Interband Dielectric Properties of Solids in an Electric Field*

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A general expression for the imaginary part of the one-electron interband dielectric function of a solid in the presence of an electric field is derived. The result is valid for all regions of **k** space and explicitly takes into account the variation of the dipole matrix element and effective mass throughout the Brillouin zone. Under certain approximations the finite-electric-field dielectric function reduces to the convolution of the zero-field dielectric function with an Airy function. This result can be used in conjunction with band-structure calculations which have already been done in order to predict the line shape of the electroreflectance spectra as a function of electric field. The convolution expression reduces further at nondegenerate critical points, and the forms for the four types of critical points are presented. In certain instances it is possible to unfold the convolution integral and obtain the zero-field dielectric function from either the finite-field dielectric constant or the electroreflectance data at isolated critical points.

I. INTRODUCTION

THE recent development of modulation techniques[1–7] in the study of energy bands has succeeded in providing much needed information for the starting points of band-structure calculations and has given further impetus to the study of band structure. These modulation techniques have involved the application of periodic perturbations such as electric field,[1–3] strain,[4] or heat[7] to the solid and have used phase sensitive detection methods to measure the periodic modulation of the optical properties of solids.

The power of these methods lies in the fact that modulation techniques enhance the signals at certain points in the band structure: the critical points or the van Hove singularities where the relative interband gradient of the energy vanishes at some point of **k** space. By measuring the piezo-optic, thermo-optic, or electro-optic spectrum, it is possible in principle to obtain the energy and position in **k** space of the critical points of the energy band spectrum.

Knowing the effect of an electric field on the band structure of a solid is a necessary prerequisite for the interpretation of experimental electro-optic results, not only to determine the critical points but also to determine the validity of the one-electron approximation for interband transitions. Following the initial work of Franz[8] and Keldysh,[9] many calculations of the effect of an electric field on the real and imaginary parts of the dielectric properties have appeared in the literature.[10–15] With few exceptions, these calculations have been done in the framework of the weak field, effective mass approximation. In every case, the results have been restricted to regions of **k** space where the energy bands are simple functions of **k**. It is the purpose of this paper to derive a general formula for the imaginary part of the interband dielectric constant $\epsilon_2(\omega,\mathcal{E})$ for a solid in an electric field which does not require the assumption of quadratic energy surfaces about particular points in **k** space. The expression derived allows direct calculation of $\epsilon_2(\omega,\mathcal{E})$ for a solid directly from the theoretical band structures calculated by pseudopotential,[16–18] $\mathbf{k}\cdot\mathbf{p}$,[19] or other methods.[20] This general formula is derived in Sec. II.

In Sec. III, it will be shown that for regions of **k** space having quadratic energy surfaces there is a very simple convolution integral relating the finite-field dielectric constant to the zero-field dielectric constant

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through an Airy function. This convolution integral form will be shown to be valid for indirect transitions as well as direct transitions near critical points. Using the fact that the electric field mixes all of the states in $\mathbf{k}$ space, it will be shown that in principle it is possible to recover the zero-field dielectric function from differential electro-optical data. Also, the above-mentioned convolution integral form for the imaginary part of the dielectric constant in an electric field is used with the Kramers-Kronig relations to calculate the real part of the dielectric constant.

Appendix A is a proof used in Sec. II and Appendix B is an independent derivation of the results of Sec. III.

II. CONVOLUTION INTEGRAL FORMULATION OF ELECTRIC-FIELD EFFECT

In this section a convolution integral representing the effect of an electric field on ϵ_2, the imaginary part of the dielectric constant, in terms of the zero-field parameters will be derived. The derivation of Callaway,[10] which is based on the work of Argyres[21] and Kane,[22] will be followed; $\epsilon_2(\omega,\mathcal{E})$ will be obtained by calculating the interaction of an electromagnetic wave with the crystal. A similar derivation has also been given by Yacoby, for both direct and indirect transitions at an M_0 edge.[14]

We consider first the absorption of light by a solid in the absence of an electric field, using the semiclassical approximation where the perturbation introduced by the photon is represented by the vector potential

$$\mathbf{A}=\hat{e}Ae^{i\mathbf{k}\cdot\mathbf{r}-i\omega t}. \tag{2.1}$$

The one-electron states of the solid are assumed nondegenerate and represented by Bloch functions which satisfy the equation

$$\left[-\frac{\hbar^2}{2m}\nabla^2+V(\mathbf{r})\right]\psi_n(\mathbf{k},\mathbf{r})=E_n(\mathbf{k})\psi_n(\mathbf{k},\mathbf{r}), \tag{2.2}$$

where the potential $V(\mathbf{r})$ is periodic in the lattice. $\mathbf{k}$ and n denote the wave vector and band index, respectively, with $\mathbf{k}$ being restricted to the first Brillouin zone. The Bloch functions are normalized to the volume V as

$$\int_V d^3r\,\psi_{n'}{}^*(\mathbf{k}',\mathbf{r})\psi_n(\mathbf{k},\mathbf{r})=\delta_{n'n}\delta_{\mathbf{k}'\mathbf{k}} \tag{2.3}$$

and can serve as a basis for expansion of a function over the volume.

By time-dependent perturbation theory, the transition rate from the filled discrete state represented by the Bloch function $\psi_n(\mathbf{k},\mathbf{r})$ to the empty discrete state $\psi_{n'}(\mathbf{k}',\mathbf{r})$ is given by[23]

$$w(\mathbf{k}'n';\mathbf{k},n;t)=\frac{1}{t}|a(\mathbf{k}',n';\mathbf{k},n;t)|^2=\frac{4}{\hbar^2}|H'_{\mathbf{k}',n';\mathbf{k},n}|^2 \times\frac{\sin^2\tfrac{1}{2}(\omega_{n'}-\omega_n-\omega)t}{t(\omega_{n'}-\omega_n-\omega)^2}, \tag{2.4}$$

where the matrix element $H'_{\mathbf{k}',n';\mathbf{k},n}$ in the dipole approximation is given by

$$H'_{\mathbf{k}',n';\mathbf{k},n}=\langle\psi_{n'}(\mathbf{k}',\mathbf{r})|\frac{ieh A}{mc}\hat{e}\cdot\nabla|\psi_n(\mathbf{k},\mathbf{r})\rangle, \tag{2.5}$$

and $\hbar\omega_n=E_n$, $\hbar\omega_{n'}=E_{n'}$. Since $\psi_{n'}(\mathbf{k}',\mathbf{r})$ and $\psi_n(\mathbf{k},\mathbf{r})$ are Bloch functions,

$$\langle\psi_{n'}(\mathbf{k}',\mathbf{r})|\nabla|\psi_n(\mathbf{k},\mathbf{r})\rangle=\frac{i}{\hbar}\mathbf{P}_{n'n}(\mathbf{k})\delta_{\mathbf{k}'\mathbf{k}}, \tag{2.6}$$

which can be taken as the equation defining $\mathbf{P}_{n'n}(\mathbf{k})$. The transition rate per unit time from the filled states $\psi_n(\mathbf{k},\mathbf{r})$ to the empty states $\psi_{n'}(\mathbf{k}',\mathbf{r})$ is therefore

$$w(\mathbf{k}',n';\mathbf{k},n;t)=\frac{4e^2A^2}{m^2c^2\hbar^2}|\hat{e}\cdot\mathbf{P}_{n'n}(\mathbf{k})|^2\delta_{\mathbf{k}'\mathbf{k}} \times\frac{\sin^2\tfrac{1}{2}(\omega_{n'}-\omega_n-\omega)t}{t(\omega_{n'}-\omega_n-\omega)^2}. \tag{2.7}$$

In the limit of large t

$$\lim_{t\to\infty}\frac{\sin^2\tfrac{1}{2}\omega t}{\omega^2 t}=\tfrac{1}{2}\pi\delta(\omega), \tag{2.8}$$

and the total transition rate over the entire solid is obtained by summing over all filled and empty states, giving

$$w_{\text{tot}}=\frac{2\pi e^2A^2}{m^2c^2\hbar}\sum_{\mathbf{k},n',n}|\hat{e}\cdot\mathbf{P}_{n'n}(\mathbf{k})|^2\delta[E_{n'n}(\mathbf{k})-\hbar\omega], \tag{2.9}$$

where the sum n' ranges over empty bands and n over filled bands, and $E_{n'n}(\mathbf{k})=E_{n'}(\mathbf{k})-E_n(\mathbf{k})$. To obtain the number of transitions per unit volume per unit time we write

$$\sum_{\mathbf{k}}\rightarrow\frac{V}{(2\pi)^3}\int_{\text{B.Z.}}d^3k, \tag{2.10}$$

where the integral is taken over the first Brillouin zone. Including the factor of 2 for spin degeneracy gives the desired rate per unit volume per unit time as

$$w=\frac{2\pi e^2A^2}{m^2c^2\hbar}\sum_{n',n}\frac{2}{(2\pi)^3} \times\int_{\text{B.Z.}}d^3k|\hat{e}\cdot\mathbf{P}_{n'n}(\mathbf{k})|^2\delta[E_{n'n}(\mathbf{k})-\hbar\omega]. \tag{2.11}$$

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Since the imaginary part of the dielectric constant is

$$\epsilon_2(\omega)=\frac{nc}{\omega}\alpha=\frac{2\pi c^2\hbar}{\omega^2 A^2}w, \tag{2.12}$$

Eq. (2.11) gives

$$\epsilon_2(\omega)=\frac{4\pi^2e^2}{m^2\omega^2}\sum_{n',n}\int_{\text{B.Z.}}\frac{d^3k}{(2\pi)^3}\times|\hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k})|^2\delta[E_{n'n}(\mathbf{k})-\hbar\omega], \tag{2.13}$$

which is the usual result for ϵ_2 in the absence of an electric field.[17] The sums n' and n are over empty and filled bands, respectively. Equation (2.12) supposes a low rate of loss of incident energy with distance (small α), but a rigorous derivation of ϵ_2 by means of the current operator also yields Eq. (2.13).[24]

In the presence of an electric field $e\mathcal{E}=\mathbf{F}$, Eq. (2.2) becomes

$$\left[-\frac{\hbar^2}{2m}\nabla^2+V(\mathbf{r})-Fx\right]\phi_{\nu,n}(\mathbf{k}_\perp,\mathbf{r})=W_{\nu,n}(\mathbf{k}_\perp)\phi_{\nu,n}(\mathbf{k}_\perp,\mathbf{r}), \tag{2.14}$$

where the direction of the electric field is chosen to be the x direction. It is assumed that this direction coincides with one of the (infinite) vectors of the reciprocal lattice.[21] If this is done, the Brillouin zone can be chosen so that the symmetry perpendicular to the field is retained, and the eigenfunctions $\phi_{\nu,n}(\mathbf{k}_\perp,\mathbf{r})$ can be expanded in terms of the Bloch functions of the unperturbed Hamiltonian as[21]

$$\phi_{\nu,n}(\mathbf{k}_\perp,\mathbf{r})=\sum_{k_x}A_{\nu,n}(\mathbf{k})\psi_n(\mathbf{k},\mathbf{r}) \tag{2.15}$$

with normalization.

