Electronic and optical properties of a double quantum dot molecule with Kane’s dispersion law

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Abstract. In the framework of the adiabatic approximation the electronic states and direct interband absorption of light in the double quantum dot molecule (QDM) are discussed. Cases of both standard parabolic and Kane’s dispersion law are considered. Analytical expressions for the wave functions and energy spectrum of the electron in the QDM are treated. The split of energy levels due to the possibility of the electron tunneling between quantum dots (QDs) in the molecule is revealed. The corresponding selection rules of quantum transitions for the direct interband absorption of light are obtained. The absorption edge behavior characteristics depending on the QDs geometrical sizes and the width of the QDs connecting region are revealed as well.

1. Introduction

QDMs are one of the modern semiconductor low-dimensional systems, which are of a great interest due to their possible applications in photonics, in the design of photovoltaic devices and various high-precision detectors, as well as the development of quantum computers [1-4]. There are many works devoted to detailed and versatile studies of vertically stacked (grown one over the other) systems of QDs. It is relatively easy to grow vertically arranged QDMs due to the internal symmetry of semiconductors. The purposeful experimental and theoretical study of horizontally stacked QDMs remains a significant challenge for the research groups [5-10]. However, during the growth of real semiconductor structures, in addition to the separately arranged QDs, both symmetric and asymmetric QDMs arise inevitably. Such original "errors" in the experiments lead to useful results, as history of science shows. The tunneling of charge carriers (CCs) from one QD to another becomes possible in closely located QDs. The splitting of the energy levels in the QDM, due to possible tunneling, allows one to consider these objects as an artificial analogue of the molecular orbitals of real molecules. Similarity of the electron orbitals of different analyte molecules to the levels of the QDM also allows the tunneling of electrons between the analyte molecules and the QDM. Practical implementation of this effect opens wide possibilities for the applications of QDMs for the design of various biochemical sensors and detectors, which may potentially lead to label free detection and identification of a wide range of analytes with a single molecule sensitivity.

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Due to the interaction between, the problem of finding of electronic states and wave functions (WFs) in the QDM is much more complicated. At the same time, their properties strongly depend on the external form, confining potential, inhomogeneity of CCs effective masses, and the number of QDs in the molecule. In order to understand QDMs properties, one needs to understand the essence of their correlations and coupling between them. Therefore, it is very important to develop clear criteria for classification and identification of QDs correlations and QDMs formations. Despite the objective difficulties, along with the correct choice of the confining potential, it is also very important task to describe an external form of the molecule as a whole. First proposed method for describing such systems are based on the following: the properties of individual QDs of different shapes are considered, and depending on their relative position (close or far) corresponding corrections due to their interaction are introduced [6]. In other words, several correlation terms are added in the Hamiltonian, which describes the interactions of the separately grown, but closely arranged QDs. However, most often there is no clear boundary between QDs in the molecule in real grown structures and a binding region – an isthmus – appears between QDs. Obviously, in such systems above mentioned description method will not be accurate. It is well known that even a small change in external shape of QDs leads to a significant change of the CC energy spectrum, and consequently to other physical properties of the sample. From this perspective, revealing a correlation between QDs, reduced to an accurate description of their binding region (isthmus), and a detailed study and estimation of the contribution of its presence on the behavior of the CC within the system. We proposed several models describing the correlation between QDs in molecules, with a single confining potential [7]. In particular, the additional splitting of the electronic levels due to the tunneling in presence of the central QDs was revealed. In the paper [11], authors considered the electronic structure of two laterally coupled Gaussian QDs filled with two particles. Their research shows that such structures have highly modifiable properties, promoting it as an interesting quantum device, showing the possible use of these states as a quantum bit gate.

