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PHYSICAL REVIEW B VOLUME 4, NUMBER 6 15 SEPTEMBER 1971

Excitonic Effects on Dielectric Properties of Solids in a Uniform Electric Field

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(Received 8 February 1971)

The imaginary part of the dielectric function for excitonic transitions and different types of critical points of a solid in a uniform electric field are presented in a closed form. The time-dependent Schrödinger equation, with a time-dependent gauge for the applied electric field which includes the electron-hole interaction forces is treated within the effective-mass approximation. In the weak fields and using the two-band approximations, the results can be expressed in terms of Airy functions. The peak position of the exciton lines is found to shift to lower energies and then move to higher energies as the electric field increases. The amplitude of the peak decreases as the electric field increases. The electron-hole interaction can be neglected in calculating the electric-field-induced change of the dielectric constant, when the applied electric field is much larger than the effective field of the electron-hole interaction.

I. INTRODUCTION

The electro-optic or Franz-Keldysh effect has been fruitful in identifying energies of solids at which critical points occur.[1] However, the one-electron uniform-electric-field theory can not explain the experimental results.[2] Some authors have pointed out that other effects, such as nonuniformity of the modulating electric field,[3] the Coulomb interaction,[2,4] and collision broadening[5] should be properly included in the theory.

In order to describe the effect of nonuniform fields, Aspnes and Frova[6] have proposed an averaging procedure, based on an approximate solution of Maxwell's equations for inhomogeneous media, and a one-electron theory and an exponentially decaying electric field[7] have been used to derive the field-induced change in dielectric function. This explains the gross features of electroreflectance. The validity of the one-electron approximation for interband transitions is based on the assumption that the Coulomb interaction between electrons and holes is weak. In order to provide an adequate interpretation of the optical experiments, a qualitative discussion by Hamakawa *et al.*[2,8] shows that the broadening of exciton lines by the electric field produces additional peaks in the field-induced change in dielectric function. Duke and Alferieff[9] have used a semiquantitative theory of this effect to discuss the optical absorption in semiconductors. Recently, Penchina, Pribram, and Sak[10] and Rowe and Aspnes[11] used the Koster-Slater model, while Ralph,[12] Dow and Redfield,[13] and Blossey[14] solved the effective-mass equation numerically to discuss the excitonic effects on the optical absorption of solids. Enderlein[15] used the Green's-function approach to solve the problem, but one of his assumptions made his result only valid in the limit of zero electron-hole interaction.[10,16]

We present here a theoretical calculation which includes the electron-hole interaction and is based on the assumptions that include the validity of the effective-mass approximation and the uniform electric field.

II. MODEL OF PROBLEM

A. Wave Functions

We consider a model of an electronic Hamiltonian

in a uniform electric field which consists of the electron-hole interaction potential and the linear electric field in nonstationary Schrödinger equation. In a static-lattice and two-band model, the nonstationary initial and final states are electric-field-perturbed valence- and conduction-bands states, respectively. The wave function of each band can be solved in terms of the wave functions in the absence of external electric field. Therefore, we achieve a solvable model giving the absorption coefficient, or dielectric function, which is expressed in a closed form.

The Schrödinger equation for the Hamiltonian in an external electric field $\vec{\mathcal{E}}$ is given as

$$H_s \psi_{n\vec{l}m}(\vec{r}, t) = i\hbar \frac{\partial}{\partial t} \psi_{n\vec{l}m}(\vec{r}, t) , \tag{1}$$

where

$$H_s = \sum_i \frac{1}{2m_i} \left(P_i - \frac{e}{c} \vec{A} \right)^2 + V(\vec{r}) , \tag{2}$$

$i = x, y, z$ and $\vec{A} = -c\vec{\mathcal{E}}t$, $V(\vec{r})$ is the electron-hole interaction potential, and the external electric field is assumed to be in x direction.

Defining

$$H_0 = -\sum_i (\hbar^2/2m_i) \nabla^2 + V(\vec{r}) \tag{3}$$

and

$$H_0 e^{-i\hbar^{-1}E_n t} \varphi_n(\vec{k}, \vec{r}) = E_n e^{-i\hbar^{-1}E_n t} \varphi_n(\vec{k}, \vec{r}) , \tag{4}$$

we have

$$\left(\hbar^{-1} H_0 - i\lambda t \frac{\partial}{\partial x} + \eta^3 t^2 \right) \psi_{n\vec{l}m}(\vec{r}, t) = i \frac{\partial}{\partial t} \psi_{n\vec{l}m}(\vec{r}, t) , \tag{5}$$

where $\lambda = e\mathcal{E}/m_x$ and $\eta^3 = (e\mathcal{E})^2/2m_x\hbar$.

It is shown in Appendix A that the wave function

$$\psi_{n\vec{l}m}(\vec{r}, t) = \exp(-\tfrac{1}{3} i \eta^3 t^3 - i\hbar^{-1} H_0 t) \times \exp[i\hbar^{-1} I(\vec{r}, t)] \varphi_n(\vec{k}, \vec{r} - \tfrac{1}{2}\lambda t^2) . \tag{6}$$

It should be noted that $I(\vec{r}, t)$ is a function of coordinates and time,

$$I(\vec{r}, t) = t \sum_{m=0}^{\infty} (-1)^m \left[\left(t^2 \lambda \frac{\partial}{\partial x} \right)^{m+1} \Big/ 3 \cdot (3+2) \cdots (3+2m) \right] V(\vec{r}) . \tag{7}$$

