Linear and saturable nonlinear optical properties of GaAs quantum dot

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Abstract

In this paper the lowest transition energy for GaAs quantum dot (QD) is calculated by solving the Schrödinger equation for rigid confinement. The shift in lowest transition energy for a QD is larger than the bulk system and this makes it useful for application from florescent labeling to light emitting diodes. Employing this transition energy its linear and saturable nonlinear optical properties such as the real and imaginary part of dielectric constant, the index of refraction and the absorption coefficient are studied. The result confirms that the absolute strength of the resonance is indeed large compared to typical absorption behavior of bulk materials. Reducing the size of a QD, results in the blue shift of the maxima of absorption coefficient, refractive index and dielectric constant. The saturable nonlinear absorption coefficient and refractive index decreases with increasing intensity. Moreover the saturable nonlinear absorption completely saturates after enough refractive index and absorption changes takes place to make a given device switch in contrast to bulk system. This property may be utilized for optical bistability and optical switching.

Keywords: GaAs QD, dielectric constant, transition energy, absorption coefficient, index of refraction.

Resumen

En este trabajo se calcula la transición de energía más baja para los puntos cuánticos (QD) del GaAs mediante la resolución de la ecuación de Schrödinger para confinamiento rígido. El cambio en la transición de energía más baja para un QD es más grande que el sistema a granel y esto hace que sea útil para la aplicación del etiquetado fluorescente en diodos emisores de luz. Se estudió el empleo de esta energía de transición, sus propiedades ópticas no lineales y lineales saturables, tales como la parte real e imaginaria de la constante dieléctrica, el índice de refracción y el coeficiente de absorción. El resultado confirma que la fuerza absoluta de la resonancia es de hecho grande en comparación con el comportamiento de absorción típico de los materiales a granel. La reducción del tamaño de un QD, resulta en el desplazamiento al azul de los máximos del coeficiente de absorción, índice de refracción y constante dieléctrica. El coeficiente de absorción lineal saturable e índice de refracción disminuye con el aumento de la intensidad. Más aún, la absorción no lineal saturable se satura por completo después de bastante índice de refracción y cambios de absorción se llevan a cabo para hacer un cambio de dispositivo dado en contraste con el sistema mayor . Esta propiedad se puede utilizar para la biestabilidad óptica y conmutación óptica .

Palabras clave: GaAs QD, constante dieléctrica, energía de transición, coeficiente de absorpción, índice de refracción.

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I. INTRODUCTION

A large number of theoretical and experimental work has been devoted to the study of optical properties in low dimensional semiconductor nanostructures with carrier confined in one direction (quantum wells), and two dimensions (quantum wires) [1, 2]. The remarkable advancement in micro fabrication technology has now made it possible to realize structures particularly with quantum confinement in all three dimensions (quantum dots) [2, 3]. Some basic linear optical properties of QD's are already understood theoretically. The interband absorption spectrum should change from continuous band to a set of discrete lines, as a size of crystallite is decreased [4, 5].

The study of optical property in such QD structures is therefore considered to be a subject of fundamental interest and significant attention has been developed to it since last decade [2, 6]. A system of electrons fully confined in all three dimensions will have discrete charge and electronic states, as do atoms and molecules [7]. As we make semiconductor crystal smaller the physical properties change in many ways. In a large crystal the overall shape and size of the crystal makes little or no difference to its internal properties [8]. As a crystal becomes smaller, however, the effect of the surface becomes increasingly important.

The most extreme consequence is that as a fraction of atoms at the surface becomes larger, the arrangement of the nuclei changes in order to relax the energy of the system as a whole [6, 9]. In the case of strong confinement the image force effects are very small and can be neglected since the effects are not large enough to distort the wave functions significantly [10]. So it is very simple to calculate the single particle energy levels of the quantum dot. The quantum confinement will split the bulk conduction and valence bands in to a series of discrete energy shells in spherical quantum dot. The Schrödinger equation [11] for three dimensional spherical motions is,

$$\frac{-\hbar^2}{2m}\nabla^2\psi(r,\theta,\varphi)+V(r)\psi(r,\theta,\varphi)=E\psi(r,\theta,\varphi), \qquad (1)$$

where,

$$V(r)_{nl}=\begin{cases}0, & for \quad r<R,\\ \infty, & for \quad r\geq R.\end{cases} \qquad (2)$$

The well known solution of this Schrödinger equation for rigid confinement is,

$$R_{nl}(K_{nl}r)=\frac{1}{R}\sqrt{\frac{2}{r}}\frac{J_{l+\frac{1}{2}}(K_{nl}r)}{J_{l+\frac{3}{2}}(K_{nl}R)}, \qquad (3)$$

where, R is the radius of a QD and K_{NL} is the n^{th} zero of $J_{l+\frac{1}{2}}(K_{nl}R)$.