Another major factor influencing the properties of the CC in the low-dimensional systems is the internal structure of matter composing the grown system. It is known that in the narrow-gap semiconductors the CCs' dispersion law is completely different from the standard parabolic law. However, there are many papers in which more complicated dependence of the CC effective mass on the energy is considered [12-16] in the framework of the Kane’s theory. For example, for the narrow-gap QDs of InSb, the dispersion law of CCs is non-parabolic and it is well described by the Kane's two-band mirror model [14,17]. Within the framework of the two-band approximation the electron dispersion law (light hole) formally coincides with the relativistic law. For more successful and realistic approximation of real grown double QDs, in the present paper, the electronic states and direct interband absorption of light in the QDM having the shape of the Cassini lemniscate of revolution for the standard and Kane's dispersion laws are calculated. The potential applications of these systems include sensors and detectors for various optoelectronic, medical and IT applications.
2. Electronic States

Consider an impermeable symmetric QDM consisting of two QDs having a shape of Cassini lemniscate of revolution (see figure 1). Then the potential energy of the particle in cylindrical coordinates can be written as

\[ U = \begin{cases} 0, & \rho^2 + \lambda Z^2 \leq \Lambda^2 \rho^2 + \lambda Z^2 - \alpha^2 + c_1^2 \leq 0, \\ \infty, & \rho^2 + \lambda Z^2 - \alpha^2 + c_1^2 > 0, \end{cases} \]

where \( c_1 \) is a focal length of the lemniscate of revolution, \( \alpha \) is a product of distances from foci to any point on the surface, and \( \lambda \) is a parameter of ellipsoidality (prolateness) of QDs. Here we will discuss the case of equal effective masses in both QDs \( m_e^* = m_h^* = m_e^* \), however, one can easily take into account their difference depending on the geometrical directions (for example, along the axis of the molecule and perpendicular to it \( m_{e1}^* \neq m_{h1}^* \)). Obtained results can be generalized to the case of the complete difference between the effective masses.

2.1. Parabolic dispersion law

In the regime of the strong size quantization (SQ), when the condition \( 2c_1 \ll a_B \) (where \( a_B \) is the effective Bohr radius) takes place, the energy of the Coulomb interaction between an electron and a hole is much less than the energy caused by the SQ contribution. In this approximation the Coulomb interaction between the electron and hole can be neglected, and the problem reduces to the determination of their energy states separately. First, we will discuss the case of the standard parabolic dispersion law of charge carriers. Then the Hamiltonian in cylindrical coordinates can be written

\[ \hat{H} = -\frac{\hbar^2}{2m_e^*} \left( \frac{\partial^2}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial}{\partial \rho} + \frac{1}{\rho^2} \frac{\partial^2}{\partial \varphi^2} \right) - \frac{k^2}{2m_{\|}^*} \frac{\partial^2}{\partial z^2} + U (\rho, \varphi, Z). \] (2)

It follows from the geometrical shape of the QDM that the particle motion in the \( OZ \) direction is faster than in the perpendicular direction. This fact allows one to apply the geometric adiabatic approximation. The system's Hamiltonian can be represented as a sum of the "fast" \( \hat{H}_f \) and "slow" \( \hat{H}_s \) subsystems Hamiltonians in dimensionless units: \( \hat{H} = \hat{H}_f + \hat{H}_s + U (r, \varphi, z) \), where

\[ \hat{H}_f = -\frac{1}{4} \left( \frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} + \frac{1}{r^2} \frac{\partial^2}{\partial \varphi^2} \right), \quad \hat{H}_s = -\frac{\beta}{4} \frac{\partial^2}{\partial z^2}, \] (3)

and the following notations are introduced: \( \hat{H} = \hat{H}_f, \quad r = \rho, \quad z = Z, \quad \beta = m_{\|}^* / m_{\|}^*, \quad E_B = \frac{k^2}{2m_e^* a_B^2} \) is the effective Rydberg energy of an electron, \( a_B = \frac{\kappa \hbar^2}{m_e^* e^2} \) is the effective Bohr radius of an electron, \( \kappa \) is a dielectric permittivity, \( e \) and \( m_e^* \) are the charge and effective mass of an electron, respectively. WFs of the problem is sought in the form

\[ \Psi (r, \varphi, z) = C e^{im\varphi} R (r; z) \chi (z), \] (4)

where \( C \) is a normalization constant. At a fixed value of the coordinate \( z \) of the "slow" subsystem the particle motion is localized in a two-dimensional potential well with an effective width (figure 2)

