Polaritonic effects in superlattices

Gerard Czajkowski
Scuola Normale Superiore, Piazza dei Cavalieri 7, I-56126 Pisa, Italy
and University of Technology and Agriculture, Kaliskiego 7, PL-85790 Bydgoszcz, Poland

Franco Bassani and Alessandro Tredicucci
Scuola Normale Superiore, Piazza dei Cavalieri 7, I-56126 Pisa, Italy
(Received 30 November 1995)

We present a method to compute the optical functions of superlattices in the excitonic energy region including the effect of the coherence between the electron-hole pair and the electromagnetic field. The electron-hole screened Coulomb potential is adopted and the valence-band structure is taken into account in the cylindrical approximation, thus separating light- and heavy-hole motions. The calculated optical functions have poles in correspondence to the polariton eigenvalues for a multiplicity of excitonic states. We also calculate the amplitudes of higher polariton branches and the line shapes of the optical functions. Numerical examples appropriate to GaAs/$Ga_{1-x}Al_xAs$ superlattices are given and the effect of coherence is displayed in the line shape of the excitation spectrum. [S0163-1829(96)09027-3]

I. INTRODUCTION

The band-edge optical properties of semiconductor quantum wells and superlattices (SL's) are dominated by the excitonic behavior. Several approaches have been applied to compute the excitonic binding energies and the correct positions of the excitonic transitions have been obtained.[1,2] The above approaches are not sufficient for describing the optical functions of SL's since they do not take into account the polaritonic aspect. A recent contribution by Andreani considers the polariton aspect in the framework of the nonlocal susceptibility[3] and with the transfer-matrix approach.[4] In this case, however, the spatial dispersion of the susceptibility is determined by the motion of the center of mass and the exciton dipole is taken to be pointlike. This implies the introduction of *additional boundary conditions* (ABC's) for computing the optical functions. The discussion of ABC and ABC-free approaches has a long history (see, for example, Refs. 5–14). It has been shown that all types of ABC approaches have weak points, such as, for example, the ambiguity of ABC's (which becomes evident when higher excitonic states are taken into account); furthermore, the separation of the relative carrier motion from the center-of-mass motion makes the use of a dead-layer necessary.

A more satisfactory approach, from an *a priori* point of view, was introduced by Stahl and Balslev,[5] who extended to crystals the *coherent density-matrix approach*. Such a point of view has been recently adopted by Meier *et al.*[15] to introduce effects of electron and hole coherence with the electric field in the Bloch oscillations and Wannier-Stark ladder problem and by Glutsch and Chemla[16] to derive Bloch equations in a magnetic field.

We have recently shown[17] that the polariton aspect can be investigated using Stahl and Balslev's density-matrix approach, including electron-hole attraction and modeling the superlattice as an anisotropic medium characterized by effective masses parallel and perpendicular to the planes of the layers. We now present a calculational scheme that takes into account the band structure (in particular, the valence-band degeneracy), the Coulomb interaction between the electron and the hole, which implies higher polariton branches, and also considers coherence between the electron-hole pair and the radiation field. The ABC problem is completely avoided in our approach. The method is applied to a GaAs/$Ga_{1-x}Al_xAs$ SL, where we obtain the dispersion rule for polaritons and compute the optical functions.

The paper is organized as follows. In Sec. II we derive the basic equations for the density matrix approach adapted to the case of superlattices. In Sec. III we give the scheme for calculating SL optical functions in the case when the total thickness of the SL is much greater than the excitonic Bohr radius. In Sec. IV we discuss results obtained for GaAs/$Ga_{1-x}Al_xAs$ SL's. We present our conclusions in Sec. V.

II. DENSITY-MATRIX FORMULATION FOR SUPERLATTICES

Band-edge optical properties of superlattices can be discussed by modeling the superlattice as an effective anisotropic medium in which the quasifree carriers propagate and interact. In the low barrier limit the electron and hole motion in the confinement direction is determined by the superlattice potential and is replaced by an effective-mass motion, with the appropriate effective masses obtained from the miniband dispersion relations.[1,2] We neglect the possible formation of localized surface states in finite-size superlattices, which is usually not relevant, contrary to the case of organic heterostructures.[18] The superlattice exciton can then be treated like an exciton in an effective anisotropic medium and some previous results obtained for such a system[17] can be applied. More generally, the optical properties in such a system will be treated analogously to the case of a two-band semiconductor, with corresponding transition dipole density. Since SL excitons are of Wannier type, the transition dipole will have a spatial extension, characterizing the interaction of radiation with electrons and holes located at different sites.

This gives a spatial coherence between the electron-hole pair and the radiation field. In analogy to bulk semiconductor excitons, SL excitons induced by an electromagnetic wave propagating through the SL will give rise to mixed modes ''SL *polaritons*.''

All the above ingredients (Wannier excitons, effective-mass approximation, and exciton-polaritons with coherence) justify the use of the so-called coherent wave approach of Stahl and Balslev[5] to describe the optical properties of superlattices. In what follows we adapt Stahl and Balslev's method to the case of superlattices and show how to calculate the optical functions.

We consider a superlattice with N wells and barriers, both of thickness $L/2$, with the external surfaces located at the $z=0$ and NL planes. We assume that the conditions of small barrier regime are satisfied. We discuss the linear response of the slab to a normally incident electromagnetic wave, linearly polarized in the x direction

$$E_i(z,t)=E_{i0}\exp(ik_0z-i\omega t), \quad k_0=\frac{\omega}{c}. \tag{1}$$

In Stahl and Balslev's approach the linear response will be described by a set of coupled equations: two constitutive equations for the coherent amplitudes $Y_H(\mathbf{r}_e,\mathbf{r}_h)$ and $Y_L(\mathbf{r}_e,\mathbf{r}_h)$ for the heavy-hole exciton (H) and the light-hole exciton (L) and a Maxwellian field equation. The constitutive equations have the form

$$\partial_t Y_{12H}+\frac{i}{\hbar}H_{ehH}Y_{12H}=\frac{1}{\hbar}[i\mathbf{M}_H(\mathbf{r})\mathbf{E}(\mathbf{R})-\Gamma_H Y_{12H}], \tag{2}$$

$$\partial_t Y_{12L}+\frac{i}{\hbar}H_{ehL}Y_{12L}=\frac{1}{\hbar}[i\mathbf{M}_L(\mathbf{r})\mathbf{E}(\mathbf{R})-\Gamma_L Y_{12L}],$$