II. QUANTUM SHIFT OF THE FIRST TRANSITION

For 2.8 nm radius of spherical GaAs quantum dot the resulting quantum shifts of the first transition are calculated employing the roots of Bessel function from the equation,

$$J_{l+\frac{1}{2}}(K_{nl}R)=0. \qquad (4)$$

The function $J_{l+\frac{1}{2}}(K_{nl}R)$ is the $(l+\frac{1}{2})^{th}$ order spherical Bessel function and the coefficient K_{nl} is the n^{th} zero of $J_{l+\frac{1}{2}}(K_{nl}R)$. The roots are numerically determined by a program written in MATLAB. The results are illustrated in Table I.

The conduction band effective electron mass for GaAs QD is $0.067m_0$ (where m_0 is free electron mass) [13, 14] and the electron energy levels are given by

$$E_{nl}=E_{00}\left(\frac{K_{NL}}{K_{00}}\right)^2=\frac{5.6144\times10^{-14}}{R^2}\left(\frac{K_{NL}}{K_{00}}\right)^2 \text{cm}^2\text{-eV}, \qquad (5)$$

It clearly verifies that reducing the size of QD increases the energy gap of the QD. The 1s electron state increases in energy with decreasing the radius R of the QD, while the 1s hole state decreases. The band gap therefore grows and can be tune over a wide range by changing the radius R. For the smallest radii, there is a shift in the lowest transition energy.

TABLE I. Roots of $J_{l+1/2}(K_{nl}R)=0$ for $l=0$, 1, 2, 3 and 4.

No of zeros	*l*=0	*l*=1	*l*=2	*l*=3	*l*=4
1	3.14159	4.49341	5.76346	6.98793	8.18256
2	6.28319	7.72525	9.09501	10.4171	11.7049
3	9.42478	10.9041	12.3229	13.6980	15.0397
4	12.5664	14.0662	15.5146	16.9236	18.3013
5	15.7080	17.2208	18.6890	20.1218	21.5254
6	18.8496	20.3713	21.8539	23.3042	24.7276
7	21.9911	23.5195	25.0128	26.4768	27.9156

Spacing between the lowest two energy levels is 0.71612 eV. This is the quantum shift of the first transition for spherical GaAs quantum dot of 2.8 nm. The variation of energy with respect to radius of GaAs QD is illustrated in Fig. 1.

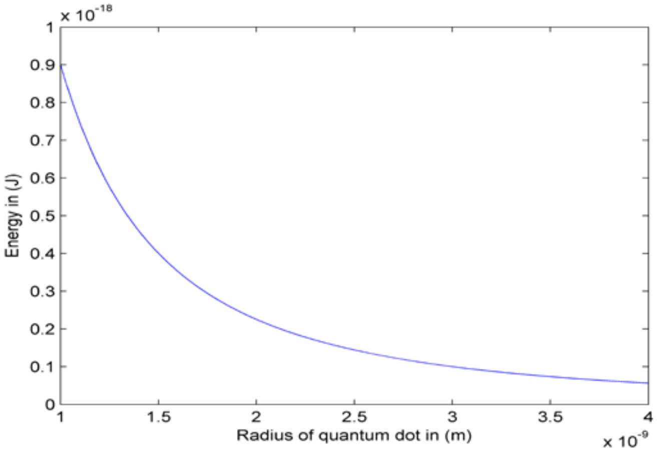

FIGURE 1. Energy versus radius of one electron GaAs quantum dot.

II. LINEAR OPTICAL PROPERTIES OF GaAs QUANTUM DOT

If the QD is subject to an oscillating electric field the perturbing Hamiltonian, $H(t)=H_p e^{i\omega t}$, induces a quantum transition from the initial state i of energy E_i to the final state f of energy E_f. Using Fermi's golden rule, the transition probability for a QD [2, 11, and 12] is,

$$W(\omega)=\frac{2\pi}{\hbar}\frac{e^2A_0^{\,2}}{m^{*2}c^2}\left|p_{cv}\right|^2\frac{1}{V}\rho^{0D}. \qquad (6)$$

Where, A_0 is the amplitude of the vector potential, p_{cv} is the optical matrix element, V the volume of the QD. The optical matrix element is described as [14]

$$\left|p_{cv}\right|^2=\frac{3m_e^{\,2}E_x\left(E_x+\Delta x_0\right)}{2\left(3E_x+2\Delta x_0\right)}\left(\frac{1}{m^*_e}-\frac{1}{m_e}\right). \qquad (7)$$