\[ r_e (z) = \sqrt{4 \lambda e^2 z^2 + a^2 - \lambda z^2 - c^2}, \] (5)

where \( a = \frac{a_1}{a_B}, \quad c = \frac{c_1}{a_B} \). After simple transformations, the Schrödinger equation
\[
-\frac{1}{4} \left( \frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} + \frac{1}{r^2} \frac{\partial^2}{\partial \theta^2} \right) R(r, z) = \varepsilon(z) R(r, z),
\]
reduces to the following Bessel equation
\[
R''(\eta, z) + \frac{1}{\eta} R'(\eta, z) + \left( 1 - \frac{m^2}{\eta^2} \right) R(\eta, z) = 0,
\]
where \( \eta = 2\sqrt{\varepsilon(z)} r \). The solutions of the equation (7) are the first kind Bessel functions:
\[
R(r, z) = A(z) J_m \left( \sqrt{\varepsilon(z)} r \right),
\]
where \( A(z) \) is a normalization constant. From the boundary conditions one obtains the following expression for the "effective" two-dimensional motion energy:
\[
\varepsilon(z) = \frac{\alpha_{n,m}^2}{4 r_0^2(z)} = \frac{\alpha_{n,m}^2}{4 \left( \sqrt{4 \lambda c^2 z^2 + a^4} - \lambda z^2 - c^2 \right)},
\]
where \( \alpha_{n,m} \) are roots of the Bessel functions. The energy (9) plays role of an effective potential energy in the Schrödinger equation of the "slow" subsystem, which obviously cannot be solved exactly. However, for the lower levels of the energy spectrum, the particle is localized in the geometrical center of one of the QDs, with coordinates \( z_0 = \pm \sqrt{4c^4 - a^4} \). After expanding (9) in a power series around these points one gets the expression (figure 3)
\[
\varepsilon(z) = \frac{\alpha_{n,m}^2 c^2}{a^4} + \frac{\lambda \alpha_{n,m}^2 (4c^4 - a^4)}{a^8} \left( z \pm \frac{\sqrt{4c^4 - a^4}}{2c} \right)^2,
\]
or
\[
\varepsilon(z) = \varepsilon_0 + \gamma \left( z \pm z_0 \right)^2,
\]
with \( \varepsilon_0 = \frac{4 \alpha_{n,m}^2 c^2}{a^4} \) and \( \gamma = \frac{4 \alpha_{n,m} \sqrt{\lambda (4c^4 - a^4)}}{a^4} \).

Note, that in the vast majority cases of the adiabatic approximation applications, in contrast to this specific one, the effective potential of the "slow" subsystem turns out parabolic. Here the electron motion in the direction of the "slow" subsystem occurs under the influence of the double parabolic effective potential as a consequence of the specific geometric form of the QDM. Further, following the geometric adiabatic approximation technique, it is necessary to solve the Schrödinger equation with the effective potential (11):
\[
\left\{ \frac{\beta}{4} \frac{\partial^2}{\partial z^2} + \varepsilon_0 + \frac{\gamma}{16} \left( z \pm z_0 \right)^2 \right\} \chi(z) = \varepsilon \chi(z).
\]
After the change of variables \( \xi = \sqrt{\frac{\gamma}{\beta}} (z \pm z_0) \), one obtains the parabolic cylinder equation for the left (\( \xi^- \)) and right (\( \xi^+ \)) sides of the double parabolic potential, respectively:

**Figure 3.** Confinement potential of the "slow" subsystem.
where \( \delta = \frac{4(e - e_0)}{\gamma \sqrt{B}} \). The solutions of the equation (13) are the parabolic cylinder functions \( D_\nu \) :

\[
\chi^\pm(z) = C^\pm D_\nu \left( \gamma \sqrt{B} (z \pm z_0) \right),
\]

where \( \nu = \delta - \frac{1}{2} = \frac{4(e - e_0)}{\gamma \sqrt{B}} - \frac{1}{2} \). Finally, in the strong SQ regime the WFs of the electron in the QDM can be written as

\[
\Psi(r, \varphi, z) = C_{\rho m} j_{\nu, m} \left( \frac{\alpha_{\rho m}}{r_0(z)} \right) D_\nu \left( \gamma \sqrt{B} (z \pm z_0) \right).
\]