where Y_{12} contains the dependence on the spatial coordinates of the hole and of the electron, $\mathbf{r}=\mathbf{r}_e-\mathbf{r}_h$ is the relative electron-hole coordinate, and $\mathbf{R}$ is the center-of-mass coordinate. We have considered relaxation times $\hbar/\Gamma$ as phenomenological quantities. The operator H_{ehH} is the SL heavy-hole exciton effective mass Hamiltonian

$$H_{ehH}=E_{gH}-\frac{\hbar^2}{2M_{zH}}\partial_Z^2-\frac{\hbar^2}{2M_{\parallel H}}\nabla_{R_\parallel}^2-\frac{\hbar^2}{2\mu_{zH}}\partial_z^2-\frac{\hbar^2}{2\mu_{\parallel H}}\nabla_\rho^2 +V_{ehH}, \tag{3}$$

where we have separated the center-of-mass coordinate $R_\parallel$ from the relative coordinate ρ on the plane x-y; a similar formula holds for the light-hole exciton Hamiltonian H_{ehL}. In the above formulas the reduced masses in the z direction are given by

$$\frac{1}{\mu_{zH}}=\frac{1}{m_{ezH}}+\frac{1}{m_{hzH}} \tag{4}$$

for the heavy-hole case, with a similar expression for the light hole, where the electron and the hole effective masses in the z direction follow from the miniband dispersion relations (one for electrons and one for H and L holes, respectively)

$$\cos(KL)=\cos(k_WL_W)\cosh(k_BL_B) -\frac{1}{2}\left(\frac{1}{\xi}-\xi\right)\sin(k_WL_W)\sinh(k_BL_B), \tag{5}$$

where

$$k_W(K)=\sqrt{\frac{2m_WE(K)}{\hbar^2}},$$

$$k_B(K)=\sqrt{\frac{2m_B[V_B-E(K)]}{\hbar^2}}, \tag{6}$$

$$\xi(K)=\frac{m_Wk_B(K)}{m_Bk_W(K)}.$$

In Eqs. (5) and (6) the subscripts W and B denote the wells or barriers, $L=L_W+L_B$ is the superlattice period, and m_W and m_B are the respective bulk effective masses. For the in-plane effective masses we will take the values calculated in Ref. 19 by making use of the cylindrical approximation

$$m_{h\parallel H}=\frac{m_0}{\gamma_1+\gamma_2}, \quad m_{h\parallel L}=\frac{m_0}{\gamma_1-\gamma_2}, \tag{7}$$

where m_0 is the free-electron mass and γ_1,γ_2 are the Luttinger parameters with $\gamma_3=0$. The quantities M_{zH} and $M_{\parallel H}$ are the total heavy-hole excitonic masses in the growth direction and parallel to the layers, respectively, and similarly for the light-hole exciton masses.

The potential term in Eq. (3) represents the Coulomb interaction in an anisotropic medium[2,20]

$$V_{eh}=-\frac{e^2}{4\pi\epsilon_0\epsilon_b[\rho^2+z^2\epsilon_\parallel/\epsilon_z]^{1/2}}, \tag{8}$$

where we have introduced the two effective dielectric constant, $\epsilon_\parallel$ and ϵ_z, respectively, and defined $\epsilon_b=\sqrt{\epsilon_\parallel\epsilon_z}$. The ratio of the effective dielectric constants is given by[2]

$$\frac{\epsilon_z}{\epsilon_\parallel}=(\epsilon_WL_W+\epsilon_BL_B)\frac{\epsilon_W^{-1}L_W+\epsilon_B^{-1}L_B}{(L_W+L_B)^2}, \tag{9}$$

where ϵ_W and ϵ_B denote the isotropic dielectric constants in each layer of the superlattice.

The transition dipole density $\mathbf{M}_{\lambda\mu}$ and its integrated strength $\mathbf{M}_{0\lambda\mu}$ are defined as[5,21]

$$\mathbf{M}_{\lambda\mu}(\mathbf{r})=-\frac{e}{\Omega}\int d\mathbf{x}\, w_{v\lambda}^*(\mathbf{x})\mathbf{x}w_{c\mu}(\mathbf{r}+\mathbf{x}), \tag{10}$$

where λ indicates the valence subbands and μ the conduction subbands, Ω is the volume of the unit cell, and $w_{v\lambda}$ and $w_{c\mu}$ are the respective Wannier functions. Inserting the relation between Wannier functions and Bloch functions, in the limit of an infinite crystal, we arrive at

$$\mathbf{M}_{\lambda\mu}(\mathbf{r})=\frac{e\hbar}{im_0(2\pi)^3}\int_{\mathrm{BZ}}\frac{\mathbf{p}_{\lambda\mu}(\mathbf{k})\exp(i\mathbf{k}\cdot\mathbf{r})}{E_{c\mu}(\mathbf{k})-E_{v\lambda}(\mathbf{k})}d^3\mathbf{k}, \tag{11}$$

where $E_{c,v}(\mathbf{k})$ are the energies of band electrons, $\mathbf{p}_{\lambda\mu}(\mathbf{k})$ is the momentum matrix element between Bloch states, and the integration is extended over the first Brillouin zone. In the spirit of the effective medium approach to superlattices, the SL periodicity L acts upon the envelope functions, so that the SL *Brillouin zone* is now extended to $2\pi/L$. Optical transitions in the SL are governed by its specific selection rules, which we consider to be known.[1,22] Here we assume that the superlattice has a direct gap at the point $\mathbf{k}=0$, and expanding the energy difference to second order in $\mathbf{k}$ we obtain

$$\mathbf{M}_\lambda(\mathbf{r}) = -\frac{e\hbar}{m_0(2\pi)^3} \times \int \frac{\exp(i\mathbf{k}\cdot\mathbf{r}) i\mathbf{p}_\lambda(\mathbf{k})}{E_{g\lambda} + \frac{\hbar^2}{2}\mu_{\parallel\lambda}^{-1}(k_x^2+k_y^2) + \frac{\hbar^2}{2}\mu_{z\lambda}^{-1}k_z^2} d\mathbf{k}, \tag{12}$$

where we restrict ourselves to the most common case of a nondegenerate conduction band. The reduced masses to be inserted in Eq. (12) are known from (4) together with the miniband dispersion relations. In type-I SL's $\mathbf{p}_\lambda(\mathbf{k})$ is, to a good approximation, independent of $\mathbf{k}$ and equal to $\mathbf{p}_\lambda(0)$.[22] In such a case, extending the limits of integration to infinity, we obtain from (12), for the considered case of a SL with heavy-hole (HH) and light-hole (LH) valence bands,