As shown in Eq. (6) the wave function $\psi_{n\vec{l}m}(\vec{r}, t)$ is equivalent to a unitary operator multiplying on the wave functions in the absence of the external electric field. For this reason we may express $\psi_{n\vec{l}m}(\vec{r}, t)$ in terms of the linear combinations of $\varphi_n(\vec{k}, \vec{r})$:

$$\psi_{n\vec{l}m}(\vec{r}, t) = \sum_\nu C_{n\nu}(\vec{k}, t) \varphi_\nu(\vec{k}, \vec{r}) , \tag{8}$$

where

$$C_{n\nu}(\vec{k}, t) = \exp[-(\tfrac{1}{3} i \eta^3 t^3 + i\hbar^{-1} E_n) \delta_{n\nu}] \times \exp[i\hbar^{-1} \langle I(\vec{r}, t) \rangle_{n\nu}] , \tag{9}$$

$$\langle I(\vec{r}, t) \rangle_{n\nu} = \int_\nu \varphi_\nu^*(\vec{k}, \vec{r}) I(\vec{r}, t) \varphi_n(\vec{k}, \vec{r} - \tfrac{1}{2}\lambda t^2) d^3r . \tag{10}$$

B. Transition Rate and Dielectric Function

We introduce the photon perturbation which is represented in a semiclassical approximation by the vector potential

$$\vec{B} = \hat{\epsilon} B_0 e^{i(\vec{q}\cdot\vec{r} - \omega t)} e^{b' t} , \tag{11}$$

where b' is a positive number which will be taken to the zero limit.

The transition rate for two-band approximation is expressed as

$$T(\vec{k}) = \lim_{b' \to 0} \frac{\partial}{\partial t} |g_{21}|^2 . \tag{12}$$

The coefficient g_{21} can be found from time-dependent perturbation theory[17]:

$$g_{21} = \frac{ieB_0}{m\hbar c} \langle \varphi_c | \hat{\epsilon} \cdot \vec{P} | \varphi_v \rangle \int_{-\infty}^{t} \exp(+i\omega t' + b' t') \times \exp[-i\hbar^{-1} E_{cv}(\vec{k}) t' - \tfrac{1}{3} i \Theta_c^3 t'^3 + i\hbar^{-1} \Delta \langle I(\vec{r}, t') \rangle_{cv}] dt' , \tag{13}$$

where

$$\Theta_c^3 = (e\mathcal{E})^2/2\mu_x\hbar , \tag{14a}$$

$$E_{cv}(\vec{k}) = E_c(\vec{k}) - E_v(\vec{k}) , \tag{14b}$$

$$\Delta \langle I(\vec{r}, t') \rangle_{cv} = \langle I(\vec{r}, t') \rangle_{cv} - \langle I(\vec{r}, t) \rangle_{vv'} , \tag{15}$$

$\mu_x = m_{xe} m_{xh}/(m_{xe} + m_{xh})$ is the effective mass of the electron and hole, and Θ_c is the characterisitic frequency. One should bear in mind that φ_v and φ_c in Eq. (13) correspond to the valence band [with effective masses $m_i = -m_{ih}$, wave vector $-\vec{k}$, and energy $-E_v(\vec{k})$] and conduction band [with effective masses $m_i = m_{ie}$, wave vector $+\vec{k}$, and energy $+E_c(\vec{k})$], respectively, and are given in Eq. (8).

The argument in Eq. (13) is the difference of eigenvalue between the conduction and valence bands. Expanding the exponential function of Eq. (13) in a Fourier integral and evaluating the t' integral, the coefficient g_{21} becomes

$$g_{21} = \frac{ieB_0}{m\hbar c} \langle \varphi_c | \hat{\epsilon} \cdot \vec{P} | \varphi_v \rangle \times \int_{-\infty}^{\infty} d\alpha \, F(\hbar^{-1} E_{cv}(\vec{k}) - \omega + 2\pi\alpha, \mathcal{E}) \times \frac{e^{i2\pi\alpha t + b' t}}{i2\pi\alpha + b'} , \tag{16}$$

where

$$F(\hbar^{-1}E_{cv}(\vec{k}) - \omega + 2\pi\alpha, \mathcal{E}) = \int_{-\infty}^{\infty} \exp[-i(\hbar^{-1}E_{cv}(\vec{k}) - \omega + 2\pi\alpha)t + i\hbar^{-1}\langle I(\vec{r}, t)\rangle_{cv}] \exp(-i\tfrac{1}{3}\Theta_c^3 t)\, dt . \quad (17)$$

The imaginary part of the dielectric function can be expressed as

$$\epsilon_2(\omega, \mathcal{E}) = (2\pi c^2\hbar/\omega^2 B_0^2)\, W_{tot} , \quad (18)$$

where W_{tot} is the total transition rate of a solid,

$$W_{tot} = \sum_{\vec{k}} T(\vec{k}) . \quad (19)$$

Since the electron-hole interaction is taken into account we have two kinds of states, i.e., bound states and continuum states.

Now we write $\epsilon_2(\omega, \mathcal{E})$ in the form

$$\epsilon_2(\omega, \mathcal{E}) = \epsilon_2(\omega, \mathcal{E})_{bound} + \epsilon_2(\omega, \mathcal{E})_{cont} , \quad (20)$$

where $\epsilon_2(\omega, \mathcal{E})_{bound}$ follows from Eq. (18) for the summation restricted to the bound states, and $\epsilon_2(\omega, \mathcal{E})_{cont}$ corresponds to the case where the summation is carried out only over continuum states.

