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Optical absorption of one-particle quantum-confined states of charge carriers in quasi-zero-dimensional nanogeterostructures: Theory

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Abstract: A theory is developed for the interaction of an electromagnetic field with one-particle quantum-confined states of charge carriers in semiconductor quantum dots. It is shown that the oscillator strengths and dipole moments for the transitions involving one-particle states in quantum dots are gigantic parameters, exceeding the corresponding typical parameters of bulk semiconductor materials. In the context of dipole approximation it is established that the gigantic optical absorption cross sections in the quasi-zero-dimensional systems make it possible to use the systems as efficient absorbing materials.

Keywords: One-Particle Quantum-Confined States of Charge Carriers, Dipole Approximation, Oscillator Strength, Absorption and Scattering of Light, Quantum Dots

1. Introduction

At present, the optical properties of quasi-zero-dimensional structures are extensively studied [1, 2, 3, 4, 5]. Such structures commonly consist of spherical semiconductor nanocrystals (the so-called quantum dots (QDs)) with a radius $a \approx 1 - 10$ nm grown in semiconductor (or dielectric) matrices. The studies in this field are motivated by the fact that such heterophase systems represent promising materials for the development of the components of nonlinear nanooptoelectronics to be used, specifically, for controlling optical signals in optical computers [3, 5] or for manufacturing active layers of injection semiconductor nanolasers [2, 3, 4, 5].

In the work [5] the optical properties of an array of InAs and InSb QDs in the GaAs and GaSb matrices and the corresponding operational characteristics of injection lasers, with the active region on the basis of this array, were studied experimentally. In these studies, a large short-wavelength shift of the laser emission line was observed for the array of QDs. In such an array, the energy spectrum of charge carriers is completely discrete [6, 7], if the QDs are smaller than $a \approx 1 - 7$ nm in size. In the first-order approximation, the spectrum of such quantum-confined states can be described as a spectrum of a charge carrier in the spherically symmetric well with infinitely high walls [7].

The present paper uses the effective mass method in the dipole approximation to study absorption and scattering of light by one-particle quantum-confined states of electron (hole) in QDs. The electron (hole) transitions between intraband energy levels in QDs are considered as well.

2. Charge Carriers Energy Spectrum in Quasi-Zero-Dimensional Nanosystems

We consider a simple model in which a quasi-zero-dimensional system is defined as a neutral spherical semiconductor QD of radius a and permittivity ε_2, embedded in a surrounding medium with permittivity ε_1. Let an electron (e) and a hole (h), whose effective masses are, correspondingly, m_e and m_h, be in motion in this QD. Let the spacing between the electron or hole and the QD center be r_e or r_h. We assume that the bands for electrons and holes are parabolic. Along with the QD radius a, the characteristic lengths of the problem are a_e, a_h, and a_{ex}, where

$$a_e = \varepsilon_2 \hbar^2 / m_e e^2, a_h = \varepsilon_2 \hbar^2 / m_h e^2, \quad (1)$$

$$a_{ex} = \varepsilon_2 \hbar^2 / \mu e^2,$$

are the Bohr radii of the electron, hole and exciton in the semiconductor with the permittivity ε_2, respectively, and $\mu = (m_e m_h)/(m_e + m_h)$ is the exciton effective mass. All of the characteristic lengths of the problem are much larger than the interatomic spacing a_0 [8]:

$$a, a_e, a_h, a_{ex} \geq a_0 \quad (2)$$

This allows us to treat the motion of the electron and hole in the QD in the effective-mass approximation.

In the context of the above-described model and approximations for a quasi-zero-dimensional system, the Hamiltonian of the electron-hole pair [9]:

$$H = -\frac{\hbar^2}{2m_e}\Delta_e - \frac{\hbar^2}{2m_h}\Delta_h + V_{eh}(\boldsymbol{r}_e, \boldsymbol{r}_h) + U(\boldsymbol{r}_e, \boldsymbol{r}_h, a) + E_g, \quad (3)$$

where the first two terms in the sum define the kinetic energy of the electron and hole; E_g is the energy band gap in the bulk (unbounded) semiconductor with the permittivity ε_2; $V_{eh}(r_e, r_h)$ is the energy of the electron-hole Coulomb interaction

$$V_{eh}(r_e, r_h) = \frac{e^2}{2\varepsilon_2 a}\frac{2a}{(r_e^2 - 2r_e r_h \cos\theta + r_h^2)^{1/2}}, \quad (4)$$

with the angle Θ between the vectors r_e and r_h, and $U(r_e, r_h, a)$ is the energy of interaction of the electron and hole with the polarization field induced by the electron and hole at the spherical interface between the two media. For arbitrary values of ε_1 and ε_2 the interaction energy $U(r_e, r_h, a)$ can be represented analytically as [4,7]:

$$U(\boldsymbol{r}_e, \boldsymbol{r}_h, a) = -\frac{e^2\beta}{\varepsilon_2 a}\frac{1}{[(r_e r_h/a^2)^2 - 2(r_e r_h/a^2)\cos\theta + 1]^{1/2}} - \frac{e^2\beta}{2(\varepsilon_1+\varepsilon_2)a}\int_0^\infty \frac{dy\,(a^2/r_h y)^\alpha \Theta(y - a^2/r_h)}{|r_e - y(r_e/r_h)|} - \frac{e^2\beta}{2(\varepsilon_1+\varepsilon_2)a}\int_0^\infty \frac{dy\,(a^2/r_e y)^\alpha \Theta(y - a^2/r_e)}{|r_h - y(r_h/r_e)|}, \quad (5)$$

where $\Theta(x)$ is the unit-step function and $\beta = (\varepsilon_1 - \varepsilon_2)/(\varepsilon_1 + \varepsilon_2)$, $\alpha = \varepsilon_1/(\varepsilon_1 + \varepsilon_2)$ are parameters. In the bulk of a QD, the electron (hole) energy levels can originate. Their energies are defined as [6, 7]:

$$E_{n,l}(a) = \frac{\hbar^2}{2m_{e(h)}a^2}\varphi_{n,l}^2, \quad (6)$$

where the subscripts (n, l) refer to the corresponding quantum size-confined states. Here, n and l are the principal and azimuthal quantum numbers for the electron (hole) and $\varphi_{n,l}$ are the roots of the Bessel function, i.e., $J_{l+1/2}(\varphi_{n,l})$. For the quantum-confined levels to originate, it is necessary that, in the Hamiltonian (3), the electron (hole) energy $E_{n,l}(a)$ (6) be much larger than the energy of the interaction of the electron (hole) with the polarization field $U(a)$ (5) generated at the spherical QD-dielectric (semiconductor) matrix interface [4,7] and the energy of the Coulomb electron-hole interaction (4):

$$E_{n,l}(a) = \frac{\hbar^2}{2m_{e(h)}a^2}\varphi_{n,l}^2 \gg U(a) \approx \frac{e^2\beta}{2\varepsilon_2 a}. \quad (7)$$

Condition (7) is satisfied for QDs of radii

$$a \ll a_s^{e(h)} = \frac{\varphi_{n,l}^2}{\beta} a_{e(h)} \quad (8)$$

At room temperature T_0, the discrete levels of the electron (hole) $E_{n,l}(a)$ (6) in the QD are slightly broadened if the energy separation between the levels is

$$\Delta E_{n,l}(a) = E_{n`,l} - E_{n,l}(a) \ll kT_0 \quad (9)$$

Taking into account (6), we can rewrite inequality (9) as

$$\frac{\hbar^2}{2m_{e(h)}a^2}\frac{\varphi_{n`,l}^2 - \varphi_{n,l}^2}{kT_0} = \eta(a) \ll 1 \quad (10)$$

Formula (6) describing the spectrum of charge carriers in a QD is applicable to the lowest states (n, l) that satisfy the inequality

$$\Delta E_{n,l}(a) \ll \Delta V_0(a), \quad (11)$$

where $E_0(a)$ is the depth of the potential well for electrons in the QD. For example, for the CdS QDs whose sizes satisfy inequality (8), the value of V_0 is 2.3 − 2.5 eV [10].

If condition (8) is satisfied, we can use, for the electron (hole) wave function in a QD, the wave function of an electron (hole) in a spherical quantum well with infinitely high walls [9]:

$$\Psi_{n,l,m}(r, \theta, \varphi) = Y_{l,m}(\theta, \varphi)\frac{J_{1+1/2}(\varphi_{n,l})\sqrt{2}}{J_{1+3/2}(\varphi_{n,l})a\sqrt{r}}, \quad (12)$$

where $r = r_e$ or $r = r_h$ is the distance of the electron or hole from the QD center, Θ and φ are the azimuthal and polar angles that define the orientation of the radius vector of the electron (hole), $Y_{l,m}$ are the normalized spherical functions (m is the magnetic quantum number of the electron or hole), and $J_v(x)$ are the Bessel functions.

