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Quantum Dot Research: Current State and Future Prospects

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Abstract
The general objective of this review is to assess the current state of research involving semiconductor quantum dots, to provide brief review of the theory behind their unique properties, and to introduce the explanation and the importance of quantum dot research. The characteristic shifting of the band gap energy with quantum dot size, as predicted from the density of states for low-dimensional structures, allows experimental measurements to determine the extent to which quantum confinement effects play a role in the resulting properties. A few of the current techniques used to measure the presence and physical characteristics of quantum dots and their energy levels is reviewed, including transmission electron microscopy, optical transmission, and Raman and photoluminescence spectroscopy. Finally, some of the more exciting applications for quantum dots currently being researched for use in the field of optoelectronics are reviewed, including quantum dot infrared photo detectors, quantum dot lasers, and quantum dot solar cells. Comments are made on the current progress and the future prospects of quantum dot research and device applications.
Keywords: Quantum dot, confinement effect, Spectroscopy, Photovoltaic device

1. INTRODUCTION

The term “quantum dots” (QDs) refers to crystal structures with extremely small dimensions. Due to its small size, the electronic states begin to resemble and its electronic characteristics and optical properties are changed [1]. Compared to bulk (three-dimensional) materials and quantum well (two-dimensional) structures, QDs are zero-dimensional systems and their electronic states are fully quantized, which are similar to a single atom or atomic system. This fully quantized system is stable against any thermal perturbation [2].

Low-dimensional nanometer-sized systems have defined a new research area in condensed matter within the last 20 years. Modern semiconductor processing techniques allowed the artificial creation of quantum confinement of only a few electrons. Such finite fermions systems have much in common with atoms, yet they are man-made structures, designed and fabricated in the laboratory. Usually they are called ‘‘quantum dots,’’ referring to their quantum confinement in all three spatial dimensions[3].

The trend in device-oriented manufacturing toward smaller and smaller dimensions has helped fuel the need for nanofabrication techniques and for understanding the nanometer scale materials. Many of the basic concepts of material behavior and properties are derived from studies in the bulk. However, research has revealed exciting and technologically important insights into science and engineering at the nanometer scale.

As discussed in a review of nanostructure materials by P. Moriarty, our ability to understand and manipulate materials of such small dimensions has been facilitated by advances in surface and subsurface imaging techniques [4]. Such techniques, including the scanning tunneling microscope (STM), atomic force microscope (AFM), near-field scanning optical microscope (NSOM), and the scanning transmission electron microscope (STEM), have allowed imaging of nanostructure materials by providing resolutions down to a few angstroms. In addition, these techniques also provide a way to fabricate nanostructures by manipulating individual atoms and Molecules.

Many techniques exist for the fabrication of nanometer scale materials like top-down approach, using processes such as etching and photolithography to create patterned nanostructures., including molecular beam epitaxy (MBE), pulsed laser deposition (PLD), sputtering and wet chemical synthesis[5]. However, more promising results have been obtained in the bottom-up approach, where nanoscale structures are built by the manipulation of individual atoms and by controlled nucleation and growth processes. Perhaps the most widely used technique currently in use is the strain-induced self-organized growth of nanoscale quantum dot structures using MBE. However, many fabrication techniques are still being studied in the hope of finding a processing method that can consistently and predictably create nanoscale structures with a high degree of control over size, such as the ability to form nano sized particles, shape, and spatial distribution.

1.1. Low-Dimensional Structure Science

Nanoscale science, as the name implies, is concerned with the study of materials with a dimension of roughly 1 to 100 nm. Such structures belong to a group known as low-dimensional structures (2D-0D). The dimensionality refers to the number of directions in which the carriers of the material act as free carriers. For example, a 3D structure is one in which the electronic carriers are free in all three directions. This is the case for a bulk material. Now imagine that a completely free bulk material is spatially confined in one direction. What results is a thin film,

whose carriers may now be confined in one of the three directions, creating the 2D structure, also known as a quantum well. Taking this spatial confinement process to the next step and further confining the thin film, one is left with a 1D system, or the quantum wire. Finally, when a material is spatially confined in all three directions, a 0D structure is formed that is, the quantum dot. This progression for low-D structures is represented in Figure 1.

