

Magneto-optics of layers of triple quantum dot molecules

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We consider theoretically the impact of the coherent manipulation of electronic states in the triple vertical lens-shaped circular InAs/GaAs quantum dot molecules on the collective magneto-optical reflection from layers of those nano-objects. The quantum dots have substantially different diameters in contrast to most of the known simulations. We demonstrate a possibility to drive molecular electronic ground states (to redistribute electronic wave functions within the molecule) by applying an external magnetic field along the system's growth direction. The change in the states leads to the redistribution of the intensity of the reflectance and absorbance peaks in the collective magneto-optical response of a layer of the triple quantum dot molecules. Varying the distance between quantum dots within the layer one can study optically the transition from "molecular" to "atomic" configuration. We show that the changes in the quantum mechanical behaviour of the molecules can be made observable by monitoring changes of the ellipsometric parameters Ψ and Δ in the magneto-optical response of layers of such nano-objects.

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1 Introduction Modern progress in semiconductor technologies makes it possible to fabricate vertically stacked quantum dots of high quality and uniformity [1]. Those semiconductor nano-objects provide us with the possibility to manipulate and reconfigure electronic wave functions in three-dimensional space. The quantum mechanical coherent coupling and forming of molecular states in the stacked quantum dots can be considered in complete analogy to real molecules. Quantum dot's molecules (QDM) have attracted much interest because they are attractive candidates for the implementation of quantum bits [2]. So, coherent dynamic control of the electronic configuration in QDMs is a key target for application of semiconductor nano-devices in quantum information technology. In the same time semiconductor nano-objects (quantum dots, quantum dot molecules, etc.) are very promising construction elements for semiconductor based metamaterials of high potential in the field of optics [3, 4]. Proper understanding of the connection between the electronic state coherent coupling in isolated nano-objects and the collective electromagnetic response from layers made from them [5] is a prerequisite to make new nano-structured metamaterials, not resembling anything in nature.

In this theoretical study we consider impact of the coherent manipulation of electronic states in the triple vertical lens-shaped circular QDM on the collective magneto-optical reflection from layers of those nanoobjects. The manipulation is performed for InAs/GaAs quantum dot molecules assembled from the dots with substantially different diameters and when an external magnetic field is along the system's growth direction. The influence of the surrounding semiconducting matrix upon the polarizability of embedded nano-objects (QDM) has been investigated using a hybrid discrete/continuum model [6].

2 Theory The system to be investigated consists of a square lattice with lattice parameter a_L , composed of InAs triple quantum dot molecules (as it is shown in Fig. 1) of characteristic size $a \ll \lambda$ (λ is wavelength of light of the frequency ω), embedded in GaAs matrix. The optical response of a single molecule is described by means of Kramers/Heisenberg type of polarizabilities. The hybrid discrete continuum method allows us to simulate the collective electromagnetic response of embedded molecules [6]. The bare embedded polarizability α_B (which was de-

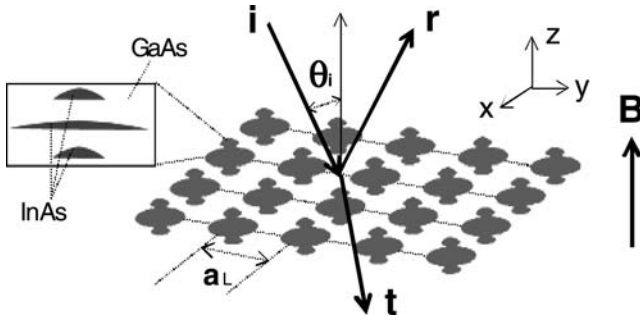


Figure 1 Schematic of magneto-optical response from a layer of triple quantum dot molecules.

