Calculation of Exciton Absorption in Arbitrary Layered Semiconductor Nanostructures with Exact Treatment of the Coulomb Singularity

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Abstract—An accurate finite-element exciton calculation is presented. The Coulomb singularity is exactly taken into account without any further assumptions because analytical expressions are calculated for the Coulomb matrix elements even for the general three-dimensional case. Due to open boundary conditions, this is an accurate and fast model for calculating the optical properties of arbitrary semiconductor structures. The advantage will be demonstrated by an investigation of bulk, quantum-well, and multiquantum-well absorption.

Index Terms—Bulk absorption, electroabsorption, excitons, quantum wells, semiconductor device modeling.

I. INTRODUCTION

In this paper, the application of a density matrix formalism in real space is demonstrated to calculate the optical properties of semiconductor microstructures. Due to the bilocal formulation, no Kramers–Kronig transformation is needed to calculate the reference index change. Real space methods describe both bound and continuum absorption, which is calculated efficiently by open boundary conditions [1]. Additionally, the coupling of all exciton states is accounted for without increasing the model complexity. This is important for coupled quantum wells (QW’s) and superlattices. For QW’s and superlattices, the equations are radially symmetric if only s-excitons are considered. As these are the only ones contributing directly to the optical absorption, higher excitons like p- and d-excitons have only an indirect influence and can be neglected [2]. Due to the very general approach, bulk properties, including the Coulomb-enhanced Franz–Keldysh effect (FKE) [3], are also modeled efficiently.

II. THEORY

The optical absorption is calculated from the density matrix for excitons which previously has been described in detail [4], [5]. We are interested in the stationary equation for the coherent electron–hole amplitude $Y$, which is also called the semiconductor Bloch equation

\[
(H_{eh} - \hbar \omega - j \Gamma_{eh}) Y = \mu E
\]

where $E$ is the driving electric field and $\mu$ the optical dipole matrix element. A phenomenological dephasing rate $\Gamma_{eh}$ is introduced to describe the reduction of $Y$ by scattering processes. $\Gamma_{eh}$ corresponds to a scattering time $T_2 = 1/\Gamma_{eh}$. $H_{eh}$ denotes the Hamiltonian, which is used for general QW structures and superlattices, allowing spatially dependent effective masses for electrons and holes.

\[
H_{eh} = -\frac{\partial^2}{\partial z_e^2} + \frac{\hbar^2}{2 m_e} \frac{\partial}{\partial z_e} - \frac{\partial^2}{\partial z_h^2} + \frac{\hbar^2}{2 m_h} \frac{\partial}{\partial z_h} - \frac{\hbar^2}{2 m_e} \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial}{\partial r} \right) + E_e(z_e) - E_v(z_h)
\]

\[
\mu = M_{eh} \frac{\delta(z_e - z_h) \delta(r)}{2 \pi r^2}.
\]

Equation (1) is solved by applying a nodal finite-element (FE) approach. The solution $Y$ is expressed as a product of a form function $N$ and nodal values $Y_0$ with

\[
Y = N^T Y_0 = \langle N | Y_0 \rangle.
\]

For a variational expression, the inner product was chosen. This has been previously used for single-particle eigenvalue calculations [6].

\[
J = \langle Y | [H_{eh} - \hbar \omega - j \Gamma_{eh} Y] - \langle Y | \mu \rangle.
\]

An integration by parts is carried out for all second-order derivatives and the interface condition [7]

\[
\frac{1}{m_{eh}} \frac{\partial Y}{\partial z_{eh}} = \text{const}
\]

is inserted. With an expansion into single-particle wavefunctions, it can easily be shown that $Y$ obeys the same interface conditions as the single particle functions. In contrast to other approaches, interface conditions are already included in
the derivation of the variational expression. Therefore, no care
must be taken during discretization.

Inserting (4) into (5) gives the variational expression which is
stationary around the correct solution. Thus, variation with
respect to \( Y^4 \) results in [6]

\[
\frac{\partial J}{\partial Y^4} = 0
\]

(7)
leading to the equation system

\[
\begin{bmatrix}
N \end{bmatrix} \begin{bmatrix}
\Phi \end{bmatrix} = \begin{bmatrix}
\Lambda \end{bmatrix} \begin{bmatrix}
\Psi \end{bmatrix}.
\]

(8)

It is a sparse linear equation system with up to 27 diagonals
when using trilinear form functions. It can be solved efficiently
with subspace iteration solvers such as conjugate gradient (CG)
methods [8]. The FE method (FEM) discretization via the inner
product shows a very interesting numerical dispersion which is
discussed in the next section. The other advantage of this
approach is that the Coulomb singularity is modeled exactly
for polynomial base functions. Dropping all constants, the
Coulomb matrix element \( M_{Coul} \) in cylindrical coordinates is

\[
M_{Coul} = \int_{r_1}^{r_2} \int_{z_1}^{z_2} \frac{N^T N}{\sqrt{\gamma^2 + (z_c - z_h)^2}} r \, dr \, dz_h \, dz_c.
\]

