

Theoretical model for Bandgap Engineering of Semiconductor Quantum Dots

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Abstract—Bandgap engineering model in semiconductor quantum dots (QDs) is here presented for technological applications. The model obtained demonstrates that the optical bandgap can be tuned to custom-designed by varying the confinement size. This results in significant increase in energy of band-to-band excitation peaks and hence a blue shift in the absorption and luminescence bandgap energies with decreasing QD size. This offers potential revolutionary solutions in many areas of modern science and engineering technology to overcome the fundamental limitation of conventional semiconductors that have their bandgap fixed. In addition, QDs display broad absorption band characteristics with narrow-emission spectra that are tunable due to size quantization effects, which contribute to advancement of medical imaging and opens up several multiplexing potentials such as multicolor detection with a single wavelength excitation source. It is found that CdSe and CdS QDs possess an optical spectrum that confer on them potential active materials for efficient light emitting diode (LED) and lasers operating over the whole range of visible region. ZnS QD possesses the widest bandgap energy which plays a vital role for absorption and emission of high energy blue photons and permits devices to operate at much higher voltages and temperatures crucial for optoelectronic device applications such as Pn junctions and power transistors. In addition, the wide bandgap absorption spectra will be relevant in high optical transmittance specifically in the range of visible to infra red (IR) spectral region. GaAs, InAs and InSb QDs show promising optical bandgap energies in the visible to near infrared (NIR) spectral region which is desirable for optoelectronic devices, operating at NIR wavelengths. We found that PbS, PbSe and PbSe QDs displayed exceptional optical characteristics that are favorable for solar cells applications owing to the fact that their absorption band are fairly good match to the solar spectrum.

Keywords—Theoretical model, Quantization effect, Bandgap engineering, Quantum confinement, quantum dot, Schrodinger equation, Spherical potential well, opto electronics.

I. INTRODUCTION

A wide range of semiconductor nano-materials are presently being explored tremendously for their novel applications in optoelectronics, primarily due to the growing need of substituting traditional materials with some alternative cost-effective materials offering commercially viable technologies. For this reason, an increasing amount of effort from both the scientific community and industry has focused on development of novel concepts and theoretical models for controlling the electronic and optoelectronic properties of semiconductors to meet the specific requirements via bandgap engineering (Smith and Nie, 2010). The process of altering bandgap energies of semiconductors for desired physical properties to meet device applications has driven research into new semiconductor materials leveraging on the principles of quantum confinement to engineer their bandgap and optoelectronic properties to have custom-designed energies, to enhance the performance of photonic devices. This current approach is more relevant, as the conventional

approach is faced with unavoidable problems of lattice mismatch.

A common approach to bandgaps engineering is to alloy two or more semiconductors with different bandgaps, and then all bandgaps between the constituent semiconductors can in principle be realized by controlling the alloy composition [1]. In general, the range of bandgaps that can be achieved in high-quality single crystal alloys practically suffers inherent limitations by the strict requirement of lattice matching in planar epitaxial growth, thus the lattice constant of the material to be grown has to match with that of the substrate [2]. Owing to the limited availability of substrates and the lattice mismatch issue, the range of achievable bandgaps is severely constrained and hence not all bandgaps are naturally available through the direct synthesis of elemental or binary individual semiconductors [3]. Revolutionary advancements require semiconductors materials with tunable bandgap characteristics that can be tailored for specific applications. When the material length scales down into the nanometric dimension approximately from 1nm to 100nm quantum effects become prominent leading to confinement of carriers (electrons and holes) [4],

which forces them to occupy space smaller than the normal equilibrium distance in the bulk matter. Precisely, as the size of semiconductors is reduced to nanoscale, their fundamental bandgap energies become size dependent [5] thereby allowing for engineering of their optical and electro-optical responses. Thus decreasing the physical size of a semiconductor into the nanoscale dimension can also be regarded as a bandgap engineering technique as it alters the bandgap due to quantum confinement effects [6]. Such bandgap engineering optimizes their optical performances considerably and provides the state – of – the- art device techniques.