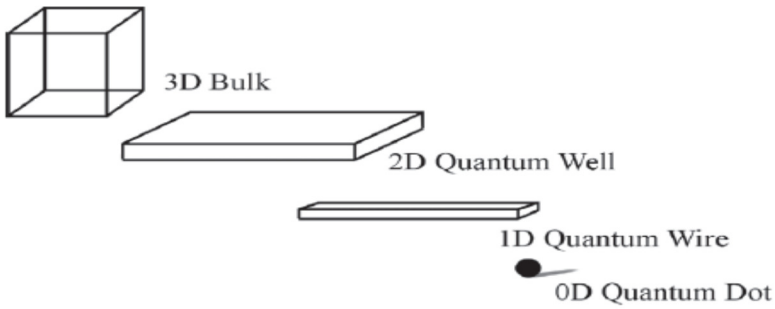

Figure1. Low-dimensional structures [6].

The significance of this spatial confinement of the electronic carriers can be seen in a plot of the density of states for each low-dimensional structure. Depicted in Figure 2 is the change in the density of states that occurs as a material evolves from a completely unconfined state to the three-dimensionally confined state. Interesting changes occur in the allowed carrier energies as a result of the confinement process. In the bulk, the carriers can exist in nearly continuous bands. This is clearly not the case for the 0D structure where the material has been spatially confined in all three directions forming a small dot of material. Instead of a band of allowable energies, the carriers within a 0D structure are restricted to a specific set of completely quantized energy states. This complete quantization of energy states makes the quantum dot a very unique structure for research.

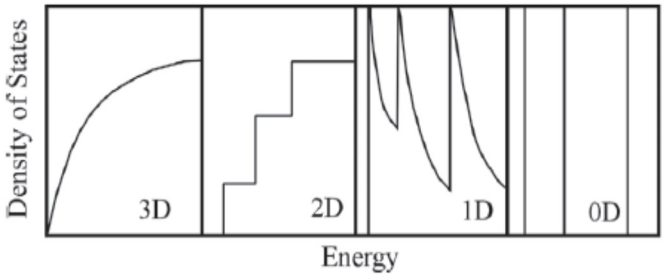

Figure2. Density of states vs. energy for a bulk material (3D), quantum well (2D), quantum Wire (1D), and quantum dot (0D) [7].

1.1.1. The Exciton

During optical excitation of carriers in a semiconductor, the minimum energy required to form free carriers is the band gap energy. Energy below that value cannot excite free carriers. However, low-temperature absorption studies of semiconductors have shown excitation just below the band gap [8]. This excitation is associated with the formation of an electron and hole bound to each other, otherwise called an exciton. Because the electron and hole remain bound to each other, the formation energy required is lower than the band gap energy. The stable structure formed consists of an electron orbiting a local potential minimum associated with a hole. In the case of spatially confined systems, such as the quantum dot, excitons are the carriers that create the resulting quantized energy states.

The structure of an electron bound to a hole is very reminiscent of the hydrogen atom. In the hydrogen atom case, a single electron is bound to a single proton by a Coulombic attractive force. The behavior of the exciton is very similar. In fact, as the electron orbits the hole, a set of hydrogen-like states is created. Just as an electron orbiting a nucleus has a characteristic dimension, called the Bohr radius, so too does the electron orbiting a hole in a quantum dot exciton. This characteristic dimension is called the exciton Bohr diameter, a_x, and essentially is a measure of the diameter (or radius) of the exciton. The exciton Bohr diameter is a critical parameter that provides a basis on which to judge the criteria for size confinement in materials. Table1 lists several different semiconductors and their corresponding exciton Bohr diameters [9].