Where, E_x is the band gap of the QD with one electron, Δx_0 the spin- orbit splitting of valence bands and m_e * is the electron effective mass and m_e is the electron mass. For 2.8 nm radius of a spherical GaAs QD $E_x=2.16$ eV, $\Delta x_0=0.34$ eV and m_e*=0.067 m_e. The optical absorption coefficient (α) is proportional to the number of optical transitions per unit volume and time. It is given by absorbed energy per unit time $\hbar\omega W(\omega)$ divided by energy flux, $\frac{A_0^{\,2}\omega}{\lambda}=\frac{A_0^{\,2}\omega^2 n(\omega)}{2\pi c}$ and $\alpha(\omega)=\frac{\hbar\omega W(\omega)}{A_0^{\,2}\omega^2 n(\omega)}2\pi c$

$$\alpha(\omega)=\frac{1}{V}\frac{4\pi^2e^2}{m^{*2}\,\omega n(\omega)c}\left|p_{cv}\right|^2\rho^{0D}. \qquad (8)$$

The optical absorption coefficient in terms susceptibility can also be written as

$$\alpha(\omega)=\frac{4\pi\omega}{cn(\omega)}\operatorname{Im}\chi\cdot \qquad (9)$$

Relating equation (15) and equation (16) we obtain,

$$\operatorname{Im}\chi=\frac{f}{V}\sum(2l+1)\delta\left(E-E_{enl}-E_{hnl}\right)\cdot \qquad (10)$$

Where $f=\frac{2\pi e^2\hbar^2}{m^{*2}E^2}\left|p_{cv}\right|^2$.

The linear optical properties such as real and imaginary part of dielectric constant, and the absorption coefficient are determined for GaAs QD. In a GaAs QD, the allowed states are separated by large energies, so that transitions due to screening, exchange and band gap renormalization are difficult and strongly inhibited [11, 12]. For the same reason, it is difficult to polarize the exciton. In general, it is possible to have two electrons or holes in any given energy level because the individual electron and hole states are spin degenerate. This means however that there is no exchange interaction between excited particles in a given energy level, as they have different spins. Hence there are identically no exchange effects if we only create electrons and holes in a given conduction level, respectively. This will be the cause for the absorption of the lowest quantum dot transition. In the vicinity of the resonance the dielectric constant ($\varepsilon=\varepsilon_1+i\varepsilon_2$) for the quantum dot [3, 4, 5, 13] is,

$$\varepsilon=\varepsilon_\infty+\frac{\beta(\delta+i)}{1+\delta^2}. \qquad (11)$$

Where $\beta=\frac{4\pi f}{\hbar\Gamma_x V}$ and $\delta=\frac{\left(\hbar\Omega_x-\hbar\omega\right)}{\hbar\Gamma_x}$ with $\hbar\omega$ being the photon energy E, $\hbar\Omega$ being the lowest transition energy E_x, $\hbar\Gamma_x$ being the broadening E_b, and ε_∞ the back ground dielectric constant. After some manipulation, the real and imaginary part of the dielectric constant for a quantum dot is given by:

$$\varepsilon_1-\varepsilon_\infty=\left(\frac{9\pi e^2\hbar^2E_x\left(E_x+\Delta x_0\right)}{E_bR^3E^2\left(3E_x+\Delta x_0\right)}\right)\left(\frac{m_e-m^*}{m^*_e\,m_e}\right)\left(\frac{\left(E_x-E\right)\big/E_b}{1+\left(\frac{E_x-E}{E_b}\right)^2}\right) \qquad (12)$$

$$\varepsilon_2-\varepsilon_\infty=\left(\frac{9\pi e^2\hbar^2E_x\left(E_x+\Delta x_0\right)}{E_bR^3E^2\left(3E_x+\Delta x_0\right)}\right)\left(\frac{m_e-m^*}{m^*_e\,m_e}\right)\left(\frac{1}{1+\left(\frac{E_x-E}{E_b}\right)^2}\right) \qquad (13)$$

With the knowledge of ε_1 and ε_2 the index of refraction and the absorption coefficient are given by;

$$n_r(\omega)=\left(\frac{1}{2}\varepsilon_1(\omega)+\frac{1}{2}\left\{\left[\varepsilon_1(\omega)\right]^2+\left[\varepsilon_2(\omega)\right]^2\right\}^{\frac{1}{2}}\right)^{\frac{1}{2}}. \qquad (14)$$

$$\alpha(\omega)=\frac{\omega\varepsilon_2(\omega)}{c\left(\frac{1}{2}\varepsilon_1(\omega)+\frac{1}{2}\left\{\left[\varepsilon_1(\omega)\right]^2+\left[\varepsilon_2(\omega)\right]^2\right\}^{\frac{1}{2}}\right)^{\frac{1}{2}}} \qquad (15)$$

The curves in Fig. 2 are drawn according to (12) and (13) for a QD, with E_x =2.16 eV, E_b =0.5 meV, $\varepsilon_\infty=10$, $\Delta x_0=0.34$ eV and the radius of QD $R=2.8$ nm. They show the real and imaginary part of the dielectric constant.