An interesting phenomenon which characterizes semiconductors in the nanoscale regime is the ability to control their electrical and optical properties by controlling their sizes known as quantization effect [7], as was demonstrated for quantum structures that have their charge carrier confined in one, two or three directions by potential barriers, such as in quantum wells, quantum wires, and quantum dots respectively, contrary to bulk absorption spectrum that maintains continuous energy band distribution with their charge carriers free to move within their respective bands in all the three directions [8]. The dream materials which will enable new applications due to size tunable bandgap has led to the emergence of QDs being the ultimate limit of carrier confinement among other confined structures [9]. The persistence of interest in QDs arises from their novel optical characteristics that are not accessible with the bulk materials [10].

QDs are semiconductors in nanometer dimensions providing strong quantum confinements on the carriers in all the three spatial directions leading to discrete and size dependent energy levels [11]. The confinement of carriers in QDs dramatically modulates the charge carrier states thereby enabling development of new materials with significant milestones in the frontier semiconductor devices [12]. The fundamental bandgap energy of QDs is tunable by varying their confinement dimension which allows engineering of novel materials that have their bandgaps at desired energies [13]. As a result, the bandgaps of these nano-materials can be widely tuned without the detrimental defects that are often unavoidable in bulk materials which are beset with limited tolerance to lattice mismatches. The transport properties of photo excited carriers can be independently and continuously engineered for a given application [14]. This class of nano-materials could have significant potential technological applications on a wide range of photonic and optoelectronic devices, including tunable lasers, solid-state lightings, artificial photosynthesis, light emitting diodes, flat panel displays, Bio-labeling, and new solar cells. In this paper quantum confinement model within the effective mass approximation is extended to analyze the possibility of bandgap engineering in QDs, and their potential applications in different technologies.

We organized the work in sections as follows. The introduction and related literature are discussed extensively

in section 1. Section 2 provides detailed related work followed by methodology described in section 3 which deals with the computation details and method of calculation. A detailed analytical results as obtained from the simple model have been provided and been thoroughly discussed in section 4. In the section 5 the conclusion of the observed results are presented.

II. RELATED WORK

QDs are particles with physical dimensions smaller than the exciton Bohr radius [15]. The small size of QDs leads to “quantum confinement” The quantum confinement effects occur when size of QD is comparable to bulk exciton Bohr radius [16]. The quantum confinement means that the motion of charge carriers is restricted within the quantum box such that the energy levels that the carriers inhabit become discrete, with a finite separation between them [17]. However, there are some energy levels that the electrons cannot occupy, which are collectively known as band gap. The versatility and usefulness of semiconductor materials lies in their fundamental band gap energy which in turn govern their photonics and optoelectronic properties [18]. The bandgap is therefore arguably the most critical parameter for almost all applications involving photon absorption and emission [19]. The fundamental bandgap energy of QDs are alter as a function of size due to quantum effect [20]. The optical bandgap is broaden with decreasing particle size leading to a blueshift in the bandgap absorption spectrum, as a consequence, optoelectronic properties can be specifically tuned to have desired energy output by varying the dot size [21].

In a quantum dot, the charge carriers are confined in all three dimensional potential well and by approximation can be described by particle in a sphere in which the photo-excited carriers (electron and hole) may be treated as particles in a spherical potential well [22]. The potential energy is zero everywhere inside the well but is infinite on its walls. We can also refer to this well as a quantum box. The simplest shapes for a three-dimensional box can be that of sphere or a cube. [23]. For a spherical QD of diameter d , the Schrödinger equation is resolved by introducing spherical coordinates and by separating the equation in a radial part and in a part that contains the angular momentum [24]. The potential in this problem is best

described mathematically in spherical coordinates, as

$$V(r, \theta, \phi) = \begin{cases} 0; & r \leq L \\ \infty; & r > L \end{cases} \quad (1)$$

This describes the region in which the particle can exist as a sphere of radius L centred at the origin; outside the ball the potential is infinite. Note that this means the potential is independent of θ and ϕ . The 3D time-independent Schrödinger equation for this system is

$$-\frac{\hbar^2}{2m} \nabla^2 \Psi(r, \theta, \phi) = E \Psi(r, \theta, \phi) \quad (2)$$

where, m = mass of particle, E = energy of particle, Ψ = wave function associated with particle. Due to the spherical symmetry of the Hamiltonian the wavefunction can be written as $\Psi(r, \theta, \phi) = R(r)\Theta(\theta)\Phi(\phi)$, and then the technique of separation of variables results in two differential equations to be solved, one for R and one for Y . It turns out that the solutions for R are given by the spherical Bessel functions $J_n(r)$, and the solutions for Y are the spherical harmonics $Y_{lm}(\theta, \phi)$. The solution to the spherical well is then

$$\Psi_{nlm}(r, \theta, \phi) = J_n(r)Y_{lm}(\theta, \phi) \quad (3)$$

Since the potential depends on the radius from a fixed point of a spherical quantum dot, the Laplacian ∇^2 of the spherical polar coordinate is independent of the angular part and is given as:

$$\nabla^2 = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial}{\partial r} \right) \quad (4)$$