Table1. Exciton Bohr diameters and band gap energies for various semiconductors

Semiconductor	Exciton Bohr Diameter	Band gap Energy
CuCl	13Å	3.4 eV
ZnSe	84Å	2.58 eV
CdS	56Å	2.53 eV
CdSe	106Å	1.74 eV
CdTe	150Å	1.50 eV
GaAs	280Å	1.43 eV
Si	37Å(longitudinal) 90Å(transverse)	1.11 eV
Ge	50Å(longitudinal) 200Å(transverse)	0.67 eV
PbS	400Å	0.41 eV

1.1.2. Confinement Regimes

The exciton Bohr diameter is a useful parameter for evaluating the conditions for creating quantum confinement effects in a specific material. In general, confinement effects must be taken into account as the material dimension is reduced to a size approaching the exciton Bohr diameter. There are different regimes of confinement, strong and weak that gives different resulting energy state equations [8]. These strong and weak states are determined by the degree of coupling between the electron and hole in the exciton. Figure 3 helps to illustrate the process that occurs as a free exciton is confined to a coupled state. As the degree of confinement is increased in a smaller dot, the exciton can no longer exist and one reverts to the free electron and hole states. This is the strong confinement case.

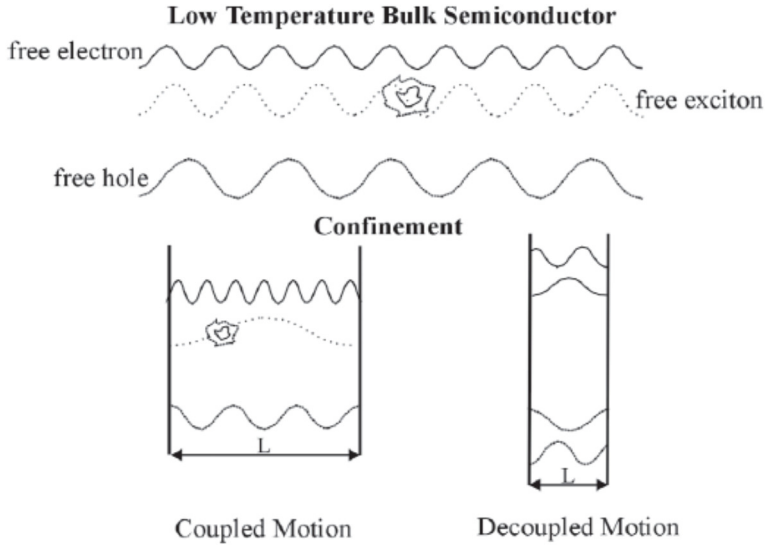

Figure3. Schematic band diagrams for a free exciton, confined, coupled exciton, and a decoupled Exciton [10].

When an exciton is created in a semiconductor quantum dot and the size of the crystal is roughly 3 to 10 times the exciton Bohr radius of the material, then the exciton is said to be in a weakly confined state. This means that the Coulomb interaction energy is on the order of the electron and hole sublevel separations and so must be considered.

2. OBJECTIVES

2.1. General Objective

- To assess the current state and future prospects of quantum dot research.

2.2. Specific Objective

- To provide brief review of the quantum dot theory.
- To introduce the explanation and the importance of quantum dot research.
- To explain the importance and application of quantum dot in relation to quantum well.

3. EXPERIMENTALLY OBSERVING QUANTUM CONFINEMENT EFFECTS

Quantum confinement effects give rise to changes in the behavior of a bulk material. This is a direct result of the change in the density of states for low-dimensional structures. To establish the existence of quantum dots within

a sample and to measure the resulting changes in material properties due to quantum confinement effects, several techniques may be used. Reviewed are four such techniques transmission electron microscopy, absorption, Raman, and photoluminescence. These specific measurements do not encompass all techniques used to characterize low-dimensional structures, but are the most commonly used. This section describes these techniques along with supporting published data.