$$\mathbf{M}_H(\mathbf{r}) = \frac{\mu_{\parallel H} e \mathbf{p}_H(0)}{2i\pi\hbar r m_0\sqrt{\alpha_H}} e^{-r/r_{0H}} = \frac{\mathbf{M}_{0H}}{4\pi\sqrt{\alpha_H} r_{0H}^2 r} e^{-r/r_{0H}}, \tag{13}$$

where

$$r = \sqrt{x^2+y^2+\frac{z^2}{\alpha_H}}, \tag{14}$$

$$\alpha_H = \mu_{\parallel H}/\mu_{zH},$$

a totally analogous expression holding for $\mathbf{M}_L(\mathbf{r})$. The integrated matrix element $\mathbf{M}_{0H}$ ($\mathbf{M}_{0L}$) is given by

$$\mathbf{M}_{0H} = \frac{\hbar e \mathbf{p}_H(0)}{i m_0 E_{gH}}, \tag{15}$$

with an analogous expression for the L case, and r_{0H} is the so-called coherence radius

$$r_{0H}^{-1} = \sqrt{\frac{2\mu_{\parallel H}E_{gH}}{\hbar^2}}. \tag{16}$$

The above expression gives the coherence radius in terms of effective band parameters, but we find it convenient to treat the coherence radii as free parameters that can be determined, for example, by fitting experimental spectra.

The coefficients $\Gamma_{H,L}$ in the constitutive equations (2) represent dissipative processes that, in general, are energy and temperature dependent.[23] In superlattices the radiative lifetime is infinite because k_z is still a good quantum number, contrary to the quantum-well case. As in bulk crystals,[24] we can expect a significant temperature-dependence of the SL spectra; microscopic analysis of damping parameters, which are the main temperature-dependent factors, requires future studies and will not be explicitly considered in this paper.

The coherent amplitudes Y_H, Y_L, together with the transition dipole densities $\mathbf{M}_{H,L}(\mathbf{r})$, give the total polarization of our effective anisotropic medium

$$\mathbf{P}(\mathbf{R}) = 2\int d\mathbf{r}\{M_H(\mathbf{r})\mathrm{Re}[Y_H(\mathbf{r},\mathbf{R})] + M_L(\mathbf{r})\mathrm{Re}[Y_L(\mathbf{r},\mathbf{R})]\}. \tag{17}$$

Equation (17), with the constitutive equations (2), connects the polarization with the electric field. Both the polarization and the electric field must obey Maxwell's equations, which must be solved to obtain the propagation modes. One advantage of the procedure presented here with respect to other approaches[1,3,4,22] is that microscopic theory and macroscopic theory are treated on the same footing and the problem of ABC's finds a natural solution in the conditions that the geometry of the problem imposes on Y_{12H}, Y_{12L}. They are obtained by requiring that the SL electron-hole pair functions $Y_{12H,L}$ decay very rapidly outside the SL, so that we can assume that

$$Y_H(\mathbf{r}_e, \mathbf{r}_h) = 0, \quad Y_L(\mathbf{r}_e, \mathbf{r}_h) = 0, \tag{18}$$

when the electron or the hole attains the SL external boundaries.

The above formulation contains all the ingredients for the calculation of all SL optical functions. They are obtained, as usual, by comparing the amplitudes of incident, reflected or transmitted electric fields, and display a dependence on the total SL thickness and on the coherence radii $r_{0H,L}$ of physical significance. In particular, the coherence of the electric field with the electron-hole pair is expected to give corrections on the results obtained from simpler approaches and we will display such effects.

III. OPTICAL FUNCTIONS OF SUPERLATTICES

The constitutive equations (2) can be solved introducing Green's functions G_H and G_L, the coherent amplitudes at the frequency ω of the electric field being

$$Y_H(1,2) = \int\int d\mathbf{r}_e' d\mathbf{r}_h' G_H(\mathbf{r}_e, \mathbf{r}_e'; \mathbf{r}_h, \mathbf{r}_h') \times \mathbf{M}_H(\mathbf{r}_e' - \mathbf{r}_h')\mathbf{E}(\mathbf{R}'), \tag{19}$$

with an analogous expression for $Y_L(1,2)$. This has the advantage that the boundary conditions can be imposed on the Green functions and are automatically satisfied by the coherent amplitudes. However, Green functions for the Hamiltonians in Eqs. (2) satisfying the boundary conditions (18) are not known in an analytical form. Such Green functions can be found for the kinetic part of the Hamiltonians. Therefore we separate the Hamiltonians of Eqs. (3) into a kinetic part H_{kin} and a potential term V and obtain from (2)

$$H_{\mathrm{kin}}Y = \mathbf{ME} - VY, \tag{20}$$

which gives the Lippmann-Schwinger equation, with the Green function G appropriate to the kinetic part

$$Y = G\mathbf{ME} - GVY. \tag{21}$$

The Green functions of the kinetic term, which take into account the dependence on $k_\parallel$ and $R_\parallel$, are[25]

$$G_H=\frac{1}{2\pi}\sqrt{\frac{m_{ezH}}{\mu_{\parallel H}}}\sum_{n=1}^{\infty} v_n(z_h)v_n(z_h')g_{nH}(z_e,z_e';\rho,\rho'), \tag{22}$$

with confinement functions

$$v_n(z)=\sqrt{\frac{2}{NL}}\sin\frac{n\pi z}{NL} \tag{23}$$

and

$$g_{nH}=\frac{2\mu_{\parallel H}}{\hbar^2}\int_0^\infty x dx J_0(x\rho)J_0(x\rho')\frac{\sinh\left(k_{nxH}\sqrt{\frac{m_{ezH}}{\mu_{\parallel H}}}z_e^<\right)\sinh\left(k_{nxH}\sqrt{\frac{m_{ezH}}{\mu_{\parallel H}}}(NL-z_e^>)\right)}{k_{nxH}\sinh\left(k_{nxH}\sqrt{\frac{m_{ezH}}{\mu_{\parallel H}}}NL\right)}, \tag{24}$$

where $z^<=\min(z,z')$, $z^>=\max(z,z')$, $J_0(x)$ are Bessel functions of zeroth order, and

$$k_{nxH}^2=\frac{2\mu_{\parallel H}}{\hbar^2}(E_{gH}-\hbar\omega-i\Gamma_H)+\frac{\mu_{\parallel H}}{M_{\parallel H}}k_\parallel^2+\frac{\mu_{\parallel H}}{m_{hzH}}\frac{n^2\pi^2}{(NL)^2}+x^2. \tag{25}$$

An analogous expression holds for the light-hole function G_L.