The full energy of the electron in the strong SQ regime in the QDM with the standard parabolic dispersion law is determined from the sewing of WFs (14) at the point \( z = 0 \):

\[
\frac{\chi^-(z)}{\chi^+(z)} \bigg|_{z=0} = \frac{\chi^-(z')}{\chi^+(z')} \bigg|_{z=0}. \tag{16}
\]

### 2.2 Kane's dispersion law

Let us consider the QDM with Kane's dispersion law \( E^2 = P^2 S^2 + m^*_e S^4 \), where \( S \sim 10^5 \text{ cm/s} \) is the parameter related to the semiconductor bandgap \( E_g = 2m^*_e S^2 \). One can write the Klein-Gordon equation [18] for a symmetric QDM in the strong SQ regime, when Coulomb interaction between the electron and the hole is neglected:

\[
(P^2 S^2 + m^*_e S^4) \Psi(\rho, \varphi, Z) = (E - U(\rho, \varphi, Z))^2 \Psi(\rho, \varphi, Z). \tag{17}
\]

After simple transformations, the (17) can be written as the reduced Schrödinger equation

\[
\frac{P^2}{2m^*_e} \Psi(\rho, \varphi, Z) = E_1 \Psi(\rho, \varphi, Z), \tag{18}
\]

where \( E_1 = \frac{E^2 - m^*_e S^4}{2m^*_e S^2} = \frac{4E^2 - E_g^2}{4E_g} \). Then, repeating the above procedure of the adiabatic approximation, one gets the following expression for the \( OZ \) direction WFs:

\[
\chi_\pm(z) = C_\pm D_\nu \left( \sqrt{\gamma} (z \pm z_0) \right), \tag{19}
\]

where \( \nu = \frac{4(e_1 - e_0)}{\gamma} - \frac{1}{2} = \frac{4E^2 - E_g^2 - e_0}{\gamma} - \frac{1}{2} \). Note, that in contrast to the parabolic dispersion law, the WFs dependence on the CC energy is non-linear in the case of the Kane's dispersion law. Finally, the WFs of the electron in the QDM with Kane's dispersion law can be written as

\[
\Psi(r, \varphi, z) = C_{\rho m} j_{\nu, m} \left( \frac{\alpha_{\rho m}}{r_0(z)} \right) D_\nu \left( \gamma \sqrt{B} (z \pm z_0) \right). \tag{20}
\]

The total energy of the electron in the \( InSb \) QDM in the strong SQ regime is determined from the sewing of WFs (19) at the point \( z = 0 \) (see (16)).
3. Direct interband absorption of light

Consider the direct interband absorption of light in the QDM in the strong SQ regime, when the Coulomb interaction between the electron and a hole is neglected. The case of a heavy hole is discussed (\( m'_h \ll m'_e \)), and the absorption coefficient is determined by the expression [19]

\[
K = A \sum_{\alpha, \alpha'} \left| \langle \Psi^\alpha | \Psi'^{\alpha'} \rangle \right|^2 \delta \left( \hbar \Omega - E^\alpha - E'^{\alpha'} \right),
\]

where \( \alpha \) and \( \alpha' \) are the quantum number (QN) sets corresponding to the electron and a heavy hole, \( E_g \) is the bandgap of a bulk semiconductor, \( \Omega \) is an incident light frequency, \( A \) is a quantity proportional to the square of the matrix element taken by the Bloch functions. After simple calculations one gets the following expression for the absorption edge (AE) \( W_{100} \):

\[
W_{100} = 1 + \frac{\mu \Delta_{100}}{E_g}, \tag{22}
\]

where \( W_{100} = \frac{\hbar \Delta_{100}}{E_g} \), \( d = \frac{\hbar}{\sqrt{2\mu E_g}} \), and \( \mu = \frac{m'_e m'_h}{m'_e + m'_h} \) is the reduced mass of the exciton. The formula (22) describes the dependence of the effective bandgap on the parameters \( a \) and \( c \) (from the expression of the energy). With increasing the parameter \( a \), the AE shifts to the long wave region, whereas the increase in the \( c \) shifts AE to the short wave region.