rived in [7] as a measurable quantity) we can present by the expression

$$\tilde{\alpha}_{BE}(\omega) = \tilde{\alpha}_{BE}^S + \frac{3}{4} \frac{e^2}{\hbar} r_{ch}^2 [\hat{x}\hat{x}^T + \hat{y}\hat{y}^T] \sum_{hi, ej} \langle F_{hi} | F_{ej} \rangle^2 f_{hi, ej}(\omega), \quad (1)$$

where $\tilde{\alpha}_{BE}^S$ is the static part of the polarizability and $f_{hi, ej}(\omega)$ is the function introduced in [5], which depends on transition energies $E_{ij} = E_{hi} + E_{ej} + E_g$ of the resonance optical transitions (from hole energy levels (hi) to electron energy levels (ej) (E_g stands for the energy gap of the dot's material). The hole–electron overlap integrals $\langle F_{hi} | F_{ej} \rangle$ should be calculated using the envelop wave functions F , and r_{ch} stands for the bulk inter-band optical matrix element.

To compute the transition energies and the wave functions of the InAs/GaAs QDM with the hard-wall confinement potential we use realistic semiconductor material parameters (for instance the band offset of the InAs/GaAs strained heterostructure, corrected to the strain conditions band parameters, etc.). This allows us to simulate the magnetic dependence of the electron energy states and wave functions in a system of very asymmetrical shape: our quantum dots have substantially different diameters and different heights in contrast to the most of known simulations (see insert in Fig. 1). The effective one-band Hamiltonian for electrons (holes) [5] is taken as the following

$$\hat{H} = \frac{1}{2} \Pi_r \frac{1}{m_{e,h}(E, \mathbf{r})} \Pi_r + V_{e,h}(\mathbf{r}) + \frac{\mu_B}{2} g_{e,h}(E, \mathbf{r}) \boldsymbol{\sigma} \cdot \mathbf{B}, \quad (2)$$

where $\Pi_r = -i\hbar\nabla_r + e\mathbf{A}(\mathbf{r})$ is the electron momentum operator, ∇_r is the spatial gradient, $\mathbf{A}(\mathbf{r})$ is the vector potential of the magnetic field $\mathbf{B} = \text{curl } \mathbf{A}$, $m_e(E, \mathbf{r})$ is the energy and position dependent electron effective mass

$$\frac{1}{m_e(E, \mathbf{r})} = \frac{2P^2}{3\hbar^2} \left[\frac{2}{E + E_g(\mathbf{r}) - V(\mathbf{r})} + \frac{1}{E + E_g(\mathbf{r}) - V(\mathbf{r}) + \Delta(\mathbf{r})} \right]$$

and

$$g_e(E, \mathbf{r}) = 2 \left\{ 1 - \frac{m_0}{m(E, \mathbf{r})} \frac{\Delta(\mathbf{r})}{3(E + E_g(\mathbf{r})) + 2\Delta(\mathbf{r})} \right\}$$

is the electronic Landé factor. For the holes we use energy-independent effective mass m_h and the Landé factor g_h . In the equations above: $V(\mathbf{r})$ is the confinement potential, $E_g(\mathbf{r})$ and $\Delta(\mathbf{r})$ stand for the position dependent band gap and spin–orbit splitting in the valence band, P is the momentum matrix element, $\boldsymbol{\sigma}$ is the vector of the Pauli matrices, μ_B is the Bohr magneton, and m_0 is the free electron mass. The hard-wall confinement potential we present as: $V(\mathbf{r}) = 0$, if \mathbf{r} is inside the dots; and $V(\mathbf{r}) = V_0$, if \mathbf{r} is outside the dots, where V_0 is the band offset in InAs/GaAs heterostructures. In our simulation the magnetic field \mathbf{B} is directed along the system z -axis, and we can treat the problem in cylindrical coordinates (ρ, ϕ, z) . The envelop wave functions can be represented as

$$F(\mathbf{r}) = \Phi(\rho, z) \exp(i l \phi), \quad (3)$$

where $l = 0, \pm 1, \pm 2, \dots$ is the orbital quantum number. This leads to a two-dimensional problem in the (ρ, z) coordinates and forms a set of the quantum numbers $i, j = \{n, l, s\}$, where n is the principal quantum number, and $s = \pm 1$ refers to the orientation of the electron spin along z -axis. Finally we use the computed wave functions and energies to simulate the polarizability tensor (1).