(9)

This integral has been evaluated analytically for trilinear form
functions \( N \) by a symbolic math processing system. This
approach does not need the assumption of a ground state [5] or
modified quadrature methods [9]. Due to the exact inclusion of
the Coulomb matrix element, the presented approach is
more accurate and flexible. It includes the quantum-confined
Stark effect (QCSE), the Wannier–Stark effect (WSE), and the
Franz–Keldysh effect (FKE).

The Coulomb term generally can be determined for spatially
dependent \( \epsilon \) by using an infinite series of image charges [10],
[11]. The effect on the exciton binding energy has been investi-
gated in the InGaAs–GaAs system with the result that the
binding energy increases by about 6% for 10-nm-wide QW’s
[11]. In the AlGaInAs system, the dielectric constant is usu-
larily higher than in AlGaAs; thus, the dielectric mismatch
results in a smaller increase of the exciton binding energy. Using
approximate formulas derived by perturbation theory [11], the
exciton binding energy will increase by about 0.3 meV which is
on the order of 3%. This holds for the examples given in Section
V. Thus, the image charges can be neglected for simplicity. It
should be mentioned that it is straightforward to include image
charges with our theory because each image charge can be rep-
resented by a Coulomb potential again.

The optical polarization \( \mathbf{P} \) results from an integration over
relative coordinates \( r \) and \( z = z_c - z_h \) with

\[
\mathbf{P}(Z) = \int \mu Y(Z, z, r) \, r \, dr \, dz
\]

(10)
and depends on the center coordinate \( Z = (z_c + z_h)/2 \). The
material susceptibility \( \chi \) is defined as

\[
\chi(Z) = \frac{\mathbf{P}(Z)}{\alpha_0 \mathbf{E}(Z)}.
\]

(11)
The effective optical waveguide properties are a solution of
Maxwell’s equations. In typical waveguide modulators, the
induced optical field deviation is only small, thus a first-order
perturbation theory is appropriate to calculate a complex
effective dielectric function

\[
\epsilon_{eff} = \int \frac{1 + \chi(Z) \mathbf{E}^2(Z)}{\mathbf{E}^2(Z)} \, dz
\]

(12)
which is directly related to the propagating wave. If the real
part \( \epsilon'_{eff} \) of \( \epsilon_{eff} \) is much larger than the imaginary part, the
usual expression for the effective index \( n_{eff} \) and the absorption \( \alpha \) of
the propagating wave are

\[
n_{eff} = \sqrt{\text{Re}(\epsilon_{eff})}
\]

\[
\alpha = \frac{k_0}{n_{eff}} \text{Im}(\epsilon_{eff})
\]

(13)
(14)
where \( k_0 = 2\pi/\lambda_0 \) is the free space wavevector.

A. Numerical Dispersion

Dispersion and band nonparabolicity in semiconductors has
been investigated for a long time. The most simple approach
is the parabolic model. This model is only valid near the \( \Gamma_{eh} \)
point. \( k \cdot p \) models extend the valid range due to the inclusion
of band coupling but increase the numerical effort. The main effect
is that band coupling reduces single-particle energies for larger
wavevectors \( k \), which is overestimated for parabolic bands.

The numerical dispersion of the FEM shows an interesting
feature, because with the present discretization scheme the particle
energy is estimated to be smaller than the parabolic energy
modeled with the Hamiltonian. Perhaps this discretization error
is small, but band mixing is usually negligible. In Fig. 1, the
band structure of GaAs at the \( \Gamma \) point for electron, heavy-, and
light-hole bands is shown. Within the FEM discretization, no
electrons with energies higher than 3 eV can propagate within
the numerical scheme. The \( k \) range corresponds to about 25% of
the first Brillouin zone. Typically, all technologically important
effects occur in the region \( |k| < 0.1 \, \text{Å}^{-1} \). For this region, the
heavy holes match the parabolic dispersion quite well. In con-
trast, the FEM dispersion gives results very similar to the empirical
formulation for electron nonparabolicity labeled “nonpar”
[13]:

\[
E(1 + \zeta E) = \frac{n^2}{2m} k^2, \quad \text{with} \quad \zeta \approx 1/E_g.
\]

(15)
Thus, the numerical solution poses an additional degree of
freedom by choosing properly adjusted discretization with \( \Delta z \)
typically ranging between 1 and 2 nm. This not only minimizes the calculation time but also improves the dispersion model.

