Physica E 66 (2015) 268–274

Contents lists available at ScienceDirect

Physica E

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

Effect of phonon confinement on one- and two-polar optical phonon capture processes in quantum dots

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HIGHLIGHTS

- The capture rate in one- and two-phonon-mediated capture processes was studied.
- The phonon confinement effect on capture rate was taken into account.
- The analytic expressions for carrier capture rates were obtained.
- The carrier capture rate shows a maximum as a function of carrier density.
- Capture rates due to emission one and two optical phonon modes were compared.

ARTICLE INFO

Article history:
Received 2 July 2014
Received in revised form
16 October 2014
Accepted 21 October 2014
Available online 23 October 2014

Keywords:
Quantum dot
Capture
Polar optical phonon
Phonon confinement

ABSTRACT

The electron capture in spherical quantum dot–quantum well structure is studied theoretically. The capture rate in one- and two-polar-optical-phonon-mediated capture processes has been studied by taking into account the phonon confinement. We have derived the explicit analytic expressions for carrier capture rates which can be conveniently applied to practical calculations for the spherical quantum dot systems. The numerical results of the capture rate as function of dot radius, lattice temperature and electron density in GaAs/AlAs QD systems are obtained and discussed. The dependence of the carrier capture rate for a fixed dot radius shows a maximum as a function of carrier density. It is shown that the capture rate of an electron from the barrier region to the quantum dot ground-state, via emission of one and two phonons, exhibits the existence of the bands of dot radii where capture is energetically allowed. We found that the height of the capture rate peaks obtained for one-phonon assisted processes is lowered as QD radius decreases when the phonon confinement is taken into account. The capture rates due to emission single and two optical phonon modes are compared. Carrier capture is shown to proceed with rates as high as $10^{10}\ s^{-1}$ at temperature$T > 100K$. A short capture time is also achieved for a low carrier density.

1. Introduction

An important characteristic of quantum dot (QD) structures is the timescale on which the excited charge carriers relax to their equilibrium state in QD. The understanding of ultrafast carrier dynamics in QDs is important for operation of optoelectronic devices such as QD lasers [1], QD infrared photodetectors [2], and QD optical amplifiers [3]. Time-resolved photoluminescence spectroscopy with picosecond [4–6] and subpicosecond resolution [7] as well as subpicosecond optical pump–probe experiments [8,9] have been applied to obtain an information on carrier dynamics in various QD systems. In a typical experiment, carriers in the surrounding QD barrier layers are initially excited. These carriers can then become captured by the QD, and then recombine, often radiatively. The processes involved carrier capture into the dots and intradot relaxation, have been under extensive theoretical study during the past decade. Two different capture and relaxation processes have been considered; via carrier–carrier interaction (Auger processes) or via carrier–phonon coupling [10–12]. The discrete density of states may also impose impediments to carrier energy relaxation by optical phonon emission. This so called "phonon bottleneck" [13] could restrict the modulation rate of QD lasers, thus limiting their use for high-speed applications. On the

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http://dx.doi.org/10.1016/j.physe.2014.10.028

other hand, reported relaxation rates vary significantly [8,14–19]. Thus the general question of whether or not carrier capture at low carrier densities in a QD structure suffers from "phonon bottleneck" effects is still hard to answer. Sanguinetti et al. have been presented picosecond time resolved photoluminescence measurements of GaAs/AlGaAs QD structures grown by modified droplet epitaxy, where no wetting layer is connecting the dots and show a fast carrier relaxation time (30 ps) to the dot ground state [20]. Most of the nanoscale microcrystals have a spherical shape. In many cases, these spherical QDs are composed of a spherical core of one (core-well) material embedded in a matrix of another (shell-barrier) material.

The inhomogeneous nature of nanostructures leads to strong modifications of the electronic properties as well as the phonon spectrum. The existence of boundaries between the constituting materials and/or vacuum introduces a coupling of the longitudinal and transverse optical phonon modes even for isotropic media. Additionally, new types of confined (LO1, LO2) interface (IO1, IO2) and surface (SO1, SO2) optical modes can occur in QD quantum wells [21].

There have been theoretical studies of electron capture [22–24] in QDs due to various phonon processes: single longitudinal-optical (LO) [22,23], LO plus acoustic (AC) phonons [25], and two LO phonons [23].

The purpose of the present study is to investigate the effect of phonon confinement on two phonon capture processes in a spherical quantum dot–quantum well structure, in the framework of the effective-mass approximation and the rectangular potential barrier model, and derive an expression for the capture rate of electrons for open spherical quantum dots with an arbitrary thickness of barrier layers (from zero to infinity). To the best of our knowledge, there are no theoretical works where the effect of phonon confinement on the two-phonon assisted capture processes in QD structures was examined.

