Nonradiative resonance energy transfer between quantum dots

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Abstract. In the work we examined the mechanism of nonradiactive resonant energy transfer between quantum dots (QD), the probability of this process was calculated. The valence band has difficult structure due to the additional matrix element connected with another polarization of heavy holes. Dependences of transfer probability on distance between quantum dots and barrier heights for electrons were studied.

1. Introduction

The modern medicine passes to new more modern diagnostics methods with usage of quantum dots for detecting of DNA helix hybridization/splitting that can help us to study a genetic code and to warn genetic diseases, to create special DNA chips, to investigate the delivery of drugs to bodies, to detect the location of cancerous growth, etc. In this case it is necessary to construct the theory of this process [1,2]. One of mechanisms of realization of this process is nonradiactive resonant energy transfer. First this process was considered by Forster for interaction between protein molecule [3].

In the work we examined the mechanism of nonradiactive resonant energy transfer between quantum dots (QD), the probability of this process was calculated. The task was considered in Kane model where the nonparabolic zones are considered. The valence band has difficult structure due to the additional matrix element connected with another polarization of heavy holes. Dependences of transfer probability on distance between quantum dots and barrier heights for electrons were studied.

The accounting of the barrier finiteness leads to transitions with participation of electron and hole conditions with different main quantum numbers.

There is a strong dependence of energy transfer probability on distance between QD: $W_{if} \propto \dfrac{1}{r^6}$. This

dependence turns out at consideration of the dipolar allowed transitions. Due to such behavior practical realization of nonradiactive resonant energy transfer is possible: detecting of linkage or not linkage of DNA helixes with the attached QD both in vitro, and in vivo

2. Nonradiactive resonant energy transfer

2.1. Model

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Let's consider the system of two QD located from each other at final distance r in the dielectric environment ε. QD are made of the same material. QD have final potential barriers to electrons and holes. In an initial condition the electron in the energy donor QD is in a conduction band (condition 1), and in the acceptor QD the electron in a valence band (condition 3). As a result of the Coulomb interaction ($V = \dfrac{e^2}{|\mathbf{r} + \mathbf{r_d} - \mathbf{r_a}|}$), where $\mathbf{r_d}$ and $\mathbf{r_a}$ - QD radius), there is the nonradiactive resonant energy transfer: the birth an electron - hole pair in the acceptor due to the transferred excitement (transition of electrons from conditions 1, 3 in conditions 2, 4 respectively – fig.1). In fact, this process can be carried to the Auger process. This process takes place only under condition of energy conjunction of the donor and the acceptor accurate within half-width.
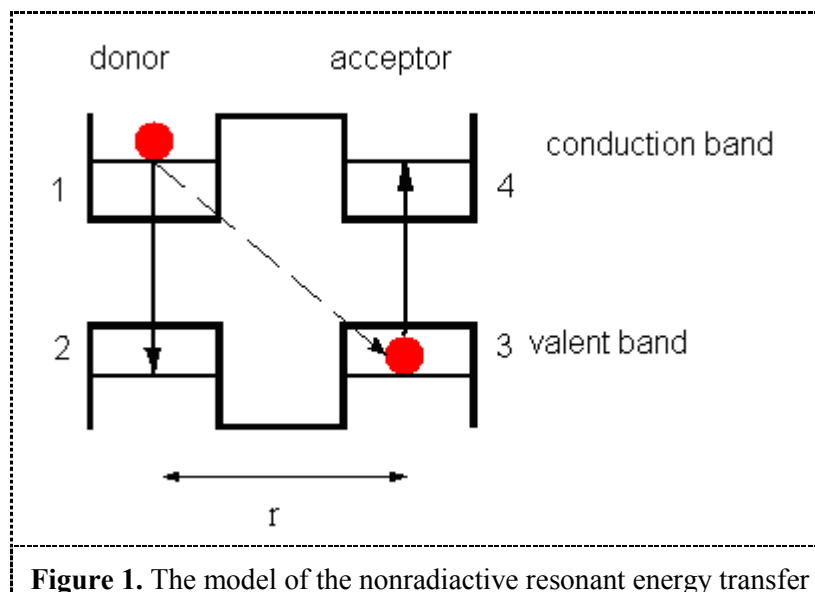