Having ready the polarizability tensor for the isolated QDM we can define the collective electromagnetic response of the layer of QDM. For the system of the embedded nano-objects each presented by the discrete dipole strength \mathbf{p}_k we have to solve the system of equations [7]:

$$\tilde{\alpha}_{BE, k}^{-1} \mathbf{p}_k - \varepsilon_m^{-1} \sum_{q \neq k} \tilde{\mathbf{t}}_{kq} \mathbf{p}_q = \mathbf{E}_0, \quad (4)$$

where $\tilde{\mathbf{t}}_{kq}$ is the frequency dependant intercellular transfer tensor, screened by the matrix dielectric constant ε_m , \mathbf{E}_0 is the external electric field of light. Solution of (4) determines the Vlieger expressions [8] for reflection (r_{ss} and r_{pp}) and transmission (t_{ss} and t_{pp}) coefficients for the reflected electric field:

$$\begin{aligned} r_{ss} &= f_k (A_y \cos \theta_i - f_k)^{-1}, \\ t_{ss} &= 1 + r_{ss}, \\ r_{pp} &= \frac{f_k \cos \theta_i}{A_x - f_k \cos \theta_i} - \frac{f_k \sin^2 \theta_i}{A_z \cos \theta_i - f_k \sin^2 \theta_i}, \\ t_{pp} &= \frac{f_k \cos \theta_i}{A_x - f_k \cos \theta_i} - \frac{A_z \cos \theta_i}{A_z \cos \theta_i - f_k \sin^2 \theta_i}, \end{aligned}$$

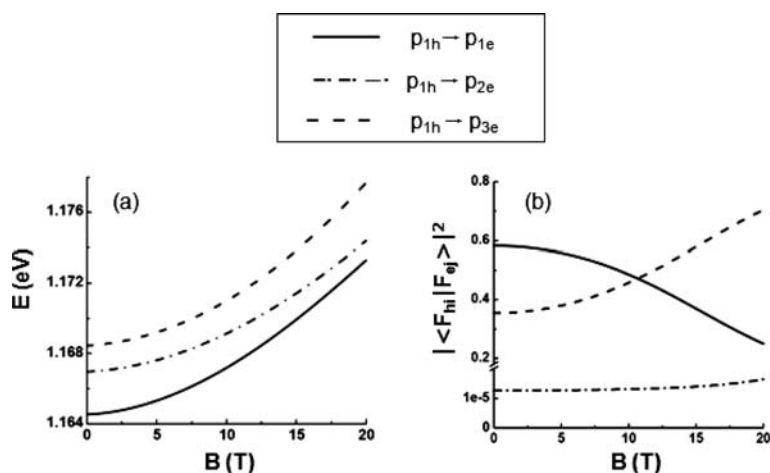


Figure 2 Optical transition $hi \rightarrow ej$ in the triple QDM at the edge of the optical absorption. Inter-dot distance: $d = 10$ nm. (a) Transition energies as functions of magnetic field for three closest to the edge transitions. (b) The modulus squared hole electron overlap integrals for the same transitions.

where subscripts “ss” and “pp” refer to the light polarization conventionally, θ_i is the angle of incidence,

$$A_u = \alpha_0 \alpha_{BE,uu}^{-1}(\omega) - \varepsilon_m^{-1}(f_u + \alpha_0 t_u),$$

$$f_k = 2\pi i a_L k,$$

$$t_u = -\frac{N_u}{\varepsilon_0 V} + \frac{ik^3}{6\pi\varepsilon_0},$$

$$k = \sqrt{\varepsilon_m} \frac{\omega}{c}, \quad \alpha_0 = 4\pi\varepsilon_0 a_L^3.$$

Here N_u ($u = x, y, z$) and V denote the depolarization factor and the volume of the molecule respectively. The planar tensor f is defined for the two-dimensional lattice of the nano-objects as $f_x = f_y = -f_z/2$, $f_z = 9.03362$ [8].