$$\int_V d^3r\phi_{\nu',n'}{}^*(\mathbf{k}_\perp{}',\mathbf{r})\phi_{\nu,n}(\mathbf{k}_\perp,\mathbf{r})=\delta_{\nu\nu'}\delta_{nn'}\delta_{\mathbf{k}_\perp\mathbf{k}_\perp'}. \tag{2.16}$$

Substitution of Eq. (2.15) into Eq. (2.14) gives the equation of the coefficients $A_{\nu,n}(\mathbf{k})$:

$$0=\sum_{k_x}\{A_{\nu,n}(\mathbf{k})[E_n(\mathbf{k})-W_{\nu,n}(\mathbf{k}_\perp)-Fx]\psi_n(\mathbf{k},\mathbf{r})\}, \tag{2.17}$$

and taking the inner product of this expression with $\psi_{n'}(\mathbf{k}',\mathbf{r})$ yields

$$0=\sum_{k_x}\{A_{\nu,n}(\mathbf{k})[E_n(\mathbf{k})-W_{\nu,n}(\mathbf{k}_\perp)]\delta_{n'n}\delta_{\mathbf{k}'\mathbf{k}}-A_{\nu,n}(\mathbf{k})F\langle\psi_{n'}(\mathbf{k}',\mathbf{r})|x|\psi_n(\mathbf{k},\mathbf{r})\rangle\}. \tag{2.18}$$

It is now necessary to consider the matrix element of x which appears in Eq. (2.18). Since the Bloch function can be written in the form

$$\psi_n(\mathbf{k},\mathbf{r})=e^{i\mathbf{k}\cdot\mathbf{r}}u_n(\mathbf{k},\mathbf{r}), \tag{2.19}$$

where $u_n(\mathbf{k},\mathbf{r})$ is cell-periodic, it follows that

$$\langle\psi_{n'}(\mathbf{k}',\mathbf{r})|x|\psi_n(\mathbf{k},\mathbf{r})\rangle=-i\delta_{n'n}\left(\frac{\partial}{\partial k_x}\delta_{\mathbf{k}'\mathbf{k}}\right)+i\delta_{\mathbf{k}'\mathbf{k}}X_{n'n}(\mathbf{k}), \tag{2.20}$$

where

$$X_{n'n}(\mathbf{k})=\int_V d^3r u_{n'}{}^*(\mathbf{k},\mathbf{r})\frac{\partial}{\partial k_x}u_n(\mathbf{k},\mathbf{r}). \tag{2.21}$$

The operation of the derivative on the Kronecker delta function has symbolic significance only and is a result of differentiating Eq. (2.19), where the wave vectors $\mathbf{k}$ form a quasicontinuum but are really discrete. However, if the volume V is large enough $\mathbf{k}$ may be considered continuous allowing interpretation of the derivative in the usual manner. The derivative of the Kronecker delta therefore is equivalent to the negative of the derivative of the function multiplying the Kronecker delta within the summation operation over the variable of differentiation. Therefore, Eq. (2.18) becomes

$$0=\sum_{k_x}\delta_{\mathbf{k}'\mathbf{k}}\left\{\delta_{n'n}\left[E_n(\mathbf{k})-W_{\nu,n}(\mathbf{k}_\perp)-iF\frac{\partial}{\partial k_x}\right]A_{\nu,n}(\mathbf{k})-iFX_{n'n}(\mathbf{k})A_{\nu,n}(\mathbf{k})\right\}, \tag{2.22}$$

and the sum over the vector $\mathbf{k}$ can be carried out immediately, eliminating the δ function of $\mathbf{k}$ and $\mathbf{k}'$.

We sum over the index n', obtaining independent equations for the coefficients $A_{\nu,n}(\mathbf{k})$:

$$0=\left[E_n(\mathbf{k})-iF\sum_{n'}X_{n'n}(\mathbf{k})-W_{\nu,n}(\mathbf{k}_\perp)-iF\frac{\partial}{\partial k_x}\right]A_{\nu,n}(\mathbf{k}). \tag{2.23}$$

The fact that the summation of $X_{n'n}(\mathbf{k})$ over the index n' results in a quantity dependent on n and $\mathbf{k}$ like the energy $E_n(\mathbf{k})$ suggests the definition of an effective energy

$$E_n'(\mathbf{k})=E_n(\mathbf{k})-iF\sum_{n'}X_{n'n}(\mathbf{k}), \tag{2.24}$$

which takes into account the interband terms $X_{n'n}(\mathbf{k})$ as well as the intraband polarization of the Bloch functions represented by $-F\partial/\partial k_x$. The interband terms represent electron transfer between bands,[22] which causes a decay in the wave-function amplitude of an initially filled state and an increase in that of an empty state. Therefore, the second term on the right of Eq. (2.24) is complex. [In fact, if the crystal has inversion symmetry, the $X_{n'n}(\mathbf{k})$ are purely real as is shown in Appendix A and thus the interband contributions to the energy are purely imaginary.] Retention of the quantities $X_{n'n}(\mathbf{k})$ means that the normalization con-

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dition of Eq. (2.16) cannot be satisfied; this difficulty is avoided by assuming that the electric field is sufficiently small so that interband tunneling is negligible.

The equation for the coefficients $A_{\nu,n}(\mathbf{k}_\perp)$ in the approximation that the interband tunneling is negligible is therefore

$$\left[E_n(\mathbf{k})-W_{\nu,n}(\mathbf{k}_\perp)-iF\frac{\partial}{\partial k_x}\right]A_{\nu,n}(\mathbf{k})=0. \quad (2.25)$$

The general solution of this equation is

$$A_{\nu,n}(\mathbf{k})=C_{\nu,n}(\mathbf{k}_\perp) \times\exp\left\{\frac{i}{F}\int_0^{k_x}[W_{\nu,n}(\mathbf{k}_\perp)-E_n(\mathbf{k}_\perp,k_x')]dk_x'\right\}, \quad (2.26)$$

where the coefficient $C_{\nu,n}(\mathbf{k}_\perp)$ and the energy $W_{\nu,n}(\mathbf{k}_\perp)$ can be determined by the normalization condition of Eq. (2.16) and the choice of the Brillouin zone which makes the end points in the x direction equivalent.[21] If the length of the Brillouin zone in the x direction is K_x, then by equivalence we have $A_{\nu,n}(\mathbf{k}+\mathbf{K}_x)=A_{\nu,n}(\mathbf{k})$, so

$$W_{\nu,n}(\mathbf{k}_\perp)=\frac{2\pi\nu F}{K_x}+\bar{E}_n(\mathbf{k}_\perp), \quad (2.27)$$

where we define

$$\bar{E}_n(\mathbf{k}_\perp)=\frac{1}{K_x}\int_0^{K_x}dk_x'E_n(\mathbf{k}_\perp,k_x'). \quad (2.28)$$

We note that since $E_n(\mathbf{k})$ is periodic in the Brillouin zone there is no need to integrate from the limits in Eq. (2.28), but in general

$$\bar{E}_n(\mathbf{k}_\perp)=\frac{1}{K_x}\int_{k_x}^{k_x+K_x}dk_x'E_n(\mathbf{k}_\perp,k_x'), \quad (2.29)$$

a result which will be used later. The coefficient $C_{\nu,n}(\mathbf{k}_\perp)$ is determined from the normalization condition: if the length of the box in the x direction is L_x and the length of the unit cell in this direction is R_x, so that there are N cells in the x direction ($L_x=NR_x$), then $C=N^{-1/2}$ and

$$A_{\nu,n}(\mathbf{k})=\frac{1}{\sqrt{N}} \times\exp\left\{\frac{i}{F}\int_0^{k_x}[W_{\nu,n}(\mathbf{k}_\perp)-E_n(\mathbf{k}_\perp,k_x')]dk_x'\right\}. \quad (2.30)$$

Equations (2.15), (2.27), (2.28), and (2.30) determine the wave function of the electron in the presence of an electric field, from which the imaginary part of the dielectric constant can be calculated as previously done with the zero-field wave functions.

At this point it may be useful to list the approximations inherent in the result given by Eq. (2.30). The neglect of the interband tunneling terms has already been discussed. The fact that the matrix element of x could be separated at all into interband and intraband terms depended on the Bloch form of the wave function, i.e., that the potential $V(\mathbf{r})$ is periodic in the lattice. This is a characteristic of the one-electron formalism, which neglects all interactions between electrons and in particular neglects the Coulomb attraction between the electron and hole which results in excitons. This approximation must necessarily limit the application of the results to cases where the electron-hole interaction is small. Finally, nondegenerate perturbation theory is used so that the formalism may break down at degeneracy points of bands.

We now proceed through the calculation of the imaginary part of the dielectric constant using the wave functions just obtained. The transition rate out of the state $\phi_{\nu,n}(\mathbf{k}_\perp,\mathbf{r})$ into the state $\phi_{\nu',n'}(\mathbf{k}_\perp',\mathbf{r})$ caused by the perturbation of Eq. (2.1) is

$$\frac{1}{t}|a_{\nu'n'\mathbf{k}_\perp';\nu n\mathbf{k}_\perp;t}|^2=\frac{4}{\hbar^2}|H'_{\nu'n'k_\perp';\nu n\mathbf{k}_\perp}|^2 \times\frac{\sin^2\frac{1}{2}(\omega_{\nu'n'\mathbf{k}_\perp'}-\omega_{\nu n\mathbf{k}_\perp}-\omega)t}{t(\omega_{\nu'n'\mathbf{k}_\perp'}-\omega_{\nu n\mathbf{k}_\perp}-\omega)^2}, \quad (2.31)$$

where $\hbar\omega_{\nu'n'\mathbf{k}_\perp'}=W_{\nu',n'}(\mathbf{k}_\perp')$, etc., the matrix element is

$$H'_{\nu'n'k_\perp';\nu n\mathbf{k}_\perp}=\langle\phi_{\nu'n'}(\mathbf{k}_\perp',\mathbf{r})|\frac{ieA}{mc}\hat{\epsilon}\cdot\nabla|\phi_{\nu n}(\mathbf{k}_\perp,\mathbf{r})\rangle =\frac{ie\hbar A}{mc}\sum_{\mathbf{k}_x}\delta_{\mathbf{k}_\perp',\mathbf{k}_\perp}\frac{i}{\hbar}\mathbf{P}_{n'n}(\mathbf{k})A_{\nu',n'}{}^*(\mathbf{k})A_{\nu,n}(\mathbf{k}), \quad (2.32)$$

and $\mathbf{P}_{n'n}(\mathbf{k})$ is defined in Eq. (2.6). Taking the limit of large t according to Eq. (2.8) and summing over all final (empty) and initial (filled) states denoted by the wave functions $\phi_{\nu',n'}(\mathbf{k}_\perp',\mathbf{r})$ and $\phi_{\nu,n}(\mathbf{k}_\perp,\mathbf{r})$, respectively, gives the total rate of transitions over the entire volume V as in Eq. (2.9):

$$w_{\text{tot}}=\frac{2\pi e^2A^2}{m^2c^2\hbar}\sum_{\substack{\nu\nu'nn'\\ \mathbf{k}q_x}}\hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k}_\perp,k_x)\hat{\epsilon}\cdot\mathbf{P}_{n'n}{}^*(\mathbf{k}_\perp,q_x) \times A_{\nu',n'}{}^*(\mathbf{k}_\perp,k_x)A_{\nu,n}(\mathbf{k}_\perp,k_x)A_{\nu,n}{}^*(\mathbf{k}_\perp,q_x)A_{\nu',n'}(\mathbf{k}_\perp,q_x) \times\delta[W_{\nu',n'}(\mathbf{k}_\perp)-W_{\nu,n}(\mathbf{k}_\perp)-\hbar\omega]. \quad (2.33)$$