3. Results and Discussion

3.1. Dipole Approximation

In the frequency region corresponding to the above-considered states of charge carriers in QD bulk, (n, l) (6), the wavelength of light is much larger than the dimensions of these states ($\approx a_e, a_h$). Therefore, the behavior of these states is adequately described in the dipole approx-imation. In this case, the operator of the dipole moment of the electron (hole) located in the QD bulk is expressed as [11]:

$$D(r) = \frac{3\varepsilon_1}{2\varepsilon_1+\varepsilon_2} D^0(r), \qquad D^0(r) = er \tag{13}$$

To estimate the value of the dipole moment, it is suffcient to consider the transition between the lowest discrete states (6), e.g., between the ground states 1*s* ($n = 1$, $l = 0$, $m = 0$) and 1*p* ($n = 1$, $l = 1$, $m = 0$). To calculate the matrix element of the dipole moment of the charge-carrier transition from the 1*s* state to the 1*p* state, $D_{1,0}(a)$, we assume that the uniform field of the light wave $E(\omega, t)$ is directed only along the axis Z (ω is the wave frequency). In this case, we take the dipole moment $D(r)$ (13) induced by the field $E(\omega, t)$ as the perturbation responsible for such dipole transition. The expression for the dipole moment of the transition $D_{1,0}(a)$ follows from formulas (13) and the expression for the dipole moment of the transition in free space

$$D^0_{1,0}(a) = \langle 1s|D^0(r)|1p\rangle = e\langle 1s|r|1p\rangle. \tag{14}$$

On substitution of (12) into formula (14) and integration, we obtain the expression for the dipole moment of the transition in free space as follows:

$$D_{1,0}^{0}(a) = 0.433\, ea. \tag{15}$$

Then, according to (15) and (13), the dipole moment of the transition in the QD with the permittivity ε_2 in the surrounding matrix with the permittivity ε_1 is

$$D_{1,0}(a) = \Lambda \times 0.433\, e\, a, \quad \Lambda = 3\varepsilon_1/(2\varepsilon_1 + \varepsilon_2). \tag{16}$$

3.2. Optical Absorption of One-Particle Quantum-Confined States

Using the above results for the matrix element of the dipole moment of the transition $D_{1,0}$ (16), we can elucidate the behavior of the semiconductor quasi-zero-dimensional systems on absorbing the energy of electromagnetic field in the frequency region corresponding to the energies of the quantum-confined states in the QD $E_{n,l}(a)$ (6). The absorption cross section of a spherical QD of radius a can be expressed in terms of the polarizability of QD, $A^{//}(\omega, a)$, as [10, 11]

$$\sigma_{abs}(\omega, a) = 4\pi(\omega/c)A^{//}(\omega, a), \tag{17}$$

where ω is the frequency of the external electromagnetic field.

The polarizability $A^{//}(\omega, a)$ can be easily determined if the QD is considered as a single giant ion. Let the radius of QD be a (8). In such QD, the quantum-confined states of charge carriers (n, l) (6) are formed. At room temperature, these states are slightly broadened, satisfying inequality (10). In this case, the polarizability of the charged QD, $A^{//}(\omega, a)$, can be expressed in terms of the matrix element of the dipole moment of the transition $D_{1,0}(a)$ (16) between the lowest 1*s* and 1*p* states as [8]:

$$A^{``}(\omega, a) = \frac{e^2}{m_{e(h)}} \frac{f_{0,1}}{\omega_1^2(a) - \omega^2 - i\omega\Gamma_1(a)}, \tag{18}$$

where

$$f_{0,1} = \frac{2m_{e(h)}}{\hbar e^2}[\omega_1(a) - \omega_0(a)]\left|D_{1,0}(a)\right|^2 \tag{19}$$

is the oscillator strength of the transition of a charge carrier with the effective mass m_e (or m_h) from the ground 1*s* state to the 1*p* state; $\hbar\omega_1(a) = E_{1,1}(a)$ and $\hbar\omega_0(a) = E_{1,0}(a)$ are, correspondingly, the energies of the discrete 1*s* and 1*p* levels by formula (6); $\Gamma_1(a)$ is the width of the 1*p* level [6,7]. Taking into account formulas (6) and (16), we can express the oscillator strength (19) of the transition as

$$f_{0,1} - \left(\varphi_{1,1}^2 - \pi^2\right)\frac{\Lambda^2\left(D^0_{1,0}(a)\right)^2}{e^2a^2} - 0{,}967 \cdot 10^{-1}\Lambda^2 \tag{20}$$

We assume that the frequency ω of the wave of light is far from the resonance frequency $\omega 1$,, of the discrete 1 *p* state and, in addition,that the broadening $\Gamma_1(a)$, of the 1*p* level is small, i.e. $(\Gamma_1/\omega_1 \leq 1)$. Then, for the qualitative estimate of the QD polarizability $A^{//}(\omega, a)$ (18), we obtain, with regard to (6), the following expression:

$$A^{``}(a) = \frac{4f_{0,1}}{\varphi_{1,1}^4}\frac{m_{e(h)}}{m_0}\left(\frac{a}{a_B}\right)^4 a_B^3, \tag{21}$$

where ($a_B = \hbar^2/m_0 e^2$) is the Bohr radius of an electron in free space.

