

Dielectric theory of interacting excitonic resonances

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The dielectric function is developed for two interacting excitonic resonances with spatial dispersion. The longitudinal-transverse energy splitting and the reflectivity of these resonances depend very strongly on the energy separation between the resonances. It is shown that simple interpretations of reflection spectra of degenerate resonances split by external fields may yield incorrect oscillator strengths or polarizabilities and resonance frequency shifts. The energy versus wave-vector polariton dispersion relation is obtained and plotted for the materials CdS and ZnO as examples. Effective refractive indices are derived to be used in the generalized multilayer Fresnel formula for oblique incidence of light polarized parallel and perpendicular to the plane of incidence.

I. INTRODUCTION

Spatial dispersion, the effect of the wave-vector-dependent dielectric function of excitons, has been treated originally by Pekar¹ and Hopfield and Thomas.² Most of the work carried out in this field, however, concentrated on the case of one single excitonic resonance, since the problem then reduces to an easily tractable one. Exceptions are limited to the case of normal incidence of light^{3,4} or to resonances without spatial dispersion.⁵

The present work treats the general case of two interacting excitonic resonances including non-normal incidence of light, spatial dispersion, and an additional exciton-free surface layer. The interaction is the Coulomb interaction which changes the longitudinal-transverse energy splitting of each resonance. The present work is necessary to describe, for instance, reflectance spectra of two excitonic transitions with small energy separation as in CdS and particularly ZnO. This theory leads further to a considerable improvement in interpreting reflectance spectra of degenerate resonances split by external fields and in extracting polarizabilities and energy shifts of resonance frequencies from experimental spectra.

II. DIELECTRIC FUNCTION

Polarizabilities α of the following form characterize insulating crystals for given wave vector k and frequency ω (Ref. 2):

$$\alpha(\omega, k) = \sum_j \frac{\alpha_{0j} \omega_{jT}^2}{\omega_{jT}^2 - \omega^2 + \beta_j k^2 - i\omega\Gamma_j}, \quad (1)$$

with α_{0j} being the polarizabilities of each excitonic resonance j at $\omega=0$ and $k=0$, ω_{jT} the transverse resonance frequencies at $k=0$, and Γ_j the empirical damping constants. The influence of spatial dispersion is described by $\beta_j k^2 = (\hbar\omega_{jT}/M_j)k^2$, where M_j is the effective exciton mass and \hbar is

Planck's constant divided by 2π .

The quantum mechanics introduces the oscillator strengths f_j instead of the polarizabilities α_{0j} . Then, one has to replace $\alpha_{0j}\omega_{jT}^2$ by $f_j n_{0j} q^2 / M_j$ in⁶ Eq. (1) (n_{0j} : density of oscillators; q : elementary charge).

The frequency is chosen in this paper near two particular resonances A and B in Eq. (1), and the sum over all other oscillators is lumped into a frequency- and wave-vector-independent background polarizability α_∞ :

$$\alpha(\omega, k) = \alpha_\infty + \frac{\alpha_{0A} \omega_{AT}^2}{\omega_{AT}^2 - \omega^2 + \beta_A k^2 - i\omega\Gamma_A} + \frac{\alpha_{0B} \omega_{BT}^2}{\omega_{BT}^2 - \omega^2 + \beta_B k^2 - i\omega\Gamma_B}. \quad (2)$$

The relation $\epsilon \vec{\epsilon}_0 E = \vec{\epsilon}_0 E + \alpha E$ between the electric field E existing in the crystal, the permittivity of vacuum $\vec{\epsilon}_0$, the polarizability α , and the dielectric function ϵ leads to

$$\epsilon(\omega, k) = \epsilon_\infty + \frac{\alpha_{0A}}{\vec{\epsilon}_0} \frac{\omega_{AT}^2}{\omega_{AT}^2 - \omega^2 + \beta_A k^2 - i\omega\Gamma_A} + \frac{\alpha_{0B}}{\vec{\epsilon}_0} \frac{\omega_{BT}^2}{\omega_{BT}^2 - \omega^2 + \beta_B k^2 - i\omega\Gamma_B}, \quad (3)$$

where ϵ_∞ is a frequency- and wave-vector-independent background dielectric constant ($\epsilon = \epsilon_\infty$ for $\omega \rightarrow \infty$).