Substituting Eq. (2.30) for the coefficients $A_{\nu,n}(\mathbf{k})$ and Eq. (2.27) for $W_{\nu,n}(\mathbf{k}_\perp)$ gives

$$w_{\text{tot}}=\frac{2\pi e^2A^2}{m^2c^2\hbar N^2}\sum_{\substack{\nu\nu'nn'\\ \mathbf{k}q_x}}\hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k}_\perp,k_x)\hat{\epsilon}\cdot\mathbf{P}_{n'n}{}^*(\mathbf{k}_\perp,q_x)\exp\left\{\frac{i}{F}\right. \times\left.\int_{q_x}^{k_x}dk_x'\left[E_{n'n}(\mathbf{k}_\perp,k_x')-\bar{E}_{n'n}(\mathbf{k}_\perp)+\frac{2\pi F}{K_x}(\nu-\nu')\right]\right\} \times\delta\left(\frac{2\pi F}{K_x}(\nu'-\nu)+\bar{E}_{n'n}(\mathbf{k}_\perp)-\hbar\omega\right). \quad (2.34)$$

Since the double sum over ν and ν' is over the combination $(\nu-\nu')$, it can be replaced with the equivalent expression[21] valid in the limit of large N:

$$\sum_{\nu,\nu'} f(\nu-\nu') \rightarrow N \sum_{l} \int_{-\infty}^{\infty} d\xi f(\xi)\, e^{-i2\pi l\xi}, \quad (2.35)$$

which enables the integration over the δ function to be performed with the result

$$w_{\text{tot}}=\frac{e^2A^2K_x}{m^2c^2\hbar NF} \sum_{\substack{lnn' \\ \mathbf{k}q_x}} \exp\left[\frac{lK_x}{iF}(\bar{E}_{n'n}(\mathbf{k}_\perp)-\hbar\omega)\right] \times \hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k}_\perp,k_x)\hat{\epsilon}\cdot\mathbf{P}_{n'n}{}^*(\mathbf{k}_\perp,q_x) \times \exp\left\{\frac{i}{F}\int_{q_x}^{k_x} dk_x'[E_{n'n}(\mathbf{k}_\perp k_x')-\hbar\omega]\right\}. \quad (2.36)$$

Since $K_x=2\pi/R_x$, the change from sums to integrals in k space, followed by multiplication by 2 for spin degeneracy and dividing by the volume of the box in order to obtain transitions per unit time per unit volume, gives

$$w=\frac{e^2A^2}{m^2c^2\hbar F}\int_{\text{B.Z.}} d^3k \int_{\text{B.Z.}} dq_x \times \sum_{ln'n} \exp\left[\frac{lK_x}{iF}[\bar{E}_{n'n}(\mathbf{k}_\perp)-\hbar\omega]\right] \times \hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k}_\perp,k_x)\hat{\epsilon}\cdot\mathbf{P}_{n'n}{}^*(\mathbf{k}_\perp,q_x) \times \frac{2}{(2\pi)^3}\exp\left\{\frac{i}{F}\int_{q_x}^{k_x} dk_x'[E_{n'n}(\mathbf{k}_\perp,k_x')-\hbar\omega]\right\}, \quad (2.37)$$

and by Eq. (2.12) the imaginary part of the dielectric constant in the presence of the field $\mathbf{F}=e\mathcal{E}$ is

$$\epsilon_2(\omega,\mathcal{E})=\frac{2\pi e^2}{m^2\omega^2F}\int_{\text{B.Z.}} d^3k \int_{\text{B.Z.}} dq \times \sum_{ln'n} \exp\left[\frac{lK_x}{iF}[\bar{E}_{n'n}(\mathbf{k}_\perp)-\hbar\omega]\right] \times \hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k}_\perp,k_x)\hat{\epsilon}\cdot\mathbf{P}_{n'n}{}^*(\mathbf{k}_\perp,q_x) \times \frac{2}{(2\pi)^3}\exp\left\{\frac{i}{F}\int_{q_x}^{k_x} dk_x'[E_{n'n}(\mathbf{k}_\perp,k_x')-\hbar\omega]\right\}. \quad (2.38)$$

Equation (2.38) is completely equivalent to the result obtained by Callaway[10] before the evaluation of his matrix element $M_{nn'}$, except that by combining the two integrations implicit in $|M_{nn'}|^2$ we have obtained $\epsilon_2(\omega,\mathcal{E})$ in a slightly different form. This form is necessary to represent the electric-field effect on ϵ_2 as a convolution-type integral in terms of zero-field quantities. It is possible to go back to Callaway's formulation by simply writing the exponent integral between the limits k_x and q_x in Eq. (2.38) as a product of two exponent integrals between the limits 0 and k_x, and q_x and 0, respectively, assuming $\mathbf{P}_{n'n}(\mathbf{k})$ is independent of $\mathbf{k}$, expanding the energy about the point $\mathbf{k}=0$, and retaining only the $l=0$ term in the sum over l.

The integrations over k_x and q_x cover the first Brillouin zone (B.Z.) in the field direction. Since the B.Z. width in this direction is K_x, these integrations can be taken between the limits $\pm\frac{1}{2}K_x$, and may be represented in (k_x,q_x) space as an integration over a square of side K_x centered at the point (0,0). We next consider the (k_x,q_x) integration over the square of side K_x centered at $(\nu K_x,\nu' K_x)$ which is, omitting irrelevant terms,

$$\int_{-(1/2)K_x+\nu K_x}^{(1/2)K_x+\nu K_x} dk_x \int_{-(1/2)K_x+\nu' K_x}^{(1/2)K_x+\nu' K_x} dq_x\, \hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k}_\perp,k_x)\hat{\epsilon}\cdot\mathbf{P}_{n'n}{}^*(\mathbf{k}_\perp,q_x)\exp\left\{\frac{i}{F}\int_{q_x}^{k_x} dk_x'[E_{n'n}(\mathbf{k}_\perp,k_x')-\hbar\omega]\right\}$$

$$=\int_{-(1/2)K_x}^{(1/2)K_x} dk_x dq_x \hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k}_\perp,k_x)\hat{\epsilon}\cdot\mathbf{P}_{n'n}{}^*(\mathbf{k}_\perp,q_x)\exp\left\{\frac{i}{F}\int_{q_x+\nu' K_x}^{q_x} dk_x'\cdots\right\} \times\exp\left\{\frac{i}{F}\int_{q_x}^{k_x} dk_x'\cdots\right\}\exp\left\{\frac{i}{F}\int_{k_x}^{k_x+\nu K_x} dk_x'\cdots\right\}$$

$$=\int_{-(1/2)K_x}^{(1/2)K_x} dk_x dq_x \hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k}_\perp,k_x)\hat{\epsilon}\cdot\mathbf{P}_{n'n}{}^*(\mathbf{k}_\perp,q_x)\exp\left\{\frac{iK_x}{F}(\nu-\nu')[\bar{E}_{n'n}(\mathbf{k}_\perp)-\hbar\omega]\right\} \times\exp\left\{\frac{i}{F}\int_{q_x}^{k_x} dk_x'[E_{n'n}(\mathbf{k}_\perp k_x')-\hbar\omega]\right\}, \quad (2.39)$$

where the periodicity of $\mathbf{P}_{n'n}(\mathbf{k})$ in $\mathbf{k}$ space has been used in the second step and Eq. (2.29) in the third. But Eq. (2.39) is the term $l=(\nu-\nu')$ of the sum over l in Eq. (2.38), so the integral over the square of side K_x centered at $(\nu K_x,\nu' K_x)$ in (k_x,q_x) space gives the $l=(\nu-\nu')$ term in the sum over l. The k_x,q_x integrations

can therefore be defined so as to absorb the sum over l, for instance, if $\nu=0$ we have $l=-\nu'$, and the sum over all l generates the strip $|k_x|\leq\frac{1}{2}K_x$, $|q_x|<\infty$ in (k_x,q_x) space as shown in Fig. 1. Conversely, the strip $|q_x|\leq\frac{1}{2}K_x$, $|k_x|<\infty$ is generated for $\nu'=0$.