III. BOUND STATES

A. Coulomb Interaction

The effects of Coulomb interaction between electrons and holes at different critical points have been studied by Elliott,[18] Velicky and Sak,[19] and Kane[20] in the effective-mass approximation and the absence of an electric field. If we consider the electron-hole Coulomb interaction in Eq. (10), we have

$$\langle I(\vec{r}, t)\rangle_{nv} = -\int_v \varphi_v^*(\vec{k}, \vec{r}) \left\{ t \sum_{m=0}^{\infty} (-1)^m \left[\left(t^2\lambda \frac{\partial}{\partial x} \right)^{m+1} \Big/ 3\cdot(3+2)\cdots(3+2m) \right] \frac{e^2}{\epsilon_0 r} \right\} \varphi_n(\vec{k}, \vec{r} - \tfrac{1}{2}\lambda t^2) , \quad (21)$$

where ϵ_0 is the dielectric constant of the medium in which the system is considered.

In the weak-field approximation we may keep the lowest-order term of Eq. (21) which describes the electric field and dipole interaction. The dipole moments will have a random orientation in space and their magnitude will have statistical distribution around some average in the direction of the electric field, so we would expect that Eq. (15) becomes

$$\Delta\langle I(\vec{r}, t)\rangle_{cv} = \tfrac{1}{3} t^3 (e^2/2\mu_x)\, \mathcal{E}\mathcal{E}_H , \quad (22)$$

where

$$\mathcal{E}_H = \frac{2\mu_x}{e} \left[\int_v \frac{1}{m_{xe}} \varphi_v^*(\vec{k}, \vec{r}) \left(\frac{\partial}{\partial x} \frac{-e^2}{\epsilon_0 r} \right) \varphi_c(\vec{k}, \vec{r})\, d^3r + \int_v \frac{1}{m_{xh}} \varphi_{v'}^*(\vec{k}, \vec{r}) \left(\frac{\partial}{\partial x} \frac{-e^2}{\epsilon_0 r} \right) \varphi_v(\vec{k}, \vec{r})\, d^3r \right] . \quad (23)$$

Furthermore, we would expect that the interaction will broaden the linewidth. Substituting Eq. (23) into Eq. (17), we obtain

$$g_{21} = \frac{ieB_0}{m\hbar c} \langle \varphi_c | \hat{\epsilon}\cdot\vec{P} | \varphi_v \rangle \times 2\pi \int_{-\infty}^{\infty} d\alpha\, \mathrm{Ai}\left(\frac{\hbar^{-1}E_{cv}(\vec{k}) - \omega + 2\pi\alpha}{\Lambda_H} \right) \times \frac{1}{\Lambda_H} \frac{e^{i2\pi\alpha t + b't}}{i2\pi\alpha + b'} , \quad (24a)$$

where

$$\Lambda_H^3 = \Theta_c^3 - (e^2\mathcal{E}\mathcal{E}_H)/2\mu_x\hbar \quad (24b)$$

and

$$\mathrm{Ai}(x) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-(i/3)s^3 - ixs}\, ds . \quad (24c)$$

Direct Transitions

Substituting Eqs. (24) into (12), we have

$$T(\vec{k}) = \frac{2\pi(eB_0)^2}{(m\hbar c)^2} |\langle \varphi_c | \hat{\epsilon}\cdot\vec{P} | \varphi_v \rangle|^2 \times \frac{1}{|\Lambda_H|} \mathrm{Ai}\left(\frac{\hbar^{-1}E_{cv}(\vec{k}) - \omega}{\Lambda_H} \right) . \quad (25)$$

From Eqs. (18) and (20), we have the imaginary part of the dielectric function

$$\epsilon_2(\omega, \mathcal{E})_{bound} = \sum_{\text{bound states}} \frac{4\pi^2 e^2}{\omega^2 \hbar m^2} |\langle \varphi_c | \hat{\epsilon}\cdot\vec{P} | \varphi_v \rangle|^2 \times \frac{1}{\hbar|\Lambda_H|} \mathrm{Ai}\left(\frac{E_{cv}(\vec{k}) - \hbar\omega}{\hbar\Lambda_H} \right) . \quad (26)$$

As the zero-field limit is approached

$$\lim_{\mathcal{E}\to 0} \frac{1}{\hbar|\Lambda_H|} \mathrm{Ai}\left(\frac{E_{cv}(\vec{k}) - \hbar\omega}{\hbar\Lambda_H} \right) = \delta(E_{cv}(\vec{k}) - \hbar\omega) . \quad (27)$$

It is easy to see that the expression for the imaginary part of the dielectric function reduces to the expressions of zero applied electric field.[18-20]

Indirect Transitions

For the indirect transition, we have to carry out a sum over the center of mass as well as the relative vector. The sum over the center-of-mass

states can be expressed as[1]

$$\sum_{\text{c.m.}} \rightarrow \frac{(2M_x M_y M_g)^{1/2}}{2\pi^2\hbar^3}\int_0^\infty dE_{\text{c.m.}}\,(E_{\text{c.m.}})^{1/2}\,, \tag{28}$$

where $E_{\text{c.m.}}$ is the center-of-mass energy of the electron-hole pair and $M_i = m_{ie} + m_{ih}$. The imaginary part of the dielectric constant in an electric field for indirect transition is given as

$$\epsilon_2^{\text{ind}}(\omega, \mathcal{E})_{\text{bound}} = D \sum_{\text{bound states}} (n_{\vec{k}_0} + \tfrac{1}{2} \pm \tfrac{1}{2}) \int_{E_g \pm \hbar\omega_{\vec{k}_0}}^\infty dE$$

$$\times (E - E_g \mp \hbar\omega_{\vec{k}_0})^{1/2} \frac{1}{\hbar|\Lambda_H|}\,\text{Ai}\left(\frac{E + E_{cv}(\vec{k}) - E_g - \hbar\omega}{\hbar\Lambda_H}\right), \tag{29}$$

$$D = \frac{(2M_x M_y M_g)^{1/2}}{2\pi^2\hbar^3}\,\frac{4\pi^2 e^2}{\omega^2\hbar m^2}\,|\langle\varphi_c|\hat{\epsilon}\cdot\vec{P}|\varphi_v\rangle|^2\,, \tag{30}$$

where, in Penchina's notation,[21] $\hbar\omega_{\vec{k}}$ is the energy of phonon and the upper and lower signs of $(n_{\vec{k}_0} + \frac{1}{2} \pm \frac{1}{2})$ refer to the emission and absorption of energy $\hbar\omega_{\vec{k}_0}$. We have defined a change of variable $E = E_{\text{c.m.}} + E_g \pm \hbar\omega_{\vec{k}_0}$.