Equation (3) contains the polarizabilities α_{0A} and α_{0B} , which are not directly determined by experimental data for a given material. The longitudinal resonance frequencies, however, are often known from absorption and emission experiments.

The dielectric function ϵ of two excitonic resonances vanishes at these longitudinal resonance frequencies ω_{AL} and ω_{BL} , and is given then ($\omega_{AT} \neq \omega_{BT}$) by

$$\epsilon(\omega, k) = \epsilon_\infty \left(1 + \frac{\omega_{BL}^2 - \omega_{AT}^2}{\omega_{BT}^2 - \omega_{AT}^2} \frac{\omega_{AL}^2 - \omega_{AT}^2}{\omega_{AT}^2 - \omega^2 + \beta_A k^2 - i\omega\Gamma_A} + \frac{\omega_{AL}^2 - \omega_{BT}^2}{\omega_{AT}^2 - \omega_{BT}^2} \frac{\omega_{BL}^2 - \omega_{BT}^2}{\omega_{BT}^2 - \omega^2 + \beta_B k^2 - i\omega\Gamma_B} \right). \quad (4)$$

Setting $\omega=0$ yields the extended Lyddane-Sachs-Teller relation

$$\epsilon_0 = \epsilon(\omega=0) = \epsilon_\infty \omega_{AL}^2 \omega_{BL}^2 / \omega_{AT}^2 \omega_{BT}^2. \quad (5)$$

The two factors accompanying the resonance expressions in Eq. (4) describe the interaction between the A and B resonance. They approach unity, if the energy separation between the two resonances is much larger than the corresponding longitudinal-transverse splittings.

III. DISPERSION RELATION

Equation (4) leads to the frequency versus wave-vector polariton dispersion relation for two excitonic resonances having spatial dispersion. The transverse solutions ϵ_T of ϵ are determined by the condition

$$\epsilon_T(\omega, k) = k^2 c^2 / \omega^2. \quad (6)$$

For given ω , Eq. (6) is cubic in k^2 . There are three transverse solutions propagating in each direction for a given principal polarization and frequency (c is the vacuum velocity of light).

The longitudinal solutions ϵ_L are given per definition by

$$\epsilon_L(\omega, k) = 0. \quad (7)$$

This equation is quadratic in k^2 , thus leads to two

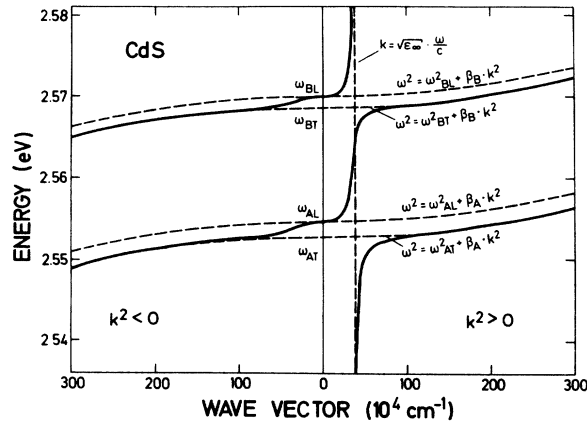


FIG. 1. Energy vs wave-vector polariton dispersion relation of two well-separated excitonic resonances A and B in CdS. Damping is omitted. (For parameters see Table I.)

TABLE I. Parameters of the A and B excitons in CdS and ZnO.

	CdS	ZnO
$\hbar\omega_{AT}$ (eV)	2.5528 ^a	3.3753 ^b
$\hbar\omega_{AL}$ (eV)	2.5547 ^a	3.3773 ^b
$\hbar\omega_{BT}$ (eV)	2.5687 ^a	3.3813 ^c
$\hbar\omega_{BL}$ (eV)	2.5700 ^a	3.3932 ^d
ϵ_∞	8.0 ^a	6.16 ^d
$M_A = M_B (m_{e0})$	0.94 ^a	0.87 ^d

^aJ. Voigt, M. Senoner, and I. Rückmann, Phys. Status Solidi B 75, 213 (1976).

^bE. Tomzig and R. Helbig, Solid State Commun. 15, 1513 (1974).

^cR. Helbig, Habilitation thesis (Erlangen, 1976) (unpublished).

^dSee Ref. 4.

longitudinal solutions for a given frequency.