3.1. Transmission Electron Microscopy

Perhaps the most obvious evidence to support the existence of quantum dots is to create a visual image or micrograph. A common technique for creating images at the nanometer scale is the transmission electron microscope (TEM). The TEM uses electrons instead of photons, but operates in a similar way to an optical microscope, with condensing and objective lenses. TEM configurations have been developed, including the high resolution TEM (HRTEM) and the scanning TEM (STEM) [6] Figure 4 presents a cross-sectional STEM image of a sample consisting of germanium, Ge, quantum dots in an indium tin oxide (ITO) matrix. The quantum dot film was deposited using a dual gun RF magnetron sputtering process to create a multilayered structure of alternating Ge and ITO layers [10]. Through post-deposition anneals, Ge quantum dots form within the Ge layer, as seen in the figure. The lighter areas represent the ITO matrix and its polycrystalline nature can be seen, while the darker regions represent the Ge quantum dots.

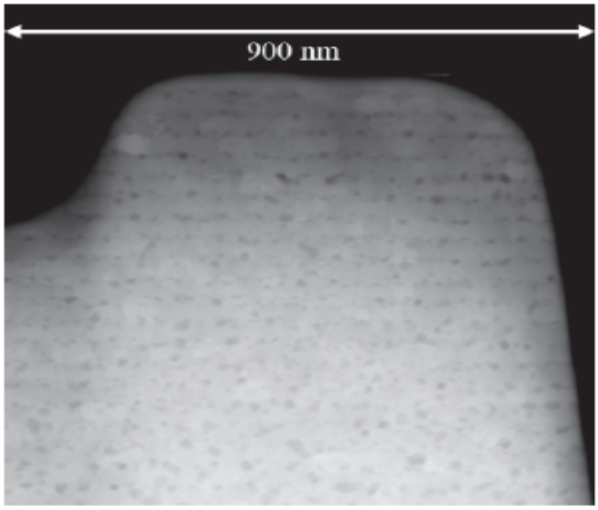

Figure4. STEM image of Ge quantum dots in an ITO matrix [10].

3.2. Optical Absorption

Measuring the absorption spectrum of semiconductor materials offers a simple way to investigate their band structure. In this way, a direct measurement of the band gap energy, Eg, of the particular film can be determined. By comparing the calculated band gap energy from the measured absorbance data for the quantum dot case with that for the bulk some insight into the ways that quantum confinement effects play a role can be obtained. Recall that in dimensionally confined systems there is a predicted blue-shift in the band gap energy with decreasing confinement length. Hence, if quantum confinement effects are present in a sample, the measured band edge should be shifted toward shorter wavelengths. This shift of band gap energy with dot size has been established experimentally. Figure 5 presents absorbance data for various sized Ge quantum dots in silica, SiO_2, matrix [11]. There is an obvious blue-shift in band edge as the confinement dimension is reduced, consistent with quantum confinement theory. In fact, Ge has one of the largest shifts reported among other semiconductors studied.

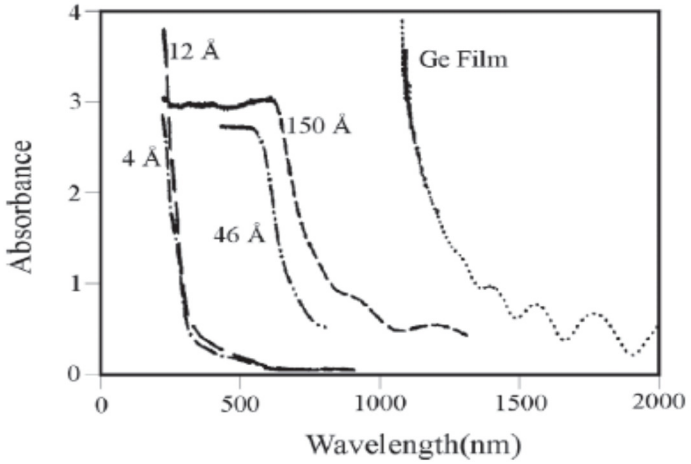

Figure5. Absorbance vs. wavelength for a Ge film and for various Ge quantum dots 150 Å, 46 Å, 12 Å, and 4 Å in diameter [11].

Figure 6 illustrates this point by comparing published data for several semiconductors. It can be seen that, over similar size ranges (from bulk down to roughly 15 nm in diameter), Ge has a much larger shift in energy with quantum dot size [12].