The convolution integral formulation follows by putting the variables k_x,q_x on an equal footing. To do this, we map the squares defined by the index $l=\nu-\nu'$ in the following way. If l is even, the square is centered on the point $(\frac{1}{2}lK_x, \frac{1}{2}lK_x)$. For odd l, the square is bisected along the face diagonal $\theta=-45°$; the upper right half (URH) is mapped on the URH of the square centered on $[\frac{1}{2}(l-1)K_x, -\frac{1}{2}(l-1)K_x]$. The sum over all l now completely maps the strip defined by $|k_x+q_x|\leq K_x$, $|k_x-q_x|<\infty$ as shown in Fig. 1, and

$$\epsilon_2(\omega,\mathcal{E})=\frac{2\pi e^2}{m^2\omega^2F}\int_{\text{B.Z.}}d\mathbf{k}_\perp\int_{-\infty}^{\infty}dk_x\int_{-K_x-k_x}^{K_x-k_x}dq_x$$
$$\times\sum_{nn'}\hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k}_\perp,k_x)\hat{\epsilon}\cdot\mathbf{P}_{n'n}{}^*(\mathbf{k}_\perp,q_x)$$
$$\times\frac{2}{(2\pi)^3}\exp\left\{\frac{i}{F}\int_{q_x}^{k_x}dk_x'[E_{n'n}(\mathbf{k}_\perp k_x')-\hbar\omega]\right\}. \quad (2.40)$$

We now define new variables a,b such that

$$a=\tfrac{1}{2}(k_x+q_x);\ b=\tfrac{1}{2}(k_x-q_x);\ dk_xdq_x=2dadb. \quad (2.41)$$

The region of integration in Eq. (2.40) is defined by these variables as

$$|a|\leq\frac{K_x}{2},\quad |b|<\infty, \quad (2.42)$$

so

$$\epsilon_2(\omega,\mathcal{E})=\frac{4\pi e^2}{m^2\omega^2F}\sum_{nn'}\int_{\text{B.Z.}}d\mathbf{k}_\perp\int_{-K_x/2}^{K_x/2}da\int_{-\infty}^{\infty}db$$
$$\times\hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k}_\perp,a+b)\hat{\epsilon}\cdot\mathbf{P}_{n'n}{}^*(\mathbf{k}_\perp,a-b)$$
$$\times\frac{2}{(2\pi)^3}\exp\left\{\frac{i}{F}\int_{a-b}^{a+b}dk_x'[E_{n'n}(\mathbf{k}_\perp,k_x')-\hbar\omega]\right\}. \quad (2.43)$$

If $k_x''=k_x'-a$ so that $k_x'=k_x''+a$, and if we represent the dummy variable a by k_x, Eq. (2.43) becomes

$$\epsilon_2(\omega,\mathcal{E})=\frac{4\pi e^2}{m^2\omega^2F}\sum_{nn'}\int_{\text{B.Z.}}d^3k\int_{-\infty}^{\infty}db$$
$$\times\hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k}_\perp,k_x+b)\hat{\epsilon}\cdot\mathbf{P}_{n'n}{}^*(\mathbf{k}_\perp,k_x-b)\frac{2}{(2\pi)^3}$$
$$\times\exp\left\{\frac{i}{F}\int_{-b}^{b}dk_x''[E_{n'n}(\mathbf{k}_\perp,k_x+k_x'')-\hbar\omega]\right\}. \quad (2.44)$$

In order to obtain a form which reduces easily to the zero-field equation and which can be readily expanded

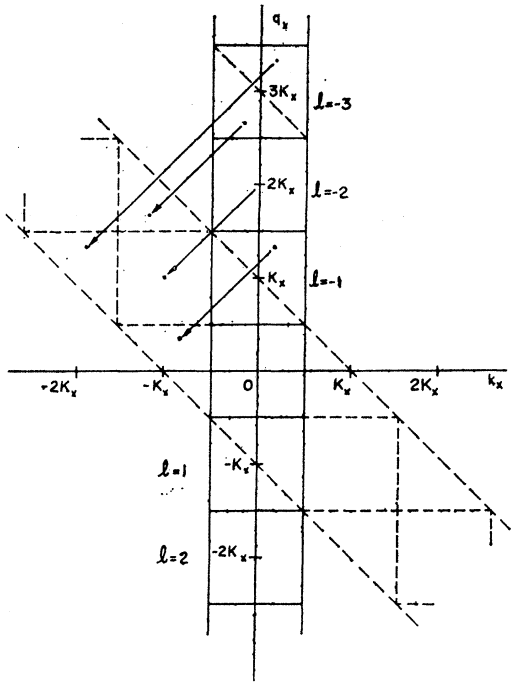

FIG. 1. Equivalent regions of integration for Eq. (2.39).

for weak fields, we change variables once more by defining s, t of reciprocal energy dimension:

$$s=\frac{b}{F},\quad t=\frac{k_x''}{F}.$$

With this substitution, Eq. (2.44) becomes the basic expression for the imaginary part of the dielectric constant in the presence of an electric field:

$$\epsilon_2(\omega,\mathcal{E})=\frac{4\pi e^2}{m^2\omega^2}\sum_{nn'}\int_{\text{B.Z.}}d^3k\int_{-\infty}^{\infty}ds$$
$$\times\hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k}+s\mathbf{F})\hat{\epsilon}\cdot\mathbf{P}_{n'n}{}^*(\mathbf{k}-s\mathbf{F})\frac{2}{(2\pi)^3}$$
$$\times\exp\left\{i\int_{-s}^{s}dt[E_{n'n}(\mathbf{k}+t\mathbf{F})-\hbar\omega]\right\}. \quad (2.45)$$

The field therefore mixes states along its direction in k space. Reduction to the zero-field case is trivial, since for $\mathbf{F}=0$ the integral in the exponent is just

$$2s[E_{n'n}(\mathbf{k})-\hbar\omega]$$

and

$$\int_{-\infty}^{\infty}ds\exp\{2is[E_{n'n}(\mathbf{k})-\hbar\omega]\}$$
$$=\pi\delta[E_{n'n}(\mathbf{k})-\hbar\omega], \quad (2.46)$$

which, when substituted into Eq. (2.45), gives Eq. (2.13), the zero-field expression for ϵ_2.

The length of the shortest reciprocal lattice vector in the direction of $\mathbf{F}$ will be designated as K_F. This appears as a periodicity is s, of period $\Delta s = K_F/F$. By explicitly including this in Eq. (2.46), the Stark steps can be obtained.[10] The integral over s can be partitioned into an integral over a fundamental line $|s| \leq K_F/2F$ which is summed over the centers lK_F/F of these lines. By Eq. (2.29), this leads to

$$\epsilon_2(\omega,\mathcal{E}) = \frac{4\pi e^2}{m^2\omega^2} \sum_{ln'n} \int_{\text{B.Z.}} d^3k \exp\left\{ i \frac{2K_F}{F} l[E_{n'n}(\mathbf{k}_\perp) - \hbar\omega] \right\} \times \frac{2}{(2\pi)^3} \int_{-K_F/2F}^{K_F/2F} ds \hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k}+s\mathbf{F}) \hat{\epsilon}\cdot\mathbf{P}_{n'n}{}^*(\mathbf{k}-s\mathbf{F}) \times \exp\left\{ i \int_{-s}^{s} dt [E_{n'n}(\mathbf{k}+t\mathbf{F}) - \hbar\omega] \right\}. \quad (2.47)$$

Since for any function $G(\mathbf{k}_\perp)$,[10]

$$\sum_l \exp\left[i \frac{2K_F}{F} l G(\mathbf{k}_\perp) \right] = \frac{\pi F}{K_F} \sum_{l'} \delta\left(G(\mathbf{k}_\perp) + \frac{\pi F}{K_F} l' \right), \quad (2.48)$$

an alternative expression is

$$\epsilon_2(\omega,\mathcal{E}) = \frac{4\pi^2 e^2 F}{m^2\omega^2 K_x} \sum_{l'nn'} \int_{\text{B.Z.}} d^3k \times \delta\left(E_{n'n}(\mathbf{k}_\perp) - \hbar\omega + \frac{\pi F}{K_x} l' \right) \frac{2}{(2\pi)^3} \int_{-K_F/2F}^{K_F/2F} ds \times \hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k}+s\mathbf{F}) \hat{\epsilon}\cdot\mathbf{P}_{n'n}{}^*(\mathbf{k}-s\mathbf{F}) \times \exp\left\{ i \int_{-s}^{s} dt [E_{n'n}(\mathbf{k}+t\mathbf{F}) - \hbar\omega] \right\}, \quad (2.49)$$

which has steplike structure in $E_{n'n}(\mathbf{k}_\perp)$ with step separation $\pi F/K_F$. Equation (2.49) is useful for large fields; for small fields Eq. (2.47) is more convenient. In the latter case the only contribution is from the $l=0$ term because of the large quantity K_F/F which causes a rapidly varying phase of the exponent for $l \neq 0$.

The simplest possible approximation of Eq. (2.45) for finite fields is to expand the energy and/or the momentum in a Taylor series in terms of the field, keeping only the lowest terms. Even though such an expansion is nonperiodic, the fact that only the $l=0$ term in Eq. (2.47) contributes appreciably suggests that the Taylor expansions should be a good approximation since they fit E and $\mathbf{P}$ in the contributing region at small s and t. The energy may be expanded in terms of the field $\mathbf{F}$ as

$$E_{n'n}(\mathbf{k}+t\mathbf{F}) = E_{n'n}(\mathbf{k}) + t(\mathbf{F}\cdot\nabla_\mathbf{k}) E_{n'n}(\mathbf{k}) + \frac{t^2}{2!}(\mathbf{F}\cdot\nabla_\mathbf{k})^2 E_{n'n}(\mathbf{k}) + \cdots, \quad (2.50)$$

Substitution of Eq. (2.50) in Eq. (2.45) and subsequent integration over t yields

$$\epsilon_2(\omega,\mathcal{E}) = \frac{4\pi e^2}{m^2\omega^2} \sum_{n'n} \int_{\text{B.Z.}} d^3\mathbf{k} |\hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k})|^2 \frac{2}{(2\pi)^3} \times \int_{-\infty}^{\infty} ds \exp\{ 2is[E_{n'n}(\mathbf{k}) - \hbar\omega] + \tfrac{1}{3}is^3(\mathbf{F}\cdot\nabla_\mathbf{k})^2 E_{n'n}(\mathbf{k}) \}, \quad (2.51)$$

where we have assumed that $\mathbf{P}(\mathbf{k}+s\mathbf{F}) \cong \mathbf{P}(\mathbf{k})$. The integration over t removes odd powers in $\mathbf{F}$. The terms of even order in $\mathbf{F}$, not included in Eq. (2.51), are of fourth order or higher and are considered negligible. In the weak-field approximation these terms are neglected. Since the Airy function (Ai) is defined as[25]

$$\text{Ai}(x) = \frac{1}{2\pi} \int_{-\infty}^{\infty} ds \exp[\tfrac{1}{3}is^3 + isx] = \frac{1}{\pi} \int_0^{\infty} ds \cos(\tfrac{1}{3}s^3 + xs), \quad (2.52)$$

where the constant of normalization $N=\pi$ so that $\lim_{\epsilon\to 0} |\epsilon|^{-1} \text{Ai}(x/\epsilon) = \delta(x)$, Eq. (2.51) becomes

$$\epsilon_2(\omega,\mathcal{E}) = \frac{4\pi^2 e^2}{m^2\omega^2} \sum_{n'n} \int_{\text{B.Z.}} d^3k \frac{2}{(2\pi)^3} |\hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k})|^2 \times \left[\frac{1}{\hbar|\Omega_{n'n}(\mathbf{k})|} \text{Ai}\left(\frac{E_{n'n}(\mathbf{k}) - \hbar\omega}{\hbar\Omega_{n'n}(\mathbf{k})} \right) \right] \quad (2.53)$$

in the weak-field approximation, where

$$\hbar\Omega_{n'n}(\mathbf{k}) = \hbar \left[\frac{F^2}{8\mu\hbar} \right]^{1/3}, \quad (2.54)$$

$$\mu = \mu_{n'n}(\mathbf{k}) = \left\{ \frac{1}{\hbar^2 F^2} (\mathbf{F}\cdot\nabla_\mathbf{k})^2 E_{n'n}(\mathbf{k}) \right\}^{-1} = \left\{ \frac{1}{\hbar^2} \frac{\partial^2}{\partial k_F{}^2} [E_{n'n}(\mathbf{k})] \right\}^{-1}, \quad (2.55)$$

and $\hbar\Omega_{n'n}(\mathbf{k}) = \hbar\theta/2^{2/3}$ where $\hbar\theta$ is the characteristic electro-optic energy.[15] $\Omega_{n'n}(\mathbf{k})$ and the mass $\mu_{n'n}(\mathbf{k})$ have the same sign and may be either positive or negative.