Figure 1 shows the frequency versus wave-vector dispersion relation for transverse modes of light coupled to a dielectric having two excitonic resonances. The parameters are chosen for CdS with two well separated resonances A and B . Damping is omitted. The wave vectors are either purely real or purely imaginary. They are plotted on the right- or left-hand sides. (See Table I.)

Parameters valid for ZnO lead to a different shape of the dispersion relation as shown in Fig. 2. This line-shape change results from the small energy separation between the A and B excitons in ZnO. Thus, the dispersion branch can no longer trail the light line between ω_{AL} and ω_{BT} (see Fig. 2), but rather crosses the light line and rapidly approaches the curve $\omega^2 = \omega_{BT}^2 + \beta_B k^2$.

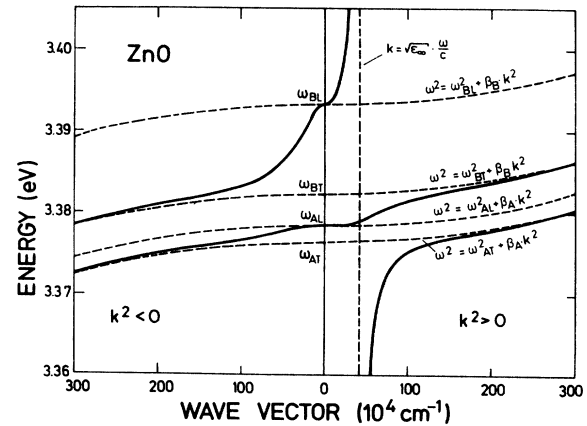


FIG. 2. Energy vs wave-vector polariton dispersion relation of two strongly interacting excitonic resonances A and B in ZnO. Damping is omitted. (For parameters see Table I.)

IV. INTERACTION BETWEEN THE RESONANCE FREQUENCIES

A. Longitudinal-transverse frequency splitting

The dielectric function ϵ is given in Eqs. (3) and (4) by different sets of parameters. Both equations show the same behavior of an excitonic system, but from different points of view. The description which has to be used depends on the respective known parameters.

Equation (3) contains the transverse resonance frequencies ω_{AT} and ω_{BT} and the polarizabilities α_{0A} and α_{0B} . Setting $\epsilon(\omega, k) = 0$ leads the longitudinal frequencies of this oscillating excitonic system. The resulting longitudinal-transverse frequency splitting of each resonance depends on the given polarizabilities *and* above all on the energy separation between the two transverse frequencies because of the interaction between the resonances. Equation (3) is advantageously applied to a material where the two transverse resonance frequencies and the excitonic polarizabilities are known.

On the other hand, Eq. (4) contains the two transverse and two longitudinal resonance frequencies of the *A* and *B* excitons. These frequencies may, for example, be found by absorption or emission experiments. Such measured longitudinal resonance frequencies already contain the interaction between the two resonances. This interaction is taken into account by the additional factors in Eq. (4) which cause $\epsilon = 0$ if $\omega = \omega_{AL}$ or $\omega = \omega_{BL}$ ($k=0$, $\Gamma_A = \Gamma_B = 0$). Therefore, the dielectric function of Eq. (4) applies best to a material where all four resonance frequencies are known.

The interaction between two resonances is shown in Fig. 3. The transverse resonance frequencies ω_{AT} and ω_{BT} change (full lines), whereas the pol-

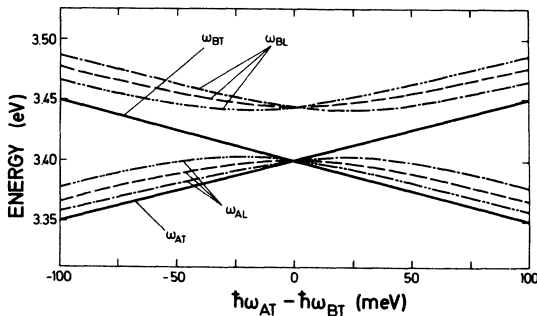


FIG. 3. Transverse and resulting longitudinal resonance frequencies for given polarizabilities of two interacting resonances. The dashed and dotted lines give the longitudinal frequencies for the following sets of parameters: Dashed line: $\alpha_{0A} = \alpha_{0B} = 8 \times 10^{-2} \tilde{\epsilon}_0$; dash-dot line: $\alpha_{0A} = 4 \times 10^{-2} \tilde{\epsilon}_0$; $\alpha_{0B} = 12 \times 10^{-2} \tilde{\epsilon}_0$; dash-double-dot line: $\alpha_{0A} = 12 \times 10^{-2} \tilde{\epsilon}_0$; $\alpha_{0B} = 4 \times 10^{-2} \tilde{\epsilon}_0$.