3 Simulation results In our simulations we use realistic semiconductor material parameters and dimensions of the dots in the molecule known in literature [9]. Our molecule consists of three quantum dots with substantially different radii $\rho_C > \rho_S$ and heights $h_C < h_S$ [C and S stand for the “Central” and “Side” dots in Fig. 1]. So, the system is non-uniform in z -direction. Here we calculate the low electron (upper hole) energy states of a system assembled from the lens-shaped dots with: $\rho_C = 25$ nm, $\rho_S = 9.5$ nm, $h_C = 3$ nm, $h_S = 4$ nm, and different inter-dot (base-to-base) distance d .

For the strained InAs inside of the dots according to the corrections done in [10] we choose: $E_{gInAs} = 0.842$ eV, $\Delta_{InAs} = 0.39$ eV, $m_{eInAs}(0) = 0.044 m_0$, $m_{hInAs} = 0.074 m_0$, $g_{hInAs} = 2.33$, and $\varepsilon_{InAs} = 15.2$. For the GaAs matrix we take from [11]: $E_{gGaAs} = 1.52$ eV, $\Delta_{GaAs} = 0.341$ eV, $m_{eGaAs}(0) = 0.067 m_0$, $m_{hGaAs} = 0.5 m_0$, $g_{hGaAs} = 3.6$, and $\varepsilon_m = \varepsilon_{GaAs} = 13.1$. The conducting band offset of the dot material is $V_0 = 0.474$ eV. The dumping parameter γ is chosen to be 1 meV for all calculations [5]. The energy states and wave functions of the electrons and holes confined in the quantum dot molecule, are found by the nonlinear iterative method [12] using the Comsol Multiphysics package (www.comsol.com). The static part of the polarizability tensor was calculated with the approach described in [13] also using Comsol Multiphysics package.

The tunnel coupling between dots creates molecular states in symmetric and anti-symmetric configurations. The non-uniformity of the QDM geometry in z -direction generates interesting features of the lowest energy states for electrons with $l = 0$ relating to the edge of the optical absorption in the system. For reason of clarity we concentrate on the lowest three energy electronic states $p_{e1} = \{1, 0, +1\}$, $p_{e2} = \{2, 0, +1\}$, $p_{e3} = \{3, 0, +1\}$, the highest heavy hole state $p_{h1} = \{1, 0, +1\}$, and inter-band optical transitions between them satisfying the selection rule: $l_h - l_e = 0$ (see Ref. [5–7, 14]). The corresponding allowed transition energies are shown Fig. 2(a). Increasing external magnetic field we can see the diamagnetic shift for the energies. The diamagnetic shifts are non-uniform – different slopes for

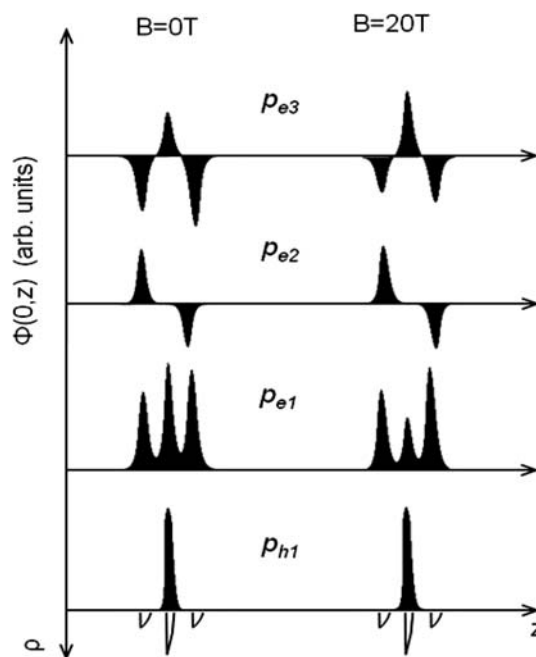


Figure 3 Wave function’s evolution in magnetic field for the lowest three electron’s energy levels and one hole level. The bottom graph is sketching the dots geometry in (ρ, z) -plane. Inter-dot distance: $d = 10$ nm.