Equations (26) and (29) provide the expressions of the optical absorption of excitons in a uniform electric field. A simple case which has been discussed in the absence of external electric field[18] and will be examined in detail (since it has a well-known analytic solution) is that of two spherical single bands with effective masses m_e and m_h, which has a same equation as hydrogen atom. For bound states,[18] $\varphi_n(\vec{k}, \vec{r})$ is the normalized hydrogen-atom wave function with reduced mass $\mu = m_e m_h/(m_e + m_h)$ and eigenvalue

$$E_{cv}(\vec{k}) = E_g - G/n^2 + \hbar^2 K^2/2(m_e + m_h)\,, \tag{31}$$

where $G = \mu e^4/2\hbar^2\epsilon_0^2$, $\vec{K} = \vec{K}_e + \vec{K}_h$, and n is the principle quantum number.

The last term of Eq. (31) is the kinetic energy of center-of-mass motion. A few features of the electric field effect on the excitonic lines can be seen from Eq. (10). If we assume $x = r\cos\Theta$ in spherical coordinates and take the dipole term into account, the ground state (1S) of hydrogen is nondegenerate and even parity; $\langle\partial V(\vec{r})/\partial x\rangle_{11} = 0$. Hence no linear Stark effect occurs, and there is no permanent dipole moment. Therefore we would expect that $\langle\partial V(\vec{r})/\partial x\rangle_{1\nu} \neq 0$ is contributed from the 2P states, and higher energy levels which satisfy the selection rule will effect the amplitude of $\langle\partial V(\vec{r})/\partial x\rangle_{cv}$. For instance, the expectation value of $\Delta\langle(\partial/\partial x)(-e^2/\epsilon_0 r)\rangle_{1S,2P}$ between 1S and 2P states is

$$\Delta\left\langle\frac{\partial}{\partial x}\left(-\frac{e^2}{\epsilon_0 r}\right)\right\rangle_{1S,2P} = \frac{2\sqrt{2}}{27}\cdot\frac{e^2}{\epsilon_0 a^2}\,, \tag{32a}$$

where

$$a = (4\pi t_0\hbar^2)/\mu e^2\,. \tag{32b}$$

It is evident that the electric field has little effect on the 1S hydrogenic level. The situation is different for the excited states, of which we shall treat the lowest one. The 2S state and three 2P states are degenerate, the former being of even parity, whereas the latter three are odd. Hence, the linear Stark effect exists. The corresponding electric field has more effect on the $n = 2$ exciton lines.

Because of conservation of momentum, direct transitions take place at $\vec{K} = 0$ states. Substituting Eq. (31) into Eqs. (26) and (29), we have

$$\epsilon_2(\omega, \mathcal{E})_{\text{bound}} = \sum_{n=1}^\infty \frac{4\pi^2 e^2}{\omega^2\hbar m^2}\,|\langle\varphi_c|\hat{\epsilon}\cdot\vec{P}|\varphi_v\rangle|^2\,\frac{1}{\hbar|\Lambda_H|}\,\text{Ai}\left(\frac{E_g - G/n^2 - \hbar\omega}{\hbar\Lambda_H}\right), \quad \text{direct transitions} \tag{33a}$$

$$\epsilon_2^{\text{ind}}(\omega, \mathcal{E})_{\text{bound}} = D\sum_{n=1}^\infty (n_{\vec{k}_0} + \tfrac{1}{2} \pm \tfrac{1}{2}) \int_{E_g \pm \hbar\omega_{\vec{k}_0}}^\infty dE\,(E - E_g \mp \hbar\omega_{\vec{k}_0})^{1/2}\,\frac{1}{\hbar|\Lambda_H|}\,\text{Ai}\left(\frac{E - G/n^2 - \hbar\omega}{\hbar\Lambda_H}\right),$$

$$\text{indirect transitions}\,. \tag{33b}$$

A few general features of the excitonic effect on the electroreflectance can be seen from the effective characteristic frequency Λ_H. We see from Eq. (33) that the exciton lines broaden in an electric field. The peak position of the line also changes in an electric field. We focus our attention on Eq. (33) where the peak position is located at

$$\frac{E_g - G/n^2 - \hbar\omega}{\hbar\Lambda_H} = -S_i\,, \tag{34}$$

where the maximum of $\text{Ai}(x)$ is located at $-S_i$.

From Eq. (24), we have

$$E_g - \frac{G}{n^2} - \hbar\omega = -\hbar\left(\frac{e^2}{2\mu\hbar}\right)^{1/3}(\mathcal{E}^2 - \mathcal{E}\mathcal{E}_H)^{1/3}S_i\,. \tag{35}$$

As shown in Eq. (23) we would expect $\mathcal{E}_H$ to be the order of electron charge divided by the Bohr radius for the exciton. To discuss the peak position of the exciton, we consider three regions, i.e., $|\mathcal{E}| \gg |\mathcal{E}_H|$, $|\mathcal{E}| \ll |\mathcal{E}_H|$, and $|\mathcal{E}|$ are comparable with $|\mathcal{E}_H|$.

