SOME STUDIES ON LINEAR AND NONLINEAR OPTICAL PROPERTIES OF INTRABAND TRANSITIONS IN SILICON QUANTUM DOTS

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SYNOPSIS

Nanotechnology can be defined as the science and engineering involved in the design, characterization, production and application of materials at smaller size as at nanometer scale. In the last few decades nanoscience and nanotechnology has become one of the most intensely studied area of research. The fabrication of devices using components of nano dimension has fuelled the need for nanofabrication techniques as well as a need for understanding the physics of materials at nanometer scale. The nanomaterials behaviour and properties have revealed many exciting and technologically important insights into science and engineering. Nano-scaled materials due to their quantum nature provide exciting opportunities to develop new technological devices e.g. tandem solar cells, infrared photodetectors, single electron transistors. A wide ranging interest in nanostructures is mainly to control and exploit quantum mechanical properties which are not observable by using bulk.

During the last few decades the study of electronic and optical properties of semiconductor nanostructures has been the subject of great interest both theoretically and experimentally. At the nanometer size of a material comparable to the characteristic de-Broglie wavelength of electron, its continuous band structure is replaced by discrete energy levels because of the quantum confinement effects. Quantum confinement effects are present in a wide variety of systems, e.g. in quantum wells, quantum wires, and quantum dots. In quantum wells and quantum wires the charge carries are confined in one and two dimensions respectively. In quantum dots (QDs) the charge carriers are confined in all three dimensions due to which they behave differently from quantum wells and quantum wires. In QDs due to 3D quantum confinement the density of states is delta like with well separated energy levels, instead of being continuous bands as in bulk or subbands of energy levels in semiconductor quantum wells and quantum wires. Hence, quantum dots behave as artificial atoms. The atom like structure of QDs has been demonstrated experimentally [1]. Due to atomic like structures the optical properties of quantum dots are of great interest. The energy levels can be monitored to different values by controlling the shape of the dot, size of the dot, dot material and the confinement potential. The electronic transitions in the discrete energy levels can be used to tailor the optical properties of quantum dot nanostructures. Their band gap energy, luminescence wavelength, transition probability, photoabsorption and emission spectra can be tuned by adjusting the dot size [2]. These features of quantum dots are very useful for the development of optoelectronic devices such as in light emitting diodes [3-4], photovoltaics [5-6] and biological imaging [7].

In the interaction of radiation with matter at low radiation intensity the induced polarization depends linearly on the electric field and linear response is obtained by reflection, refraction, scattering or absorption. However, when the intensity is increased nonlinear terms in induced polarization become important which lead to nonlinear optical properties. There are many nonlinear optical processes e.g. second harmonic generation, four wave mixing, optical Kerr effect, third harmonic generation, and multiphoton absorption processes [8]. In the presence of intense field the induced polarization become a nonlinear function of the applied field. Theoretical prediction of the nonlinear optical process, the simultaneous absorption of two photons by the same molecule was made by Goppert-Mayer [9]. The experimental demonstration of the nonlinear optical process became possible only after the invention of laser in 1960 by Maiman [10]. The experimental demonstration of the second harmonic generation by Franken et al. [11] in 1961 is considered as the milestone of nonlinear optics. In the second and the third harmonic generation processes the doubling and the tripling of the incident frequency are obtained respectively. The nonlinear optical response by a material depends on the symmetry in its structure e.g. the second order optical nonlinearity occurs only in noncentrosymmetric material and the third order nonlinear optical process is possible for the both centrosymmetric and noncentrosymmetric materials.

In semiconductor quantum dots, the nonlinear optical properties are known to be greatly enhanced as compared to the bulk semiconductors due to the quantum confinement effect [12]. The nonlinear optical properties of quantum dots have attracted considerable interest due to their relevance to several applications such as in optical power limiting [13-14], frequency upconversion [15].

The interaction of radiation with quantum confined systems results in interband and intraband optical transitions. The interband transitions occur between the conduction band and the valence band involving two types of charge carriers (electrons and holes). The first evidence of strong intersubband absorption in multiquantum wells was observed by West et al. [16]. Based on the results of intersubband absorption Levin et al. [17] demonstrated the first quantum well infrared photodetector. From the physical aspect and device application point of view the interband optical properties of quantum dots have been widely studied [18-20]. The intraband transitions take place between quantized levels within the valence band or in conduction band [21]. The studies on intraband transitions in semiconductor QDs have attracted researchers about two decades back. The first observation of intraband transitions in semiconductor QDs was reported in 1994 by Drexler et al. [22] in InGaAs self assembled quantum dots. The intraband transitions in semiconductor QDs are of particular interest due to their transition energies being in the infrared region and involving only one type of charge carrier which allows studying separately the dynamics of electrons and holes. In quantum dots, the intraband transitions are possible for any polarization of light due to the confinement in all three directions. The intraband transitions in quantum dot are used in the developments of infrared photo detectors and lasers.