The above equations, with the addition of Maxwell's equations, give the solutions for the optical functions in closed form. It can be shown that the poles of the functions Y_H and Y_L, and consequently of the susceptibility, correspond to the eigenvalues of the corresponding anisotropic Schrödinger equation. When the superlattice thickness goes to infinity, we find the polariton modes. However, the solutions of Eqs. (21), together with Eq. (17) and the corresponding Maxwell equations, represent a nontrivial computational problem for realistic SL data. After exploiting the symmetry, we are left with a system of integral equations in a three-dimensional configurational space. We solved similar equations in Refs. 24 and 25, where a special choice of the interaction potential and of the dipole density was adopted. Here we will compute the fields $E(Z)$ and polarizations $P(Z)$ with the realistic electron-hole potential (8) and for the dipole density (13).

We consider a superlattice where $N\gg 1$ so that the total SL thickness is much greater than the Bohr radius of the SL exciton. In this case we distinguish a SL *bulk*, where the polariton waves propagate, and near surface regions, where SL excitons are created. Consequently, we divide the SL of thickness NL into two parts in the following way: (i) *surface range*, $0\leqslant Z\leqslant Z_0$ and $NL-Z_0\leqslant Z\leqslant NL$; (ii) *internal bulk region*, $Z_0\leqslant Z\leqslant NL-Z_0$, Z_0 being of the order of a few SL excitonic Bohr radii. We do not consider the possibility of two different transition layer thicknesses, because in our approach they represent the termination of the crystal and are larger than the SL period. The method consists of two steps. In the first step we solve equations (21) in the surface regions (i) neglecting the effect of electron-hole interaction. The neglected effects, which are essential for the formation of exciton-polaritons, are instead considered when solving the problem in the region (ii). Using a calculation scheme similar to that proposed in Refs. 27 and 28 we calculate the values of all functions (field and polarization) at a number of points in the surface regions, while in the bulk region we express the fields as a sum of K propagating waves

$$E(Z)=\sum_{j=1}^{K} E_j\exp(ik_{zj}Z)+\sum_{s=1}^{K} E_{K+s}\exp(ik_{z,K+s}Z), \tag{26}$$

$$P(Z)=\sum_{j=1}^{K} P_j\exp(ik_{zj}Z)+\sum_{s=1}^{K} P_{K+s}\exp(ik_{z,K+s}Z), \tag{27}$$

which have to be counted twice because of the two directions of propagation: $k_{z,K+j}=-k_{zj}$, $j,s=1,\ldots,K$; wave vectors k_{zj} follow from the bulk polariton dispersion relation derived in Ref. 17. The number of polariton waves is related to the number n of excitonic states considered, so we have $K=n+1$. The values $E(0),E(Z_1),\ldots,E(NL)$ and the amplitudes E_j,P_j of the bulk polariton modes are obtained by solving a set of equations that is given in the Appendix. Having the field $E(z)$ inside the SL and making use of the incident wave intensity E_{i0}, we obtain the SL optical functions from the relations

$$R=\left|\frac{E(0)}{E_{i0}}-1\right|^2,\quad T=\left|\frac{E(NL)}{E_{i0}}\right|^2,\quad A=1-R-T. \tag{28}$$

The above-described method, by an adequate choice of band parameters (energy gaps, effective masses, and dielectric function), can be used also for anisotropic semiconductor slabs when the slab thicknesses are much greater than the excitonic Bohr radii.

IV. RESULTS FOR GaAs/$Ga_{1-x}Al_x$As AND DISCUSSION

We have computed the optical functions of the most regular GaAs/$Ga_{1-x}Al_x$As SL with a chosen total thickness of 2000 Å (20 periods) in the belief that the total thickness is sufficient to obtain results that are independent of the number of layers. The values of the relevant parameters are well known and are given in Table I. In our scheme the field and

TABLE I. Parameter values for a GaAs/$Ga_{1-x}Al_xAs$ superlattice (SL) with $x=0.2$ and $L=100$ Å and for bulk GaAs (bulk) (data for bulk effective masses are from Ref. 19, energy is in units of meV, and mass is in m_0).

Quantity	SL	Bulk	Quantity	SL	Bulk
m_e	0.13	0.0665	α_H	0.336	0.75
m_{hzH}	3.12	0.34	α_L	0.769	1.282
M_{zH}	3.25	0.406	R_H^*	3.64	3.64
m_{hzL}	0.13	0.094	R_L^*	4.34	4.34
M_{zL}	0.2	0.161	$\lvert E_{1H}\rvert$	4.971	3.992
μ_{zH}	0.125	0.056	$\lvert E_{1L}\rvert$	4.758	3.984
μ_{zL}	0.06	0.039	E_{gH}	1596.8	1519.2
$\mu_{\parallel H}$	0.042	0.042	E_{gL}	1612.15	1519.2
$\mu_{\parallel L}$	0.05	0.050	Δ_{LTH}		0.08
ϵ_b	12.257	12.53			

the polarization in the surface regions are connected by a depth-dependent polarization (A1), which gives a depth-dependent susceptibility $\chi(Z)$, whose shape is defined by expression (36) and is displayed in Fig. 1 for two values of the coherence radius r_0. The field and the polarization in the bulk region (ii) of the SL define the polariton modes, whose dispersion follows from the relation

$$\frac{c^2k_z^2}{\omega^2}=\epsilon_b+\sum_n \frac{\epsilon_b\Delta_{LTH}F_{nH}}{E_{gH}+E_{nH}-\hbar\omega-i\Gamma_H+(k_za_H^*)^2(\mu_{\parallel H}/M_{zH})R_H^*}+\sum_n \frac{\epsilon_b\Delta_{LTL}F_{nL}}{E_{gL}+E_{nL}-\hbar\omega-i\Gamma_L+(k_za_L^*)^2(\mu_{\parallel L}/M_{zL})R_L^*}, \tag{29}$$

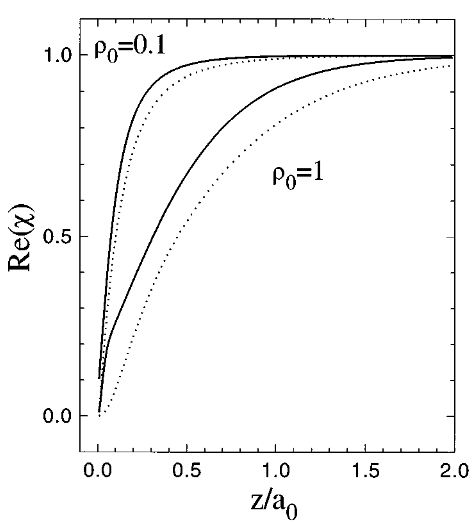

FIG. 1. Depth-dependent susceptibility $\chi_H(Z)$ for a GaAs/$Ga_{0.8}Al_{0.2}As$ SL (continuous line) and bulk GaAs $\chi(Z)$ (dotted line) for two values of the coherence radius $\rho_0=r_0/a^*$ and energy near the lowest excitonic resonance. Curves are normalized to the $Z\rightarrow\infty$ value.

