

## TRANSIENT PUMP–PROBE ABSORPTION SPECTROSCOPY OF SEMICONDUCTOR NANOTETRAPODS

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### Abstract

We develop a theoretical description of transient pump-probe absorption spectroscopy of semiconductor nanotetrapods. The absorption of the probe pulse is assumed to be induced by the pump pulse resonant to some interband transition of the nanotetrapod. We show that the absorption of the probe is governed by a sum of exponential functions with exponents proportional to the energy relaxation rates of electronic states in the nanotetrapod and the delay time between pump and probe pulses. The derived analytical expression for the absorption efficiency can be used for studying the dynamics of quantum transitions in semiconductor nanotetrapods.

**Keywords:** Transient pump–probe spectroscopy, semiconductor nanotetrapods, density matrix formalism.

### 1. INTRODUCTION

Semiconductor nanostructures of complex geometries are a topical subject of nanophotonics due to the possibility of flexible manipulation of their optical response. Nanotetrapods [1, 2] are of particular interest not only due to this feature—associated with the possibility to alter the lengths and diameters of the nanorods, the sizes and shape of the dot, as well as the composition of both kinds of elements—but also owing to their strong optical anisotropy. The linear optical response and the associated electronic dynamics of nanotetrapods can be studied using a variety of optical methods, including the luminescence and absorption spectroscopies [3-6], Raman spectroscopy [7], coherent control of secondary emission [8], as well as stationary and time-resolved pump–probe spectroscopies [9-12]. This study could facilitate the development of accurate theoretical models of nanotetrapods, in particular, by exploring the dominant relaxation mechanisms of the nanotetrapod's electronic subsystem. This is quite a challenging task due to a wide variety of relaxation channels present in complex semiconductor nanostructures. These may include the photon-assisted relaxation [13-17], the relaxation on plasmons or plasmon–phonons [18-20], relaxation on defects [21, 22], Auger processes [23], and various kinds of nonradiative energy transfer [24, 25].

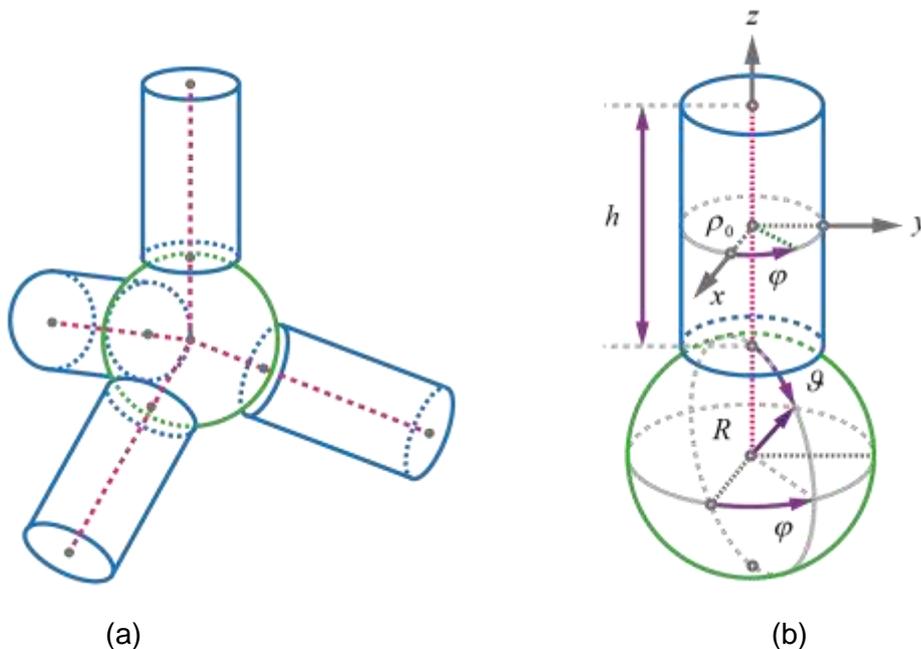
In this work we develop a unified theory of transient pump–probe absorption spectroscopy of a semiconductor nanotetrapod. The physical processes underlying this spectroscopic method are treated using the density matrix formalism. We assume that absorption of the probe pulse is induced by the pump pulse resonant to some interband transition of the nanotetrapod. We report on the physical conditions under which the dependence of the absorbed energy of the probe on the delay time between the pump and probe pulses is described by a sum of exponential functions with exponents proportional to the energy relaxation rates of the nanotetrapod's electronic states. We also show that the transient absorption spectroscopy allows one to reliably determine the energy relaxation rates of the nanotetrapod's electronic states. The derived analytical expressions can be used for studying the dynamics of quantum transitions in semiconductor nanotetrapods.