Figure 1. The model of the nonradiactive resonant energy transfer

For resonant energy transfer studying it is necessary to find wave functions of charge carriers. We will calculte wave functions within Kane's three-band model. Energy is counted from a bottom of the conduction zone. backs - orbital interaction is neglected,therefore $\Delta_{SO} = 0$.
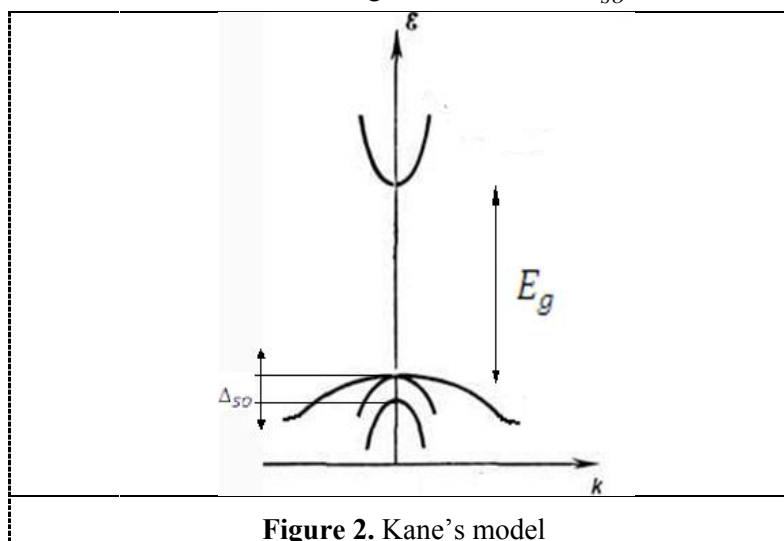

Figure 2. Kane's model

Wave functions can be written down in the following look:

$$\psi = \psi_s |s\rangle + \boldsymbol{\psi}|\mathbf{p}\rangle, \tag{1}$$

where $|s\rangle$ and $|\mathbf{p}\rangle$ are Bloch wave functions. Functions of s-type describe a condition in a conduction band, and p-type describe a condition of a valence band. Functions ψ_s and $\boldsymbol{\psi}$ are envelope wave functions. Kane's equations for envelope functions have the following appearance:

$$\begin{cases} \left(E_g - E\right)\psi_s - i\hbar\gamma\nabla\boldsymbol{\psi} = 0 \\ -E\boldsymbol{\psi} - i\hbar\gamma\psi_s - \dfrac{\hbar^2}{2m}\left(\gamma_1 + 4\gamma_2\right)\nabla(\nabla\boldsymbol{\psi}) - \dfrac{\hbar^2}{2m}\left(\gamma_1 - 2\gamma_2\right)\nabla\times\left(\nabla\times\boldsymbol{\psi}\right) = 0, \end{cases} \tag{2}$$

where γ- Kane's matrix element, γ_1 and γ_2 – Lattinzher's generalized parameters, m- free electron mass.

As we have spherical QD, so the equation (3) can be written is spherical coordinates, in this case variables are divided and thewave function can be presented in a radial part and spherical function:

$$\psi_s = R(r)Y_{lm}(\theta,\varphi) \tag{3}$$

The following equation for the radial part $R(r)$ takes place:

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial R}{\partial r}\right) + \left(k^2 - \frac{l(l+1)}{r^2}\right)R = 0 \tag{4}$$

For envelope functions of electrons in QD we received the following expressions (wave functions of electrons and light holes):

$$\psi_s = Aj_l(k_e r)Y_{lm}(\theta,\varphi)$$

$$\boldsymbol{\psi} = -\frac{i\hbar\gamma}{E}Ak_e\left(\left(\frac{l+1}{2l+1}\right)^{1/2}j_{l+1}(k_e r)\mathbf{Y}_{lm}^{l+1}(\theta,\varphi) + \left(\frac{l}{2l+1}\right)^{1/2}j_{l-1}(k_e r)\mathbf{Y}_{lm}^{l-1}(\theta,\varphi)\right) \tag{5}$$