The absolute strength of the resonance is indeed large as compared to typical absorption and refraction behavior in the bulk material. Fig. 3A and 3B demonstrate the linear refractive index and absorption coefficient respectively. These graphs are drawn according to (14) and (15). From Fig. 4 one can see that reducing a size of the QD increases the energy gap and results in the blue shift of the maxima of the above mensioned physical quantities. This property makes the QD applicable from florescent labeling to light emitting diodes.

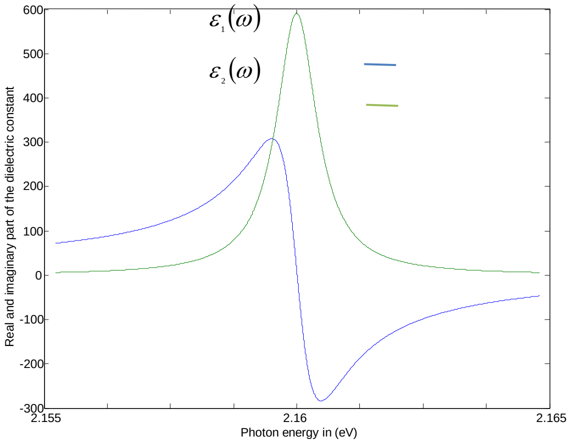

FIGURE 2. Real and Imaginary part of linear dielectric constant for (GaAs QD).

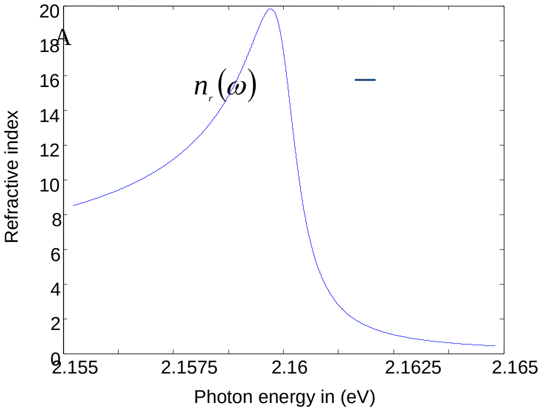

FIGURE 3(A). Linear refractive index versus photon energy for GaAs QD with one electron.

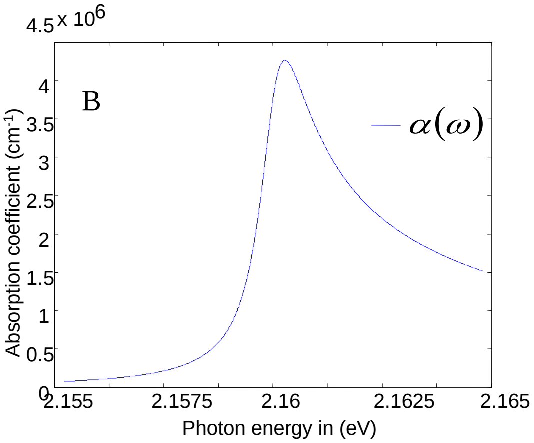

FIGURE 3(B). Linear refractive index versus photon energy for GaAs QD with one electron.

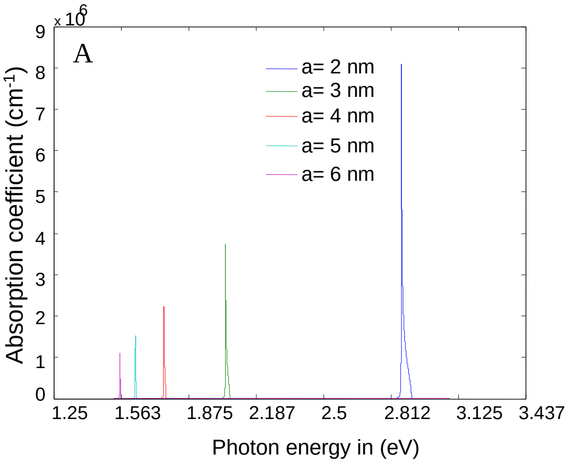

FIGURE 4. Linear absorption coefficient for one electron GaAs QD

IV. NONLINEAR OPTICAL PROPERTIES OF GaAs QUANTUM DOT

Since exchange and Coulomb effects are rigorously negligible, the absorption saturation of the lowest transition in the QD can be treated like a pair of two level systems, one for each spin. As the result the absorption saturates as [12, 13],