Region 1: $|\mathcal{E}| \ll |\mathcal{E}_H|$

In this region we can express Eq. (35) approximately as

$$\hbar\omega \approx E_g - \frac{G}{n^2} - \hbar\left(\frac{e^2}{2\mu\hbar}\right)^{1/3} (\mathcal{E}\mathcal{E}_H)^{1/3}\left(1 - \frac{1}{3}\frac{\mathcal{E}}{\mathcal{E}_H}\right). \tag{36}$$

Since $|\mathcal{E}/\mathcal{E}_H| \ll |$, the peak position of the line is shifted to lower energies and the amplitude of the peak decreases as the electric field increases. These results agree qualitatively with the numerical calculation of Ralph,[12] Dow and Redfield,[13] and Blossey,[14] where equation is solved numerically. It is obvious that the peak moves to higher energies as electric field increasing after the peak position of the line shifted to lower energies.

Region 2: $|\mathcal{E}| \approx |\mathcal{E}_H|$

Since the amplitudes of $|\mathcal{E}|$ and $|\mathcal{E}_H|$ are comparable we have to take the higher-order terms of $\Delta\langle I(\vec{r}, t)\rangle_{cv}$ into account for the electroabsorption of the exciton. As shown in Appendix B the imaginary part of the dielectric function can be calculated from the Fourier transformation

$$F(\hbar^{-1}E_{cv}(\vec{k}) - \omega, \mathcal{E}).$$

Region 3: $|\mathcal{E}| \gg |\mathcal{E}_H|$

In this region Eq. (35) becomes

$$\hbar\omega \approx E_g - \frac{G}{n^2} + \hbar\left(\frac{e^2}{2\mu\hbar}\right)^{1/3} (\mathcal{E}^2)^{1/3}\left(1 - \frac{1}{3}\frac{\mathcal{E}_H}{\mathcal{E}}\right) S_i . \tag{37}$$

It should be mentioned here that the electron-hole interaction can be neglected in this region. Therefore the expression for the imaginary part of the dielectric function, Eqs. (26) and (29), can be reduced to the exact expression of reference[1] where the electron-hole interaction is neglected.

Enderlein[15] has performed a Green's-function approach to solve the problem and has used the time-dependent gauge. But one of his assumptions, i.e., $\partial V(\vec{r})/\partial z = 0$, made his results only valid in the limit $\mathcal{E} \gg \mathcal{E}_H$ and therefore the excitonic effect on the electroabsorption can be neglected.[22] This point can be seen clearly in Eq. (21).

B. Koster-Slater Model

The electron-hole interaction in this case is of extremely short-range nature, in contrast to the Coulomb forces. There is the advantage that this problem is exactly soluble, and both one- and three-dimensional cases of the Koster-Slater type have been solved[10,11] and the results expressed in relatively well-known functions. In this section we will show that the effect of nonhydrogenic excitons on electroabsorption can be expressed in terms of Airy functions with a slightly different argument from the hydrogenic excitons.

We shall however treat the simplest model of a Koster-Slater contact interaction potential $V(\vec{r}) = g\delta(\vec{r})$, $g < 0$, which equals zero except when the Wannier electron and hole are on the same unit cell, as a limiting case of the Coulomb-interaction potential. The influence of the electron-hole interaction on the expansion coefficient $C_{n\nu}(\vec{k}, t)$ is just contained in $\langle I(\vec{r}, t)\rangle_{n\nu}$. The differentiation of a Dirac's function has the property[23]

$$\delta^{(m)}(x) = \int_{-\infty}^{\infty} (-\gamma 2\pi i)^m e^{-2\pi i\gamma x} d\gamma . \tag{38}$$

Substituting Eq. (38) into Eq. (7) and evaluating the power series, we obtain

$$I(\vec{r}, t) = -g\,\delta(y)\,\delta(z)$$
$$\times\left[t\int_{-\infty}^{\infty}(e^{i\pi t^2\lambda\gamma}\int_0^1 e^{-i\pi t^2\lambda\gamma\eta'^2} d\eta' - 1)\, e^{-2\pi i\gamma x} d\gamma\right], \tag{39}$$

where we have used the relation $\delta(\vec{r}) = \delta(x)\,\delta(y)\,\delta(z)$. Evaluating the integral over γ, we have

$$I(\vec{r}, t) = -t\left(\frac{g}{2t}\int_{-t}^{t}\delta(\vec{r} - \tfrac{1}{2}\lambda t^2 + \tfrac{1}{2}\lambda\xi^2)\, d\xi - g\,\delta(\vec{r})\right). \tag{40}$$

Substituting Eq. (40) in (10), we have

$$\langle I(\vec{r}, t)\rangle_{n\nu} = -g\,t\left(\frac{1}{2t}\int_{-t}^{t}\varphi_n^*(-\tfrac{1}{2}\lambda\xi^2)\right.$$
$$\left.\times\varphi_\nu(\tfrac{1}{2}\lambda t^2 - \tfrac{1}{2}\lambda\xi^2)\, d\xi - \varphi_n^*(-\tfrac{1}{2}\lambda t^2)\,\varphi_\nu(0)\right). \tag{41}$$

The simplest possible approximation of Eq. (41) for finite fields is to expand $\langle I(\vec{r}, t)\rangle_{n\nu}$ in a Taylor series in terms of the field and keeping only the lowest-order term. Equation (41) becomes

$$\langle I(\vec{r}, t)\rangle_{n\nu} = -g\tfrac{1}{3}(e\mathcal{E}/\mu_x)$$
$$\times t^3(\varphi_n'(0)\,\varphi_\nu(0) + \varphi_n(0)\,\varphi_\nu'(0)) + \cdots . \tag{42}$$