Here $\mathbf{Y}_{lm}^{l\pm1}(\theta,\varphi)$ is vector spherical harmonicas, l and m – values of the full angular moment and its projection to an axis z, $j_l(kr)$ – Bessel's spherical function, k_e – a wave vector of electrons, A – a normalizing constant.

By analogy we received wave functions under a barrier. For heavy holes we receive:

$$\psi_s = 0$$

$$\boldsymbol{\psi}_{h2} = Bj_l(k_h r)\mathbf{Y}_{lm}^l(\theta,\varphi) \tag{6}$$

$$\boldsymbol{\psi}_{h1} = C\left(\left(\frac{l}{2l+1}\right)^{1/2}j_{l+1}(k_h r)\mathbf{Y}_{lm}^{l+1}(\theta,\varphi) + \left(\frac{l+1}{2l+1}\right)^{1/2}j_{l-1}(k_h r)\mathbf{Y}_{lm}^{l-1}(\theta,\varphi)\right)$$

where k_h – wave vector of holes. B and C – normalizing constants.
Here are boundary conditions fo electrons and light holes:

$$\begin{cases} \psi_s^< = \psi_s^> \\ \dfrac{1}{E_g - E}\dfrac{\partial\psi_s^<(R)}{\partial r} = \dfrac{1}{E_g - E - V}\dfrac{\partial\psi_s^>(R)}{\partial r} \end{cases} \tag{7}$$

for heavy holes

$$\begin{cases} \psi_s \equiv 0, \quad \boldsymbol{\psi}^<(R) = \boldsymbol{\psi}^>(R), \quad \dfrac{d\psi_\perp^<(R)}{dr} = \dfrac{d\psi_\perp^>(R)}{dr} \end{cases} \tag{8}$$

Indexes «<» and «>» means that parametrs undertake respectively at the left and the right side of heterobarrier.

2.2. Matrix element

For finding of probability of nonradiactive resonant energy transfer we will use Fermi's "gold" rule within a first order perturbation theory:

$$W_{if} = \frac{2\pi}{\hbar} \left| M_{if} \right|^2 \delta \left(E_i - E_f \right) \qquad (9)$$

Our task is reduced to find a matrix element of transition from an initial condition (in the donor there is an electron - hole pair, the acceptor is empty) to the final (the donor is empty, in an acceptor there is an electron - hole pair):

$$M_{if} = \left\langle \psi_4(r_d)\psi_2(r_2) \middle| V \middle| \psi_1(r_d)\psi_3(r_a) \right\rangle =$$
$$\frac{e^2}{\varepsilon} \iint d^3 r_d d^3 r_a \frac{\psi_3 \psi_4^* \psi_1 \psi_2^*}{\left| \mathbf{r} + \mathbf{r_d} - \mathbf{r_a} \right|} \qquad (10)$$

where r_d and r_a are coordinates of the electron - hole pairs in the donor and the acceptor respectively, ψ_1 and ψ_2 wave functions of the initial and final conditions of the electron in the donor, ψ_3 and ψ_4 wave functions of the initial and final conditions of the electron in the acceptor. In our work we consider two types of the hole polarization, two matrix elements are possible, they answer to different polarization: M_{if1} and M_{if2}. Let's consider a matrix element with polarization for heavy holes. For the calculation of a matrix element we will use Fourier representation for Coulomb interaction:

$$\frac{e^2}{\left| \mathbf{r} + \mathbf{r_d} - \mathbf{r_a} \right|} = \frac{4\pi e^2}{\varepsilon} \int \frac{d^3 q}{(2\pi)^3 q^2} \exp \left(i\mathbf{q}(\mathbf{r} + \mathbf{r_d} - \mathbf{r_a}) \right) \qquad (11)$$