We note that the reduced mass defined in Eq. (2.55) is just the interband reduced mass of the pair of states between which the transition is taking place, calculated in the direction of the electric field. If the band structure of a solid is known, i.e., if $E_{n'n}(\mathbf{k})$ is known for every $\mathbf{k}$

[25] H. A. Antosiewicz, in *Handbook of Mathematical Functions*, edited by M. Abramowitz and I. A. Stegun (U. S. Department of Commerce, National Bureau of Standards, Washington, D. C., 1964), Appl. Math. Series **55**, 446 ff.

in the B.Z. and if values for $\mathbf{P}_{n'n}(\mathbf{k})$ can be computed and are slowly varying in the field direction, then $\epsilon_2(\omega,\mathcal{E})$ can be calculated directly from Eq. (2.53) since $\hbar\Omega_{n'n}(\mathbf{k})$ can be derived from $E_{n'n}(\mathbf{k})$ using Eqs. (2.50) and (2.51). Equation (2.53), in contrast to previous calculations,[8–15] is not limited to a region in $\mathbf{k}$ space where the energy surfaces are quadratic, but includes changes in the curvature of the energy surfaces explicitly in the form $\hbar\Omega_{n'n}(\mathbf{k})$. It should be remembered that the difference $\Delta\epsilon_2(\omega,\mathcal{E})=\epsilon_2(\omega,\mathcal{E})-\epsilon_2(\omega,0)$ is of interest experimentally and may be calculated from the band structure of a solid using Eqs. (2.13) and (2.53) and subtracting the resulting curves.

To emphasize the similarity between the zero-field and weak-field equations, let us rewrite Eq. (2.13) as

$$\epsilon_2(\omega,0)=\sum_{n'n}\int_{\text{B.Z.}} d^3k C_{n'n}(\mathbf{k})\delta[E_{n'n}(\mathbf{k})-\hbar\omega] \quad (2.56)$$

and Eq. (2.53) as

$$\epsilon_2(\omega,\mathcal{E})=\sum_{n'n}\int_{\text{B.Z.}} d^3k C_{n'n}(\mathbf{k}) \times\left[\frac{1}{|\hbar\Omega_{n'n}(\mathbf{k})|}\,\mathrm{Ai}\left(\frac{E_{n'n}(\mathbf{k})-\hbar\omega}{\hbar\Omega_{n'n}(\mathbf{k})}\right)\right], \quad (2.57)$$

where

$$C_{n'n}(\mathbf{k})=\frac{4\pi^2e^2}{m^2\omega^2}\left[\frac{2}{(2\pi)^3}\right]|\hat{\epsilon}\cdot\mathbf{P}_{n'n}(\mathbf{k})|^2. \quad (2.58)$$

Comparing Eqs. (2.56) and (2.57), it is seen that the effect of the electric field, in the approximations used to derive Eq. (2.53), is simply to replace the δ function by the Airy function with the appropriate prefactor. At any point $\mathbf{k}$ the electric field thereby mixes contributions from other points in the neighborhood of $\mathbf{k}$. In the limit that the field goes to zero, this mixing should vanish: we note that in this limit $\mathbf{F}\to 0$, $\hbar\Omega_{n'n}(\mathbf{k})\to 0$, and since

$$\lim_{\hbar\Omega\to 0}\left[\frac{1}{\hbar|\Omega|}\,\mathrm{Ai}\left(\frac{E_{n'n}(\mathbf{k})-\hbar\omega}{\hbar\Omega}\right)\right]=\delta[E_{n'n}(\mathbf{k})-\hbar\omega]. \quad (2.59)$$

Equation (2.57) reduces as it must to Eq. (2.56).

Within the given approximations, Eq. (2.53) relates the band structure of a solid to its interband dielectric properties, but it is of value to simplify Eq. (2.53) about certain points in $\mathbf{k}$ space, where the relative gradient of the transition energy vanishes. About these points, $\hbar\Omega_{n'n}(\mathbf{k})$ is independent of $\mathbf{k}$ thus further simplifying the results. The equations for the interband dielectric properties of a solid in an electric field near critical points are derived and their applications discussed in the following section.

III. CONVOLUTION INTEGRAL FORMULATION NEAR CRITICAL POINTS

In this section, it is our purpose to show that, for direct transitions near critical points and also for indirect transitions, the imaginary part of the dielectric constant may be expressed in convolution integral form as

$$\omega^2\epsilon_2(\omega,\mathcal{E})=\int_{-\infty}^{\infty}d\omega'\omega'^2\epsilon_2(\omega',0)\left\{\frac{1}{|\Omega|}\,\mathrm{Ai}\left(\frac{\omega'-\omega}{\Omega}\right)\right\}, \quad (3.1)$$

where Ω is a constant and is given by

$$\Omega^3=\frac{F^2}{8\mu_F\hbar}=\frac{1}{8\hbar^3}(\mathbf{F}\cdot\nabla_{\mathbf{k}})^2E_{n'n}(\mathbf{k}), \quad (3.2)$$

where μ_F is parallel to the direction of the field $\mathbf{F}$. For direct transitions, Eq. (3.1) is a simplification of Eq. (2.53) near critical points with the assumptions that the momentum matrix element and $\hbar\Omega_{n'n}(\mathbf{k})$ are independent of $\mathbf{k}$ and only transitions between two bands need be considered. For indirect transitions, we consider only phonon-assisted transitions between a maximum in the valence band and a minimum in the conduction band. Equation (3.1) will then be used to derive the optical density of states in terms of finite-field quantities and then to calculate the real part of the dielectric constant using the Kramers-Kronig relations. An independent derivation for direct transitions is given in Appendix B which also results in Eq. (3.1).

A. Direct Transitions

Near critical points, the transition energy is a quadratic function of $\mathbf{k}$ and may be expressed in terms of reduced effective masses as

$$E(\mathbf{k})=E_g+\tfrac{1}{2}\hbar^2\left(\frac{k_x^2}{\mu_x}+\frac{k_y^2}{\mu_y}+\frac{k_z^2}{\mu_z}\right), \quad (3.3)$$

where E_g is the gap energy.

Depending on the signs of the reduced masses there are four independent types of critical points which may be defined as

M_0: μ_x,μ_y,μ_z positive,

M_1: μ_x,μ_y positive, μ_z negative,

M_2: μ_x,μ_y negative, μ_z positive,

M_3: μ_x,μ_y,μ_z negative.

Assuming that $\hbar\Omega_{n'n}(\mathbf{k})$ and $\mathbf{P}_{n'n}(\mathbf{k})$ are independent of $\mathbf{k}$ near critical points and only two bands are involved, we may write Eq. (2.53) as

$$\epsilon_2(\omega,\mathcal{E})=\frac{e^2|\hat{\epsilon}\cdot\mathbf{P}|^2}{\pi m^2\omega^2}\int_{\text{B.Z.}}d^3k\frac{1}{|\hbar\Omega|}\,\mathrm{Ai}\left(\frac{E(\mathbf{k})-\hbar\omega}{\hbar\Omega}\right). \quad (3.4)$$

Using Eq. (3.3) and assuming the B.Z. boundaries

may be extended to positive and negative infinity in the weak-field limit, we may change the integral over $\mathbf{k}$ space to an integral over energy. The signs of the effective masses may now be introduced explicitly and the equations for the four types of critical points may be written in condensed form as

$$\epsilon_2(\omega,\mathcal{E})=\frac{e^2|\hat{\epsilon}\cdot\mathbf{P}|^2}{\pi m^2\omega^2}\left(\frac{2}{\hbar^2}\right)^{3/2}|\mu_x\mu_y\mu_z|^{1/2}\int_0^\infty\frac{d\epsilon_x d\epsilon_y d\epsilon_z}{(\epsilon_x\epsilon_y\epsilon_z)^{1/2}}$$
$$\times\left\{\frac{1}{|\hbar\Omega|}\,\mathrm{Ai}\left[\frac{(\pm)^{01}(\epsilon_x+\epsilon_y)(\pm)^{02}\epsilon_z+(E_g-\hbar\omega)}{\hbar\Omega}\right]\right\}, \tag{3.5}$$

where $\epsilon_i=\hbar^2k_i^2/2|\mu_i|$ and the notation $(\pm)^{ij}$ means that the sign is positive for the M_i and M_j edges and negative for the other two edges, e.g., $(\pm)^{02}$ is positive for the M_0 and M_2 edges and negative for the M_1 and M_3 edges.