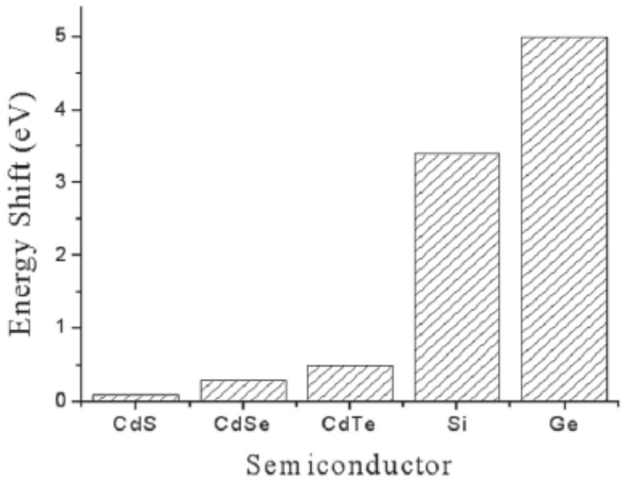

Figure 6. Relative degree of band gap energy shifts due to quantum confinement for several direct and indirect semiconductors [10].

3.3. Raman Vibrational Spectroscopy

Raman spectroscopy takes advantage of the Raman Effect, so named after the man who dis covered it in 1928[13]. Simply stated, the Raman Effect is the inelastic scattering of light by matter. When a photon of visible light too low in energy to excite an electronic transition interacts with a molecule, it can be scattered in one of three ways. It can be elastically scattered, retaining its incident energy (also known as Rayleigh scattering), or it can be inelastically scattered by either giving energy to the molecule (Stokes scattering) or taking energy away from the molecule (anti-Stokes scattering) [14].

The Raman technique is also a good tool for monitoring the crystallinity of a material. For example, in going from an amorphous to a fully crystalline sample of the same material, the Raman peak can be seen to evolve from a broad peak to a strong, narrow crystalline peak and combinations of the two, depending on the degree of crystallinity. However, quantum confined materials can also cause a once strong crystalline peak to broaden [15]. The presence of small spherical particles of a material introduces another phenomenon observed in Raman spectra. Small particles, such as quantum dots, can actually shift the Raman line toward the low frequency end, that is, toward the laser line. This has been seen experimentally in various semiconductor quantum dot materials, including silicon and germanium [11, 16].

Many researchers have reported Raman spectra for quantum dots [5, 15, 17].For example, Stella et al. reported in 1998 Raman data for Ge quantum dots deposited onto an alumina film using an Evaporation-condensation technique [17] Their Raman results as a function of Ge dot size, d, are shown in Figure 7. As the Ge dot size decreases, the Raman peak is seen to broaden, soften, and the central peak position shifts toward smaller frequencies. All of these observed effects are characteristic of quantum confinement.

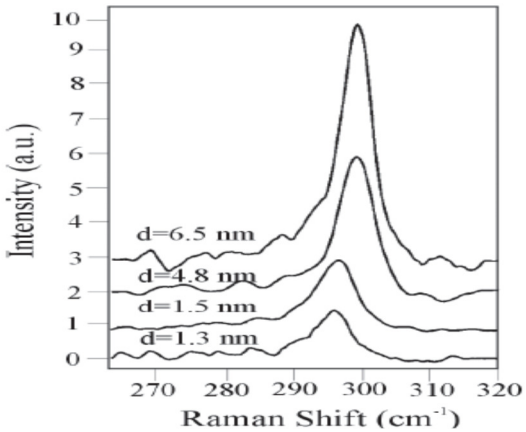

Figure7. Raman spectra for films of various Ge quantum dot diameters, d [17].

The fact that nanoparticles have little material to interact with means that the vibrational strength of the molecules with the incident radiation is diminished somewhat. As a result, less energy is given to the molecule during excitation producing in a small shift in the bulk crystalline Raman line toward the low frequency end. So, Raman spectroscopy can be used as a tool for establishing the presence of quantum dots within a film. Raman can be a useful tool in experimentally supporting the presence of quantum confinement effects in quantum dot films. However, care must be taken for films that contain stresses.

