Electrooptical Functions and Ellipsometric Parameters of Excitons in Cylindrical Quantum Dots

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Abstract.
We show how to compute the optical functions (the complex electrousceptibility tensor, dielectric function, electoreflection spectra and ellipsometric parameters) for semiconductor quantum dots (QD) exposed to a uniform electric field in the growth direction, including the excitonic effects. The method uses the microscopic calculation of the QD excitonic wave functions and energy levels, and the macroscopic real density matrix approach (RDMA) to compute the electromagnetic fields and susceptibilities. The electron-hole screened Coulomb potential is adapted and the valence band structure is taken into account in the cylindrical approximation. In the microscopic calculations we solve the 6-dimensional two-particles Schrödinger equation by transforming it into an infinite set of coupled second order 2-dimensional differential equations with the appropriate boundary conditions. These differential equations are solved numerically giving the eigenfunctions and the energy eigenvalues. Then we used the RDMA and computed the frequency- and electric field strength dependent complex excitonic susceptibility tensor. The above approach enables us to determine the relative oscillator strength connected with excitonic resonances and to find the averaged susceptibilities for light- and heavy-holes excitons. Having the frequency dependent complex susceptibility tensor, we calculate the electrooptical functions for a QD. Numerical calculations have been performed for a InGaAs QD with a constant electric field applied in the growth direction. The optical Stokes parameters and ellipsometric parameters Ψ and Δ as functions of the frequency and the angle of incidence are also determined. A good agreement with experiment is obtained.

1. Introduction
The semiconductor nanostructures are manufactured in such a way that the carriers (electrons and holes), in the strong confinement regime, move inside the nanostructure (of the 1st type) upon confinement potentials of various shapes (for example hard-wall, parabolic). At the same time they interact by the dielectrically screened Coulomb potential. The difficulties in the theoretical description of the optical properties consist of the fact, that the Coulomb interaction is of the spherical symmetry, and depends on the relative coordinate, whereas the nanostructure confinement potentials act on each quasiparticle separately. This rules out an analytical solution of the corresponding Schrödinger equation, and various approximations have been proposed (for example, [1], [2]). In the method used below the cylindrical symmetry is exploited which enables to reduce the dimension of the relevant configuration space. The resulting two-dimensional Schrödinger equation is solved numerically to obtain the eigenvalues and eigenfunctions [3], [4]. Then, having them, and in the long-wave approximation, we can compute the optical functions. We show how this method works for a specific type of cylindrically shaped Quantum Dot.
2. Eigenvalues and eigenfunctions for a Quantum Disk

In what follows we first solve the 6-dimensional two-particles Schrödinger equation for a cylindrical Quantum Dot with hard-wall potentials in all directions. Such a QD is called Quantum Disk (QDisk). When a constant electric field \( \mathbf{F} \parallel z \) is applied, the two-band effective mass Hamiltonian has the form

\[
H = \frac{p_z^2}{2m_e} + \frac{p_{\parallel}^2}{2m_{\parallel}} + V_e(z_e) + V_h(z_h) + eF(z_e - z_h) + \frac{e^2}{4 \pi \varepsilon_0 \varepsilon_b \sqrt{p_e^2 + p_h^2 - 2p_e p_h \cos(\varphi_e - \varphi_h) + (z_e - z_h)^2}},
\]

where \( m_{\parallel}, m_{\perp} \) are the in-plane and \( z \)-hole effective masses, the electron effective mass \( m_e \) is assumed to be isotropic, \( F \) is the applied field strength, and \( \varepsilon_b \) is the static dielectric constant of the QDisk material. We assume the eigenfunctions in the form

\[
\psi^{e,h}(\mathbf{r}) = \sum_n \psi_{n1m1}^e (\rho_e, \varphi_e) \psi_{n2m2}^h (\rho_h, \varphi_h) f_n(z_e, z_h),
\]

where \( \psi_{n1m1}^e, \psi_{n2m2}^h \) are the known eigenfunctions with the corresponding one-particle eigenenergies \( \varepsilon^{e,h}(n) \) of the electron (hole) motion in a two-dimensional cylindrical quantum well [e.g., Ref. [5], p. 135], and \( f_n(z_e, z_h), n = (n_1, m_1, n_2, m_2) \) unknown functions to be determined; \( z_e \) and \( z_h \) are the carriers (electron- and hole) coordinates in the \( z \)-direction, respectively. The functions \( f_n \) and the total energy \( \varepsilon \) obey an infinite system of 2-dimensional differential equations

\[
\sum_{n'} V_{nn'} f_{n'}(z_e, z_h) = \varepsilon f_n,
\]

where \( \varepsilon = \varepsilon^{e,h}(n) - \mu_{\parallel} \partial^2 / \partial z^2 - \mu_{\perp} \partial^2 / \partial \rho^2 \) is the rescaled strength of the applied electric field, \( \mu_{\parallel} \) the in-plane reduced mass, and \( V_{nn'}(z_e - z_h) \) are the Coulomb interaction matrix elements between the in-plane confinement eigenfunctions.