If we define the change of variables

$$u=\epsilon_x+\epsilon_y \quad\text{and}\quad \epsilon_z=\epsilon_z, \tag{3.6}$$

and evaluate the integration over ϵ_x, the result is

$$\epsilon_2(\omega,\mathcal{E})=\frac{e^2|\hat{\epsilon}\cdot\mathbf{P}|^2}{m^2\omega^2}\left(\frac{8|\mu_x\mu_y\mu_z|}{\hbar^6}\right)^{1/2}\int_0^\infty du\frac{d\epsilon_z}{\sqrt{\epsilon_z}}$$
$$\times\left\{\frac{1}{|\hbar\Omega|}\,\mathrm{Ai}\left[\frac{(\pm)^{01}u(\pm)^{02}\epsilon_z+(E_g-\hbar\omega)}{\hbar\Omega}\right]\right\}. \tag{3.7}$$

It is known that, for the M_1 and M_2 edges, $\epsilon_2(\omega,\mathcal{E})$ is divergent but that $\Delta\epsilon_2(\omega,\mathcal{E})=\epsilon_2(\omega,\mathcal{E})-\epsilon_2(\omega,0)$ is finite.[15] Thus it should be possible to extract a field-independent divergent term from Eq. (3.7) for the M_1 and M_2 edges. With this in mind, we define $u=(\pm)^{03}u'$ and add and subtract identical terms in the M_1 and M_2 equations so that we may use the formula[25]

$$\int_{-\infty}^{\infty}dt\,\mathrm{Ai}(t)=1 \tag{3.8}$$

to extract the divergent term. Following this procedure, Eq. (3.7) may be rewritten as

$$M_0\text{: }\epsilon_2(\omega,\mathcal{E})=\frac{B}{2\omega^2\sqrt{\hbar}}\int_0^\infty\frac{d\epsilon_z}{\sqrt{\epsilon_z}}\int_0^\infty\frac{du'}{|\hbar\Omega|}$$
$$\times\mathrm{Ai}\left[\frac{(u'+\epsilon_z)+(E_g-\hbar\omega)}{\hbar\Omega}\right]; \tag{3.9a}$$

$$M_1\text{: }\epsilon_2(\omega,\mathcal{E})=\frac{-B}{2\omega^2\sqrt{\hbar}}\int_0^\infty\frac{d\epsilon_z}{\sqrt{\epsilon_z}}\left\{\int_0^\infty\frac{du'}{|\hbar\Omega|}\right.$$
$$\times\mathrm{Ai}\left[\frac{-(u'+\epsilon_2)+(E_g-\hbar\omega)}{\hbar\Omega}\right]$$
$$\left.-\int_{-\infty}^\infty\frac{du'}{|\hbar\Omega|}\,\mathrm{Ai}\left[\frac{-(u'+\epsilon_z)+(E_g-\hbar\omega)}{\hbar\Omega}\right]\right\}; \tag{3.9b}$$

$$M_2\text{: }\epsilon_2(\omega,\mathcal{E})=\frac{-B}{2\omega^2\sqrt{\hbar}}\int_0^\infty\frac{d\epsilon_z}{\sqrt{\epsilon_z}}\left\{\int_0^\infty\frac{du'}{|\hbar\Omega|}\right.$$
$$\times\mathrm{Ai}\left[\frac{(u'+\epsilon_z)+(E_g-\hbar\omega)}{\hbar\Omega}\right]$$
$$\left.-\int_{-\infty}^\infty\frac{du'}{|\hbar\Omega|}\,\mathrm{Ai}\left[\frac{(u'+\epsilon_z)+(E_g-\hbar\omega)}{\hbar\Omega}\right]\right\}; \tag{3.9c}$$

$$M_3\text{: }\epsilon_2(\omega,\mathcal{E})=\frac{B}{2\omega^2\sqrt{\hbar}}\int_0^\infty\frac{d\epsilon_z}{\sqrt{\epsilon_z}}\int_0^\infty\frac{du'}{|\hbar\Omega|}$$
$$\times\mathrm{Ai}\left[\frac{-(u'+\epsilon_z)+(E_g-\hbar\omega)}{\hbar\Omega}\right], \tag{3.9d}$$

where

$$B=\frac{2e^2|\hat{\epsilon}\cdot\mathbf{P}|^2}{m^2\hbar}\left(\frac{8|\mu_x\mu_y\mu_z|}{\hbar^3}\right)^{1/2}. \tag{3.10}$$

Using Eq. (3.8) and defining the change of variables

$$v=u'+\epsilon_z \quad\text{and}\quad \epsilon_z=\epsilon_z \tag{3.11}$$

reduces Eqs. (3.9) to

$$M_0\text{: }\epsilon_2(\omega,\mathcal{E})=\frac{B}{\omega^2\sqrt{\hbar}}\int_0^\infty dv\sqrt{v}$$
$$\times\left\{\frac{1}{|\hbar\Omega|}\,\mathrm{Ai}\left[\frac{v+(E_g-\hbar\omega)}{\hbar\Omega}\right]\right\}; \tag{3.12a}$$

$$M_1\text{: }\epsilon_2(\omega,\mathcal{E})=\frac{-B}{\omega^2\sqrt{\hbar}}\int_0^\infty dv\sqrt{v}$$
$$\times\left\{\frac{1}{|\hbar\Omega|}\,\mathrm{Ai}\left[\frac{-v+(E_g-\hbar\omega)}{\hbar\Omega}\right]\right\}$$
$$+\frac{B}{2\omega^2\sqrt{\hbar}}\int_0^\infty\frac{d\epsilon_z}{\sqrt{\epsilon_z}}; \tag{3.12b}$$

$$M_2\text{: }\epsilon_2(\omega,\mathcal{E})=\frac{-B}{\omega^2\sqrt{\hbar}}\int_0^\infty dv\sqrt{v}$$
$$\times\left\{\frac{1}{|\hbar\Omega|}\,\mathrm{Ai}\left[\frac{v+(E_g-\hbar\omega)}{\hbar\Omega}\right]\right\}$$
$$+\frac{B}{2\omega^2\sqrt{\hbar}}\int_0^\infty\frac{d\epsilon_z}{\sqrt{\epsilon_z}}; \tag{3.12c}$$

$$M_3\text{: }\epsilon_2(\omega,\mathcal{E})=\frac{B}{\omega^2\sqrt{\hbar}}\int_0^\infty dv\sqrt{v}$$
$$\times\left\{\frac{1}{|\hbar\Omega|}\,\mathrm{Ai}\left[\frac{-v+(E_g-\hbar\omega)}{\hbar\Omega}\right]\right\}, \tag{3.12d}$$

where the second terms on the right-hand side of Eqs. (3.12b) and (3.12c) are indeed field-independent

divergent terms, as expected, and all the other terms on the right-hand side of Eq. (3.12) are field-dependent finite terms. Using Eq. (2.59), we have the zero-field limit of the above equations which may be written as

$$M_0\colon \epsilon_2(\omega,0)=\frac{B}{\omega^2}(\omega-\omega_g)^{1/2}, \quad \omega>\omega_g$$
$$=0, \qquad \omega<\omega_g; \tag{3.13a}$$

$$M_1\colon \epsilon_2(\omega,0)=\frac{-B}{\omega^2}(\omega_g-\omega)^{1/2}$$
$$+\frac{B}{\omega^2}\frac{1}{2\sqrt{\hbar}}\int_0^\infty\frac{dE}{\sqrt{E}}, \qquad \omega<\omega_g$$
$$=\frac{B}{\omega^2}\frac{1}{2\sqrt{\hbar}}\int_0^\infty\frac{dE}{\sqrt{E}}, \qquad \omega>\omega_g; \tag{3.13b}$$

$$M_2\colon \epsilon_2(\omega,0)=\frac{-B}{\omega^2}(\omega-\omega_g)^{1/2}$$
$$+\frac{B}{\omega^2}\frac{1}{2\sqrt{\hbar}}\int_0^\infty\frac{dE}{\sqrt{E}}, \qquad \omega>\omega_g$$
$$=\frac{B}{\omega^2}\frac{1}{2\sqrt{\hbar}}\int_0^\infty\frac{dE}{\sqrt{E}}, \qquad \omega<\omega_g; \tag{3.13c}$$

$$M_3\colon \epsilon_2(\omega,0)=\frac{B}{\omega^2}(\omega_g-\omega)^{1/2}, \quad \omega<\omega_g.$$
$$=0, \qquad \omega>\omega_g \tag{3.13d}$$

where $\hbar\omega_g=E_g$. Since the sign of Ω is that of μ_F, which depends on the field direction for M_1 and M_2 edges, Eqs. (3.12b) and (3.12c) are actually two equations —one with Ω positive and one with Ω negative. These two signs for Ω are equivalent to the parallel and transverse field effects calculated previously.[15] It may easily be shown that Eqs. (3.12) reduce to the previous results and in fact, the same starting point as used by Aspnes is used in Appendix B to derive Eqs. (3.12). With the change of variable $v=(\pm)^{0\text{2}}(\hbar\omega'-\hbar\omega_g)$, Eqs. (3.12) may also be expressed as

$$M_0\colon \epsilon_2(\omega,\mathcal{E})=\frac{B}{\omega^2}\int_{\omega_g}^\infty d\omega'(\omega'-\omega_g)^{1/2}$$
$$\times\left\{\frac{1}{\Omega}\mathrm{Ai}\left(\frac{\omega'-\omega}{\Omega}\right)\right\}; \tag{3.14a}$$

$$M_1\colon \epsilon_2(\omega,\mathcal{E})=\frac{-B}{\omega^2}\int_{-\infty}^{\omega_g}d\omega'(\omega_g-\omega')^{1/2}$$
$$\times\left\{\frac{1}{|\Omega|}\mathrm{Ai}\left(\frac{\omega'-\omega}{\Omega}\right)\right\}$$
$$+\frac{B}{\omega^2}\frac{1}{2\sqrt{\hbar}}\int_0^\infty\frac{dE}{\sqrt{E}}; \tag{3.14b}$$

$$M_2\colon \epsilon_2(\omega,\mathcal{E})=\frac{-B}{\omega^2}\int_{\omega_g}^\infty d\omega'(\omega'-\omega_g)^{1/2}$$
$$\times\left\{\frac{1}{|\Omega|}\mathrm{Ai}\left(\frac{\omega'-\omega}{\Omega}\right)\right\}$$
$$+\frac{B}{\omega^2}\frac{1}{2\sqrt{\hbar}}\int_0^\infty\frac{dE}{\sqrt{E}}; \tag{3.14c}$$

$$M_3\colon \epsilon_2(\omega,\mathcal{E})=\frac{B}{\omega^2}\int_{-\infty}^{\omega_g}d\omega'(\omega_g-\omega')^{1/2}$$
$$\times\left\{\frac{1}{|\Omega|}\mathrm{Ai}\left(\frac{\omega'-\omega}{\Omega}\right)\right\}. \tag{3.14d}$$

In the form of Eqs. (3.14), we can easily see that comparison of Eqs. (3.13) and (3.14) gives the result

$$\omega^2\epsilon_2(\omega,\mathcal{E})=\int_{-\infty}^\infty d\omega'\omega'^2\epsilon_2(\omega',0)\left\{\frac{1}{|\Omega|}\mathrm{Ai}\left(\frac{\omega'-\omega}{\Omega}\right)\right\}, \tag{3.15}$$

where we have used Eq. (3.10) to include the divergent terms in the convolution integral form of Eq. (3.15) which is identical with Eq. (3.1).

We will now show that Eq. (3.1) is applicable to indirect transitions as well as direct transitions near critical points.