Putting equation (4) into equation (2) for $V = 0$ we have

$$-\frac{\hbar^2}{2\mu} \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial}{\partial r} \right) = E\Psi(r, \theta, \phi) \quad (5)$$

The radial part of equation (5) by the method of separation of variables yields:

$$\frac{1}{R} \frac{d}{dr} \left(r^2 \frac{dR}{dr} \right) + \frac{2\mu R r^2}{\hbar^2} = l(l+1) \quad (6)$$

where l = orbital angular momentum, the equation (6) is simplified as:

$$\frac{d^2}{dr^2} + \frac{2}{r} \frac{dR}{dr} + \left[k^2 - \frac{l(l+1)}{r^2} \right] R(r) = 0 \quad (7)$$

where $k = \frac{\sqrt{2\mu E}}{\hbar}$

Equation (7) is reminiscent of spherical Bessel differential equation. The solutions are the spherical Bessel function of order l , $j_l(kr)$ and the spherical Neumann function of order l , $n_l(kr)$. The general solution is:

$$R_{n,l}(r) = C_l j_l(kr) + D_l n_l(kr) \quad (8)$$

where C_l and D_l are constants. Contrary to the spherical Neumann function, the behavior of the Bessel function is such that it is finite at the origin [25]. The finite requirement of the wave function suggests that D must be equal to zero. This simplifies equation (8) to:

$$R_{n,l}(r) = C_l j_l(kr) \quad (9)$$

where, C_l = normalization constant and $R_{n,l}$ = eigen function. The wave function must vanish at the boundary. However, due to mathematical expediency, a confining potential that is central was used, which implies that there exist an explicit hard core potential at the centre. But, according to the physical description of the system, no boundary exist at $r = 0$, which would introduce a node in

where 1.8 is a coefficient due to wave function overlap between the electron and hole in the first excited state whereas the minus signifies the opposite charge polarities that exist between the electron and hole. This term can be fairly significant because the average distance between an

the wave function at that point. Rewriting equation (9) in consonance with [26] yields:

$$R_{n,l}(d) = C_l j_l(kd) \quad (10)$$

where, d = diameter of the sphere
Also, the infinite potential barrier requires that $R(d) = 0$. This evaluates

$$j_l(kd) = 0 \quad (11)$$

Where kd is a zero of the l th-order spherical Bessel function. Unfortunately, the zeros are not located at good points. The boundary condition requires that:

$$K = \frac{1}{d} X_{n,l} \quad (12)$$

Where $X_{n,l}$ is the n th zero of the l th order spherical Bessel function. Comparing equation (12) to k parameter above yields:

$$\frac{1}{d^2} X_{n,l}^2 = \frac{2\mu E}{\hbar^2} \quad (13)$$

Following equation (13), the allowed energy states E_n become

$$E_n = \frac{\hbar^2}{2\mu d^2} X_{n,l}^2 \quad (14)$$

Putting $\mu = m_e^*$ into equation (14) yields the electron confinement energy E_{en} as:

$$E_{en} = \frac{\hbar^2}{2m_e^* d^2} X_{n,l}^2 \quad (15)$$

where m_e^* is the effective mass of electron. Similarly, the hole confinement energy E_{hn} is obtained as:

$$E_{hn} = \frac{\hbar^2}{2m_h^* d^2} X_{n,l}^2 \quad (16)$$

where m_h^* is the effective mass of hole. Adding equations (15) and (16) gives the total confinement energy E_c of photo-excited carriers (electron and hole) as:

$$E_c = \frac{\hbar^2}{2d^2} \left(\frac{1}{m_e^*} + \frac{1}{m_h^*} \right) X_{n,l}^2 \quad (17)$$

where, n = radial quantum number that labels the different confined energy states of the particle in the QDs. The ground state corresponds to $n = 1$ and $l = 0$. $X_{1,0} = 3.142$ which coincides with the well-known constant π [27]. Therefore equation (17) becomes:

$$E_c = \frac{\pi^2 \hbar^2}{2R^2} \left(\frac{1}{m_e^*} + \frac{1}{m_h^*} \right) \quad (18)$$