Now we write the expression for the cross section of elastic scattering of the electromagnetic wave with frequency ω by the QD of radius a [11] as:

$$\sigma_{sc}(\omega, a) = 2^7 3^{-3}\pi^3(\omega/c)^4[A^{//}(a)]^2 \tag{22}$$

4. Conclusions

In conclusion, we briefly discuss possible physical situations in which the results obtained above can be used for interpreting the experimental data. Similar to work [10] we can assume that under the experimental conditions in the work [5] the annealing of the arrays of InAs and InSb QDs in the GaAs and GaSb matrices at the temperature T = 293K induces the thermal emission of a light electron, so that a hole alone remains in the QD bulk. In this case, the electron may be localized at a deep trap in the matrix. If the distance d from this trap to the QD center is large compared to the QD radius a ($d \geq a$), the Coulomb electron-hole interaction $V_{eh}(r_e, r_h)$ (4) in the Hamiltonian (3) can be disregarded. As a result, the one-particle quantum-confined hole states (n, l), with the energy spectrum $E_{n,l}(a)$ described by formula (6), appear in the QD bulk.

Now we roughly estimate the cross sections of optical absorption σ_{abs} (17) and (21) and scattering σ_{sc} (22) at the quantum-confined hole state in the QDs for the selected (1s → 1 p) transition under the experimental conditions of [5]. For the rough estimation of the cross sections of optical absorption and scattering, we use expressions (17), (21) and (22) on the assumptions that the frequency of the light wave ω is far from the resonance frequency ω_1 of the discrete hole state in the QD and that the broadening $\Gamma 1(a)$ of the energy level $E_{11}(a) = \hbar\omega_1(a)$ (6) is small [6, 7] ($\Gamma_1/\omega_1 \leq 1$). In this case, the absorption cross section σ_{abs} and the scattering cross section σ_{sc} take the following forms:

$$\sigma_{abs}(\omega, a) = \frac{16\pi f_{0,1}}{\varphi_{1,1}^4}\frac{\omega}{c}\frac{m_h}{m_0}\left(\frac{a}{a_B}\right)^4 a_B^3, \tag{23}$$

$$\sigma_{sc}(\omega, a) = \frac{1^{11}\pi^3 {f_{0,1}}^2}{3^3\varphi_{1,1}^8}\left(\frac{\omega}{c}\right)^4\left(\frac{m_h}{m_0}\right)^2\left(\frac{a}{a_B}\right)^8 a_B^6. \tag{24}$$

The estimated parameters of the hole states in QDs dispersed in the III-V semiconductors are listed in the table1. It is worth noting that, at room temperature, the quantum-confined hole states are slightly broadened; the parameter $\eta(a)$ (10) does not exceed 18%.

The quasi-zero-dimensional systems considered here are new essential nonlinear media with respect to infrared radiation [2, 4, 12]. In fact, the dipole moments of the transitions in QDs of radii $a \approx 2.0 - 5.0$ nm are gigantic in magnitude: $D_{1,0} \approx 10D_0$ (see table, where D is the unit of measure of the dipole moment in Debye (D_0)), being many times larger than the values $D \approx 0.1D_0$ typical for the bulk III-V semiconductors [5, 12]. In addition, according to the selection rules for QDs in the electromagnetic field, the dipole transitions between the nearest levels $E_{n,l}(a)$ (6) are allowed. Under these transitions, the azimuthal quantum number changes by unity [4, 12].

From the estimates presented in the table it follows that, for QDs of radii $a \approx 2.0 - 5.0$ nm, the absorption cross section can be as gigantic as $\sigma_{abs} \approx 10^{-16}$ sm^2. This value is eight orders of magnitude larger than the typical absorption cross sections for atoms [12]. Since the scattering cross sections σ_{sc} (24) under the experimental conditions in the work [5] are negligible compared to the corresponding absorption cross sections σ_{abs} (23) ($\sigma_{sc}/\sigma_{abs} \approx 10^{-12}$) the estimates for σ_{sc} are not included in the table [12].

The gigantic optical absorption cross sections in the quasi-zero-dimensional systems treated above allow for the use of such nanostructures as new efficient absorbers of electromagnetic waves in a wide wavelength range variable over wide limits in accordance with the nature of the materials in contact.

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