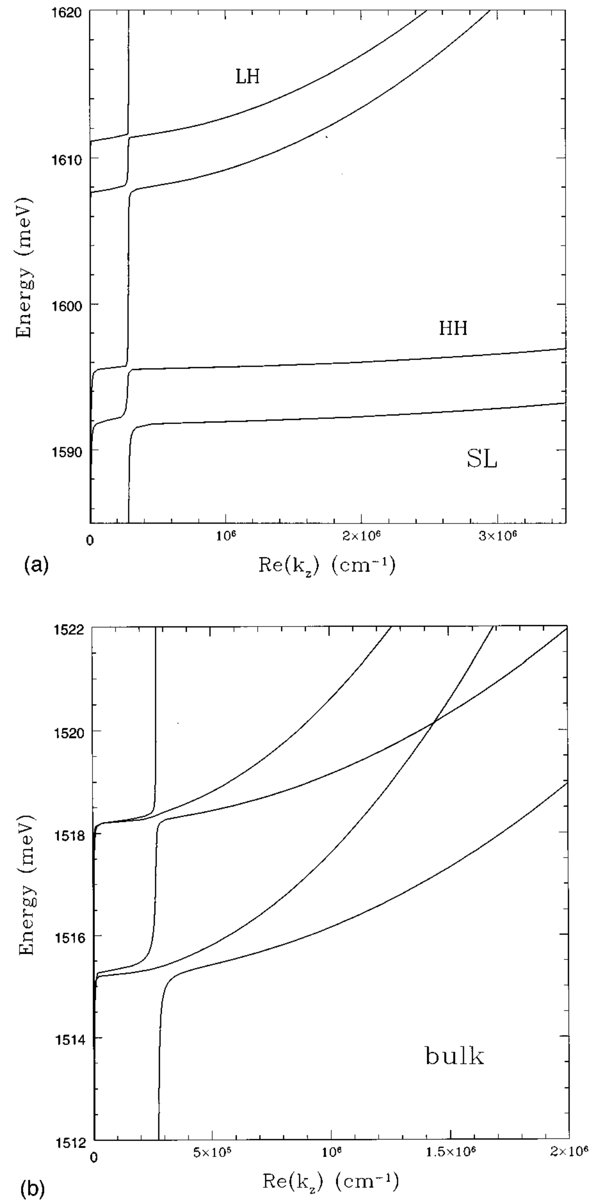

FIG. 2. Polariton dispersion relation for (a) a GaAs/$Ga_{1-x}Al_xAs$ SL ($N=20$, $L=100$ Å , $x=0.2$) and (b) bulk GaAs. We have used damping coefficients $\Gamma_H=\Gamma_L=0.01$ meV and coherence radii $\rho_{0H}=\rho_{0L}=0.1$ (other data are from Table I).

where the gaps E_{gH} and (E_{gL}) refer to the superlattice and are taken to be different for heavy-hole and light-hole excitons, $R_{H,L}^*, a_{H,L}^*$ are effective excitonic Rydbergs and Bohr radii, respectively, and $\Delta_{LTH,L}$ are L-T splittings. Oscillator strengths $F_{nH,L}$ and excitonic binding energies E_{nH}, E_{nL} have been calculated as shown in Ref. 17, and the effective masses follow from the miniband dispersions.[1,2] We consider the $1S$ and $2S$ excitonic states for both HH and LH excitons, so that the number of polaritonic states considered is $K=5$ and an isotropic background dielectric constant $\epsilon_z=\epsilon_\parallel=\epsilon_b$.

The resulting polariton dispersion in k_z is displayed in Fig. 2 and can be compared with the dispersion of a bulk GaAs layer of the same thickness, which is also shown in Fig. 2. We can observe in the case of the bulk dispersion the crossing of LH1 and HH2 polariton branches. This effect disappears in SL because of the great mass anisotropy (the total HH exciton mass is equal to $3.25m_0$ for the case considered).

The next step in our procedure is the computation of the electric fields of the different modes to obtain the optical functions. We have determined the electric field at a number of points in the surface region and ten field amplitudes in the inside region (ii) E_j ($j=1,\ldots,5$ for the ingoing and $j=6,\ldots,10$ for the outgoing waves), solving equations (A12) and (A19) and making use of the depth-dependent susceptibility (A7).

The optical functions for a GaAs/Ga$_{1-x}$Al$_x$As superlattice have been computed and can be compared with those of a bulk GaAs slab. The results for the reflectance R are displayed in Fig. 3 for different values of $\rho_0=r_0/a^*$. The effects of excitonic center-of-mass quantization appear as a fine structure above the first peak of each transition in the bulk, but only above the LH transition in the superlattice because of the largeness of the HH total mass M_{zH}.

While the resonance energies do not depend particularly on the coherence, it can be observed from the results of Fig. 3 that variations of the coherence radius change substantially the line shapes. The observed changes in the spectra are mostly due to the fact that an increase of r_0 diminishes the oscillator strengths, as already apparent in the model discussed in Ref. 17. Though the available experiments do not yet allow a sufficiently detailed comparison with the theoretical line shapes, we are confident that the coherence effect here described may be observed.