The use of the numerical nonparabolicity should be performed with care, because it has two drawbacks. First, it is material-dependent. Thus, the discretization must be adjusted to achieve the same result for different materials. Second, it can only be used for those structures that fit into the discretization range. If the geometry varies, either the numerical band structure or the device structure is altered.

III. 3-D BULK EXCITONS

The parabolic three-dimensional (3-D) bulk semiconductor absorption can be formulated with spherical coordinates. The Hamiltonian and the dipole matrix element for this case are

\[ \mathbf{H}_{\text{ch}} = -\frac{\hbar^2}{2m_e} \nabla^2 - \frac{e^2}{4\pi\varepsilon r} \]

\[ \mu = e \frac{\delta(r)}{4\pi\varepsilon r^2} \]

In this case, the formulation of the Coulomb matrix element is trivial because the singularity is compensated by the integration over the volume element \(4\pi r^2 dr\).

This formulation admits an analytical solution in terms of the hypergeometric functions \([4]\) which is used here to analyze the influence of the nonparabolicity introduced by the finite discretization. To obtain the analytical solution, the abbreviations

\[ C = -\hbar \omega - j\hbar \Gamma_{\text{elr}} \]

\[ \gamma^2 = \frac{2m_e}{\hbar^2} C \]

\[ k = \frac{m_e c^2}{4\pi\hbar^2 \gamma} \]

and the transformations

\[ z = 2\gamma r \]

\[ \psi(r) = e^{-\gamma r} F(r) \]

lead to the Kummer equation

\[ z \frac{\partial^2 F(z)}{\partial z^2} + (2 - z) \frac{\partial F(z)}{\partial z} - (1 - k) F(z) = 0 \]  

This equation is solved by the hypergeometric functions \(M(1 - k, 2, z)\) and \(U(1 - k, 2, z)\) \([14]\) and the Wronski determinant \(W\) belonging to this fundamental system. We obtain \([4]\)

\[ \chi = -\frac{2m_e M_e^2}{e_0 \hbar^2} \cdot \lim_{r \to 0} e^{-2\gamma r} M(1 - k, 2, 2\gamma r) U(1 - k, 2, 2\gamma r) \cdot \]

Recently, analytical solutions of the Schrödinger equation with the Hulthén potential, which describes a screened Coulomb potential, have been published \([15], [16]\).

In Fig. 2, the susceptibility \(\chi\) for GaAs is calculated for transitions between electrons and heavy, light, and split-off holes. The energy is given in terms of the exciton Rydberg energy \(R\) which in this case is 4.5 meV. Because no higher bands were modeled, a background susceptibility \(\chi_B\) has been added to the real part of \(\chi\) to match literature values at \(E = -50R\). The discretization is very fine to be sure to describe the same problem as solved analytically. The relative error (see Fig. 3) is below 1% for the heavy- and light-hole transitions which are usually of technical interest and below 2% for the split-off holes. This is due to the different masses. These figures show that the Coulomb singularity is resolved with high accuracy. In Fig. 4, the influence of discretization on the electron–heavy hole absorption is shown. The absorption edge is correctly described when using a fine or a coarse discretization. This even holds for the form of the
real part of the susceptibility near the bandgap. The coarse discretization gives a higher absorption for medium energies because the bandgap for finite $k_{||}$ is smaller due to numerical dispersion (see Fig. 1). For higher energies, no waves can propagate, thus reducing the absorption.

IV. ANISOTROPIC MASSES AND COULOMB-ENHANCED FKE

Anisotropic masses and an electrical bias can be described within a cylindrical coordinate system

$$H_{\text{eh}} = -\frac{\hbar^2}{2m_z} \frac{\partial^2}{\partial z^2} + \frac{\hbar^2}{2m_r} \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial}{\partial r} \right) + \frac{e^2}{4\pi\epsilon r^2 + z^2} + V(z) \delta(z) \delta(r)$$

$$\mu = M_{\text{eh}} \frac{\delta(z) \delta(r)}{2\pi r}$$

where the reduced mass $\mu$ now depends on $r$ and $z$ direction. $V(z)$ is an additional potential due to external bias. This case no longer allows an analytical treatment, but, for $m_z = m_r$ and $V = 0$, the problem reduces to (16) which is reproduced numerically with high accuracy. The absorption of bulk InP with anisotropic masses and Coulomb-enhanced Franz–Keldysh effect (FKE) is shown at room temperature which was modeled using $\Gamma_{\text{eh}} = 7$ meV for heavy and $\Gamma_{\text{eh}} = 8$ meV for light holes. All other material parameters were taken from [17]. In Fig. 5, the exciton function $Y$ is shown for a photon energy of 1.45 eV with an electric field of 40 kV/cm and a discretization $\Delta z = 0.5$ nm. Due to the use of open boundary conditions (perfectly matched layer, PML) [1], a 40-nm-wide computation window is sufficient to calculate the complete absorption characteristic including the continuum. The discretization and boundary errors are on the order of 1%. Due to the electrical bias, the exciton wave propagates to the right boundary of the computation window. This can be interpreted as an increased oscillatory behavior that results in shifted contours also. These effects are typical for the FKE resulting from the applied field and the slanted band edge. For vanishing Coulomb potential, the wavefunctions can be approximated by Airy functions [18], [19].