Silicon is the most important material for variety of applications in the optical and electronics industry due to its low cost, nontoxic, biocompatible and natural abundant advantages. The study of silicon quantum dot embedded in dielectric matrix has received strong attention due to interesting fundamental physical properties of these mesoscale objects and several technological applications in advanced electronics devices and optoelectronic devices [23-24] and as fluorescent markers in bio-imaging [25]. Also silicon quantum dots are excellent material for the development of third generation photovoltaic devices. The silicon quantum dots improve the light emission properties of their bulk counterpart [26]. The nonlinear optoelectronic responses in silicon nanostructures are of particular interest due to their promising applications such as in optical switching [27]. Due to rapid advancements in different synthesis techniques silicon quantum dots of different sizes can be fabricated [28-31]. In the literature several studies are reported on the linear and the nonlinear optical properties of Si nanostructures [32-37]. Sousa et al. [32-33] investigated the intraband absorption in Si/SiO_2 quantum dot using a tridimensional quantum mechanical model. Ma et al. [34] reported the absorption and photoluminescence spectra of nanocrystalline silicon embedded in SiO_2 matrix. Yildirim and Bulutay [35] used an atomistic pseudopotential approach to study nonlinear optical properties of Si and Ge nanocrystals embedded in SiO_2 matrix. Prusty et al. [36] calculated intensity dependent changes in the refractive index for various sizes of nanocrystallites. Using the z-scan and optical Kerr gate methods, Imakita et al. [37] studied the nonlinear optical properties of boron (B)-doped silicon nanocrystals embedded in borosilicate glass. Most of these studies are related to the interband transitions and there are only few experimental [38] and theoretical studies [32-33] on the intraband transitions in silicon nanostructures. Since the crystalline Si is centrosymmetric, the second order susceptibility vanishes and the third order is the lowest nonlinearity.

OBJECTIVES

The main objective of the present work is to use quantum mechanical formulation to study the linear and the third order nonlinear optical properties associated with the intraband transitions in the single electron charged silicon quantum dots. The work is divided into two categories:

(i) Linear optical properties of Si QDs

- To study the photo absorption process.
- To study the photoelectric process.

(ii) Nonlinear optical properties of Si QDs

- To study the third order nonlinear optical process and to calculate the third order nonlinear photo absorption coefficient and nonlinear refractive index change.
- To study the third harmonic generation process.
- To study the two photon absorption and photoelectric processes.
- To study the effect of the change in dot size and surrounding matrix on the linear and nonlinear optical properties.

The thesis is organized in seven chapters.

CHAPTER 1

In the Chapter 1 the brief introduction is given of (i) nanostructures (quantum wells, wires, and quantum dots), (ii) theoretical methods for the electronic structure calculation, (iii) interband and intraband transitions, (iv) the linear and the nonlinear optical processes and properties, and (v) some fabrication methods of silicon quantum dots.

CHAPTER 2

A study is reported on the photoabsorption and photoelectric process in single electron charged spherical Si semiconductor quantum dot embedded in amorphous SiO_2 matrix. Using the effective mass approximation (EMA) the energy levels of the dot, the photoabsorption coefficient, and photoelectric cross sections are investigated by considering the potential barrier at the interface of the dot and the matrix as of (i) infinite and (ii) finite height. The effect of self energy which is associated with the surface polarization due to the charging of the quantum dot is also considered. The present theoretical results provide a good agreement with the available the experimental data for optical transitions in the conduction band. Since these transitions are mainly in the infrared region this study will be useful in the development of infrared devices using quantum dots (e.g. infrared detectors). Further, the study on the photoelectric process in Si quantum dot will be useful in the development of photovoltaic devices (solar cell with an array of Si quantum dots). The results of this Chapter are reported in our publication [39].

CHAPTER 3

A study is reported on the linear and the nonlinear optical processes in single electron charged spherical Si semiconductor quantum dot. The density-matrix formulation has been used for the calculation of the linear and the third order nonlinear optical properties. The intraband linear, third order nonlinear and total photoabsorption coefficient of Si QD surrounded by oxide (SiO_2, Al_2O_3), nitride (Si_3N_4), and carbide (SiC) matrix have been investigated. Also the linear and the third order nonlinear refractive index changes of Si QD have been investigated. In this study within the framework of EMA, we consider the finite barrier height at the interface of the dot and matrix, the self-energy, and the size dependent dielectric constant of the dot. We study the effect of variation in size of the dot and the change in the surrounding matrix on the optical properties of Si QDs. The present theoretical study shows that the third order nonlinear photoabsorption coefficient, refractive index change and the third order nonlinear susceptibility are significantly influenced by the optical intensity and the size of the dot. It is noted that the decrease in size of the dot leads to blue shift in the peak positions of the photoabsorption coefficient, the refractive index change and the third order nonlinear susceptibility. A decrease in the total photoabsorption coefficient and refractive index change is obtained with increasing optical intensity. It is observed that the higher absorption and higher photoelectric cross sections are obtained in Si quantum dot embedded in Al_2O_3 matrix as compared to SiO_2 matrix. Also it is found that all the linear and nonlinear parameters in photoabsorption process and photoelectric process are strongly dependent on the confinement potential and the relative values of the dielectric strengths of the dot material and the surrounding matrix. The role of the surrounding matrix is to change the local field factor and the barrier height which leads to changes in the magnitude and peak position of the absorption coefficient. We expect that the present work will stimulate experimental and theoretical studies useful to develop optoelectronic devices based on nonlinear optical properties of silicon quantum dots. The results of this Chapter are reported in our publication [40-41].