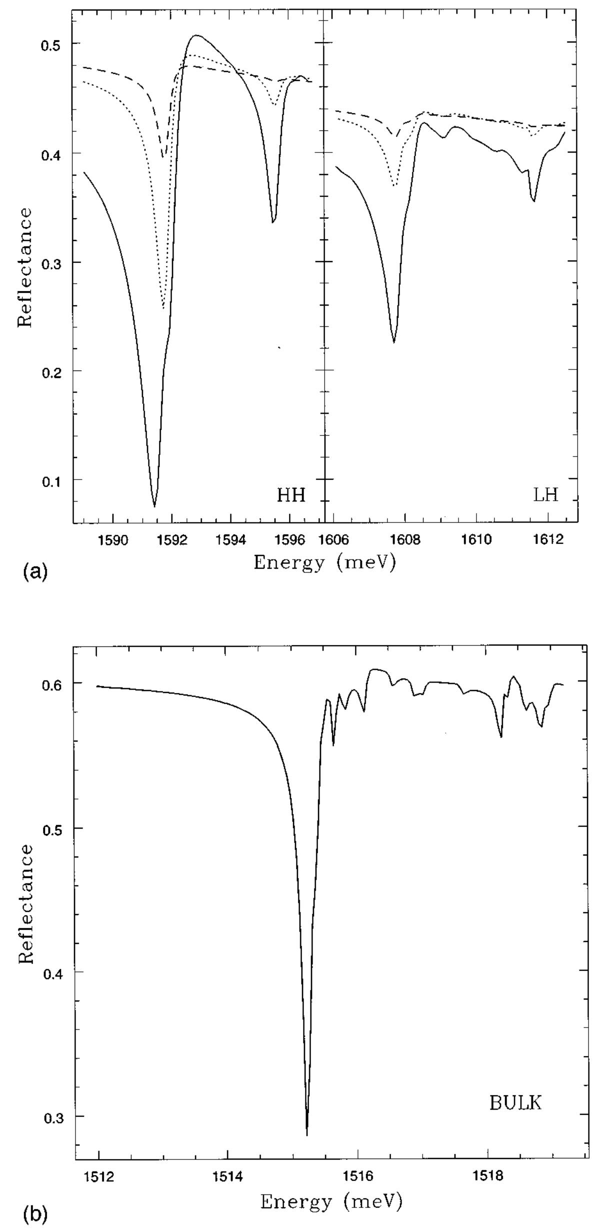

FIG. 3. (a) Reflectance of a GaAs/Ga$_{1-x}$Al$_x$As SL ($N=20$, $L=100$ Å, $x=0.2$) for three values of ρ_0: $\rho_{0H}=\rho_{0L}=0.1$ (continuous line), $\rho_{0H}=\rho_{0L}=0.5$ (dotted line), and $\rho_{0H}=\rho_{0L}=1$ (dashed line), displayed for energies near HH $n=1,2$ and LH $n=1,2$ excitonic states. We have used damping coefficients $\Gamma_H=\Gamma_L=0.2$ meV and a transition layer thickness $Z_0=150$ Å. For comparison we also give (b) the reflectance of a 2000-Å GaAs slab (data are from Table I). We have used damping coefficients $\Gamma_H=\Gamma_L=0.05$ meV and coherence radii $\rho_{0H}=\rho_{0L}=0.5$.

V. CONCLUSION

We have developed a simple mathematical procedure to calculate the optical functions of superlattices. The above model describes the propagation modes and the optical properties of a superlattice slab without the requirement of ABC's, taking into account the Coulomb interaction between electrons and holes and the existence of higher excitonic states. Our treatment includes anisotropic dispersion, as the *fractional dimensionality approach*,[29,30] but it also takes into account coherence of the electron-hole pair with the radiation field. The present method has been used to investigate the optical functions of GaAs/Ga$_{1-x}$Al$_x$As superlattices for the case of radiation incidence parallel to the growth direction.

ACKNOWLEDGMENTS

This research was based on work supported by Istituto Nazionale per la Fisica della Materia and by Consiglio Nazionale delle Ricerche. One of us (G.C.) wishes to acknowledge NATO and CNR for financial support and the Scuola Normale Superiore for the invitation and hospitality.

APPENDIX: CALCULATION OF SL OPTICAL FUNCTIONS

We show how to compute the optical functions of a semiconductor SL using the calculational scheme described in Sec. III. In particular, we show that the solutions for the electric field and the polarization through the slab can be obtained solving a set of integral equations numerically, by replacing them with a set of algebraic equations.

In the first step we calculate a depth-dependent susceptibility that connects the polarization and the field in the surface region (i). Making use of Eqs. (17), we obtain

$$P_1(Z)=P_{1H}(Z)+P_{1L}(Z) \quad \text{for } 0\leqslant Z\leqslant Z_0,$$

$$P_{1H,L}=\epsilon_0\chi_{H,L}(Z)E(Z), \tag{A1}$$

$$P_2(Z)=P_{2H}(Z)+P_{2L}(Z) \quad \text{for } NL-Z_0\leqslant Z\leqslant NL,$$

$$P_{2H,L}=\epsilon_0\chi_{H,L}(NL-Z)E(Z),$$

where

$$\chi_H(Z)=\frac{2\sqrt{\alpha_H}}{\epsilon_0}\int_0^\infty \rho d\rho\int_0^\infty \rho' d\rho'\int_{-A_H Z}^{B_H Z}dz\int_{-A_H Z}^{B_H Z}dz' \times\int_0^{2\pi}d\phi\int_0^{2\pi}d\phi' M_H(\mathbf{r})G_H(\mathbf{r},\mathbf{r}')M_H(\mathbf{r}'), \tag{A2}$$

with a similar expression for $\chi_L(Z)$. We make use of the dipole density (13). The Green function that we substitute into the above formula is proper to the kinetic part of the Hamiltonian (2) when neglecting the spatial dispersion term proportional to ∂_Z^2

$$G_H(\mathbf{r},\mathbf{r}')=G_H(z,z';\rho,\rho')=\frac{\mu_{\parallel H}}{\pi\hbar^2}\int_0^\infty x\,dx J_0(x\rho)J_0(x\rho') \times\frac{\sinh k_{xH}(A_H Z+z^<)\sinh k_{xH}(B_H Z-z^>)}{k_{xH}\sinh k_{xH}C_H Z}, \tag{A3}$$

where

$$k_{xH}^2=\frac{2\mu_{\parallel H}}{\hbar^2}(E_{gH}-\hbar\omega-i\Gamma_H)+\frac{\mu_{\parallel H}}{M_{\parallel H}}k_\parallel^2+x^2 \tag{A4}$$

and

$$A_H=\sqrt{\alpha_H^{-1}}\frac{M_{zH}}{m_{hzH}},\quad B_H=\sqrt{\alpha_H^{-1}}\frac{M_{zH}}{m_{ezH}},\quad C_H=\sqrt{\alpha_H^{-1}}\frac{M_{zH}}{\mu_{zH}}. \tag{A5}$$