Defining $\varphi_{n\nu}(x) = \varphi_n^*(x)\,\varphi_\nu(x)$ and substituting Eq. (42) into Eq. (15), we obtain

$$\Delta\langle I(\vec{r}, t)\rangle_{cv} = \tfrac{1}{3}t^3(e^2\mathcal{E}\mathcal{E}_N/2\mu_x), \tag{43}$$

where

$$\mathcal{E}_N = -\frac{2\mu_x}{e}g\left(\frac{\varphi_{cv}'(0)}{m_{xe}} + \frac{\varphi_{vv}'(0)}{m_{xh}}\right). \tag{44}$$

We shall now compare Eq. (43) for the case of Koster-Slater model with that of the Coulomb interaction. At weak fields, if

$$\Lambda_N = (e\mathcal{E})^2/2\mu_x\hbar - (e^2\mathcal{E}/2\mu_x\hbar)\,\mathcal{E}_N , \tag{45}$$

the imaginary part of the dielectric function for the exciton lines can be expressed as

$$\epsilon_2(\omega, \mathcal{E})_{\text{bound}} = \sum_{\text{bound states}} \frac{4\pi^2 e^2}{\omega^2 \hbar m^2} |\langle \varphi_c | \hat{\epsilon} \cdot \vec{P} | \varphi_v \rangle|^2 \frac{1}{\hbar |\Lambda_N|} \text{Ai}\left(\frac{E_{cv}(\vec{k}) - \hbar\omega}{\hbar \Lambda_N}\right), \quad \text{direct transitions} \tag{46a}$$

$$\epsilon_2^{\text{ind}}(\omega, \mathcal{E})_{\text{bound}} = D \sum_{\text{bound states}} (n_{\vec{k}_0} - \tfrac{1}{2} \pm \tfrac{1}{2}) \int_{E_g \pm \hbar\omega_{\vec{k}_0}}^{\infty} dE\, (E - E_g \mp \hbar\omega)^{1/2} \frac{1}{\hbar |\Lambda_N|} \text{Ai}\left(\frac{E + E_{cv}(\vec{k}) - E_g - \hbar\omega}{\hbar \Lambda_N}\right),$$

$$\text{indirect transitions} \tag{46b}$$

where $E_{cv}(\vec{k})$ is the peak position of the exciton lines in the absence of the electric field.

We have evaluated the imaginary part of the dielectric function, for direct and indirect transitions, and of exciton lines in the presence of a uniform electric field including the electron-hole contact potential $g\,\delta(\vec{r})$. The results are expressed in the same form as the Coulomb potential, but the argument of the Airy function is slightly different for the effective electric field $\mathcal{E}_N$.

IV. CONTINUUM STATES

As shown in Sec. III, the effect of electron-hole interaction on the imaginary part of the dielectric function appears in terms of $\langle I(\vec{r}, t)\rangle_{nv}$ in Eq. (10). The imaginary part of the dielectric function for continuum states can be written[11,22] as

$$\epsilon_2(\omega, \mathcal{E})_{\text{cont}} = \frac{8\pi^3 e^2}{\omega^2 m^2 \hbar} \frac{2}{(2\pi)^3} \int_{\text{BZ}} d^2 k_\perp \int_{-Kx/2}^{Kx/2} dk_x \times \int_{-\infty}^{\infty} d\beta \frac{1}{|\Lambda_i|^2} |\langle \varphi_c | \hat{\epsilon} \cdot \vec{P} | \varphi_v \rangle|^2 \text{Ai}\left(\frac{E_{cv}(\vec{k}) - \hbar\omega}{\hbar \Lambda_i}\right) \times \text{Ai}\left(\frac{E_{cv}(\vec{k}) - \hbar\omega - 2\pi\beta\hbar}{\hbar \Lambda_i}\right) e^{-2\pi i \beta t}, \tag{47}$$

where Λ_i stands for Λ_H of Coulombic potential or Λ_N of Koster-Slater contact potential and K_x is the width of Brillouin zone (BZ) in x direction. We assume that $\vec{\mathcal{E}}$ is along the direction of some reciprocal-lattice vector; under this condition the translational symmetry of the unperturbed wave function perpendicular to $\vec{\mathcal{E}}$ is preserved and the electric field changes momentum,[24]

$$\hbar \dot{k}_x = e\mathcal{E} . \tag{48}$$

We change the variable of integration from dk_x to $(e\mathcal{E}/\hbar)\,dt$. For the weak-field approximation one can extend the limits of integration $(\hbar K_x)/2e\mathcal{E}$ to ∞; Eq. (47) becomes

$$\epsilon_2(\omega, \mathcal{E})_{\text{cont}} = \frac{8\pi^3 e^2}{\omega^2 m^2 \hbar} \frac{2}{(2\pi)^3} |\langle \varphi_c | \hat{\epsilon} \cdot \vec{P} | \varphi_v \rangle|^2 \frac{e\mathcal{E}}{\hbar} \times \int_{\text{BZ}} d^2 k_\perp \frac{1}{|\Lambda_i|^2} \text{Ai}^2\left(\frac{E_{cv}(\vec{k}) - \hbar\omega}{\hbar \Lambda_i}\right) . \tag{49}$$

This expression is valid for any band model as far as the validity of effective-mass approximation is preserved and similar to the expression which is given by Aspnes, Handler, and Blossey[1] for one-electron and uniform field theory. If the band structure of a solid is known, i.e., if $E_{cv}(\vec{k})$ is known for every $\vec{k}$ in the BZ and if values of $|\langle \varphi_c | \hat{\epsilon} \cdot \vec{P} | \varphi_v \rangle|^2$ can be computed and are slowly varying in the field direction, then the imaginary part of the dielectric function can be derived.

