Superlattices and Microstructures 111 (2017) 166–172

Contents lists available at ScienceDirect

Superlattices and Microstructures

journal homepage: www.elsevier.com/locate/superlattices

Resonant electronic excitation energy transfer by exchange mechanism in the quantum dot system

O.P. Chikalova-Luzina [a], D.M. Samosvat [a, *], V.M. Vyatkin [b], G.G. Zegrya [a]

[a] Ioffe Institute, 26 Politekhnicheskaya, St Petersburg 194021, Russian Federation
[b] Saint Petersburg Electrotechnical University "LETI", 5 Prof. Popova Str., St Petersburg 197376, Russian Federation

ARTICLE INFO

Article history:
Received 20 May 2017
Received in revised form 9 June 2017
Accepted 10 June 2017
Available online 12 June 2017

Keywords:
Quantum dots
Dexter interaction
Coulomb potential
Exchange interaction

ABSTRACT

A microscopic theory of nonradiative resonance energy transfer between spherical A_3B_5 semiconductor quantum dots by the exchange mechanism is suggested. The interdot Coulomb interaction is taken into consideration. It is assumed that the quantum dot-donor and the quantum dot-acceptor are made from the same A_3B_5 compound and are embedded in the matrix of another material that produces potential barriers for electrons and holes. The dependences of the energy transfer rate on the quantum-dot system parameters are found in the frame of the Kane model that provides the most adequate description of the real spectra of A_3B_5 semiconductors. The analytical treatment is carried out with using the density matrix method, which enabled us to perform an energy transfer analysis both in the weak-interaction approximation and in the strong-interaction approximation. The numerical calculations showed the saturation of the energy transfer rate at the distances between the donor and the acceptor approaching the contact one. The contributions of the exchange and direct Coulomb intractions can be of the same order at the small distances and can have the same value in the saturation range.

1. Introduction

Methods based on nonradiative energy transfer between quantum dots conjugated with biomolecules are in considerable use in biology and medicine [1,2]. The strong dependence of the energy transfer rate on the distance between the energy donor and energy acceptor makes it possible to study the structure and dynamics of proteins and nucleic acids, where measurements of small distance within the molecule are required, and to detect the formation of biomolecule complexes. Results of the investigations are of great importance for diagnostics and therapy of diseases, including cancer. With particular interest in biosensing applications, the foundational and theoretical works on Forster energy transfer between quantum dot donors and acceptors are discussed in review [3]. However, to enable a correct interpretation of experimental data including the specific cases of short interdot distances, the development of an energy transfer theory taking into account both the direct Coulomb interaction between the donor and acceptor and their exchange interaction is needed.

The early theory of the nonradiative energy transfer between molecules due dipole-dipole interaction was elaborated by Forster [4]. Subsequently, Dexter developed the theory to describe the dipole-quadrupole and exchange mechanisms of the

* Corresponding author.
E-mail addresses: o_chikalova@mail.ru (O.P. Chikalova-Luzina), samosvat@yandex.ru (D.M. Samosvat), vadim.vyatkin@gmail.com (V.M. Vyatkin), zegrya@theory.ioffe.ru (G.G. Zegrya).

http://dx.doi.org/10.1016/j.spmi.2017.06.029
0749-6036/

energy transfer between impurity atoms or ions in an insulating crystal [5]. It was demonstrated that the exchange contribution in the energy transfer can be of importance in two situations: (i), distance between donor and acceptor is short and the donor and acceptor wave functions are not strongly localized, and (ii) dipole-dipole transitions in the acceptor are forbidden.

Theoretical explanation of the energy transfer between quantum dots due to the direct Coulomb interaction including all multipole terms was elaborated in Refs. [6,7].

In the present study a microscopic theory of the nonradiative resonance energy transfer between two spherical A_3B_5 semiconductor quantum dots by the exchange mechanism is developed. To our knowledge, the works on the nonradiative energy transfer between semiconductor quantum dots by the exchange mechanism are not available in the literature. The analytical treatment and numerical calculations of the exchange energy transfer rate for quantum dot system are first performed in our study. We used the density matrix method that enabled us to carry out an energy transfer analysis both in the weak-interaction approximation, where the first order perturbation theory may be applied, and in the strong-interaction approximation. The Kane model is also implemented because it describes most adequately the true spectra of A_3B_5 semiconductors.

It is assumed that the quantum dot donor of radius R_D and the quantum dot acceptor of radius R_A are made from the same A_3B_5 compound and embedded in a matrix from another material producing potential barriers of finite heights for electrons (V_{cD}, V_{cA}) and holes (V_{hD}, V_{hA}). The quantum dots are separated by a finite distance d between their centers. The energy transfer process is sketched in Fig. 1.

In the initial state of the system the electron of the donor is in an excited state (in the conduction band), and the electron of the acceptor is in the ground state (in the valence band). As a result of the Coulomd interaction between the donor and acceptor electrons, the system passes into the final state where the electron of the acceptor is in the conduction band and the electron of the donor is in the valence band.

2. Theory

To find the rate of the nonradiative energy transfer between two quantum dots we need to calculate the energy transfer matrix element, that is, the Coulomb interaction matrix element for transition of the system from the initial state into final state (see Fig. 1). It can be expressed as

$$M_{if} = \sum_{\boldsymbol{\sigma}_1,\boldsymbol{\sigma}_2} \int d^3r_1 d^3r_2 \psi_f^*(\xi_1,\xi_2) \frac{e^2}{\varepsilon|\mathbf{d}+\mathbf{r}_1-\mathbf{r}_2|} \psi_i(\xi_1,\xi_2), \tag{1}$$

where $\xi_i = (\mathbf{r_i}, \sigma_i)$, σ_i are spin variables, $\mathbf{r}_1$ and $\mathbf{r}_2$ are the radius-vectors of electrons in the donor and acceptor, respectively, measured from the center of the corresponding quantum dot, d is the distance between centers of quantum dots, ε is the static permittivity, ψ_i and ψ_f is the initial and final wave functions of the system.

The antisymmetrization of the wave functions describing the initial and final states of the donor and of the acceptor results in the following expression for the matrix element:

$$M_{if} = M_{coul} - M_{ex} \tag{2}$$

where M_{coul} is the matrix element of the direct Coulomb interaction and M_{ex} is the matrix element of the exchange interaction, which are given by

$$M_{coul} = \int d^3r_1 d^3r_2 \psi_{cD}(\mathbf{r}_1)\psi_{hD}^*(\mathbf{r}_1) \frac{e^2}{\varepsilon|\mathbf{d}+\mathbf{r}_1-\mathbf{r}_2|} \tag{3}$$

$$\psi_{cA}^*(\mathbf{r}_2)\psi_{hA}(\mathbf{r}_2) \sum_{\boldsymbol{\sigma}_1,\boldsymbol{\sigma}_2} \chi_{hD}^*(\boldsymbol{\sigma}_1)\chi_{cA}^*(\boldsymbol{\sigma}_2)\chi_{cD}(\boldsymbol{\sigma}_1)\chi_{hA}(\boldsymbol{\sigma}_2), \tag{4}$$

and

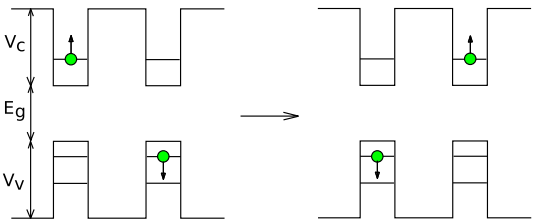

Fig. 1. Schematic drawing of the energy transfer between identical quantum dots by exchange mechanism.