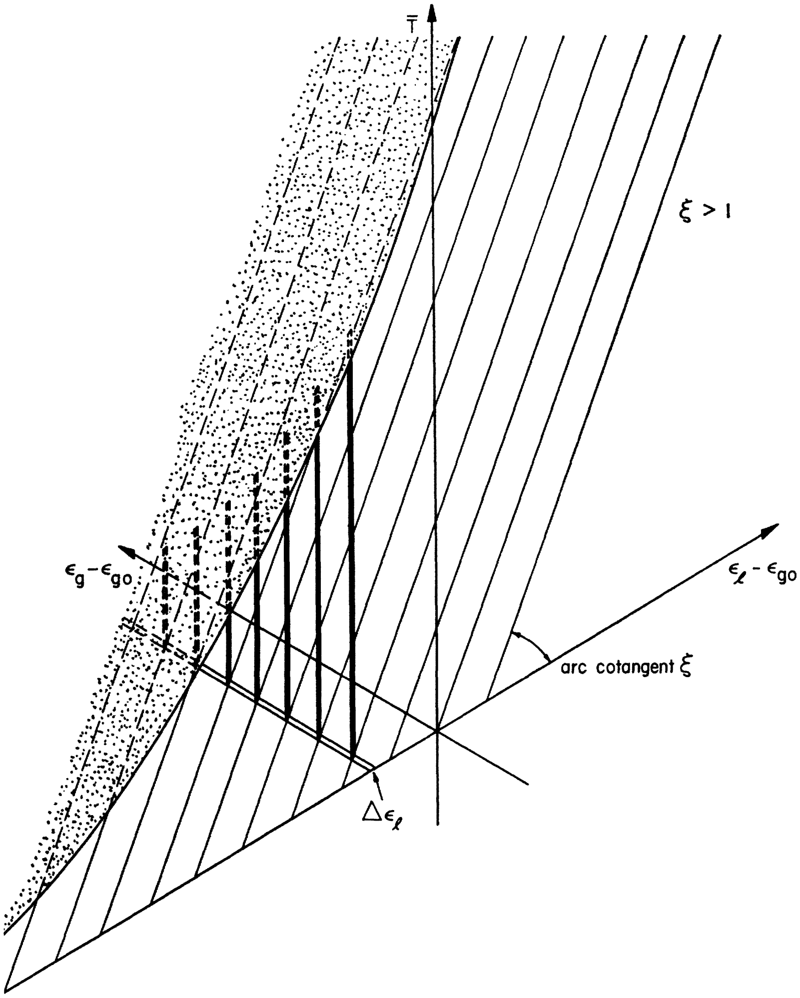

FIG. 4. Evaluation of the transition-rate fine structure for $\xi > 1$.

ing on the details of the band structure. The values of ϵ_g for which transitions are allowed for a given value of ϵ_l are

$$\epsilon_g = \epsilon_{g0} + \frac{\epsilon_l - \bar{\epsilon}_0}{\xi(\varphi)} - \frac{n\hbar\omega_0}{\xi(\varphi)}. \tag{34}$$

The density of states is zero for $\epsilon_g < \epsilon_{g0}$. Thus for $\xi > 0$ n has a maximum, $n_{\max} = -m_1$; for $\xi < 0$ n has a minimum $n_{\min} = -m_2$; m_1 and m_2 depend on ϵ_l but are independent of $|\xi(\varphi)|$.

From Eqs. (17) and (21) and using the fact that $J_\nu(\chi)$ depends on $k_\perp$ only through ϵ_g, we obtain

$$T_d = \frac{2\pi}{\hbar^4} \int_{\varphi=\varphi_1}^{\varphi_2} \left\{ \sum_{n=-\infty}^{-m_1} J_\nu^2(\chi) |g_d|^2 \int_{\epsilon_{g0}}^{\infty} \int_0^{\infty} \left[\delta\left(\frac{\bar{\epsilon}}{\hbar} + n\omega_0 - \frac{\epsilon_l}{\hbar} \right) I(\epsilon_l) \right] \frac{d\epsilon_l}{\hbar} d\epsilon_g Q(\varphi) \right\} d\varphi$$
$$+ \frac{2\pi}{\hbar^4} \int_{\varphi=\varphi_2}^{\varphi_1} \left\{ \sum_{n=-m_2}^{\infty} J_\nu^2(\chi) |g_d|^2 \int_{\epsilon_{g0}}^{\infty} \int_0^{\infty} \left[\delta\left(\frac{\bar{\epsilon}}{\hbar} + n\omega_0 - \frac{\epsilon_l}{\hbar} \right) I(\epsilon_l) \right] \frac{d\epsilon_l}{\hbar} d\epsilon_g Q(\varphi) d\varphi, \tag{35}$$

where $Q(\varphi)d\epsilon_g d\varphi$ is the density of states as a function of φ, which is independent of ϵ_g since this is the direct transition case. The integration over φ is separated into regions in which ξ is either only positive or only negative. Integrating over ϵ_g first and then over ϵ_l we obtain

$$T_d = \frac{2\pi}{\hbar^4} \Delta\epsilon_l \int_{\varphi_1}^{\varphi_2} \sum_{n=-\infty}^{-m_1} J_\nu^2(\chi) |g_d|^2 \frac{Q(\varphi)}{\xi(\varphi)} d\varphi + \frac{2\pi}{\hbar^4} \Delta\epsilon_l \int_{\varphi_2}^{\varphi_1} \sum_{n=-m_2}^{\infty} J_\nu^2(\chi) |g_d|^2 \frac{Q(\varphi)}{\xi(\varphi)} d\varphi; \tag{36}$$