## 2. NANOTETRAPOD MODEL

Consider a semiconductor nanotetrapod assembled of one spherical quantum dot attached to the bases of four circular cylinder nanorods, as shown in Fig. 1. Suppose that the nanotetrapod is embedded in a dielectric matrix and its electronic spectrum can be modelled using the approximation of infinitely high potential barriers. For the sake of definiteness, we focus on nanotetrapods with the nanorods touching the quantum dot at one point, as shown in Fig. 1(a). Much like in quantum-dot molecules [24, 25], the coupling between the electronic subsystems of the quantum dot and nanorods is relatively weak as it is caused by the nonradiative transfer of electronic excitation energy through the barriers. This allows us to calculate the energy spectra and wave functions of the nanotetrapod elements separately.

Let us assume that the quantum dot and nanorods are in the regime of strong quantum confinement, which implies that their characteristic sizes are less than the exciton Bohr radii of their corresponding bulk materials, and neglect of the Coulomb interaction between the confined electrons and holes. Then the envelope wave functions and the confinement energies of electrons and holes in the spherical quantum dot

of radius  $R$  are given by



**Fig. 1** Schematic of a semiconductor nanotetrapod composed of a spherical quantum dot touching the bases of four cylindrical nanorods;  $(R_n, \vartheta_n, \varphi_n)$  and  $(\rho_0, \varphi, h)$  are the spherical and cylindrical coordinates used for calculation of the wave functions and energy spectra of the nanotetrapod's elements.

$$\psi_{nlm}(r, \vartheta, \varphi) = \sqrt{\frac{2}{R^3}} \frac{j_l(\frac{\xi_{nl}r}{R})}{j_{l+1}(\xi_{nl})} Y_{lm}(\vartheta, \varphi), \quad E_{nl} = \frac{\hbar^2 \xi_{nl}^2}{2m_{c(v)} R^2} \quad (1)$$

where  $j_l(x)$  is spherical Bessel function;  $Y_{lm}$  is the spherical harmonic;  $j_l(\xi_{nl}) = 0$ ;  $n = 1, 2, 3, \dots$ ,  $l = 0, 1, 2, 3, \dots$ , and  $m = -l, -l+1, \dots, 0, 1, \dots, l$  are the principal quantum number, angular momentum, and momentum projection, respectively; and  $m_{c(v)}$  is the effective mass of the electron (hole). It is seen that the three-dimensional spatial confinement of electrons and holes results in splitting of their continuous energy spectra into a countable set of discrete quantum states, with energies inversely proportional to the square of the quantum dot's radius.

We also assume that the nanotetrapod's semiconductors have  $T_d$  or  $O_h$  symmetry and direct bands of bandgap  $E_g$  and restrict ourselves to the two-band model of energy spectrum. Then the energy spectrum and full wave functions of electrons and holes inside the quantum dot are given by

$$\mathcal{E}_{nl}^{(c)} = E_g + E_{nl}, \quad \mathcal{E}_{n'l'}^{(v)} = -E_{n'l'}, \quad (2)$$

$$\Psi_{nlm}^{(c)}(\mathbf{r}) = u_c(\mathbf{r})\psi_{nlm}(\mathbf{r}), \quad \Psi_{n'l'm'}^{(v)}(\mathbf{r}) = u_v(\mathbf{r})\psi_{n'l'm'}(\mathbf{r}), \quad (3)$$

where the quantum numbers with and without primes correspond to holes and electrons, respectively,  $u_c(\mathbf{r})$  and  $u_v(\mathbf{r})$  are the Bloch amplitudes, and the zero-point energy coincides with the top of the valence band. The electron–hole-pair energies

$$\mathcal{E}_{nl,n'l'} = E_g + E_{nl} + E_{n'l'} \quad (4)$$

are thus characterized by a set of four quantum numbers  $\{n, l, n', l'\}$  each.