CHAPTER 4

This Chapter deals with the study of the effect of the barrier height on the linear and the third order nonlinear photoabsorption coefficient and the refractive index change in the intraband transitions in spherical single electron Si quantum dots embedded in dielectric matrix. The present study shows that the increase in barrier height leads to blue shift in peak positions of absorption coefficient and refractive index change. The peak value of the total photoabsorption coefficient increases with the increase of the barrier height. However, the total refractive index change, and absolute value of third order nonlinear optical susceptibility decrease with the increase of the barrier height. Also the total photoabsorption coefficient and the refractive index change decrease in magnitude with the increase of optical intensity. The present study about the effect of the barrier height on the nonlinear optical properties would be useful in optoelectronic devices based on Si quantum dots. The results of this Chapter are reported in our publication [42].

CHAPTER 5

This Chapter presents the study on the third harmonic generation (THG) process associated with the intraband transitions in the conduction band of the singly charged spherical silicon quantum dots embedded in surrounding matrix of silicon dioxide, silicon nitride, silicon carbide, and alumina. Using the density matrix approach we study the third harmonic generation. We also study the effect of the variation of the dot size and the surrounding matrix on the THG coefficient for Si QD. The effective mass approximation has been used. The present study shows that the THG coefficient for spherical Si quantum dot increases with the size of dot. The resonant peak positions of THG coefficient show red shift with increasing dot radius. The surrounding matrix strongly affects the THG coefficient. The results of this Chapter are reported in our publication [43].

CHAPTER 6

This Chapter deals with the investigation of the two photon absorption by an electron in the conduction band of a singly charged Si quantum dot embedded in SiO_2 and Al_2O_3 matrix. Two cases are considered viz: (i) the two photon excitation process in which the electron is excited from its initial bound 1s state to a higher energy bound state (1d, 2s) by absorbing two photons (TPBB), and (ii) the two photon photoelectric process in which the electron is ejected out of the dot material to a free state by absorbing two photons in its initial bound state (TPBF). The effect of the variation of the dot size and the surrounding matrix on the TPBB and TPBF processes is studied in the framework of the EMA and the finite barrier height at the interface of the dot and the matrix. The present study shows that the TPBB and TPBF processes strongly depend on the dot size. The increase in the dot size leads to a red shift in peak positions of the TPBB coefficient and enhancement of its magnitude while in the TPBF process it leads to a blue shift in the peak position of the TPBF cross section and a decrease in its magnitude. The TPBB coefficient and the TPBF cross section are relatively higher for the Al_2O_3 as compared to the SiO_2 matrix. The effect of the polarization of the incident photon on the TPBF has been investigated and the variation with photon energy of the ratio of the TPBF cross sections for circularly and linearly polarized photon is presented. The results of this Chapter are reported in our publication [44].

CHAPTER 7

This Chapter summarizes all the results reported in the thesis.

LIST OF PUBLICATIONS

International Journals

[1] Anchala, Purohit S. P., Mathur K. C., *"Photoabsorption and photoelectric process in Si nanocrystallites"*, Appl. Phys. Lett., vol. 98, pp. 043106-1-043106-3, 2011[Impact factor 3.844, H index 290].

[2] Anchala, Purohit S. P., Mathur K. C., *"Linear and nonlinear intersubband optical properties of Si quantum dot embedded in oxide, nitride, and carbide matrix"*, J. Appl. Phys., vol. 110, pp. 114320-1-114320-8, 2011 [Impact factor 2.169, H index 207].

[3] Anchala, Purohit S. P., Mathur K. C., *"Third order nonlinear optical properties of Si nanodot in Al_2O_3"*, IEEE J. Quantum Electron., vol. 48, pp. 628-634, 2012 [Impact factor 2.48, H index 92].

[4] Anchala, Purohit S. P., Mathur K. C., *"Effect of barrier height on linear and nonlinear photoabsorption and refractive index change in Si quantum dots"*, Adv. Mat. Lett., vol. 3, pp. 498-503, 2012 [H index 12].

[5] Anchala, Purohit S. P., Mathur K. C., *"Third harmonic generation in intraband transitions of spherical silicon quantum dots"*, J. Appl. Phys., vol. 112, pp. 094306-1-094306-5, 2012 [Impact factor 2.169, H index 207].

[6] Anchala, Purohit S. P., Mathur K. C., *"Two photon absorption processes in silicon quantum dot embedded in SiO_2 and Al_2O_3 matrix"*, J. Comput. Theor. Nanosci., vol. 11 (in press) [Impact factor 0.981, H index 23].

International Conferences

[1] Anchala, Purohit S. P., Mathur K. C., *"Photoabsorption in Si semiconductor quantum dot nanostructure"*, AIP Conf. Proc. 1313 International Conference on Physics of Emerging Functional Materials, BARC, Mumbai, India 230-32 (Sept. 22-24, 2010).

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