B. Indirect Transitions

For indirect transitions, i.e., phonon-assisted transitions, a sum over the center-of-mass as well as the relative wave vector must be carried out due to the extra degree of freedom introduced by the inclusion of phonons.[12] The sum over the center-of-mass states can be expressed as a properly normalized integral as

$$\sum_{\text{c.m.}}\rightarrow\frac{(2M_xM_yM_z)^{1/2}}{2\pi^2\hbar^3}\int_0^\infty dE_{\text{c.m.}}[E_{\text{c.m.}}]^{1/2}, \tag{3.16}$$

where $E_{\text{c.m.}}$ is the center-of-mass energy of the electron-hole pair and, for the ith direction, $M_i=m_{ei}+m_{hi}$, where m_e and m_h are the electron and hole effective masses, respectively. The equation for direct transitions may be changed to indirect transitions by the addition of the above sum, by replacing $\hbar\omega$ by $[\hbar\omega-E_{\text{c.m.}}\mp\hbar\omega_{\kappa 0}]$, where $\hbar\omega_{\kappa 0}$ is the energy of the phonon, and multiplying by $(n_{\kappa 0}+\frac{1}{2}\pm\frac{1}{2})$ in Penchina's[12] notation. The upper and lower signs refer to the emission and absorption of phonons of energy $\hbar\omega_{\kappa 0}$, respectively. Since indirect transitions are generally taken to be from maxima in the valence band to minima in the conduction band, all effective masses will be taken as positive in the following. Also, for the above replacement to be valid, we have assumed that the square of the matrix element which now contains an energy denominator does not change appreciably over the range of integration. Following the above procedure and using Eq. (3.12a), the imaginary

part of the dielectric constant in an electric field for indirect transition is given by

$$\epsilon_2^{\rm ind}(\omega,\mathcal{E})=\frac{(2M_xM_yM_z)^{1/2}}{2\pi^2\hbar^3}\int_0^\infty dE_{\rm c.m.}[E_{\rm c.m.}]^{1/2}\times(n_{\kappa 0}+\tfrac{1}{2}\pm\tfrac{1}{2})\frac{B}{\omega^2\sqrt{\hbar}}\int_0^\infty E^{1/2}dE\times\left\{\frac{1}{\hbar|\Omega|}\,\mathrm{Ai}\left[\frac{E+E_{\rm c.m.}+E_g\pm\hbar\omega_{\kappa 0}-\hbar\omega}{\hbar\Omega}\right]\right\}. \quad (3.17)$$

Defining a change of variables

$$\hbar\omega'=E_{\rm c.m.}+E+E_g\pm\hbar\omega_{\kappa 0} \quad \text{and} \quad E=E \quad (3.18)$$

and evaluating the E integral gives the result

$$\epsilon_2^{\rm ind}(\omega,\mathcal{E})=\frac{Q}{\omega^2}\int_{\omega_g\pm\omega_{\kappa 0}}^\infty d\omega'(\omega'-\omega_g\mp\omega_{\kappa_0})^2\times\left\{\frac{1}{|\Omega|}\,\mathrm{Ai}\left(\frac{\omega'-\omega}{\Omega}\right)\right\}, \quad (3.19)$$

where

$$Q=\frac{e^2|\hat{\epsilon}\cdot\mathbf{P}|^2(n_{\kappa 0}+\frac{1}{2}\pm\frac{1}{2})}{2\pi m^2\hbar^4}\times(m_{ex}m_{ey}m_{ez}m_{hx}m_{hy}m_{hz})^{1/2}. \quad (3.20)$$

Again, as with the direct transitions, we may use Eq. (2.56) to find the zero-field limit of Eq. (3.19) which may be expressed as

$$\begin{aligned}\epsilon_2^{\rm ind}(\omega,0)&=(Q/\omega^2)(\omega-\omega_g\mp\omega_{\kappa 0})^2 && \omega>\omega_g\pm\omega_{\kappa 0}\\ &=0 && \omega<\omega_g\pm\omega_{\kappa 0}.\end{aligned} \quad (3.21)$$

Thus again we arrive at the desired result by comparing Eqs. (3.19) and (3.21) which is

$$\omega^2\epsilon_2(\omega,\mathcal{E})=\int_{-\infty}^\infty d\omega'\omega'^2\epsilon_2(\omega',0)\left\{\frac{1}{|\Omega|}\,\mathrm{Ai}\left(\frac{\omega'-\omega}{\Omega}\right)\right\} \quad (3.22)$$

and is the same as Eq. (3.15) and Eq. (3.1).

We will now show how the above equation may be employed to calculate zero-field quantities from finite-field quantities.

C. Optical Density of States from Finite-Field Data

The fact that Eq. (3.1) is applicable to both direct and indirect transitions in the limit of constant effective reduced mass, i.e., constant Ω, indicates that it is, to a certain extent, a general formula. In this light, we will then proceed to invert Eq. (3.1) and thereby derive equations that relate finite-field data to zero-field data. The zero-field imaginary part of the dielectric constant is proportional to the optical density of states; for example, for direct transitions, we have

$$\omega^2\epsilon_2(\omega,0)=\frac{4\pi^2e^2}{m^2}|\hat{\epsilon}\cdot\mathbf{P}|^2N(\hbar\omega), \quad (3.23)$$

where $N(\hbar\omega)$ is the density of states for the transition. Thus if we can calculate $\omega^2\epsilon_2(\omega,0)$, we have calculated the optical density of states within a constant of proportionality.

Using the completeness relation of the Airy function,[15]

$$\int_{-\infty}^\infty dt\,\mathrm{Ai}(t+x)\,\mathrm{Ai}(t+y)=\delta(x-y), \quad (3.24)$$

and assuming Ω is a constant, Eq. (3.22) may be inverted resulting in

$$\omega^2\epsilon_2(\omega,0)=\int_{-\infty}^\infty d\omega'\omega'^2\epsilon_2(\omega',\mathcal{E})\left\{\frac{1}{|\Omega|}\,\mathrm{Ai}\left(\frac{\omega-\omega'}{\Omega}\right)\right\}. \quad (3.25)$$

Another result may be derived relating differential data to zero-field data. Starting with either Eq. (3.22) or Eq. (3.25) it may be shown that

$$\frac{d^3}{d\omega^3}[\omega^2\epsilon_2(\omega,0)]=\frac{1}{|\Omega|^2}\int_{-\infty}^\infty d\omega'\omega'^2\frac{3F}{2}\left\{\frac{\partial}{\partial F}\Delta\epsilon_2(\omega',\mathcal{E})\right\}\times\left\{\frac{1}{|\Omega|}\,\mathrm{Ai}\left(\frac{\omega-\omega'}{\Omega}\right)\right\}, \quad (3.26)$$

where $\Delta\epsilon_2(\omega,\mathcal{E})=\epsilon_2(\omega,\mathcal{E})-\epsilon_2(\omega,0)$. Since the differential data has finite amplitude about critical points and is zero elsewhere, we might expect the integration over all energies in Eq. (3.26) to give more accurate results than Eq. (3.25). It is possible to integrate Eq. (3.26) explicitly to evaluate $\omega^2\epsilon_2(\omega,0)$. The sum rule[26]

$$\int_0^\infty\omega\epsilon_2(\omega)d\omega=\tfrac{1}{2}\pi\omega_p^2 \quad (3.27)$$

requires $\epsilon_2(\omega)$ to fall off faster than $(1/\omega^2)$ as ω approaches infinity. Thus the constants of integration must all be zero and integration of the Airy function three times gives

$$\omega^2\epsilon_2(\omega,0)=-\int_{-\infty}^\infty d\omega'\omega'^2\frac{3F}{4}\left\{\frac{\partial}{\partial F}\Delta\epsilon_2(\omega',\mathcal{E})\right\}\times\left\{\frac{1}{|\Omega|}[\mathrm{Ai}(r)+r\,\mathrm{Ai}'(r)+r^2\,\mathrm{Ai}_1(r)]\right\}, \quad (3.28)$$

where

$$r=\left(\frac{\omega-\omega'}{\Omega}\right), \quad \mathrm{Ai}'(x)=\frac{d}{dx}[\mathrm{Ai}(x)],$$

[26] D. Pines, *Elementary Excitations in Solids* (W. A. Benjamin, Inc., New York, 1964), p. 136.

and

$$\mathrm{Ai}_1(x)=\int_0^\infty \mathrm{Ai}(t)dt. \tag{3.29}$$

Thus since the differential data $\Delta\epsilon_2(\omega,\mathcal{E})$ is finite over a limited energy range, Eq. (3.28) is easily applicable to numerical integration.

D. Real Part of Dielectric Constant

From the Kramers-Kronig or dispersion equations,[27] the real part of the dielectric constant is related to the imaginary part of the dielectric constant as

$$\epsilon_1(\omega)=1+\frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{d\omega'}{\omega'-\omega}[\epsilon_2(\omega')-\epsilon_2(-\omega')]. \tag{3.30}$$

Using Eq. (3.1) for $\epsilon_2(\omega,\mathcal{E})$ gives

$$\epsilon_1(\omega,\mathcal{E})=1+\frac{2}{\pi}\int_{-\infty}^{\infty}d\omega''\omega''^2\epsilon_2(\omega'',0)\times\mathcal{P}\int_{-\infty}^{\infty}\frac{d\omega'}{\omega'(\omega'^2-\omega^2)}\left\{\frac{1}{|\Omega|}\mathrm{Ai}\left(\frac{\omega''-\omega'}{\Omega}\right)\right\}. \tag{3.31}$$

It may be shown that

$$\mathcal{P}\int_{-\infty}^{\infty}\frac{dt}{t}\mathrm{Ai}(t+x)=-\pi\,\mathrm{Gi}(x), \tag{3.32}$$

where[25]

$$\mathrm{Gi}(x)=\frac{1}{\pi}\int_0^\infty ds\,\sin(sx+\tfrac{1}{3}s^3). \tag{3.33}$$

With a partial fraction expansion of the integrand of Eq. (3.31) and the employment of Eq. (3.32), we may express the real part of the dielectric constant in a uniform electric field near a critical point as

$$\epsilon_1(\omega,\mathcal{E})=1+\frac{1}{\omega^2}\int_{-\infty}^{\infty}d\omega'\omega'^2\epsilon_2(\omega',0)\left\{\frac{1}{\Omega}\mathrm{Gi}\left(\frac{\omega'-\omega}{\Omega}\right)\right\}-\frac{2}{\omega^2}\int_{-\infty}^{\infty}d\omega'\omega'^2\epsilon_2(\omega',0)\left\{\frac{1}{\Omega}\mathrm{Gi}\left(\frac{\omega'}{\Omega}\right)\right\}+\frac{1}{\omega}\int_{-\infty}^{\infty}d\omega'\omega'^2\epsilon_2(\omega',0)\left\{\frac{1}{\Omega}\mathrm{Gi}\left(\frac{\omega'+\omega}{\Omega}\right)\right\}. \tag{3.34}$$

As ω approaches zero, we have the limiting form

$$\epsilon_1(0,\mathcal{E})=1+\int_{-\infty}^{\infty}d\omega'\omega'^2\epsilon_2(\omega',0)\frac{1}{\Omega^3}\mathrm{Gi}''\left(\frac{\omega'}{\Omega}\right). \tag{3.35}$$

[27] J. M. Ziman, *Principles of the Theory of Solids* (Cambridge University Press, London, 1964), p. 222.