arizabilities α_{0A} and α_{0B} remain fixed. The dashed and dotted lines show the longitudinal resonance frequencies resulting from Eqs. (3) and (7) with $k=0$ and no damping. The *L-T* splittings of well-separated *A* and *B* resonances are not equal because they depend on the absolute value of the respective transverse frequency.

A decrease of the energy separation between the two transverse resonances alters the longitudinal-transverse frequency splitting of each resonance. This change of the *L-T* splitting does *not* result from a change of the (classical) polarizability or of the (quantum-mechanical) oscillator strength, but is merely a consequence of the interaction between the two resonances.

B. Reflection spectra

Investigations of energy splittings of one single resonance into different components under external fields are important tools to evaluate excitonic properties. The increasing energy separation may be described by the present model of two excitonic resonances starting from one single resonance ($\omega_{AT} = \omega_{BT}$).

Reflectance experiments are sometimes the only feasible technique under external fields. Unfortunately, only the longitudinal resonance frequencies are (approximately) given by the reflectivity minima in reflection spectra, whereas the transverse resonance frequencies cannot be determined easily. Thus, the energy shifts of the longitudinal resonance frequencies are used for the analysis of many experiments of field-induced energy splittings. As shown in Fig. 3, the change of the longitudinal frequencies, however, is not the same as that of the important transverse resonance frequencies, which was pointed out also by Skettrup and Balslev.⁷

An improved fit of field-induced splittings is possible using the model of this paper. Figure 3 shows directly the splitting of the transverse frequencies, if that of the longitudinal frequencies is known. Certainly, this approximation is better to obtain energy splittings originating from external fields than the approximation by simply using the reflectivity minima.

Oscillator strengths or polarizabilities of different components of excitonic transitions in external fields can provide much information about the band structure of semiconducting materials. Most of the data are obtained in this field by reflection experiments. However, reflection spectra and reflectivity amplitudes do not inform directly about polarizabilities. The most important influence on reflection spectra is given by the longitudinal-transverse energy splitting. First of all,

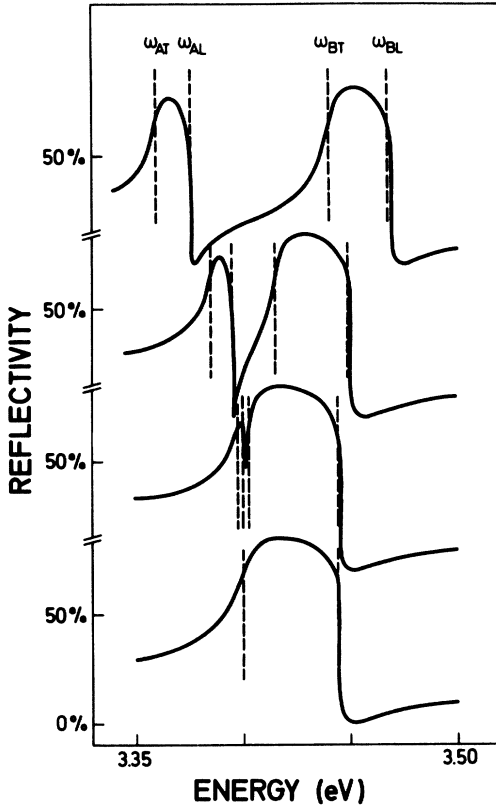


FIG. 4. Calculated reflection spectra with different transverse resonance frequencies, but given constant polarizabilities. All the spectra are calculated with $\alpha_{0A} = \alpha_{0B} = 8 \times 10^{-2} \epsilon_0$ ($\theta = 0$).

this splitting (for more than one single resonance) depends on the energy separation between the resonance frequencies, whereas the polarizabilities carry less importance.