The envelope wave functions and confinement energies of electrons and holes in a circular-cylinder nanorod of height  $h$  and base radius  $\rho_0$  are:

$$\psi_{nl n_z}(\rho, \varphi, z) = \sqrt{\frac{2}{\pi h \rho_0^2}} \frac{\exp(i l \varphi) j_l(\xi_{nl} \rho / \rho_0)}{j_{l+1}(\xi_{nl})} \begin{cases} \sin(k_{n_z} z), & n_z - \text{even}, \\ \cos(k_{n_z} z), & n_z - \text{odd} \end{cases} \quad (5)$$

$$E_{nl n_z} = \frac{\hbar^2}{2m_{c(v)}} \left( \frac{\xi_{nl}^2}{\rho_0^2} + \frac{\pi^2 n_z^2}{h^2} \right), \quad (6)$$

where  $J_l(x)$  is the cylindrical Bessel function,  $J_l(\zeta_{nl}) = 0$ ,  $n = 1, 2, 3, \dots$ ,  $l = 0, \pm 1, \pm 2, \pm 3, \dots$ , and  $n_z = 1, 2, 3, \dots$  are the quantum numbers. The energy spectrum and full wave functions of electrons and holes confined to the nanorod are therefore given by

$$\varepsilon_{nln_z}^{(c)} = E_g + E_{nln_z}, \quad \varepsilon_{n'l'n'_z}^{(v)} = -E_{n'l'n'_z}, \quad (7)$$

$$\Psi_{nln_z}^{(c)}(\mathbf{r}) = u_c(\mathbf{r})\psi_{nln_z}(\mathbf{r}), \quad \Psi_{n'l'n'_z}^{(v)}(\mathbf{r}) = u_v(\mathbf{r})\psi_{n'l'n'_z}(\mathbf{r}), \quad (8)$$

whereas the spectrum of the electron–hole pairs is of the form

$$\varepsilon_{nln_z, n'l'n'_z} = E_g + E_{nln_z} + E_{n'l'n'_z}. \quad (9)$$

To calculate the efficiency of transient light absorption by the nanotetrapod, we need to know the matrix element of the electron–photon interaction. The generation rate of electron–hole pairs in the quantum dots and nanorod is independent of geometric parameters and determined by the matrix element

$$|V_{\eta,0}^{(q)}| = \delta_{v,v'} e Z_{cv} E_q, \quad (10)$$

where  $\delta_{v,v'}$  is the product of three Kronecker deltas ( $\delta_{n,n'}\delta_{l,l'}\delta_{m,m'}$  for the quantum dot and  $\delta_{n,n'}\delta_{l,l'}\delta_{n_z,n'_z}$  for the nanorods),  $Z_{cv} = \sqrt{2}P/E_g$ ,  $P$  is the Kane's parameter,  $E_q$  is the electric field strength of the pump ( $q = pu$ ) or probe ( $q = pr$ ) pulse. Since the electron–hole pairs are seen to be generated with equal quantum numbers of electrons and holes, their energies in the quantum dot and nanorods are of the form

$$\varepsilon_{nl,nl} = E_g + \frac{\hbar^2 \zeta_{nl}^2}{2\mu R^2} \quad \text{and} \quad \varepsilon_{nln_z, nln_z} = E_g + \frac{\hbar^2}{2\mu} \left( \frac{\zeta_{nl}^2}{\rho_0^2} + \frac{\pi^2 n_z^2}{\hbar^2} \right), \quad (11)$$

where  $\mu = m_c m_v / (m_c + m_v)$ .