This is confinement energy of a spherical QDs with an electron particle and a corresponding hole in QDs. Electrostatic interaction energy between electron and hole is

$$E = \frac{-1.8e^2}{4\pi\epsilon_0\epsilon_r R} \quad (19)$$

electron and a hole in a quantum dot can be small. The last energy term contribution is the bandgap energy of the bulk semiconductor $E_{g(bulk)}$ which is characteristic of the material. We can now model the size-dependent energy gap of a spherical semiconductor quantum dot as the sum of the

bandgap energy of the bulk crystal, confinement energies of electron and the hole and the bound energy of exciton given as

$$E_{g(QD_s)} = E_{g(bulk)} + E_{confinement} + E_{exciton} \quad (20)$$

Then, by inserting equations (18) and (19) into equation 20, we obtain

$$E_{g(QD_s)} = E_{g(bulk)} + \frac{\hbar^2}{8R^2} \left(\frac{1}{m_e^*} + \frac{1}{m_h^*} \right) - \frac{1.8 e^2}{4\pi\epsilon_0\epsilon_r R} \quad (21)$$

The model predicts that the band gap energy increases as size of the semiconductor QDs decreases and therefore can be engineered. The size-dependent energy gap can be a useful tool for designing materials with well-controlled optical properties. Thus tunable bandgap energies with respect to changes in dot size allows the engineering of

bandgap to desired of optical responses for technological applications.

III. MATERIALS AND METHODS

The experimental parameters we used for this study are: effective masses of electron, M_e^* and hole, M_h^* band gap of bulk semiconductors $E_{g(bulk)}$, specifically for group II-IV (CdSe, CdS and ZnS), group III-V (GaAs, InAs and InSb) and the group IV-VI (PbSe, PbS and PbTe) QDs acquired at Advance Physics Laboratory, Sheda Science and Technology Complex Abuja as shown in the Tables 1, 2 and 3. The variation of bandgap energies as a function QD size was computed using equation 21 to ascertain the impact of confinement on bandgap engineering. The input parameters used in the theoretical calculation have been listed in Tables 1, 2 and 3

Table 1: The Group II-IV QD materials parameters used for the study

Quantum dots	M_e^x	M_h^x	$E_{g(bulk)}$ at 300k
CdSe	0.13 m_0	0.45 m_0	1.74eV
ZnS	0.34 m_0	0.23 m_0	3.68eV
CdS	0.21 m_0	0.80 m_0	2.42eV

Table 2: The Group III-V QD materials experimental parameters used for the study

Quantum dots	M_e^x	M_h^x	$E_{g(bulk)}$ at 300k
InAs	0.02 m_0	0.40 m_0	0.36eV
InSb	0.02 m_0	0.40 m_0	0.17eV
GaAs	0.06 m_0	0.51 m_0	1.42eV

Table 3: The Group IV-VI QD materials experimental parameters used for the study

Quantum dots	M_e^x	M_h^x	$E_{g(bulk)}$ at 300k
PbSe	0.05 m_0	0.04 m_0	0.27eV
PbS	0.25 m_0	0.25 m_0	0.37eV
PbTe	0.17 m_0	0.20 m_0	0.32eV

IV. RESULTS AND DISCUSSION

The bandgap energies as a function of dot sizes for group II-IV (CdSe, CdS and ZnS), group III-V (GaAs, InAs and InSb) and the group IV-VI (PbSe, PbS and PbTe) QDs using the equation 21 are plotted in Figures 3, 4 and 5 respectively. The observed spectra generally indicate substantial increase in bandgap energy and hence the separation between the valence and conduction bands and the band levels become quantized signifying widely spaced energy levels with decreasing crystal size as shown in the Figure 1