Knowing that $\mathrm{Gi}(x)$ satisfies the differential equation[25]

$$\mathrm{Gi}''(x)-x\,\mathrm{Gi}(x)+\frac{1}{\pi}=0 \tag{3.36}$$

leads us to the equation

$$\epsilon_1(0,\mathcal{E})=1-\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{d\omega'\omega'^2\epsilon_2(\omega',0)}{\Omega^3}+\int_{-\infty}^{\infty}\frac{d\omega'\omega'^2\epsilon_2(\omega',0)}{\Omega^3}\left\{\frac{\omega'}{\Omega}\mathrm{Gi}\left(\frac{\omega'}{\Omega}\right)\right\}. \tag{3.37}$$

From any of the above equations, it may be shown that

$$\epsilon_1(0,0)=1+\frac{2}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{d\omega'\epsilon_2(\omega',0)}{\omega'}. \tag{3.38}$$

Since $\epsilon_1(0,\mathcal{E})$ is the static dielectric constant in an electric field, Eq. (3.37) and Eq. (3.38) suggest that by measuring $\Delta\epsilon_1(0,\mathcal{E})=\epsilon_1(0,\mathcal{E})-\epsilon_1(0,0)$ we may derive information about the optical density of states of the solid.

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APPENDIX A: PROOF

In this Appendix we show that the quantities $X_{n'n}(\mathbf{k})$ defined by Eq. (2.21) are real if the crystal satisfies inversion symmetry. To do this, we expand the cell-periodic part of the Bloch function, $u_n(\mathbf{k},\mathbf{r})$ as

$$u_n(\mathbf{k},\mathbf{r})=\sum_m B_n{}^m(\mathbf{k})u_m(\mathbf{0},\mathbf{r}). \tag{A1}$$

The equations for the coefficients $B_n{}^m(\mathbf{k})$ are determined by the Hamiltonian

$$\left[-\frac{\hbar^2}{2m}\nabla^2+V(\mathbf{r})\right]e^{i\mathbf{k}\cdot\mathbf{r}}u_n(\mathbf{k},\mathbf{r})=E_n(\mathbf{k})\,e^{i\mathbf{k}\cdot\mathbf{r}}u_n(\mathbf{k},\mathbf{r}). \tag{A2}$$

The usual $\mathbf{k}\cdot\mathbf{p}$ perturbation treatment gives

$$0=B_n{}^\mu(\mathbf{k})\left[\frac{\hbar^2k^2}{2m}+E_\mu(\mathbf{0})-E_n(\mathbf{k})\right]+i\frac{\hbar^2}{m}\sum_\nu B_n{}^\nu(\mathbf{k})\mathbf{k}\cdot\langle u_\mu(\mathbf{0},\mathbf{r})|\nabla|u_\nu(\mathbf{0},\mathbf{r})\rangle, \tag{A3}$$

where $E_\mu(\mathbf{0})$ is the energy of the band μ at $\mathbf{k}=\mathbf{0}$.

If the phases of the wave functions at $\mathbf{k}=\mathbf{0}$ are chosen so that $u_n(\mathbf{0},\mathbf{r})$ is real for even parity and imaginary for odd parity,[19] then all coefficients of the unknown $B_n{}^\mu(\mathbf{k})$ are real and it therefore follows that the

$B_n{}^\mu(\mathbf{k})$ are also real. By definition,

$$X_{n'n}(\mathbf{k})=\int d^3r u_{n'}{}^*(\mathbf{k},\mathbf{r})\frac{\partial}{\partial k_x}u_n(\mathbf{k},\mathbf{r})$$
$$=\sum_{\mu\nu} B_{n'}{}^\mu(\mathbf{k})\frac{\partial}{\partial k_x}B_n{}^\nu(\mathbf{k})\langle u_\mu(\mathbf{0},\mathbf{r})\,|\,u_\nu(\mathbf{0},\mathbf{r})\rangle$$
$$=\sum_{\nu} B_{n'}{}^\nu(\mathbf{k})\frac{\partial}{\partial k_x}B_n{}^\nu(\mathbf{k}). \quad \text{(A4)}$$

The last expression is real, hence the quantities $X_{n'n}(\mathbf{k})$ are also real.

APPENDIX B: ALTERNATIVE DERIVATION OF $\epsilon_2(\omega,\mathcal{E})$ NEAR CRITICAL POINTS

In Sec. III A, Eq. (2.52) was modified for photon energies near critical point energies. In the following we propose to show that the same equations result from a different approach. Using the results of Elliott,[28] Tharmalingam,[11] and Aspnes,[15] the imaginary part of dielectric constant near critical points may be expressed as[15]

$$\epsilon_2(\omega,\mathcal{E})=\frac{4\pi^2B}{\hbar^2\omega^2|\theta_x\theta_y\theta_z|^{1/2}}\int_{-\infty}^{\infty}d\epsilon_x d\epsilon_y d\epsilon_z$$
$$\times\mathrm{Ai}^2\left(\frac{-\epsilon_x}{\hbar\theta_x}\right)\mathrm{Ai}^2\left(\frac{-\epsilon_y}{\hbar\theta_y}\right)\mathrm{Ai}^2\left(\frac{-\epsilon_z}{\hbar\theta_z}\right)$$
$$\times\delta(\hbar\omega_g-\hbar\omega+\epsilon_x+\epsilon_y+\epsilon_z), \quad \text{(B1)}$$

where $\hbar\omega_g$ is the energy gap and for the ith coordinate $\theta_i{}^3=F_i{}^2/2\hbar\mu_i$, where $F_i=e\mathcal{E}_i$, $\mathcal{E}_i$ is the component of the field in the ith direction, and μ_i is the reduced effective mass in the same direction. The constant B is defined in Eq. (3.10). We may use the δ function to evaluate the ϵ_z integral and the result may be expressed as

$$\epsilon_2(\omega,\mathcal{E})=\frac{4\pi^2B|\theta_x\theta_y|}{\omega^2|\theta_x\theta_y\theta_z|^{1/2}}\int_{-\infty}^{\infty}dr\,ds\,\mathrm{Ai}^2(r)\,\mathrm{Ai}^2(s)$$
$$\times\mathrm{Ai}^2\left(\frac{\omega_g-\omega}{\theta_z}-\frac{\theta_x r}{\theta_z}-\frac{\theta_y s}{\theta_z}\right), \quad \text{(B2)}$$

where $r=-(\epsilon_x/\hbar\theta_x)$ and $s=-(\epsilon_y/\hbar\theta_y)$. The function $\mathrm{Ai}^2(x)$ is related to a single Airy function by the integral[15]

$$\mathrm{Ai}^2(x)=\frac{1}{\kappa\pi}\int_0^\infty\frac{dt}{\sqrt{t}}\mathrm{Ai}(t+\kappa x), \quad \text{(B3)}$$

where $\kappa=2^{2/3}$. Let us now define for the ith coordinate

$$\Omega_i=\frac{\theta_i}{\kappa}, \quad \text{(B4)}$$

and apply Eq. (B3) to Eq. (B2) with the result

$$\epsilon_2(\omega,\mathcal{E})=\frac{B|\Omega_x\Omega_y|}{2\pi\omega^2|\Omega_x\Omega_y\Omega_z|^{1/2}}\int_0^\infty\frac{d\alpha d\beta d\gamma}{(\alpha\beta\gamma)^{1/2}}$$
$$\times\int_{-\infty}^{\infty}dr'ds'\,\mathrm{Ai}(\alpha+r')\,\mathrm{Ai}(\beta+s')$$
$$\times\mathrm{Ai}\left(\gamma+\frac{\omega_g-\omega-\Omega_x r'-\Omega_y s'}{\Omega_z}\right), \quad \text{(B5)}$$

where

$$r'=\kappa r \quad \text{and} \quad s'=\kappa s, \quad \text{(B6)}$$

and α, β, γ are dummy variables resulting from the use of Eq. (B4). The integrals over r' and s' may be evaluated using the formula[15]

$$\int_{-\infty}^{\infty}dt\,\mathrm{Ai}(x+t)\,\mathrm{Ai}(y-zt)=\frac{1}{|1+z^3|^{1/3}}\mathrm{Ai}\left[\frac{y+zx}{(1+z^3)^{1/3}}\right], \quad \text{(B7)}$$

which reduces the three Airy functions in Eq. (B5) to a single Airy function

$$\epsilon_2(\omega,\mathcal{E})=\frac{B|\Omega_x\Omega_y\Omega_z|^{1/2}}{2\pi\omega^2}\int_0^\infty\frac{d\alpha d\beta d\gamma}{(\alpha\beta\gamma)^{1/2}}$$
$$\times\left\{\frac{1}{|\Omega|}\mathrm{Ai}\left[\frac{\Omega_x\alpha+\Omega_y\beta+\Omega_z\gamma+(\omega_g-\omega)}{\Omega}\right]\right\}, \quad \text{(B8)}$$

where

$$\Omega^3=\Omega_x{}^3+\Omega_y{}^3+\Omega_z{}^3. \quad \text{(B9)}$$

Equation (B8) is essentially the same form of equation as Eq. (3.5) and may be put in the same form by defining

$$\epsilon_x=\hbar|\Omega_x|\alpha,\ \epsilon_y=\hbar|\Omega_y|\beta, \quad \text{and} \quad \epsilon_z=\hbar|\Omega_z|\gamma, \quad \text{(B10)}$$

and introducing the signs of the reduced masses, i.e., the signs of the Ω_i, explicitly into the equation as was done in Eq. (3.5). Since we have shown that Eq. (B9) is equivalent to Eq. (3.5) we need not repeat the computations between Eq. (3.5) and the final result, but may just express the final result for the four types of critical points as given in Eqs. (3.14).

Note added in proof. In a recent treatment of the effect of electric field on the absorption edges of insulating crystals, Rees [H. D. Rees, J. Phys. Chem. Solids (to be published); Solid State Commun. **5**, 365 (1967)] has derived a relation connecting the field effect and zero-field spectrums similar to Eq. (3.1). Rees, using a representation in which the electric field appears as a force term in the equation of motion, treats low-angle scattering by an adiabatic approximation.

[28] R. J. Elliott, Phys. Rev. **108**, 1384 (1957).