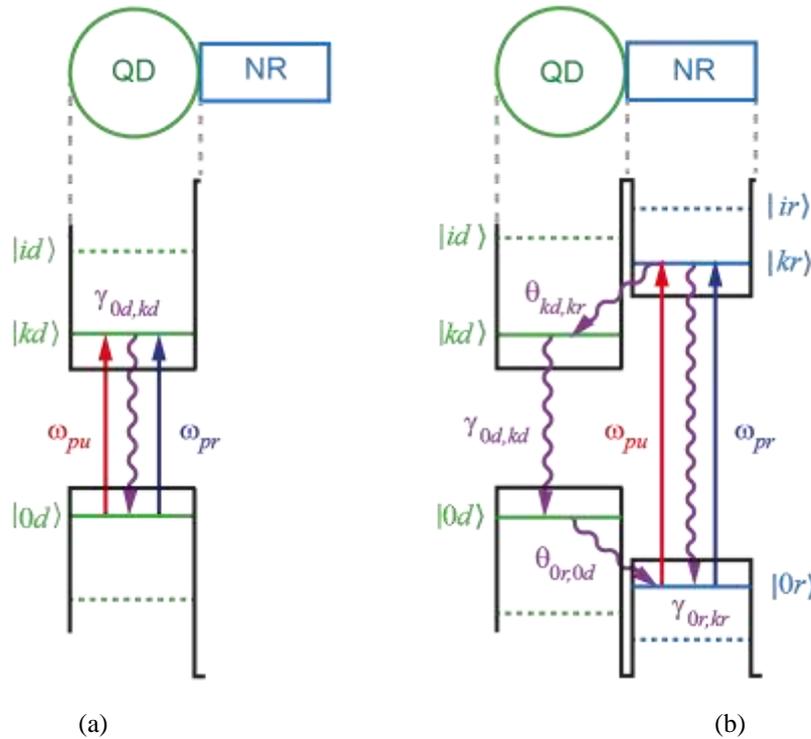
### 3. TRANSIENT INTERBAND ABSORPTION

Consider a symmetric nanotetrapod consisting of a quantum dot and four identical nanorods. It can be represented as a structure composed of a quantum dot and a single nanorod [see Fig. 1(b)] with four-fold degenerate electron and hole states. The absorption of the pump and probe pulses can be caused by generation of electron–hole pairs in the quantum dot or the nanorod. To calculate the rates of interband transitions between the states of electrons and holes, we consider two schemes of the two-pulse pump–probe method, involving interband absorption by a quantum dot [see Fig. 2(a)] or nanorod [see Fig. 2(b)].

When absorption occurs in the quantum dot, the pump pulse of carrier frequency  $\omega_{pu}$  resonantly excites electrons in state  $|kd\rangle$ , which then recombine at rate  $\gamma_{0d,kd}$  with holes in state  $|0d\rangle$ . If the probe pulse of carrier frequency  $\omega_{pr}$  (delayed by time  $\tau$  with respect to the pump pulse) is nearly resonant to the transition between states  $|0d\rangle$  and  $|kd\rangle$ , then it is absorbed, generating the electron–hole pairs. The kinetics of state populations and the absorption efficiency of probe heavily depend on relaxation rate  $\gamma_{0d,kd}$ , which can be found through the kinetics analysis.

We describe the considered absorption process mathematically using the two-level model of quantum dot. An exact solution to the problem of interband absorption shows that the absorption kinetics is generally described by a multi-exponential function that depends on the pulse shape. We restrict ourselves to cryogenic temperatures at which the spectral widths  $\sigma_{pu}$  and  $\sigma_{pr}$  of the pump and probe pulses can be chosen such that  $\tau \gg \sigma_{pr}^{-1}, \sigma_{pu}^{-1}$  and  $\sigma_{pu}, \sigma_{pr} \gg \gamma_{0d,kd}, \gamma_{kd,0}$ , where  $\gamma_{kd,0}$  is the total dephasing rate of transition  $kd \leftrightarrow 0d$ . In this case, the differential absorption of the probe by the quantum dot is found to be given by

$$\Delta E_{dot}^{(pr)}(\tau) = 2\hbar\omega_{pr}A_{dot}^{(pu)}A_{dot}^{(pr)}\left(\frac{2\sigma_{pu}}{\sigma_{pu}^2+\Delta_{pu}^2}\right)^2\left(\frac{2\sigma_{pr}}{\sigma_{pr}^2+\Delta_{pr}^2}\right)^2e^{-\gamma_{0d,kd}\tau}, \quad (12)$$



**Fig. 2** Electronic transitions in (a) quantum dot and (b) nanorod comprising a nanotetrapod being

successively excited by pump and probe pulses of frequencies  $\omega_{pu}$  and  $\omega_{pr}$ . Straight and wavy arrows show the optical and relaxation transitions, respectively.

where  $\Delta_{pr} = \omega_{kd} - \omega_{pr}$ ,  $\Delta_{pu} = \omega_{kd} - \omega_{pu}$ ,  $A_{dot}^{(pr)} = |V_{kd,0d}^{(pr)}|^2 / \hbar^2$ ,  $A_{dot}^{(pu)} = |V_{kd,0d}^{(pu)}|^2 / \hbar^2$ , and  $V_{kd,0d}^{(pr)}$  and  $V_{kd,0d}^{(pu)}$  are given in Eq. (10). This expression shows that the relaxation rate

$$\gamma_{0d,kd} = \frac{1}{\tau_2 - \tau_1} \ln \left( \frac{\Delta E_{dot}^{(pr)}(\tau_1)}{\Delta E_{dot}^{(pr)}(\tau_2)} \right). \quad (13)$$

can be found by measuring  $\Delta E_{dot}^{(pr)}(\tau)$  for two different time delays.