The rapid damping on the right-hand side is due to the eight-point-thick PML layer surrounding the calculation domain. Only a small distortion of the contours occurs for higher $r$ due to the strong anisotropic damping.

No reflections from the boundaries are visible, though the PML layer thickness is only eight points. These results show the efficiency of the PML, which is independent of the angle of incidence on the boundaries. In Fig. 6, the absorption edge is shown for different bias values. The exciton transition can be identified by the sharp band edge and the small plateau near the absorption edge. The electrical field broadens the absorption and results in oscillations above the gap, which is typical for the FKE. A sparse linear equation system of the order of 550 had to be solved 1000 times which needed less than 4 min on a standard PC with an AMD K6/400 CPU. Thus, the method proposed is a fast and efficient way to determine the unbound state absorption as well.

V. QUANTUM WELLS

Because no assumption was made about the behavior of the solution, the general 3-D formulation [2] contains all cases mentioned in earlier sections. It is valid for bulk material, QW’s, and superlattices, both for type I and II structures. To show the wide application range, we calculated a multiquantum-well (MQW) structure (Fig. 7) consisting of Al$_{0.44}$In$_{0.56}$As barriers and a four-layer well of 3-nm Al$_{0.45}$Ga$_{0.55}$As$_{2}$, 2-nm Ga$_{0.45}$Al$_{0.55}$As$_{2}$, and 6-nm Ga$_{0.45}$Al$_{0.55}$As$_{2}$ (from left to right). Material parameters were taken from [17], [20]. The whole structure consists of five QW’s. In the following, coupling effects between the QW’s are investigated by a variation of the barrier width.

In this QW structure, both electron and light holes contribute to the absorption, which is known as absorption edge merging [2], [21], [22]. In contrast to common strained QW’s with a light-hole-dominated structure, this structure has a dominant heavy-hole band edge for low bias. Therefore, this structure is better suited for integration into a monolithic laser-modulator module using the identical layer approach [23], [24]. Absorption edge merging is achieved by an enhanced light-hole Stark shift which results from the multi-layer QW [2]. Typically, in a single-layer QW, the heavy-hole shift is much stronger due to the higher confinement of the wavefunction.

Due to the small active layer, the electric field $E$ is nearly constant over the QW’s; thus, the calculation of effective properties (12) reduces to a simple spatial average. The absorption $\alpha$ is shown in Figs. 8 and 9 without and with an electric field, respectively. The enhanced light-hole QCSE is clearly seen. The barrier width of 6 nm leads to an acceptable decoupling of the wells due to the high barriers for electrons and holes in the AlGaInAs system. Coupling between different QW’s results in a smaller blue shift of the electron–light-hole absorption spectrum in comparison to a single QW, which is not shown here. Due to QW coupling, an additional light absorption peak occurs for applied bias at about 0.82 eV (see Fig. 9).

For thinner barriers, the coupling increases, leading to additional effects such as the WSE in superlattice structures. The WSE and the QCSE are both present due to the large well width, resulting in a broadening of the absorption spectrum for higher electric fields. In Fig. 10, the absorption spectra of structures...
with 6-nm- and 1-nm-wide barriers are compared. Because the real space method leads to position-dependent dielectric functions, both spectra were averaged over the position. To facilitate the comparison, the absorption of the 1-nm structure has been scaled by 5/6 due to a different barrier width. The absorption for an electric field of 0 and 100 kV/cm is shown. The broadening of the absorption edge occurs gradually with decreasing barrier thickness. For barriers below 3 nm, the exciton peak vanishes. The absorption characteristic shows a quite different behavior for thin barriers, which is further illustrated in Figs. 11 and 12. Independent of the bias, light and heavy holes contribute to the absorption with an equal Stark shift. The exciton peak in Fig. 12
VI. CONCLUSIONS

We have presented an efficient approach for calculating the absorption of arbitrary semiconductor microstructures by an exact treatment of the Coulomb singularity. The real space formulation with open boundary conditions allows for fully coupled calculations of very large QW structures.

REFERENCES


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