Consider now the selection rules in quantum transitions. Quantum transitions for the energy levels allowed for the magnetic QNs \( m = -m' \), and for the "fast" subsystem QNs \( n = n' \). The continuity of logarithmic derivatives of the WFs (16) leads to the complete removal of the "slow" subsystem QNs selection rules. Note, that the analytical expression (22) is given taking into account above mentioned selection rules.

4. Discussion

When the geometric adiabatic approximation is applied, there is usually a family of the "slow" subsystem energy sublevels for each energy level of the "fast" subsystem. In most cases, the "slow" subsystem's energy levels turn equidistant, since the effective confinement potential is parabolic. As it is seen from the (11), the confinement potential of the "slow" subsystem has the form of a double parabola, in contrast to the other cases of the geometric adiabatic approximation application. Obviously, the family of the "slow" subsystem energy levels are not equidistant due to the formation of that kind of the confinement potential. Moreover, levels are double split due to the tunneling
possibility between two QDs. In other words, split levels of the "slow" subsystem positioned on each energy level of the "fast" subsystem. Note that the magnitude of the splitting depends on the geometrical parameters of the QDM and the "fast" subsystem QN as well. The first circumstance is conditioned by the electron probability of tunneling between quantum wells, and the latter one is the consequence of the external shape of the QDM, which allows the application of the adiabatic approximation.

Figure 4 represents the dependence of the first family of the energy levels in the QDM on the parameter $a$ at a given value of the lemniscate focal length $c$. As it is seen from the figure, with the increase in the parameter $a$, the energy curves decrease. This is a consequence of the QDM geometrical sizes increasing, which is in turn a result of the increase in the isthmus connecting two QDs. At the same time, the magnitude of the splitting of levels increases. This is due to the fact that the probability of the electron tunneling from one QD to another increases for large values of $a$. It should be noted that for small values of the width of the isthmus, the tunneling probability decreases, curves of the singlet and triplet states merge, and the distance between them becomes equidistant. In other words, in the case of weakly interacting QDs (low tunneling probability), the equidistant character of the energy sublevels' families restores. The tunneling probability increases with increasing energy, as expected. The splitting magnitude is more for the higher positioned levels. Thus, at $a = 2$ and $N = 0$, the singlet-triplet energy levels splitting is $\Delta E \approx 0.35E_r$, whereas at $N = 1$ the splitting is $\Delta E \approx 1.22E_r$. Numerical calculations for the parabolic dispersion law are performed for the GaAs QDM with the following parameters: $m_e^* = 0.067m_e$, $m_h^* = 0.12m_h$, where $m_e$ is a free electron mass, $\kappa = 13.8$, $E_r = 5.275meV$, $a_e = 104A^*$ and $a_h = 15A^*$ are the effective Bohr radii for the electron and a hole, respectively, $E_g = 1.43eV$ is a bandgap of a bulk semiconductor. Opposite behavior of the energy levels is observed in Fig. 5., which shows the dependence of the first family of the electron energy levels in the QDM on the lemniscate focal length $c$ at a given value of the parameter $a$. Thus, increasing the parameter $c$ leads to the increase in the energy levels, conditioned by the increase in the distance between two QDs and narrowing of the isthmus connecting them. At the same time, the increase in the lemniscate focal length also leads to the decrease in the electron tunneling probability. As a consequence, the magnitude of the splitting of the energy levels decrease. Note, that at the value $a = c$, the Cassini lemniscate becomes the Bernoulli lemniscate, which corresponds to the case of zero width isthmus between closely spaced QDs. However, the absence of the isthmus does not result in a complete removal of the splitting. Thus, at the value of $a = c = 2$ the excited levels are still split. This is due to a non-zero probability of the electron tunneling. Further increasing of the focal parameter leads to the final isolation of the QD. Note, that just in this case there is a recovery of the equidistant
energy levels of the individual ellipsoidal QDs [20]. It is also important to note that the increase in the splitting of levels with the QN increasing, occurs only for the symmetric QDM. When the symmetry is violated, the picture changes dramatically. In the case of non-identical QDs, the energy "zone" is formed for the probable tunneling, which position depends on the difference of sizes of QDs and the QDM (see e. g. [21]).