Matrix element has the following view

$$M_{if1} = \frac{4\pi e^2}{(2\pi)^3} \int \frac{d^3 q}{q^2} \exp(i\mathbf{q}\mathbf{r}) \int d^3 r_d \psi_1 \psi_2^* \exp(i\mathbf{q}\mathbf{r_d}) \left(\int d^3 r_a \psi_3 \psi_4^* \exp(i\mathbf{q}\mathbf{r_d}) \right)^* \qquad (12)$$

Substituting of wave functions for electrons and heavy holes in this expression, we receive the following type of a matrix element for heavy holes:

$$M_{if1} = \frac{2e^2}{\varepsilon} \left(\frac{P}{E_g} \right)^2 A_1 A_2 A_3 A_4 \frac{1}{r^3}$$

$$\int_0^{R_d} r_d^2 dr_d \left(\left(\frac{l_{hd}}{2l_{hd}+1} \right)^{1/2} C_{l_{hd}+1,m_{hd},1,0}^{l_{hd},m_{hd}} j_{l_{hd}+1}(k_{hd}r_d) j_{l_{ed}}(k_{ed}r_d) \delta_{l_{ed},l_{hd}+1} \delta_{m_{ed}m_{hd}} + \right.$$

$$\left. + \left(\frac{l_{hd}+1}{2l_{hd}+1} \right)^{1/2} C_{l_{hd}-1,m_{hd},1,0}^{l_{hd},m_{hd}} j_{l_{hd}-1}(k_{hd}r_d) j_{l_{ed}}(k_{ed}r_d) \delta_{l_{ed},l_{hd}-1} \delta_{m_{ed}m_{hd}} \right)^* \qquad (13)$$

$$\int_0^{R_a} r_a^2 dr_a \left(\left(\frac{l_{ha}}{2l_{ha}+1} \right)^{1/2} C_{l_{ha}+1,m_{ha},1,0}^{l_{ha},m_{ha}} j_{l_{ha}+1}(k_{ha}r_a) j_{l_{ea}}(k_{ea}r_a) \delta_{l_{ea},l_{ha}+1} \delta_{m_{ea}m_{ha}} + \right.$$

$$\left. + \left(\frac{l_{ha}+1}{2l_{ha}+1} \right)^{1/2} C_{l_{ha}-1,m_{ha},1,0}^{l_{ha},m_{ha}} j_{l_{ha}-1}(k_{ha}r_a) j_{l_{ea}}(k_{ea}r_a) \delta_{l_{ea},l_{ha}-1} \delta_{m_{ea}m_{ha}} \right)$$

The similar expression we received for other polarization of heavy holes is too enormous and doesn't shown

2.3. Discussion of results

Let's consider dependence of probability of nonradiactive resonant energy transfer on the following parameters: distances between QD, height of a potential barrier. Carried out above calculation showed that the probability of nonradiactive resonant energy transfer depends on distance between quantum dots as: $W_{if} \sim \dfrac{1}{r^6}$ for the dipolar allowed transitions (fig. 3).
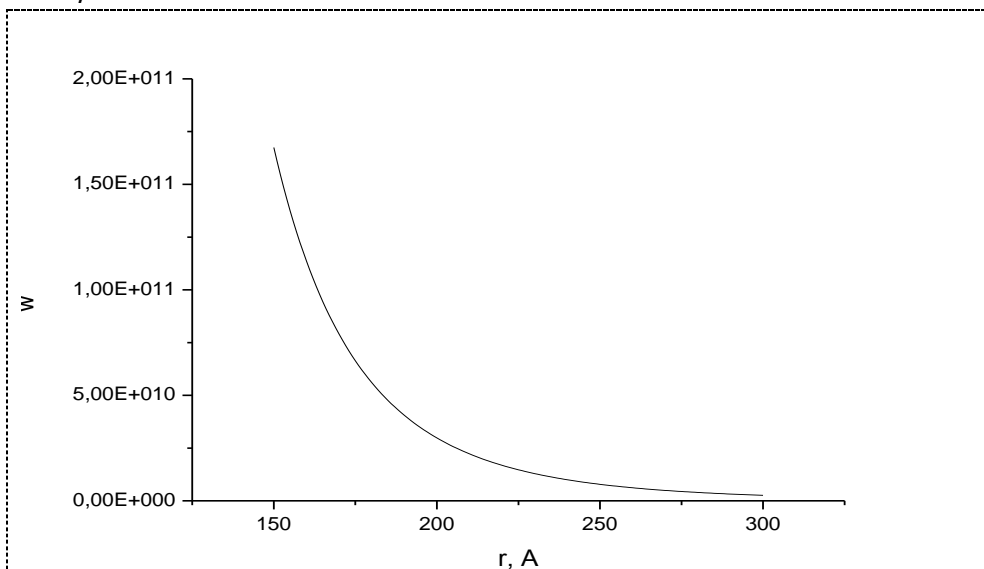