Figure 4 shows calculated reflection spectra with varying transverse resonance frequencies but constant polarizabilities. (For details of calculation see Sec. V.) The reflectivity amplitudes of the two resonances depend very strongly on the energy separation between the two transverse resonance frequencies according to the interaction between the resonances. The polarizability or oscillator strength, however, is the same for all reflection spectra of Fig. 4.

Let us consider for illustration one special example for incorrect conclusions about oscillator strengths deduced from reflection spectra. Figure 5 shows experimental reflection spectra (upper part) of the 1S exciton in InP under different magnetic fields for π polarization. An extra resonance emerges with the magnetic field additionally to the shift to higher energies. The reflec-

tivity amplitude of this extra resonance increases with the magnetic field.

Several authors deduced from this increase a field-dependent ratio of the oscillator strengths of the two split resonances.⁸ This conclusion led to essential statements about the electron-hole exchange interaction.

The lower part of Fig. 5 shows reflection spectra calculated using the model of this paper. The polarizabilities α_{0A} and α_{0B} of the two split resonances are set equal in each spectrum. The common value $\alpha_{0A} = \alpha_{0B}$ is changed from one spectrum to another because of the magnetic field. The comparison of the theoretical spectra (lower part of Fig. 5) with the experimental spectra (upper part) shows good agreement. The extra resonance emerges at the high-energy side. The reflectivity amplitude of this extra resonance increases also in the calculated spectra, although the ratio of the oscillator strengths of the two split resonances is taken independent of the magnetic field. Therefore, the earlier conclusions⁸ about the electron-hole exchange interaction are incorrect because the oscillator strengths ratio is in reality field independent.

These exemplary spectra of InP show how carefully one has to be in taking oscillator strengths or polarizabilities from reflection experiments whenever two resonances have nearly the same energy.

V. GENERALIZED FRESNEL FORMULA

This section derives generalized Fresnel formula to calculate reflection or attenuated total reflection spectra including exciton-free surface layers. We consider the general case of non-normal incident light reflected from a crystal having two excitonic resonances with spatial dispersion. The angle of incidence is θ , the electric field amplitudes of the incoming and reflected light waves are E_I and E_R , respectively, and n_0 is the refractive index for the medium in which the incoming wave travels.

The incoming light wave excites in general electric field amplitudes E_j in the crystal of three transverse ($j=1, 2, 3$) and two longitudinal polariton modes ($j=L1, L2$). Equations (6) and (7) with Eq. (3) or (4) yield the respective refractive indices n_j ($n_j^2 = \epsilon_j$).

The angles of refraction ϕ_j are given for each transverse or longitudinal wave j by Snell's law

$$\sin \phi_j = (n_0/n_j) \sin \theta. \quad (8)$$

We consider the two polarizations of light parallel and perpendicular to the plane of incidence.