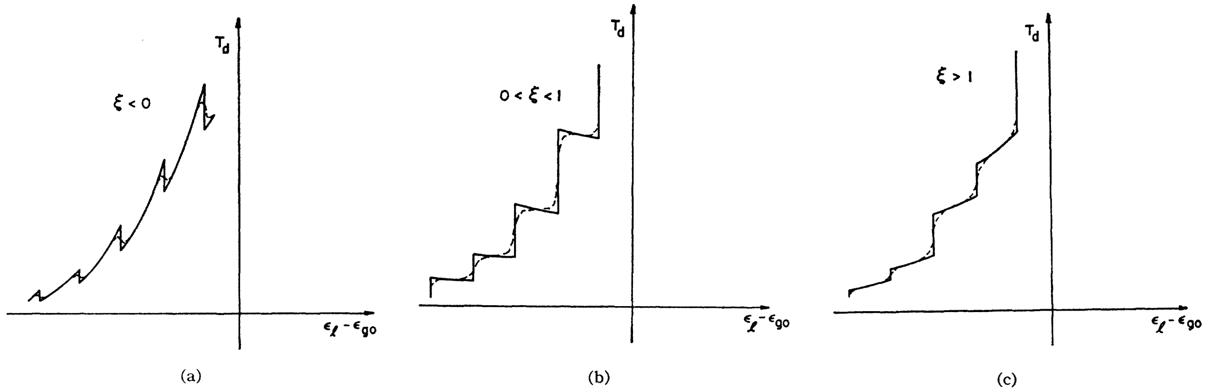

FIG. 5. Form of fine structure in the transition rate (a) $\xi<0$ (b) $0<\xi<1$ (c) $\xi>1$.

here

$$\nu=\frac{\epsilon_{g0}-\epsilon_l+(\epsilon_l-\bar{\epsilon}_0/\xi(\varphi))+(\hbar^2/\bar{m}\lambda^2)}{\hbar\omega_0}-\frac{n}{\xi}.$$

To obtain a more visual notion of the kind of results which may be obtained we use Figs. 2, 3, 4. In these figures we plot $J_\nu^2(\chi)$ as a function of $\epsilon_g-\epsilon_{g0}$ and $\epsilon_l-\epsilon_{g0}$. From the form of $J_\nu^2(\chi)$ it is clear that the lines for which $J_\nu^2(\chi)$ is constant are $(\epsilon_g-\epsilon_{g0})-(\epsilon_l-\epsilon_{g0})=$ constant. The points at which contributions to the transition rate are made for given ϵ_l are given by the intercepts of the lines $(\epsilon_l-\epsilon_{g0})=$ constant and the lines

$$(\epsilon_l-\epsilon_{g0})=\bar{\epsilon}_0-\epsilon_{g0}+\xi(\varphi)(\epsilon_g-\epsilon_{g0})+n\hbar\omega_0.$$

Thus the transition rate for a given value of φ is the sum of the shaded columns. We now consider three cases. Case 1: $\xi<0$, shown in Fig. 2. This results in a transition rate below the absorption edge of the form shown in Fig. 5(a). Note that as ϵ_l increases T_d on the average increases, but the steps are negative whereas in between the step T_d increases. Case 2: $0<\xi<1$; shown in Fig. 3. This results in a transition rate as shown in Fig. 5(b). Here the steps are positive and T_d also decreases between the steps. Case 3: $1<\xi$; shown in Fig. 4 which gives a result as shown in Fig. 5(c). The only case discussed by Callaway[4] corresponds to $\xi=1$.

When integrating over φ we note that $\bar{\epsilon}_0$ is independent of φ. Thus the position of the steps is unaltered, and the overall value of T_d will resemble one of the forms shown in Fig. 5.

In conclusion, we should make a few remarks on the problem of actually observing the fine structure. In addition to the high resolution and low temperatures necessary there is an additional difficulty. As seen from Eq. (36) and the discussion that follows, the positions of the steps are given by

$$\epsilon_l=\bar{\epsilon}_0-n\hbar\omega_0.$$

For ϵ_l in the vicinity of the energy gap, $\bar{\epsilon}_0-\epsilon_l$ is of the order of electron volts whereas $\hbar\omega_0$ for fields of 10^5 V/cm is of the order of millielectron volts. Thus n will usually be of the order of 100 or more. Accordingly, in order to observe the steps the electric field should be uniform to better than 1%. Moreover, to obtain large values of $\hbar\omega_0$ the electric field should be applied in the direction of small periodicity of the reciprocal lattice. However, if the field is applied slightly off such a direction the value of $\bar{\epsilon}$ will be changing. It can be shown that if the variation of $\bar{\epsilon}$ within a period is small compared to $\hbar\omega_0$ this will result in only small deformation of the steps as shown in the dotted curves of Fig. 5 which will become worse as the variation of $\bar{\epsilon}$ increases. In conclusion, in order to observe the fine structure, the applied electric field should be much larger than the existing internal field and it should be very uniform and well oriented.

ACKNOWLEDGMENTS

I wish to thank Professor R. B. Adler for the supervision of this work. I am deeply indebted to Professor G. W. Pratt for critically reading the work. Thanks also go to E. L. A. Courtens for a helpful suggestion in the machine computation of the Bessel function and to Dr. L. Gruenberg for a helpful discussion.

FIG. 2. Evaluation of the transition-rate fine structure for $\xi<0$.