Figure 3. Dependence of probability of nonradiactive resonant energy transfer on distance between quantum dots. Y axis denote energy transfer probability in s^{-1}
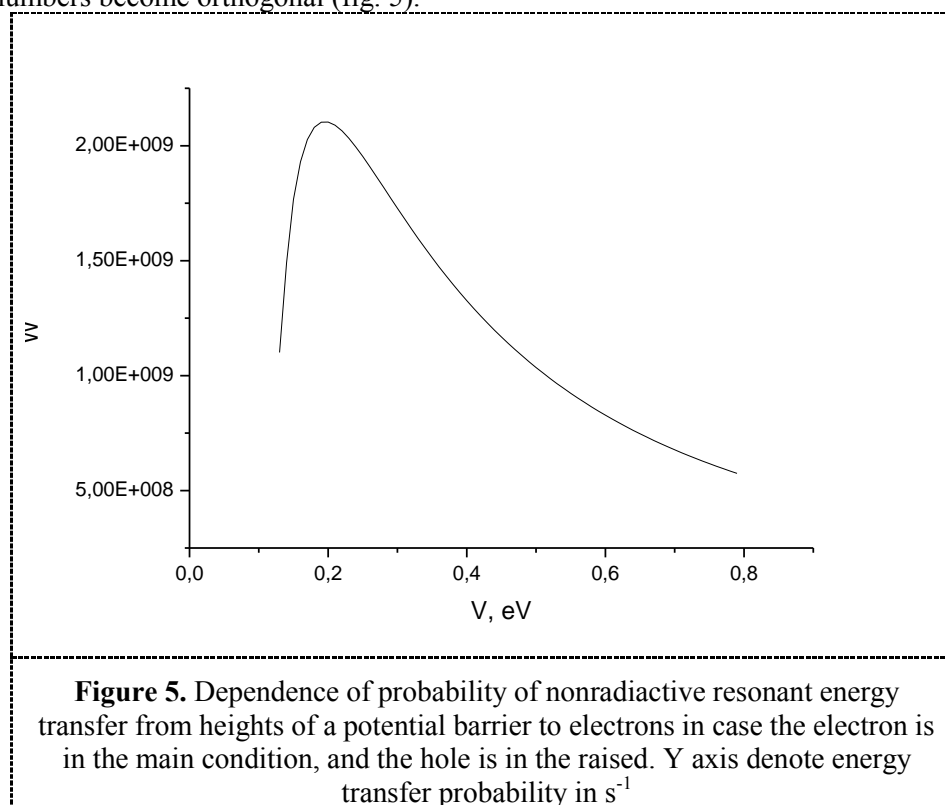
Dependence of probability of a recombination on height of a potential barrier. We consider two different cases.
• the electron and the hole are in the basic condition;
• the electron is in the basic condition, and the hole is in the raised.
In case the electron and the hole are in the main condition, dependence of recombination probability grows with increase in height of a barrier. It occurs at the expense of the following effect: if the barrier is higher, the electrons in QD localized stronger therefore the overlap integral becomes stronger and the probability of the process increases (fig. 4).