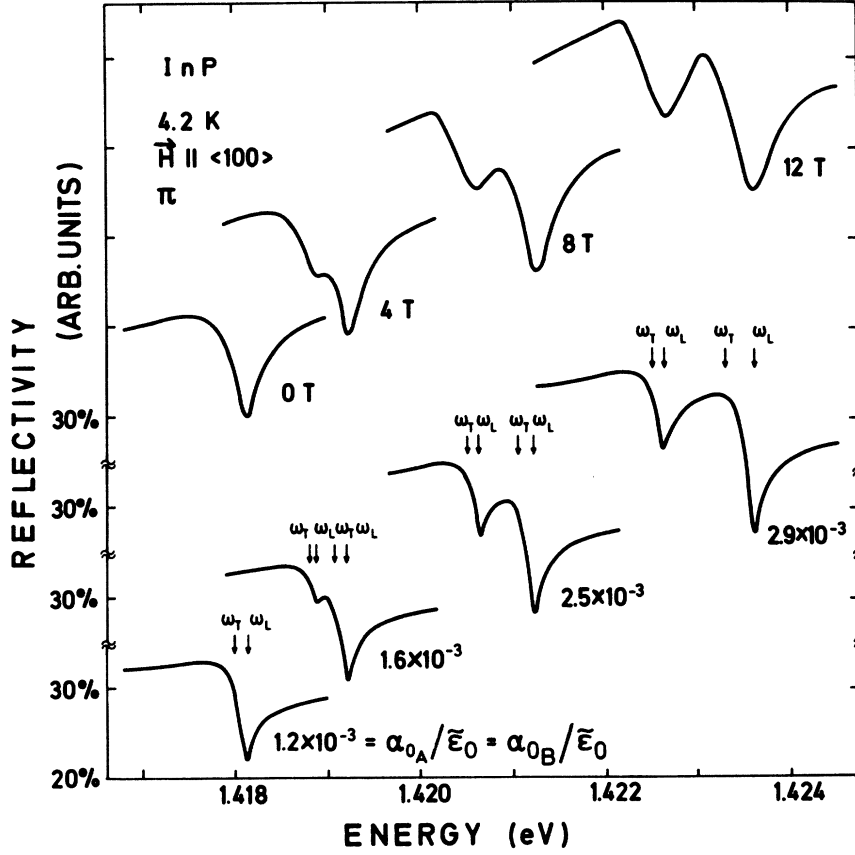


FIG. 5. Upper part: experimental reflection spectra of the 1S exciton in InP with different magnetic fields in π polarization. Lower part: calculated reflection spectra with polarizabilities $\alpha_{0A} = \alpha_{0B}$. The transverse and the longitudinal resonance frequencies are marked. [All other parameters were taken from F. Evangelisti, J. U. Fischbach, and A. Frova, Phys. Rev. B **9**, 1516 (1974)]. All the spectra are measured and calculated for normal incidence of light. [The experimental spectra were taken from D. Bimberg, K. Hess, N. O. Lipari, J. U. Fischbach, and M. Altarelli, Physica B **81**, 19 (1977).]

A. Light polarized parallel to plane of incidence

In this geometry, three transverse and two longitudinal polariton modes are excited. The conservation of the tangential component of the electric field amplitudes may then be written

$$E_I \cos \theta + E_R \cos \theta = E_1 \cos \phi_1 + E_2 \cos \phi_2 + E_3 \cos \phi_3 + E_{L1} \sin \phi_{L1} + E_{L2} \sin \phi_{L2} \quad (9)$$

or

$$(E_I + E_R) \cos \theta = E_1 [B_1 + F_{21p} B_2 + F_{31p} B_3 + F_{L1} \cos \theta (n_0/n_{L1}) + F_{L2} \cos \theta (n_0/n_{L2})], \quad (10)$$

with

$$B_{1,2,3} = \frac{(n_{1,2,3}^2 - n_0^2 \sin^2 \theta)^{1/2}}{n_{1,2,3}}, \quad (11a)$$

$$F_{21p} = E_2/E_1, \quad F_{L1} = \tan \theta (E_{L1}/E_1) \quad (11b)$$

$$F_{31p} = E_3/E_1, \quad F_{L2} = \tan \theta (E_{L2}/E_1).$$

The magnetic field is already tangential in this geometry, therefore

$$n_0 E_I - n_0 E_R = n_1 E_1 + n_2 E_2 + n_3 E_3 \quad (12)$$

or

$$(E_I - E_R) n_0 = E_1 (n_1 + n_2 F_{21p} + n_3 F_{31p}). \quad (13)$$

The longitudinal modes carry no magnetic field.

Equations (10) and (13) yield the reflection coefficient

$$r_p = \frac{E_R}{E_I} = \frac{A_p - \cos \theta / n_0}{A_p + \cos \theta / n_0}, \quad (14)$$

with

$$A_p = (B_1 + F_{21p} B_2 + F_{31p} B_3 + F_{L1} n_0 \cos \theta / n_{L1} + F_{L2} n_0 \cos \theta / n_{L2}) (n_1 + n_2 F_{21p} + n_3 F_{31p})^{-1}. \quad (15)$$

Comparison of Eq. (14) with the Fresnel formula for non-normal incidence of light polarized parallel to the plane of incidence,

$$r_p = \frac{\cos \theta / n_{\text{eff}p} - \cos \theta / n_0}{\cos \theta / n_{\text{eff}p} + \cos \theta / n_0} = \frac{(n_{\text{eff}p}^2 - n_0^2 \sin^2 \theta)^{1/2} / n_{\text{eff}p}^2 - \cos \theta / n_0}{(n_{\text{eff}p}^2 - n_0^2 \sin^2 \theta)^{1/2} / n_{\text{eff}p}^2 + \cos \theta / n_0}, \quad (16)$$

yields the angle-dependent effective refractive

index $n_{\text{eff}p}$ for light polarized parallel to the plane of incidence:

$$n_{\text{eff}p}^2 = [1 \pm (1 - 4A_p^2 n_0^2 \sin^2 \theta)^{1/2}] / 2A_p^2. \quad (17)$$

The sign has to be chosen such that $\text{Im}(n_{\text{eff}p}) > 0$.