An analogous expression holds for the light-hole function G_L. It can be proved that the Green function (A3) (and thus the H-exciton amplitude Y_H) satisfies the boundary conditions (18), which in the relative and center-of-mass coordinates have the form

$$G_H=0 \quad \text{for } z=-\frac{M_{zH}Z}{m_{ezH}},\quad z=\frac{M_{zH}Z}{m_{hzH}}, \tag{A6}$$

with similar relations for G_L. Performing the integrations in (A2), we arrive at the following expression for the susceptibility:

$$\chi_H(Z)=\frac{\mu_{\parallel H}M_{0H}^2}{2\pi\sqrt{\alpha_H}\epsilon_0\hbar^2 r_{0H}^4(r_{0H}^{-2}-k_{0H}^2)^2}I_H(Z) +\frac{\mu_{\parallel H}M_{0H}^2}{2\pi\sqrt{\alpha_H}\epsilon_0\hbar^2 r_{0H}^4(r_{0H}^{-2}-k_{0H}^2)}\Big[-r_{0H}+\frac{r_{0H}}{2} \times(e^{-2A_H Z/r_{0H}}+e^{-2B_H Z/r_{0H}})+A_H Z \times\mathrm{Ei}(-2A_H Z/r_{0H})+B_H Z\,\mathrm{Ei}(-2B_H Z/r_{0H})\Big], \tag{A7}$$

where $\mathrm{Ei}(Z)$ is the exponential-integral, and

$$I(Z)=\int_0^\infty x\,dx\bigg\{2\left(\frac{2\sinh k_x AZ\sinh k_x BZ}{k_x\sinh k_x CZ}-\frac{1}{R_x}\right) +4\frac{e^{-R_x AZ}\sinh k_x BZ+e^{-R_x BZ}\sinh k_x AZ}{R_x\sinh k_x CZ} -\frac{e^{-2R_x AZ}+e^{-2R_x BZ}}{R_x}+\frac{k_x}{R_x^2\sinh k_x CZ}[2e^{-R_x CZ} -(e^{-2R_x AZ}+e^{-2R_x BZ})\cosh k_x CZ]\bigg\}, \tag{A8}$$

where we have omitted the index H and used the abbreviation

$$R_x=\sqrt{r_0^{-2}+x^2}. \tag{A9}$$

A totally analogous formula holds for the susceptibility $\chi_L(Z)$. Having the suceptibility and thus the polarization, we solve the Maxwell equation for obtaining the electric field in the surface regions.

The field and the polarization in the bulk region (ii) are superpositions of polariton waves with amplitudes $\mathbf{E}_j$ and wave vectors k_{zj}:

$$E(Z)=\sum_{j=1}^{10}E_j e^{ik_{zj}Z},$$

$$P(Z)=P_H(Z)+P_L(Z), \tag{A10}$$

$$P_{H,L}(Z)=\epsilon_0\sum_{j=1}^{10}\chi_{jH,L}E_j e^{ik_{zj}Z},$$

where

$$\chi_{jH}=\sum_n\frac{\epsilon_b\Delta_{LTH}F_{nH}}{E_{gH}+E_{nH}-\hbar\omega-i\Gamma_H+(k_{zj}a_H^*)^2(\mu_{\parallel H}/M_{zH})R_H^*}, \tag{A11}$$

with an analogous expression for χ_{jL}. Thus, taking into account HH $n=1,2$ and LH $n=1,2$ excitonic states and exploiting the continuity of $P(Z)$, $E(Z)$, and dP/dZ, we obtain the following set of equations for determining the SL optical functions:

$$E(Z)+\frac{k_b^2}{\epsilon_0\epsilon_b}\left[\int_0^{Z_0}du G(Z,u)P_1(u) +\int_{NL-Z_0}^{NL}du G(Z,u)P_2(u)\right]+\sum_{j=1}^{10}E_j F_{js}(Z)=E_{\mathrm{hom}}(Z), \tag{A12}$$

where $s=1$ for $0\leqslant Z\leqslant Z_0$ and $s=2$ for $NL-Z_0\leqslant Z\leqslant NL$,

$$\frac{k_b^2}{\epsilon_0\epsilon_b}\left[\int_0^{Z_0}du G(Z_0,u)P_1(u)+\int_{NL-Z_0}^{NL}du G(Z_0,u)P_2(u)\right] +\sum_{j=1}^{10}E_j[F_{j1}(Z_0)+e^{ik_{zj}Z_0}]=E_{\mathrm{hom}}(Z_0), \tag{A13a}$$

$$\frac{k_b^2}{\epsilon_0\epsilon_b}\left[\int_0^{Z_0}duG(NL-Z_0,u)P_1(u)+\int_{NL-Z_0}^{NL}duG(NL-Z_0,u)P_2(u)\right]+\sum_{j=1}^{10}E_j[F_{j2}(NL-Z_0)+e^{ik_{zj}(NL-Z_0)}]=E_{\rm hom}(NL-Z_0), \quad (A13b)$$

$$\chi_H(Z_0)E(Z_0)-\sum_{j=1}^{10}\chi_{jH}E_je^{ik_{zj}Z_0}=0, \quad (A14a)$$

$$\left.\frac{d[\chi_H(Z)E(Z)]}{dZ}\right|_{Z=Z_0}-\sum_{j=1}^{10}\chi_{jH}E_jik_{zj}e^{ik_{zj}Z_0}=0, \quad (A14b)$$

$$\chi_L(Z_0)E(Z_0)-\sum_{j=1}^{10}\chi_{jL}E_je^{ik_{zj}Z_0}=0, \quad (A14c)$$

$$\left.\frac{d[\chi_L(Z)E(Z)]}{dZ}\right|_{Z=Z_0}-\sum_{j=1}^{10}\chi_{jL}E_jik_{zj}e^{ik_{zj}Z_0}=0, \quad (A14d)$$

and four additional equations obtained from Eqs. (A14) when replacing

$$Z_0 \text{ by } \mathrm{NL}-\mathrm{Z}_0, \quad (A15)$$

where

$$F_{j1}=-\sum_\lambda \frac{k_bf(Z)\chi_{j\lambda}}{2\epsilon_bW}\left[C_j^{(+)}e^{-ik_bNL}+\frac{k_b-k_0}{k_b+k_0}C_j^{(-)}e^{ik_bNL}\right], \quad (A16a)$$

$$F_{j2}=-\sum_\lambda \frac{k_bf(Z)\chi_{j\lambda}}{2\epsilon_bW}\left[C_j^{(+)}e^{-ik_bNL}+\frac{k_b-k_0}{k_b+k_0}C_j^{(-)}e^{ik_bNL}\right]+\sum_\lambda\frac{k_b\chi_{j\lambda}}{2\epsilon_b}[C_j^{(+)}e^{-ik_bZ}-C_j^{(-)}e^{ik_bZ}], \quad (A16b)$$

$$C_j^{(+)}=\frac{\exp[i(k_{zj}+k_b)(NL-Z_0)]-\exp[i(k_{zj}+k_b)Z_0]}{k_{zj}+k_b}, \quad (A16c)$$

$$C_j^{(-)}=\frac{\exp[i(k_{zj}-k_b)(NL-Z_0)]-\exp[i(k_{zj}-k_b)Z_0]}{k_{zj}-k_b}, \quad (A16d)$$