Figure 4. Dependence of probability of nonradiactive resonant energy transfer from heights of a potential barrier to electrons in case the electron and the hole are in the main conditions. Y axis denote energy transfer probability in s^{-1}
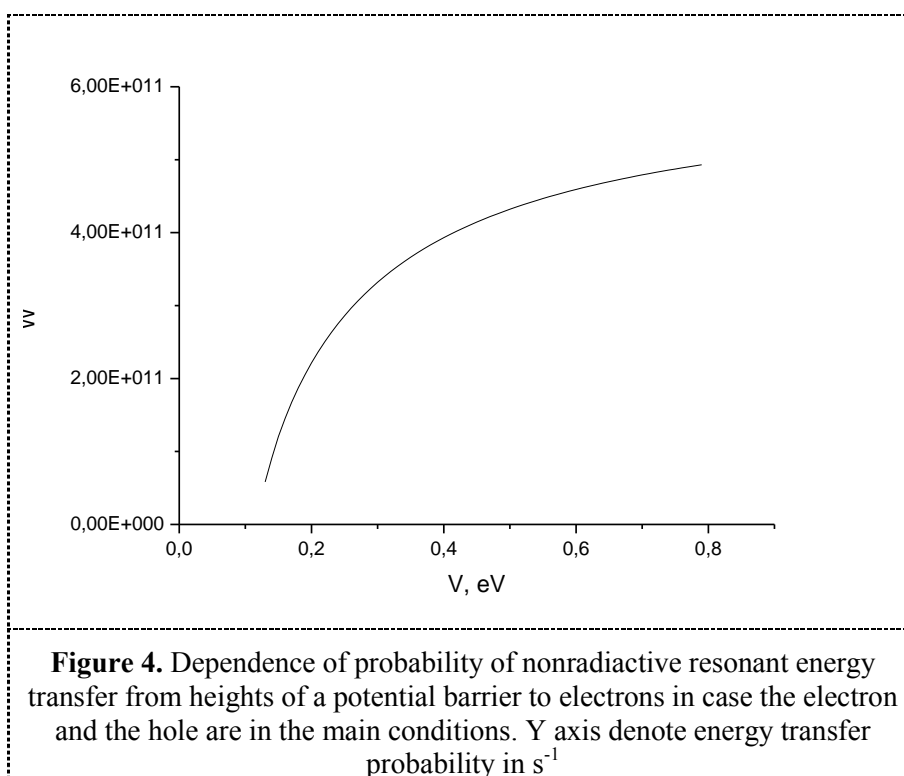
In other case if the electron is in the main condition, the hole is in the excited state at some limited height of the potential barrier, the following result turns out. On the graphics the maximum is observed: after some value of barrier height bending-around wave functions with different main quantum numbers become orthogonal (fig. 5).

Figure 5. Dependence of probability of nonradiactive resonant energy transfer from heights of a potential barrier to electrons in case the electron is in the main condition, and the hole is in the raised. Y axis denote energy transfer probability in s^{-1}

5. Conclusion

During the carried-out work it was shown that nonradiactive resonant energy transfer from one QD to another is possible. Process has many common features with Forster's theory constructed for interaction between protein molecules. The process was considered in the Kane model where the nonparabolic dispersion law of electrons is considered. The probability of energy transfer was received. Due to the nonparabolic matrix element additional sum appears, it increases probability of the transfer process.

The dependence of recombination probability on distance between QD gives . Also in connection with complcated structure of the valent band there is the additional matrix element connected with other polarization of heavy holes. The accounting of a barrier extremity leads to possibility of transitions with participation of electrons and holes with different main quantum numbers.

The phenomenon of nonradiactive resonant energy transfer finds practical application in a modern science. This theory describes experiments with the colloidal QD surrounded with a layer of a semi-conductor material which creates a potential barrier. Such structures can be used as sensors for indication of biological objects as by in vitro, and in vivo.

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