The function $\text{Ai}^2(x)$ is related to a single Airy function by the integral

$$\text{Ai}^2(x) = \frac{1}{(2)^{2/3}\pi} \int_0^{\infty} \frac{dS}{S^{1/2}} \text{Ai}(S + (2)^{2/3} x) . \tag{50}$$

In the limits of parabolic bands, the imaginary part of dielectric function $\epsilon_2(\omega, \mathcal{E})_{\text{cont}}$ becomes

$$\epsilon_2(\omega, \mathcal{E})_{\text{cont}} = \frac{2e^2 |\hat{\epsilon} \cdot \vec{P}|^2}{m^2 \hbar} \left(\frac{8 |\mu_x \mu_y \mu_z|}{\hbar^3}\right)^{1/2} \frac{1}{\omega^2 \hbar^{1/2}} \times \int_0^{\infty} d\nu\, \nu^{1/2} \left[\frac{1}{\hbar |\Lambda_{ic}|} \text{Ai}\left(\frac{\nu + E_g - \hbar\omega}{\hbar \Lambda_{ic}}\right)\right], \tag{51}$$

where $\Lambda_{ic} = \Lambda_i/(2)^{2/3}$ and $|\hat{\epsilon} \cdot \vec{P}|^2 = |\langle \varphi_c | \hat{\epsilon} \cdot \vec{P} | \varphi_v \rangle|^2$. Employing the procedure of Sec. III, after some simple calculations,[1] we have the imaginary part of the dielectric function at any critical points for indirect transitions

$$\epsilon_2^{\text{ind}}(\omega, \mathcal{E})_{\text{cont}} = \frac{Q}{\omega^2} \int_{\omega_g \pm \omega_{\vec{k}_0}}^{\infty} d\omega' (\omega' - \omega_g \mp \omega_{\vec{k}_0})^2 \times \left[\frac{1}{|\Lambda_{ic}|} \text{Ai}\left(\frac{\omega' - \omega}{\Lambda_{ic}}\right)\right], \tag{52}$$

where

$$E_g = \hbar\omega_g$$

and

$$Q = \frac{e^2 |\hat{\epsilon} \cdot \vec{P}|^2 (n_{\vec{k}_0} + \frac{1}{2} \pm \frac{1}{2})}{2\pi m^2 \hbar^4} \times (m_{xe} m_{ye} m_{ze} m_{xh} m_{yh} m_{zh})^{1/2} .$$

As shown in Sec. III the relative amplitude of the applied and the effective electric field of electron-hole interaction is an important factor of electric field modulation exciton line shapes. It is evident that the effective characteristic frequency Λ_{ic} would change sign from region 1 to 3, i.e., $|\mathcal{E}| \ll |\mathcal{E}_H|$

or $|\mathcal{E}_N|$ to $|\mathcal{E}| \gg |\mathcal{E}_H|$ or $|\mathcal{E}_N|$ and therefore the type of critical point would change or appear as mixtures of different types in the field-induced change of dielectric function. This result agrees qualitatively with recent result of Rowe and Aspnes[11] which used Koster-Slater model and time-independent gauge for applied electric field.

V. CONCLUSIONS

A systematic method of evaluating the imaginary part of the dielectric function in the presence of a uniform electric field has been presented which is solved in the nonstationary wave function and time-dependent gauge, and the result can be applied to direct and indirect transitions at any critical points and to the exciton lines. The change in absorption upon inclusion of the electron-hole interaction has been shown to depend on the relative amplitude of $\mathcal{E}$ and $\mathcal{E}_H$ or $\mathcal{E}_N$. The electron-hole interaction is important when the applied electric field is smaller compared with the amplitude $\mathcal{E}_H$ or $\mathcal{E}_N$.

APPENDIX A

From Eq. (5), we have

$$\left(\hbar^{-1}H_0 - i\lambda t\frac{\partial}{\partial x} + \eta^3 t^2\right)\psi_{nlm}(\vec{r},t) = i\frac{\partial}{\partial t}\psi_{nlm}(\vec{r},t)\,. \tag{A1}$$

Let us write the solution of Eq. (A1):

$$\psi_{nlm}(\vec{r},t) = \exp\left(-\frac{i\eta^3 t^3}{3} - \frac{iH_0 t}{\hbar}\right)\exp\left(\frac{i}{\hbar}\sum_n P_n\right) \times \varphi_n(\vec{k}, \vec{r} - \tfrac{1}{2}\lambda t^2)\,, \tag{A2}$$

where P_n is an operator to be defined. By using the identity between operators $e^A B e^{-A}$, we obtain

$$\lambda t\frac{\partial}{\partial x}e^{-iH_0 t/\hbar} = e^{-iH_0 t/\hbar} \times\left(\lambda t\frac{\partial}{\partial x} - \frac{i}{\hbar}\lambda t^2\frac{\partial V(\vec{r})}{\partial x} + \cdots\right)\,. \tag{A3}$$

If we keep the dipole term and neglect the higher moments in Eq. (A3), Eq. (A1) becomes

$$\left(\frac{\partial}{\partial t} + t\lambda\frac{\partial}{\partial x} - \frac{i}{\hbar}\lambda t^2\frac{\partial V(\vec{r})}{\partial x}\right) \times\exp\left(\frac{i}{\hbar}\sum_n P_n\right)\varphi_n(\vec{k}, \vec{r} - \tfrac{1}{2}\lambda t^2) = 0\,. \tag{A4}$$

Using the identity of the operators, Eq. (A4) becomes

$$\left\{\frac{\partial}{\partial t} + \lambda t\frac{\partial}{\partial x} - i\frac{1}{\hbar}\lambda t^2\frac{\partial V(\vec{r})}{\partial x} + \left[-\frac{i}{\hbar}\sum_n P_n\,,\ \frac{\partial}{\partial t} + \lambda t\frac{\partial}{\partial x} - \frac{i}{\hbar}\lambda t^2\frac{\partial v(\vec{r})}{\partial x}\right] + \cdots\right\}\varphi_n(\vec{k}, \vec{r} - \tfrac{1}{2}\lambda t^2) = 0\,. \tag{A5}$$