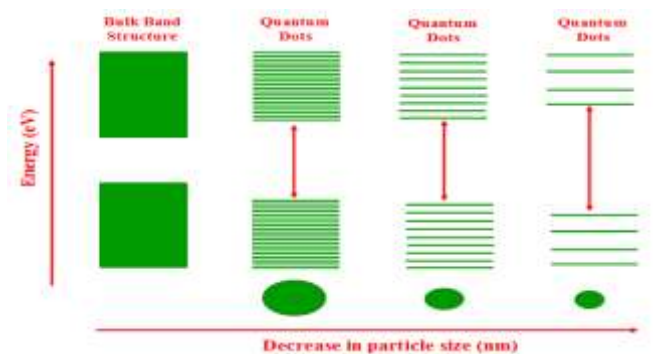


Figure 1: Energy Levels of QDs with Decreasing Size

This results in significant increase in energy of band-to-band excitation peaks and hence a blue shift in the absorption and luminescence bandgap energies as shown in Figure 2 compared with the parental bulks materials that beset with composition-dependent bandgap energy. These optical characteristics suggest that more excitation energy is needed to move electron from valence band to conduction and also more energy will be released during recombination. In this way the electrical properties, optical absorbance and transition energies of the QDs are modulated via size modification.

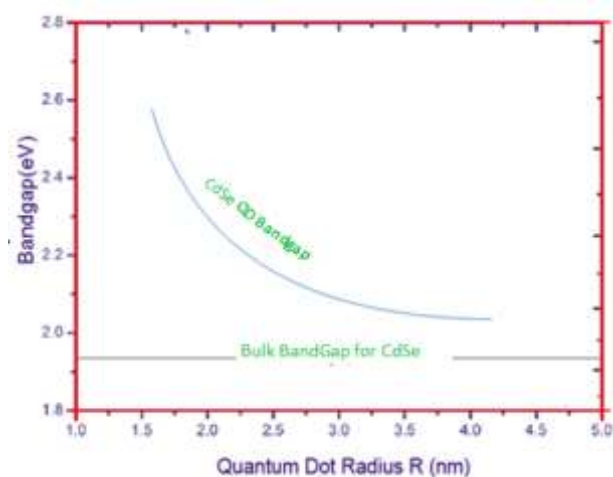


Figure 2: Showing Optical Blue Shift for CdSeQD

The tunable bandgap energies with respect to changes in particle size results in multiple applications. This quantum potential effect on bandgap has significant impact in enhancing performance of photonic devices. A QD that was transparent in infrared region, on increasing band gap may become transparent in visible region. Thus, multi-wavelength photo-detectors would be enabled by semiconductors QDs with such larger range of bandgapstunability. However, this effect demonstrates the consequences of confining the electrons and electron-hole pair (or the excitons) within a dimension which approaches the critical quantum limit, often termed as the Bohr exciton radius.

It is found that confinement of charge carriers in QDs results in discretization of energy levels and as a result electron transfer can only occur between these discrete electronic states. Consequently, only photon energies of certain dimension can be absorbed leading to monochromatic emissions. Thus QDs can uniquely emit size-dependent single-color fluorescence with high efficiency. In particular, QDs display broad absorption characteristics with narrow-emission spectra that are continuous and tunable due to quantum size effects. The narrow emission and broad absorption bands spectra of the QDs allow single wavelength excitation of emission from different-sized QDs. These fluorescent properties of QDs offer number of advantages that will help to advance biotechnologies in probing several markers at the same time particularly in biosensing/optical imaging and open up

several multiplexing potentials such as multicolor detection with a single a single wavelength excitation source.