$$\varepsilon = \varepsilon_\infty + \frac{\beta(\delta + i)}{1 + \delta^2 + I/I_s} \qquad (16)$$

where $I = E_0^2$ is the intensity with E_0 being the peak amplitude of the oscillating electric field inside the QD. $I_s = \hbar/(\beta\tau V)$ is the saturation intensity, with τ being the recombination time. The absolute changes in absorption and consequently in the refractive index of a QD can be much larger before the transition saturates in contrast to bulk system. The real and imaginary part of the dielectric constant for this case is,

$$\varepsilon_1 - \varepsilon_\infty = \left(\frac{9\pi e^2\hbar^2 E_x(E_x + \Delta x_0)}{E_b R^3 E^2(3E_x + \Delta x_0)}\right)\left(\frac{m_e - m_e*}{m_e*_e m_e}\right)\left(\frac{(E_x - E)/E_b}{1 + \left(\frac{E_x - E}{E_b}\right)^2 + I/I_s}\right). \qquad (17)$$

$$\varepsilon_2 - \varepsilon_\infty = \left(\frac{9\pi e^2\hbar^2 E_x(E_x + \Delta x_0)}{E_b R^3 E^2(3E_x + \Delta x_0)}\right)\left(\frac{m_e - m_e*}{m_e*_e m_e}\right)\left(\frac{1}{1 + \left(\frac{E_x - E}{E_b}\right)^2 + I/I_s}\right). \qquad (18)$$

The intensity dependent refractive index and absorption coefficient according to (14), (15), and (17) and (18) are described in Fig. 5 A and 5B. From the figures one can observe that the saturable nonlinear refractive and absorption coefficients decrease with increasing intensity. For QDs the nonlinear absorption completely saturates after enough refractive index changes or absorption changes takes place to make a given device switch in contrast to the bulk system. This property may be important to utilize a QD as a system for optical bistability and optical switching.

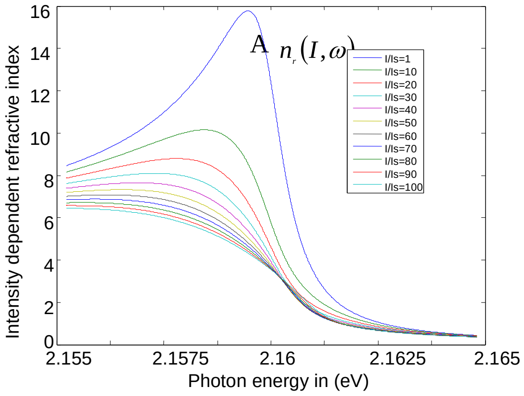

FIGURE 5A. Intensity dependent refractive index for one electron GaAs QD.

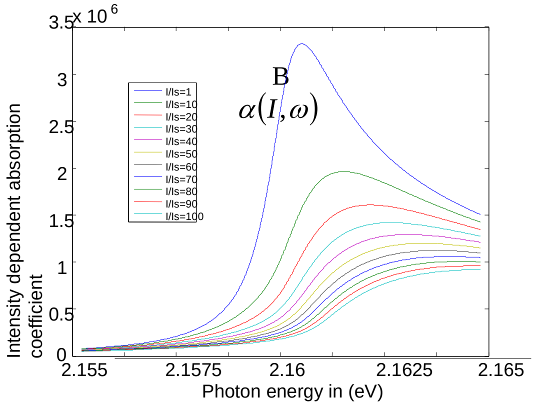

FIGURE 5B. Intensity dependent absorption coefficients for one electron GaAs QD.

V. CONCULUSION

For a 2.8 nm radius GaAs QD the lowest transition energy is about 2.16 eV and is much larger compared to its counter bulk system. This implies reducing the size of a QD results in the blue shift of the máxima of the absorption coefficient, refractive index and dielectric constant. It is also observed that the absolute strength of the saturable nonlinear refractive index and absorption coefficient decrease with increasing intensity. Moreover the saturable nonlinear absorption completely saturates after enough refractive index and absorption coefficient changes takes place to make a given device switch in contrast to the bulk system. This property may be utilized for optical bistability and optical switching.

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