Linear and saturable nonlinear optical properties of GaAs quantum dot

Menberu Mengesha
Department of Physics, College of Natural and Computational Science, Mizan-Tepi University, P. O. Box 121, Tepi, Ethiopia

E-mail: mmennbb@yahoo.com

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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 fluorescent 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.

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.

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 bistabilidad óptica y conmutación óptica.

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

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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,

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

where,

\[ V(r) = \begin{cases} \infty, & \text{for } r < R, \\ 0, & \text{for } r \geq R. \end{cases} \]  

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

\[ R_\nu(K_\nu R) = \frac{1}{R} \sqrt{\frac{2}{r}} \frac{J_{\nu+\frac{1}{2}}(K_\nu r)}{J_{\nu+\frac{1}{2}}(K_\nu R)}, \]  

where, \( R \) is the radius of a QD and \( K_n \) is the \( n \)th zero of \( J_{-\nu+\frac{1}{2}}(K, R) \).

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,

\[ J_{\nu+\frac{1}{2}}(K_\nu R) = 0. \]  

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

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</table>

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.

II. LINEAR OPTICAL PROPERTIES OF GaAs QUANTUM DOT

If the QD is subject to an oscillating electric field the perturbing Hamiltonian, \( H(t) = H_0 e^{i\omega t} \), 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,
\[ W(\omega) = \frac{2\pi}{\hbar} \frac{e^2 A^2}{m^*} \left[ \rho_{cv} \right]^2 \frac{1}{V} \delta^{\omega_0} \cdot \] (6)

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

\[ |\rho_{cv}|^2 = \frac{3m_e^2 E_x (E_x + \Delta x_0)}{2(3E_x + 2\Delta x_0)} \left( \frac{1}{m_e^*} - \frac{1}{m_e} \right). \] (7)

Where, \( E_x \) is the band gap of the QD with one electron, \( \Delta x_0 \) 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 \( (\alpha) \) is proportional to the number of optical transitions per unit volume and time. It is given by absorbed energy per unit time \( h\omega W(\omega) \) divided by energy flux, \( A_r \omega \overline{n}(\omega) \) and \( \alpha(\omega) = \frac{h\omega W(\omega)}{A_r \omega \overline{n}(\omega)} \cdot \]

\[ \frac{A_r \omega \overline{n}(\omega)}{2m} \] (9)

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

\[ \alpha(\omega) = \frac{4\pi \omega}{cn(\omega)} \text{Im} \chi. \] (9)

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

\[ \text{Im} \chi = \frac{f}{V} \sum (2l+1) \delta(E - E_{cv} - E_{inl}). \] (10)

Where \( f = \frac{2\pi^2 \hbar^2}{m^*^2 E^2} |\rho_{cv}|^2 \cdot \]

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 \( (\epsilon = \epsilon_1 + i\epsilon_2) \) for the quantum dot \([3, 4, 5, 13] \) is,

\[ \epsilon = \epsilon_\infty + \frac{\beta(\delta + i)}{1 + \delta^2}. \] (11)

Where \( \beta = \frac{4\pi e^2}{\hbar \Gamma_{\epsilon}} \) and \( \delta = \frac{(\hbar \Omega_{\epsilon} - \hbar \omega)}{\hbar \Gamma_{\epsilon}} \) with \( \hbar \omega \) being the photon energy \( E_\epsilon \), \( \hbar \Omega_{\epsilon} \) being the lowest transition energy \( E_{\epsilon x} \), \( \hbar \Gamma_{\epsilon} \) being the broadening \( E_{b \epsilon} \), and \( \epsilon_\infty \) the background dielectric constant. After some manipulation, the real and imaginary part of the dielectric constant for a quantum dot is given by:

\[ \epsilon_1 - \epsilon_\infty = \left( \frac{9\pi^2 \hbar^2 E_x (E_x + \Delta x_0)}{E_{b \epsilon} R^2 E_x^2 (3E_x + \Delta x_0) \left( m_e^* - m_e \right) m_e^* m_e} \right) \left( \frac{E_x - E_{b \epsilon}}{E_{b \epsilon}} \right)^2 \] (12)

\[ \epsilon_2 - \epsilon_\infty = \left( \frac{9\pi^2 \hbar^2 E_x (E_x + \Delta x_0)}{E_{b \epsilon} R^2 E_x^2 (3E_x + \Delta x_0) \left( m_e^* - m_e \right) m_e^* m_e} \right) \left( \frac{1}{1 + \left( \frac{E_x - E_{b \epsilon}}{E_{b \epsilon}} \right)^2} \right)^2 \] (13)

With the knowledge of \( \epsilon_1 \) and \( \epsilon_2 \) the index of refraction and the absorption coefficient are given by:

\[ n_\epsilon(\omega) = \left( \frac{1}{2} \epsilon_\epsilon(\omega) + \frac{1}{2} \left[ \epsilon_\epsilon(\omega) \right]^2 + \left[ \epsilon_\epsilon(\omega) \right]^2 \right)^{\frac{1}{2}}. \] (14)

\[ \alpha(\omega) = \frac{\omega \epsilon_\epsilon(\omega)}{c} \epsilon_\epsilon(\omega) \left( \frac{1}{2} \epsilon_\epsilon(\omega) + \frac{1}{2} \left[ \epsilon_\epsilon(\omega) \right]^2 + \left[ \epsilon_\epsilon(\omega) \right]^2 \right)^{\frac{1}{2}} \] (15)

The curves in Fig. 2 are drawn according to (12) and (13) for a QD with \( E_x = 2.16 \) eV, \( E_{\epsilon x} = 0.5 \) meV, \( \epsilon_\infty = 10 \), \( \Delta x_0 = 0.34 \) eV and the radius of QD \( R = 2.9 \) 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 mentioned physical quantities. This property
makes the QD applicable from fluorescent labeling to light emitting diodes.

![Graph of linear and saturable nonlinear optical properties of GaAs quantum dot](image)

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

![Graph of linear refractive index versus photon energy for GaAs QD with one electron](image)

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

![Graph of linear absorption coefficient for one electron GaAs QD](image)

**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],

\[
\epsilon = \epsilon_0 + \frac{\beta(\delta + i)}{1 + \delta^2 + I/I_s},
\]

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 \( \tau \) 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,

\[
\epsilon_r - \epsilon_i = \frac{9 \pi e^2 \hbar E_0 (E_0 + \Delta x)}{E_0 R E (3E_0 + \Delta x)} \left( \frac{m - m^*}{m^* m} \right) \left( \frac{E_0 - E}{E_0} \right) \left( \frac{E_0 - E}{E_0} + 1 \right)
\]

(17)

\[
\epsilon_r - \epsilon_i = \frac{9 \pi e^2 \hbar E_0 (E_0 + \Delta x)}{E_0 R E (3E_0 + \Delta x)} \left( \frac{m - m^*}{m^* m} \right) \left( \frac{1}{E_0 - E} \right) \left( \frac{1}{E_0 - E} + 1 \right)
\]

(18)

The intensity dependent refractive index and absorption coefficient according to (14), (15), and (17) and (18) are described in Fig. 5A 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.

This property may be utilized for optical bistability and optical switching.

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