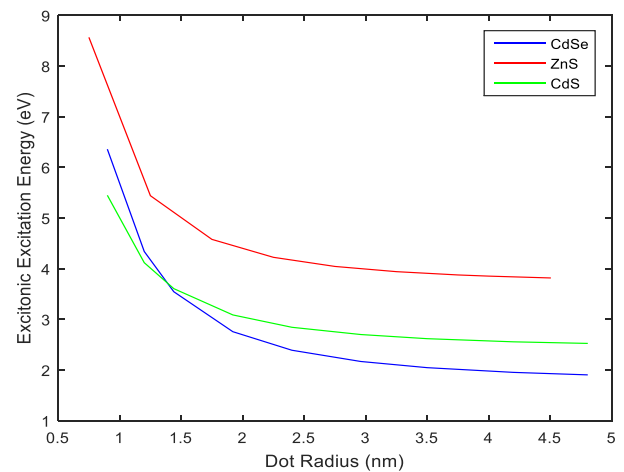


Figure 3: Excitonic Excitation Energy versus Dot Radius for CdSe, ZnS and CdSQDs

The tunable bandgap energy allows QDs to be optimally engineered for the different applications particularly in photonics and optoelectronics by providing active material for devices. It is apparent from Figure 2 that CdSe and CdSQDsposses an optical spectrum that span nearly the entire visible spectrum suitable in solid-state lighting in which illumination or multicolour display is desired. This makes them potential active materials for efficient light emitting diode LED and lasers operating over the whole of visible spectrum. Among group II-IV QD materials considered ZnS possesses the widest bandgap energy and hence the largest tunable spectra range which plays a vital role for absorption and emission of high energy ultraviolet (UV) blue spectrum and permits devices to operate at much higher voltages and temperatures crucial for applications in optoelectronic devices such as Pn junctions and power transistors. In addition, the wide bandgap absorption spectra will be relevant in high optical transmittance specifically in the range of visible to infra red (IR) spectral region.

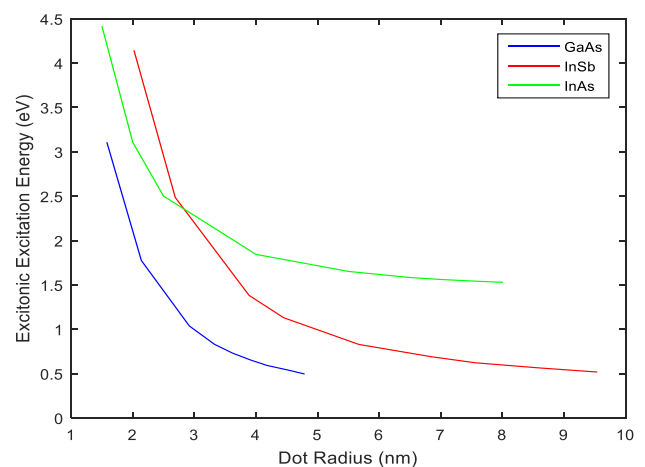


Figure 4: Excitonic Excitation Energy versus Dot Radius for GaAs, InSb and InAs QDs

GaAs, InAs and InSb QDs show promising optical trait in the visible to near infrared (NIR) spectral region which is desirable for optoelectronic applications, including solar cell, dye imaging and light emitting diodes operating at NIR wavelengths.

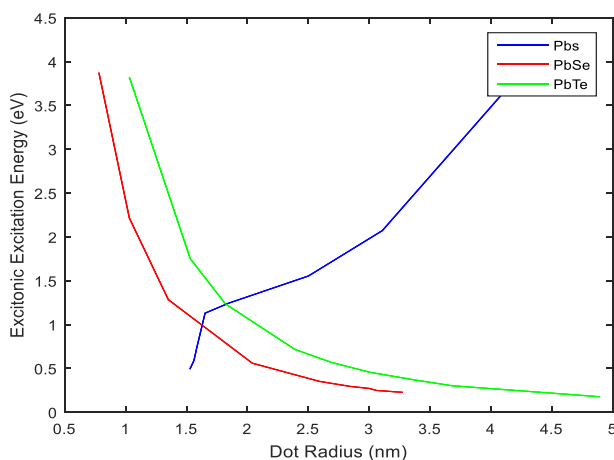


Figure 5: Excitonic Excitation Energy versus Dot Radius for PbS, PbSe and PbSeQDs

We found that PbS, PbSe and PbSe QDs display absolutely exceptional optical characteristics that can be tuned in broad range of UV to IR spectral regions. Solar cell materials selection is largely chosen on the basis of how well their absorption wavelengths match the solar spectrum. This makes PbS, PbSe and PbTe QDs suitable materials for solar cell applications owing to the fact that their absorption characteristics are a fairly good match to the solar spectrum.

V. CONCLUSION

This work described the successful bandgap engineering in QDs. A simple model is formulated to engineer the band gap energy of semiconductor QDs in different sizes. It is found that the band gap energy of QDs is inversely depends upon the dot size and the band levels become quantized signifying widely spaced energy levels. This produces enhanced optical and electrical properties than that of the bulk materials, and provides opportunities to substantially expand the range of achievable bandgap energies for growing new ranges of materials. One of the most important consequences of the spatial confinement effect observed is an increase in energy of the band-to-band excitation peaks (blue shift) with decreasing dot size in relation with the Bohr radius. Consequently, the electrical properties, optical absorbance and transition energies of the QDs are modulated via size modification. The size-dependent absorption and emission spectra with non-zero discrete electronic transition energies in QDs have potential technological applications in optoelectronic devices, including light emitting diodes, solar cells, displays, biological labeling and quantum computing etc.

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