Numerical calculations in the case of the Kane's dispersion law are performed for the InSb QDM with the following parameters: $m_e = 0.013m_e$, $m_e^* \approx m_n^*$, where $m_e$ is a free electron mass, $E_g^K = 6 \times 10^{-4} eV$, $a_n^K = 500\AA$ is the electron effective Bohr radius, $E_g^C = 0.23eV$ is a bandgap of a bulk semiconductor. As it is seen, the strong SQ regime is achievable much easier for the semiconductor with the Kane's dispersion law, due to the small effective mass of the electron and the big value of effective Bohr radius.

Figure 6 illustrates the dependence of the first family of the electron energy levels in the QDM with the Kane's dispersion law on the parameter $a$ at the given value of the focal parameter $c$. As can be seen, with an increase in the parameter $a$, the splitting of levels increases. However, due to the dispersion law, the magnitude of the splitting is different. Thus, at $a = 0.2$ and $c = 0.17$ the splitting is $\Delta E^K \approx 11.32E_g^K$, or $\Delta E^C \approx 6.79meV$, whereas for the parabolic dispersion law one gets $\Delta E \approx 1.846meV$. This significant difference is due to the fact that in the strong SQ regime the difference between CC becomes one of the key factors affecting the motion of the particle [16]. The similar dependence of the energy on the focal parameter $c$ at the given value of the parameter $a$ is shown on the figure 7. Here, with an increase in the parameter $c$, the splitting of levels decreases, as expected.

Figure 8 plots the dependence of the edge frequencies of the direct interband absorption of light in the QDM versus the width of the isthmus between two QDs in the strong SQ regime. As it is seen from the figure, the increase in the isthmus width results in the decrease in the AE due to the reduction of the SQ effect (redshift). The curve corresponding to the small value of the parameter $c$ is positioned lower, since in this case the electron localization area in the QDM is wider. The opposite pattern is observed in the figure 9, showing the dependence of the AE on the focal length distance $c$. As expected, the increase in $c$ results in the blueshift of the edge frequencies, due to the SQ effect impact increase. For this reason, the curve corresponding to the small value of the $a$ is positioned higher. Note also, that at the small values of the focal length, the AE curves are positioned closer for the different values of the parameter $a$. The increase in the parameter $c$ leads to the increase in the curves discrepancy. This is due to the fact that for small values of the focal length, the width of the isthmus feels weaker due to the closely positioned QDs. At the big values of the $c$, the width of the isthmus becomes more influential on the motion and tunneling of the electron in the QDM.

![Figure 8](image_url)  
**Figure 8.** The dependence of the AE on the parameter "$a$" at the given values of the parameter "$c$".

![Figure 9](image_url)  
**Figure 9.** The dependence of the AE on the parameter "$c$" at the given values of the parameter "$a$".
5. Conclusion
During the growth of QDM nanostructures, it is possible to control the value of the energy of the electron and the splitting magnitude of the energy levels by manipulating sizes and the distance between QDs. The latter may promote the design of a new generation of the highly sensitive and selective sensors. In case of the coincidence of the split levels of the QDM and potentially detectable molecules of the gaseous medium, it is possible to achieve a successful tunneling of electrons from the gas molecules to the QDM. It is important also, that this tunneling is possible (most probable) only in the case of the full concurrence of interlevel distances of the QDM and a detectable molecule. This ensures high selectivity of the device, which will not fix another molecule by mistake. For the design of high-precision devices the most accurate measurements of the splitting of the energy levels are needed. For this purpose, the measurement of the light absorption frequencies in the QDM allows to establish the differences between the energy levels and the amount of the splitting.

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