3.4. Photoluminescence Spectroscopy

A very common way to experimentally probe the energy levels in a quantum dot is through photoluminescence measurements. In photoluminescence studies, a laser, tuned to an appropriate wavelength, is made incident onto the quantum dot sample. Excitons or free carriers are created within the quantum dot, which recombine radiatively to create photons. What results is a plot of the intensity of the signal measured from the radiative recombination as a function of the wavelength being detected. This allows a direct measurement of the different energy states present in the quantum dots [7]. When studying quantum confinement effects of semiconductor quantum dots, photoluminescence measurements are commonly used to help determine the energy states present in the fabricated structures, as evidenced by the literature [18-20].

Figure 8 presents PL data on quantum dots as a function of dot size [21]. In this case, spectrum (a) represents the quantum well from which the quantum dots were made. The most obvious trend is the shift of the main peak toward shorter wavelengths with decreasing dot size, as predicted from quantum confinement theory. What is also evident is the emergence of small, sharp peaks as the dot size becomes very small (in this case 80 nm in diameter). The presence of such peaks is characteristic of systems having a completely quantized energy spectrum. However, instead of a few strong and clearly separate peaks at energies corresponding to the discrete energy levels, many narrowly spaced peaks are observed. The reason for this is likely the inhomogeneity of sizes of the different quantum dots within the sample.

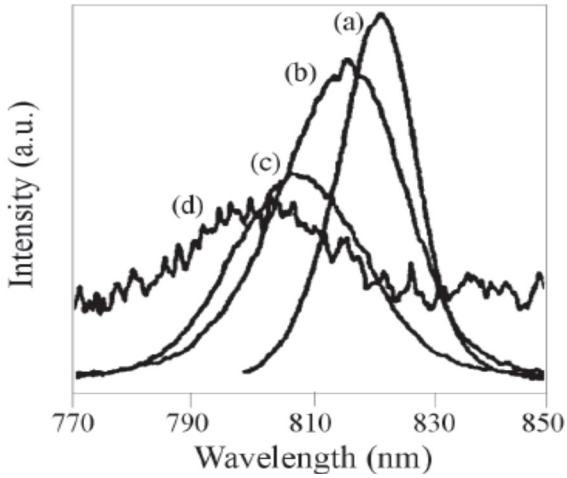

Figure8. Photoluminescence spectra of quantum dots with diameters (b) d=140 nm, (c) d=110 nm, (d) d=80 nm, with spectrum (a) being that collected for the quantum well from which the dots were grown [21].

Fortunately, advances in fabrication processes for creating quantum dots have been made over the last decade in order to produce dots with a relatively narrow size distribution. Having a strong, narrow PL peak for quantum dot samples can be very important with regard to specific performance in applications such as quantum dot lasers,

which is discussed in more detail in the following section.

4. QUANTUM DOT DEVICE APPLICATIONS

The study of semiconductor quantum dot systems has revealed very interesting changes in the behavior of 0-D structures. Many ideas have been put forth in an effort to utilize the unique characteristics of quantum dots for specific applications. In many instances, the incorporation of quantum wells into device design has already helped advance the performances of many important applications. By replacing or adding quantum dots to such devices, it is expected that performances will excel far beyond the current state. There are several areas in which quantum dots are being considered and investigated. Among these areas of study are chemical gas sensors, nonlinear optics, and optoelectronics [22-25].Outlined in this section are the three applications involving optoelectronics for which the authors deem to be the most promising the quantum dot infrared Photodetectors, the quantum dot laser, and the quantum dot solar cell.

4.1. Quantum Dot Infrared Photodetectors

Quantum well structures, i.e., 2-D structures, have been widely studied for devices such as pollution monitors and thermographers, primarily for military and medical applications. The material currently used in such devices is HgCdTe. These quantum well infrared photo detector

(QWIP) sensors typically operate in the wavelength range of 3 to 14 μm. The reason for this is that there is little atmospheric absorption from 3 to 5 μm and 8 to 14 μm. However, most objects do produce irradiance within this range, the so called black body spectral emission. The problem with the current QWIP devices is that their efficiency is reduced under conditions of normal incidence. This is due to weakened absorption within the semiconducting well layer in that direction.