If we assume P_n is a function of space coordinates and time, we have the commutator

$$\left[-\frac{i}{\hbar}\sum_n P_n\,,\ \frac{\partial}{\partial t} + \lambda t\frac{\partial}{\partial x} - \frac{i}{\hbar}\lambda t^2\frac{\partial v(\vec{r})}{\partial x}\right] = +\frac{i}{\hbar}\sum_n\left(\frac{\partial P_n}{\partial t} + \lambda t\frac{\partial P_n}{\partial x}\right)\,, \tag{A6}$$

and Eq. (A5) becomes

$$\left[\frac{\partial}{\partial t} + \lambda t\frac{\partial}{\partial x} - \frac{i}{\hbar}\lambda t^2\frac{\partial v(\vec{r})}{\partial x} + \frac{i}{\hbar}\sum_n\left(\frac{\partial P_n}{\partial t} + \lambda t\frac{\partial P_n}{\partial x}\right)\right] \times\varphi_n(\vec{k}, \vec{r} - \tfrac{1}{2}\lambda t^2) = 0\,. \tag{A7}$$

Letting

$$\frac{\partial P_0}{\partial t} = \lambda t^2\frac{\partial v(\vec{r})}{\partial x} \tag{A8}$$

and

$$\lambda t\frac{\partial P_{n-1}}{\partial x} = -\frac{\partial P_n}{\partial t}\,, \tag{A9}$$

the operator $\sum_n P_n$ can be solved by

$$I(\vec{r},t) = \sum_n P_n = t\sum_{m=0}^{\infty}(-1)^m\left[\left(t^2\lambda\frac{\partial}{\partial x}\right)^{m+1}\Big/ 3\cdot(3+2)\cdots(3+2m)\right]V(\vec{r})\,. \tag{A10}$$

APPENDIX B: HIGHER-ORDER TERMS

We have derived the imaginary part of the dielectric function of solids in the presence of a uniform electric field which includes the interaction between the electrons and holes. We see from Eqs. (26), (29), (46), and (49) that the expressions for the imaginary part of the dielectric functions for bound states and continuum states are valid at any critical points of the energy band. It should be noted that the Airy function in our results is the Fourier transformation of the coefficient $C^*_{n\nu}C_{n'\nu'}$ where we keep the electric field and dipole interaction term, and the higher-order terms of Eq. (21) are neglected. If the higher-order terms are included, the Airy function in Eqs. (26), (46), and (49) should be replaced by

$$F(\hbar^{-1}E_{cv}(\vec{k}) - \omega, \mathcal{E}) = \frac{1}{2\pi}\int_{-\infty}^{\infty}\exp\left(-\frac{i}{3}\Theta_c^3 t^3 - i\hbar^{-1}E_{cv}(\vec{k})\right)$$

$$\times \exp[i\hbar^{-1}\Delta\langle I(\vec{r},t)\rangle_{cv} + i\omega]\,dt\,. \quad \text{(B1)}$$

As the zero-field limit is approached

$$\lim_{\mathcal{E}\to 0} F(\hbar^{-1}E_{cv}(\vec{k})-\omega,\mathcal{E}) = \delta(\hbar^{-1}E_{cv}(\vec{k})-\omega)\,. \quad \text{(B2)}$$

Therefore the imaginary part of the dielectric functions becomes

$$\epsilon_2(\omega,\mathcal{E})_{\text{bound}} = \sum_{\text{bound states}} \frac{4\pi^2 e^2}{\omega^2\hbar m^2}\,|\langle\varphi_c|\hat{\epsilon}\cdot\vec{P}|\varphi_v\rangle|^2 F(\hbar^{-1}E_{cv}(\vec{k})-\omega,\mathcal{E})\,, \quad \text{(B3a)}$$

$$\epsilon_2(\omega,\mathcal{E})_{\text{cont}} = \frac{8\pi^3 e^2}{\omega^2 m^2\hbar}\,\frac{2}{(2\pi)^3}\int_{BZ} d^2k_\perp \frac{e\mathcal{E}}{\hbar}\,F^2(\hbar^{-1}E_{cv}(\vec{k}_\perp)-\omega,\mathcal{E})\,, \quad \text{(B3b)}$$

$$\epsilon_2^{\text{ind}}(\omega,\mathcal{E})_{\text{bound}} = D\sum_{\text{bound states}} (n_{\vec{k}_0}+\tfrac{1}{2}\pm\tfrac{1}{2})\int_{E_g\pm\hbar\omega_{\vec{k}_0}}^{\infty} dE(E-E_g\mp\hbar\omega_{\vec{k}_0})^{1/2}\,F(\hbar^{-1}E_{cv}(\vec{k})-\omega-\omega_g+E,\mathcal{E})\,, \quad \text{(B4a)}$$

$$\epsilon_2^{\text{ind}}(\omega,\mathcal{E})_{\text{cont}} = \frac{(2M_xM_yM_z)^{1/2}}{2\pi^2\hbar^3}\int_0^{\infty} dE_{\text{c.m.}}\,(E_{\text{c.m.}})^{1/2}\,(n_{\vec{k}_0}+\tfrac{1}{2}\pm\tfrac{1}{2})$$

$$\times\frac{8\pi^3 e^2}{\omega^2 m^2\hbar}\,\frac{2}{(2\pi)^3}\int_{BZ} d^2k_\perp \frac{e\mathcal{E}}{\hbar}\,F^2(\hbar^{-1}E_{cv}(\vec{k}_\perp)-\omega,\mathcal{E})\,. \quad \text{(B4b)}$$

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