$\lambda=H,L$, and

$$G(Z,Z')=G_1(Z,Z')+\frac{\Theta(Z-Z')\sin k_b(Z-Z')}{k_b}, \quad (A17a)$$

$$G_1(Z,Z')=-\frac{i}{2k_bW}f(Z)f(NL-Z'), \quad (A17b)$$

$$f(Z)=e^{-ik_bZ}+\frac{k_b-k_0}{k_b+k_0}e^{ik_bZ}, \quad (A17c)$$

$$W=e^{-ik_bNL}-\left(\frac{k_b-k_0}{k_b+k_0}\right)^2e^{ik_bNL}, \quad (A17d)$$

$$E_{\rm hom}(Z)=E_{i0}\frac{2k_0f(NL-Z)}{(k_b+k_0)W}, \quad (A17e)$$

$$k_b=\sqrt{\epsilon_b}\,\omega/c, \quad (A17f)$$

where $\Theta(Z)$ is the Heaviside function. The notations follow those of Ref. 27. Equations (A12) to (A15) correspond to a set of $2p+10$ algebraic equations for $2p+10$ unknowns $E(Z_1),\dots,E(Z_p)$ $(0<Z_1,\dots,Z_p=Z_0)$, $E(Z_{p+1}),\dots,E(Z_{2p})$ for $Z_{p+1}=NL-Z_0,Z_{p+2},\dots,Z_{2p}<NL$, and ten amplitudes $E_1,\dots,E_{10}$. Having $E(Z)$ and $P(Z)$, we calculate the electric field on both sides of the SL:

$$E(0)=E_{\rm hom}(0)-\frac{k_b^2}{\epsilon_0\epsilon_b}\left[\int_0^{Z_0}duG(0,u)P_1(u)+\int_{NL-Z_0}^{NL}duG(0,u)P_2(u)\right]-\sum_{j=1}^{10}E_jF_{j1}(0), \quad (A18)$$

$$E(NL)=E_{\rm hom}(NL)-\frac{k_b^2}{\epsilon_0\epsilon_b}\left[\int_0^{Z_0}duG(NL,u)P_1(u)+\int_{NL-Z_0}^{NL}duG(NL,u)P_2(u)\right]-\sum_{j=1}^{10}E_jF_{j2}(NL) \quad (A19)$$

and thus the optical functions by relations (28).

[1] G. Bastard, *Wave Mechanics Applied to Semiconductor Heterostructures* (Les Editions de Physique, Paris, 1989).

[2] M. F. Pereira, Jr., I. Galbraith, S. W. Koch, and G. Duggan, Phys. Rev. B **42**, 7084 (1990).

[3] L. C. Andreani, Phys. Lett. A **192**, 99 (1994).

[4] L. C. Andreani, Phys. Status Solidi B **188**, 29 (1995).

[5] See, for instance, A. Stahl and I. Balslev, *Electrodynamics of the Semiconductor Band Edge* (Springer-Verlag, Berlin, 1987), and references therein.

[6] J. L. Birman, in *Excitons, Modern Problems in Condensed Matter Sciences*, edited by E. I. Rashba and M. G. Sturge (North-Holland, Amsterdam, 1982), Vol. 2, p. 27.

[7] S. I. Pekar, *Crystal Optics and Additional Light Waves* (Benjamin-Cummings, Menlo Park, CA, 1983).

[8] V. M. Agranovich and E. L. Ginzburg, *Crystal Optics with Spatial Dispersion and Excitons* (Springer-Verlag, Berlin, 1984).

[9] A. D'Andrea and R. Del Sole, Phys. Rev. B **32**, 2337 (1985).

[10] K. Cho, J. Phys. Soc. Jpn. **55**, 4113 (1986).

[11] A. D'Andrea and R. Del Sole, Phys. Rev. B **38**, 1197 (1988).
[12] F. Bassani and L. C. Andreani, in *Excited States Spectroscopy in Solids*, Proceedings of the Società Italiana di Fisica, Course No. XCVI, edited by U. Grassano and N. Terzi (North-Holland, Amsterdam, 1987), p. 1.
[13] H. Ishihara and K. Cho, Phys. Rev. B **41**, 1424 (1990).
[14] V. M. Axt and A. Stahl, Solid State Commun. **77**, 189 (1991).
[15] T. Meier, G. Von Plessen, P. Thomas, and S. W. Koch, Phys. Rev. B **51**, 14 490 (1995).
[16] S. Glutsch and D. S. Chemla, Phys. Rev. B **52**, 8317 (1995).
[17] F. Bassani, G. Czajkowski, and A. Tredicucci, Z. Phys. B **98**, 39 (1995).
[18] N. B. An and E. Hanamura, Mod. Phys. Lett. B **9**, 1609 (1995).
[19] R. Atanasov and F. Bassani, Solid State Commun. **84**, 71 (1992).
[20] F. Bassani, and G. Pastori-Parravicini, *Electronic States and Optical Transitions in Solids* (Pergamon, Oxford, 1975).
[21] G. Czajkowski and I. Balslev, Phys. Status Solidi B **130**, 655 (1985).
[22] P. Voisin, G. Bastard, and M. Voos, Phys. Rev. B **29**, 935 (1984).
[23] W. C. Tait, Phys. Rev. B **5**, 648 (1972).
[24] A. Tredicucci, Y. Chen, F. Bassani, J. Massies, C. Deparis, and G. Neu, Phys. Rev. B **47**, 10 348 (1993).
[25] G. Czajkowski and A. Tredicucci, Nuovo Cimento D **14**, 1203 (1992).
[26] F. Bassani, Y. Chen, G. Czajkowski, and A. Tredicucci, Phys. Status Solidi B **180**, 115 (1993).
[27] F. Bassani, G. Czajkowski, A. Tredicucci, and P. Schillak, Nuovo Cimento D **15**, 337 (1993).
[28] G. Czajkowski, A. Tredicucci, and F. Bassani, Nuovo Cimento D **17**, 1417 (1995).
[29] H. Mathieu, P. Lefebvre, and P. Christol, Phys. Rev. B **46**, 4092 (1992).
[30] P. Lefebvre, P. Christol, and H. Mathieu, Superlatt. Microstruct. **17**, 19 (1995).