The paper is organized as follows. In Sec. 2 the theoretical framework used in the calculation is given. Subsection 2.1 briefly recalls the main aspects of the bound and unbound states of QDs hosting one electron. In the next subsection (2.2), we present the polar optical phonon modes in a spherical core-shell QD system. The one- and two-phonon capture rate will be calculated in the subsection 2.3. In Section 3, we present and discuss the numerical results for one- and two-phonon-assisted capture rates in GaAs quantum dot. Section 4 summarizes the present studies.

2. Theory

2.1. The single-particle states in a spherical quantum Dot

Let us briefly describe the model and fundamental theory applied in our calculations. We consider a spherical quantum dot-quantum well structure consisting of quantum dot of radius r_c embedded in a dielectric medium. We shall use the one-band approximation for the conduction band electrons and ignore the effects of band mixing as well as the mass anisotropy caused by strain in the barrier material and the QD. To come closer to the real situation, the electronic confinement is modeled by a finite potential well. Within the effective mass approximation the Schrödinger equation is solved exactly. As a result the complete set of orthonormalized wave functions is

$$\psi_{l,m,n}^{bound}(r,\vartheta,\varphi) = AY_{l,m}(\vartheta,\varphi)\left(j_l(k_0r)\theta(r_c-r) + \frac{j_l(k_0r_c)}{h_l^{(+)}(i\kappa r_c)}h_l^{(+)}(i\kappa r)\theta(r-r_c)\right), \quad (1)$$

$$\psi_{l,m,\mathbf{k}}^{unbound}(r,\vartheta,\varphi) = BY_{l,m}(\vartheta,\varphi)\left(j_l(k_1r)\theta(r_c-r) + \frac{j_l(k_1r_c)}{f_l(kr_c)}f_l(kr)\theta(r-r_c)\right), \quad (2)$$

where $\psi_{l,m,n}^{bound}$ are the wave functions of the stationary states of the discrete spectrum, the energy levels ($E_{lm}<0$) of which are defined by the dispersion equation [26]

$$\frac{i\kappa m_1^*}{k_0m_2^*}\left(\frac{l}{i\kappa r_c} - \frac{h_l'^{(+)}(i\kappa r_c)}{h_l^{(+)}(i\kappa r_c)}\right) = \frac{l}{k_0r_c} - \frac{j_l'(k_0r_c)}{j_l(k_0r_c)}. \quad (3)$$

Here, m_1^* and m_2^* denote the effective masses in the dot and the barrier, respectively,

$$k_0^2 = \frac{2m_1^*}{\hbar^2}(V_0+E_{lm}), \quad \kappa^2 = -\frac{2m_2^*}{\hbar^2}E_{lm}, \quad h_l^{(+)}(x) = n_l(x) + ij_l(x), \quad (4)$$

V_0 is the depth of the confining potential well, $j_l(x)$ and $n_l(x)$ are the Bessel and Neumann spherical functions, respectively, $\theta(x)$ is the unit step function, $Y_{l,m}(\theta,\varphi)$are the spherical harmonic functions with $m=0,\pm1,\ldots,\pm l$. $\psi_{l,m,\mathbf{k}}^{unbound}$are the wave functions of the stationary states of the continuous spectrum with the energy $E_{\mathbf{k}}>0$, and

$$k_1^2 = \frac{2m_1^*}{\hbar^2}(V_0+E_{\mathbf{k}}), \quad k^2 = \frac{2m_2^*}{\hbar^2}E_{\mathbf{k}}, \quad f_l(kr) = \cos(\delta_l)j_l(kr) + \sin(\delta_l)n_l(kr), \quad (5)$$

$$\tan\delta_l = \frac{kj_l'(kr_c)j_l(k_1r_c) - k_1j_l(kr_c)j'_l(k_1r_c)}{k_1j_l'(k_1r_c)n_l(kr_c) - kj_l(k_1r_c)n'_l(k_1r_c)}. \quad (6)$$

The normalization constant of Eq. (1) can be determined by the condition

$$\int_0^\infty drr^2\int_0^\pi d\theta\sin\theta\int_0^{2\pi}d\varphi\left|\psi_{l,m}^{bound}(r,\vartheta,\varphi)\right|^2 = 1, \quad (7)$$

and Bin Eq. (2) is given by

$$B = 2k\frac{f_l(kr_c)}{j_l(k_1r_c)}, \quad (8)$$

and is defined from the normalizing condition

$$\int_0^\infty F_{kl}^*(r)F_{k',l}(r)r^2dr = \delta(k-k'), \quad (8a)$$

$F_{kl}(r)$ are the radial wave functions of the stationary states of the continuous spectrum.