In case of spatial dispersion there are more than two unknown electric field amplitudes at the boundary, whereas the Maxwell boundary conditions yield only the two, Eqs. (10) and (13). Other equations are necessary to solve the system, the so-called additional boundary conditions (ABC).

There has been a long discussion in the literature about the ABC's beginning with papers by Pekar¹ and Hopfield and Thomas.² This discussion is still continuing.⁹

The ABC determines F_{21p} , F_{31p} and F_{L1} , F_{L2} which connect the electric field amplitudes of the upper two transverse and the longitudinal polariton branches, respectively, with that of the lowest transverse branch. Most of the formalism developed in this paper is independent of the special ABC which is chosen. One has to use only the respective relations of F_{21p} , F_{31p} , F_{L1} , and F_{L2} .

We use the ABC proposed by Pekar¹ and Hopfield and Thomas² and derived more generally by Zeyher, Birman, and Brenig.¹⁰ They claimed that the excitonic polarizability vanishes at the surface. This ABC is originally valid for one single resonance and must be expanded to two resonances.³ Then, the polarization contribution of each resonance A and B to all polariton branches j vanishes:

$$\begin{aligned} \epsilon_1^A \vec{E}_1 + \epsilon_2^A \vec{E}_2 + \epsilon_3^A \vec{E}_3 + \epsilon_{L1}^A \vec{E}_{L1} + \epsilon_{L2}^A \vec{E}_{L2} &= 0, \\ \epsilon_1^B \vec{E}_1 + \epsilon_2^B \vec{E}_2 + \epsilon_3^B \vec{E}_3 + \epsilon_{L1}^B \vec{E}_{L1} + \epsilon_{L2}^B \vec{E}_{L2} &= 0, \end{aligned} \quad (18)$$

with

$$\epsilon_j^{A,B} = \frac{\alpha_{0A,B}}{\epsilon_0} \frac{\omega_{A,B}^2}{\omega_{A,B}^2 T - \omega^2 + \beta_{A,B} (\omega^2 / c^2) n_j^2 - i\omega \Gamma_{A,B}}. \quad (19)$$

Splitting the electric field vectors of Eq. (18) into components parallel and perpendicular to the boundary yields a system of four linear equations. Herefrom, the unknowns F_{21p} , F_{31p} , F_{L1} , and F_{L2} may be determined.

B. Light polarized perpendicular to plane of incidence

Three transverse and no longitudinal modes are excited in this geometry. The conservation of the tangential component of the electric field amplitude requires

$$E_I + E_R = E_1(1 + F_{21s} + F_{31s}), \quad (20)$$

with

$$F_{21s} = E_2/E_1, \quad F_{31s} = E_3/E_1. \quad (21)$$

Conservation of the tangential component of the

magnetic field amplitude yields

$$(E_I - E_R)n_0 \cos \theta = E_1(n_1 B_1 + n_2 B_2 F_{21s} + n_3 B_3 F_{31s}). \quad (22)$$

The resulting reflection coefficient is

$$r_s = E_R/E_I = (n_0 \cos \theta - A_s)/(n_0 \cos \theta + A_s), \quad (23)$$

with

$$A_s = (n_1 B_1 + n_2 F_{21s} B_2 + n_3 F_{31s} B_3) / (1 + F_{21s} + F_{31s}) \quad (24)$$

Comparison of Eq. (23) with the Fresnel formula for non-normal incident light polarized perpendicular to the plane of incidence,

$$\begin{aligned} r_s &= \frac{n_0 \cos \theta - n_{\text{eff}s} \cos \phi}{n_0 \cos \theta + n_{\text{eff}s} \cos \phi} \\ &= \frac{n_0 \cos \theta - (n_{\text{eff}s}^2 - n_0^2 \sin^2 \theta)^{1/2}}{n_0 \cos \theta + (n_{\text{eff}s}^2 - n_0^2 \sin^2 \theta)^{1/2}}, \end{aligned} \quad (25)$$

yields the angle-dependent effective refractive index $n_{\text{eff}s}$ for light polarized perpendicular to the plane of incidence

$$n_{\text{eff}s}^2 = A_s^2 + n_0^2 \sin^2 \theta. \quad (26)$$

The electric field is already tangential in this geometry. Thus, the additional boundary condition is

$$\begin{aligned} \epsilon_1^A + \epsilon_2^A F_{21s} + \epsilon_3^A F_{31s} &= 0, \\ \epsilon_1^B + \epsilon_2^B F_{21s} + \epsilon_3^B F_{31s} &= 0. \end{aligned} \quad (27)$$