We now consider absorption by the nanorod shown in Fig. 2(b). In this case, the electrons generated in state  $|kr\rangle$  can recombine with holes in state  $|0r\rangle$  at rate  $\gamma_{0r,kr}$  or transfer their energy to the quantum-dot electrons in state  $|kd\rangle$  at rate  $\theta_{kd,kr}$ . The electrons residing in the quantum dot then recombine with holes in state  $|0d\rangle$  at rates  $\gamma_{0d,kd}$ . Finally, states  $|0d\rangle$  decay to state  $|0r\rangle$  at rate  $\theta_{0r,0d}$ . The total energy relaxation rate of state  $|kr\rangle$  is the sum  $\gamma_{kr,kr} = \gamma_{0r,kr} + \theta_{kd,kr}$ .

As in the previous case, the relaxation rate of the excited electron state can be found by studying the efficiency of probe absorption as a function of the delay time between the pulses. To calculate this efficiency, we use the four-level model of the nanotetrapod shown in Fig. 2(b). By assuming that  $\tau \gg \sigma_{pr}^{-1}, \sigma_{pu}^{-1}$  and  $\sigma_{pu}, \sigma_{pr} \gg \gamma_{kr,kr}, \gamma_{kr,0}$ , where  $\gamma_{kr,0}$  is the total dephasing rate of transition  $kr \leftrightarrow 0r$ , we arrive at the following functional dependency:

$$\Delta E_{rod}^{pr}(\tau) = 4\hbar\omega_{pr} A_{rod}^{(pu)} A_{rod}^{(pr)} \left( \frac{2\sigma_{pu}}{\sigma_{pu}^2 + \Delta_{pu}^2} \right)^2 \left( \frac{2\sigma_{pr}}{\sigma_{pr}^2 + \Delta_{pr}^2} \right)^2 \times \quad (14)$$

$$\left( \alpha_{kr,kr} e^{-\gamma_{kr,kr}\tau} - \alpha_{kd,kd} e^{-\gamma_{0d,kd}\tau} - \alpha_{0r,0d} e^{-\theta_{0r,0d}\tau} \right),$$

where  $\Delta_{pu} = \omega_{kr,kr} - \omega_{pu}$ ,  $\Delta_{pr} = \omega_{kr,kr} - \omega_{pr}$ ,  $\alpha_{kr,kr} = 2 + \alpha_{kd,kd} + \alpha_{0r,0d}$ ,  $\alpha_{kd,kd} = \theta_{kd,kr} / (\gamma_{0d,kd} - \gamma_{kr,kr})$ ,  $\alpha_{0r,0d} = \gamma_{0d,kd} / (\theta_{0r,0d} - \gamma_{kr,kr})$ ,  $A_{rod}^{(pr)} = |V_{kr,0r}^{(pr)}|^2 / \hbar^2$ , and  $A_{rod}^{(pu)} = |V_{kr,0r}^{(pu)}|^2 / \hbar^2$ ; and where  $V_{kr,0r}^{(pr)}$  and  $V_{kr,0r}^{(pu)}$  are given by Eq. (10). The analysis of the obtained multiexponential function is simplified by the fact that the

relaxation rates  $\gamma_{0d,kd}$  can be found from an independent experiment performed according to the scheme in Fig. 2(a). The above analysis shows that the relaxation parameters of the nanotetrapod can be determined experimentally using a combination of two considered schemes in Figs. 2(a) and 2(b).

#### 4. CONCLUSIONS

We have developed a theory of the pump–probe absorption spectroscopy, which allows determining the energy relaxation rates of the excited electronic states of a semiconductor nanotetrapod. Two schemes of transitions were considered, with the pump and probe pulses nearly resonant to the interband transition of the nanotetrapod. The pump-induced absorption of the probe was analyzed as a function of time delay between pulses. When the pulses are relatively short, this function is a sum of exponentials regardless of the

pulse shapes. The exponents are proportional to the energy relaxation rates of the excited states. This allows studying the kinetics of transitions and determining of relaxation parameters of semiconductor nanotetrapods, which may prove useful for the development of novel optical methods for nondestructive analysis of quantum nanostructures.

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