We performed numerical calculations for InGaAs (disk)/ GaAs (barrier) QDisks, having in mind the experimental results by Oulton et al. [6]. As we noticed in Ref. [4], we can restrict the number of functions \( f_n \) used, obtaining a good convergence. Below the excitonic probability densities for the ground state for two chosen disk heights and two electric field strengths are displayed. In Figs. 1 the effects of the field strength for the disk of 30 nm height are shown, whereas in Fig. 2 the height 150 nm is considered. The effect is more pronounced for the larger height.

To check the validity of our approach we have computed the lowest state eigenenergy as a function of the applied external electric field. The comparison of our calculated results and the experimental results by Oulton et al. [6] is shown in Fig. 3. The best fit was obtained for the disk height of 6.2 nm and the radius equal to 15 nm, which agrees very well with the dimensions given in [6].

It is worth to mention that our approach does not contain any free parameters and the relevant material parameters for In0.5Ga0.5As quantum dot were calculated by linear interpolation of InAs data taken from Ref. [10] and GaAs data from Ref. [11]. The numerical values are: \( m_e = 0.04475 m_0, m_{\parallel} = 0.075 m_0, m_{\perp} = 0.34 m_0, \varepsilon_b = 13.84, m_0 \) - free electron mass.

3. Electrooptical functions

We now use the RDMA [7], [8] to compute the QDisk electrooptical functions. To this end we solve the so-called constitutive equations

\[
(\mathbf{H} - \hbar \omega - i \Gamma) \mathbf{Y} = \mathbf{ME}
\]
for the two-point correlation functions $\mathcal{Y}$ (excitonic amplitudes); $H$ is the above Hamiltonian (1), $\Gamma$ is a life-time broadening parameter, $E$ is the electric field amplitude of the incoming electromagnetic wave, and $M$ the interband transition dipole density. Expressing the amplitudes $\mathcal{Y}$ in terms of the obtained eigenfunctions $\Psi$ (2), we calculate the QDisk polarization from

$$P(R, t) = 2 \sum_\nu \int d^3r M^*_{\nu}(r) \mathcal{Y}_{\nu}(r, R, t)$$

(5)

the summation is over the allowed interband transitions, $R$ is the center-of-mass and $r$ the relative electron-hole pair coordinate. In the long-wave approximation we can compute the effective QDisk susceptibility $\chi$ and thus the effective dielectric function $\epsilon$. From the latter the QDisk ensemble optical functions are obtained. The real and imaginary part of the susceptibility are displayed in Figs 4 and 5, respectively. When the external static field is applied, we observe the diminishing of the oscillator strengths and the new peaks appearance due to the additional energy levels connected with the change of the effective e-h interaction potential. Having the

Figure 1. Probability density projection on the $(z_e, z_h)$ subspace: a) without the electric field, b) with the applied electric field $F = 20 F_1$ ($F_1$ - ionization field). QDisk height is equal to 30 nm and the radius $R=20$ nm.

Figure 2. The same as above, the disk height is equal to 150 nm, radius $R=20$ nm and $F = 0.8 F_1$.

Figure 3. Comparison of the calculated Stark shift (solid line) with the experimental results by Oulton et al. [6] (symbols).

Figure 4. The real part of the excitonic susceptibility.

Figure 5. The imaginary part of the excitonic susceptibility.
dielectric function, all optical functions can be calculated. For the purpose of illustration, we have chosen the electroreflectance (Fig. 6), the ellipsometric parameters $\Psi$ (Fig. 7), $\Delta$ (Fig. 8), and the Stokes parameters: here is shown the parameter $s_3$ - Fig. 9 (for their definitions see, for example, Ref. [9] and caption to Fig. 9). The ellipsometric parameters are relevant for the oblique incidence and the dependence on the incidence angle is shown. In particular, the parameter $s_3$ reflects the behaviour of the imaginary part of the susceptibility. In conclusion, we have shown how electrooptical functions for cylindrical Quantum Dots in the excitonic energy region can be computed with a high degree of accuracy.

**Figure 6.** Normal incidence reflectance spectra of a medium with distributed Quantum Disks for two values of the applied electric field.

**Figure 7.** The ellipsometric parameter $\Psi$.

**Figure 8.** The ellipsometric parameter $\Delta$.

**Figure 9.** The Stokes parameter $s_3 = 2 \text{Im}[E_{r}^{(p)} E_{r}^{(s)}]$. The parameters $\Psi$ and $\Delta$ from Figs. 7 and 8 describe the change of the polarization state of an electromagnetic plane wave: $(E_{r}^{(p)}/E_{r}^{(s)}) (E_{0}^{(p)}/E_{0}^{(s)})^{-1} = \tan \Psi \exp(i\Delta)$, the amplitudes $E_{0}$ stand for incident, $E_{r}$ for exiting waves, $p$ stands for the polarization parallel, $s$ for the perpendicular to the plane of incidence.

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