The two cases (Secs. V A and V B) yield generalized Fresnel formula. The effective refractive indices $n_{\text{eff}p}$ and $n_{\text{eff}s}$ describe the optical behavior of a crystal having two excitonic resonances with spatial dispersion. They are similar to those of one single resonance.¹¹

C. Multilayer system

The use of these effective refractive indices in their general form is not restricted to the direct reflectance at the boundary of a medium with excitonic resonances. They may be used in this form in any Fresnel formula, even of a multilayer system.¹² The angle of incidence θ on the spatially dispersive medium and the refractive index n_0 have then to be replaced by the angle of incidence α on the first additional layer and the refractive index n_α of the upper half space:

$$n_0 \sin \theta = n_\alpha \sin \alpha. \quad (28)$$

In this case θ may be complex also. The only condition is that the sequence of the layers is non-

spatially dispersive media—spatially dispersive media.

The generalized Fresnel formula of two excitonic resonances with spatial dispersion facilitate strongly the calculation of reflection spectra with exciton-free surface layers¹³ or of attenuated total reflection spectra of excitonic surface polaritons including exciton-free surface layers.¹⁴

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²J. J. Hopfield and D. G. Thomas, Phys. Rev. **132**, 563 (1963).

³T. Skettrup and I. Balslev, Phys. Rev. B **3**, 1457 (1971).

⁴J. Lagois and K. Hümmer, Phys. Status Solidi B **72**, 393 (1975).

⁵See, for instance, L. Merten, in *Festkörperprobleme—Advances in Solid State Physics*, edited by O. Madelung (Pergamon, Oxford, 1972), Vol. XII, p. 343.

⁶See, for instance, C. Kittel, *Introduction to Solid State Physics*, 4th ed. (Wiley, New York, 1971), p. 463.

⁷T. Skettrup and I. Balslev, Phys. Status Solidi **40**, 93 (1970).

⁸F. Willmann, S. Suga, W. Dreybrodt, and K. Cho, Solid State Commun. **14**, 783 (1974); D. Bimberg, K. Hess, N. O. Lipari, J. U. Fischbach, and M. Altarelli, Phys. Rev. B (to be published).

⁹See, for instance, M. J. Frankel and J. L. Birman, Phys. Rev. B **13**, 2587 (1976); D. L. Johnson and

P. R. Rimbey, *ibid.* **14**, 2398 (1976).

¹⁰R. Zeyher, J. L. Birman, and W. Brenig, Phys. Rev. B **6**, 4613 (1972).

¹¹J. Lagois and B. Fischer, Solid State Commun. **18**, 1519 (1976).

¹²H. Wolter, in *Handbuch der Physik*, edited by S. Flügge (Springer, Berlin, 1956), Vol. XXIV, p. 461. In his Eq. (2) or (3) on p. 472, ρ_1 and ρ_2 have to be replaced by $-\rho_1$ and $-\rho_2$ according to our ansatz of the plane wave $E \exp[i(kx - \omega t)]$ against that of Wolter's $E \exp[-i(kx - \omega t)]$. This replacement has also been done in Ref. 11.

¹³Reference 4 contains a detailed list of references; also see J. U. Fischbach, W. Dreybrodt, S. Suga, and F. Willmann, Phys. Status Solidi B **66**, 263 (1974); A. Frova, F. Evangelisti, and M. Zanini, Phys. Status Solidi A **24**, 315 (1974); J. U. Fischbach, W. Rühle, D. Bimberg, and E. Bauser, Solid State Commun. **18**, 1255 (1976).

¹⁴J. Lagois and B. Fischer, *Proceedings of the Thirteenth International Conference on the Physics of Semiconductors, Rome, 1976*, edited by F. G. Fumi, (Tipografia Marves, Rome, 1976) p. 788.