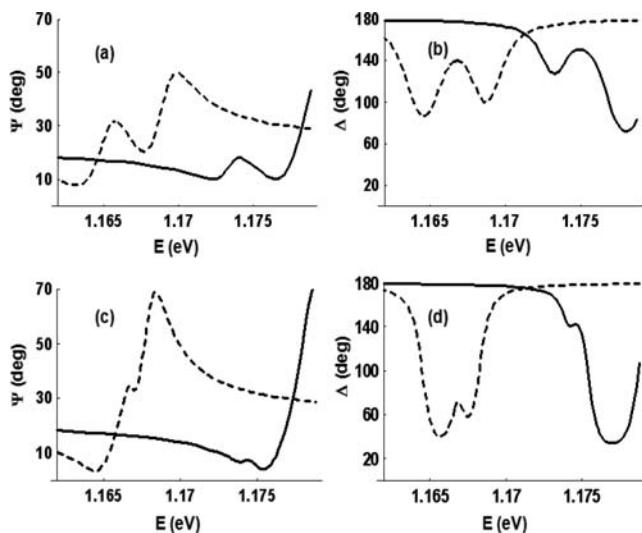


Figure 4 Magnetic field dependence of the ellipsometric angles for the layer of triple QDM, $a_L = 80$ nm: (a) and (b) inter-dot distance $d = 10$ nm; (c) and (d) inter-dot distance $d = 12$ nm. Dashed curve: $B = 0$ T; Solid curve: $B = 20$ T.

different transitions. More importantly, it can be seen from Fig. 3 that the electronic wave functions are redistributing within the molecule. For instance, the probability density of the state p_{e1} changes from the on-centre concentrated configuration to the on-side concentrated one and the probability density of the state p_{e3} changes in the opposite direction. In the same time, the probability density of the hole state p_{h1} remains concentrated on the central dot. Clearly, the magnetic field changes the overlap integrals for the corresponding optical transitions as it is shown in Fig. 2(b).

This clear quantum mechanical effect obviously shows itself in the collective magneto-optical response of the layer of QDM. Here we present results of our simulation for the ellipsometric angles Ψ and Δ , which follow from the commonly used definition:

$$\frac{r_{pp}}{r_{ss}} = \tan \Psi e^{i\Delta}.$$

For the angle of incidence $\theta_i = 60^\circ$, close to the Brewster angle, the ellipsometric angles Ψ and Δ clearly change their profile when the magnetic field changes from zero to 20 T as it is shown in Fig. 4(a, b). We present only peaks corresponding to the transitions considered in Fig. 3. Our calculation results show that we always can distinguish them if $\gamma = 1$ meV [5–7]. We stress that Ψ -angle peak-to-peak ratio replicates the peak-to-peak ratio for the probability density of the electronic states corresponding to the transitions (see Fig. 3). So, the ellipsometric data consists of important information on the quantum mechanical configuration of the molecular states in the QDM.

To demonstrate the connection between the reconfiguration of the probability density of the molecular states and the ellipsometric parameters we present in Fig. 4(c, d) the ellipsometric angles Ψ and Δ for the layers of QDM with

the inter-dot distance $d = 12$ nm. For this distance the tunnel coupling between dots with larger distance is weaker and the molecular states gradually “dissociate” into “atomic” states of isolated dots. For this configuration the “atomic” electronic states located mainly in the central dot should play the main role in the overlap integrals. Accordingly, for $d = 12$ nm we can recognize one main peak in Ψ -ellipsometric data.

In conclusion, we studied theoretically the magneto-optical response from a layer of embedded semiconductor triple quantum dot molecules. The calculation results clearly suggest measurable values for the ellipsometric angles for any modern ellipsometric setup. We emphasize that the magneto-ellipsometric data reproduce important information on the quantum mechanics of the molecules. Varying magnetic field and the distance between quantum dots within the layer we can investigate optically the transition from “molecular” to “atomic” behaviour of the system. The approach can be potentially useful for the design of new nano-structured metamaterials.

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