It has been predicted that quantum dot infrared photo detectors (QDIPs) would have advantages over QWIPs, including reduced dark current and higher electric gain. Quantum dots, being

0-D structures, have confinement of the carriers in all three directions, which can allow absorption of the normally incident IR light. This can enhance the efficiency of the device by increasing the responsivity. In addition, it is expected that the QDIPs can produce enhanced photo excited carrier lifetimes, which could also lead to higher detector responsivity as well as higher operating temperatures.

4.2. Quantum Dot Lasers

The concept of utilizing quantum dots for improved performance in solid state laser devices has been an intriguing and much researched area during the last decade. The density of states in a completely confined system, such at the quantum dot, is made up of discrete, quantized energy levels. This provides the opportunity for a laser with a single, narrow excitation mode. The ability of quantum dot materials to vary their band gap energy with size makes the lasing wavelength tailorable. In addition, it has been realized that quantum dot (QD) lasers have the potential for very low threshold current densities. This idea was first observed in 1994 when a QD laser consisting of self-organized quantum dots produced a threshold current density of 120 A/cm^2 at 77 K. This was a dramatic improvement over quantum well laser devices whose best threshold current density was as high as7.6 kA/cm^2 at 77 K. This initial report sparked great interest among researchers and since that time many advances, in addition to the low threshold current densities discovered, have been made, including above room temperature operation, high temperature stability, high power operation and an extended wavelength range.

Currently, the most reported advances in quantum dot laser diode performance are in threshold current density. The experimental device design is being engineered to further reduce its threshold value. The incorporation of quantum dot structures into the laser diodes, instead of quantum wells, has allowed advances in this area. Figure 9 illustrates the trends observed in the measured threshold current densities for semiconductor laser diodes over the last 30 years [22].Comparing the threshold current density measured for a double heterostructure (DH) laser diode in 1969 to that measured for a quantum dot laser diode in 1999, it is clear that the semiconductor quantum dot structures dominate in this category.

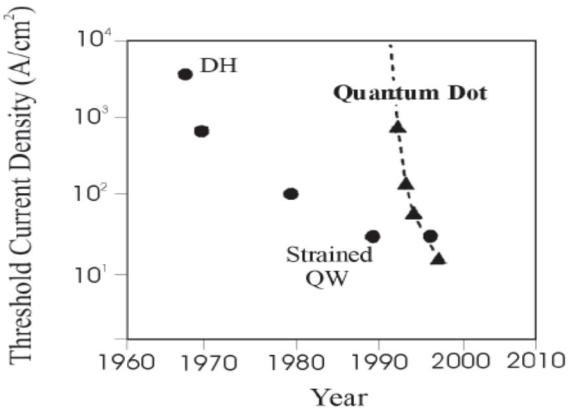

Figure9. Historic development of threshold current density for semiconductor laser diodes, comparing the trends for devices containing quantum wells (QW) and quantum dots [22].

The most studied materials system for quantum dot laser applications is the InGaAs/AlGaAs on a GaAs (001) substrate. The substrate material and orientation has been found to play a role in the resulting performance of the lasers. Substrate effects are still under study as they will likely be a factor in the further lowering of threshold current densities. The quantum dot material itself affects the emission range of the resulting lasers. For the InGaAs based QD lasers, emission wavelengths from ~900 nm in the IR to beyond 1000 nm have been achieved. By varying the dot composition this range can be expanded.

Currently, the best lasing behavior has been observed in devices grown using the Stranski- Krastanow technique to create self-organized quantum dots, typically done with MBE process. The underlying principle of such a fabrication technique is the formation of islands during epitaxy due to the lattice mismatch in the quantum dot and substrate materials. This process is preferred because it has the ability to produce high quantum dot densities, a narrow dot size distribution and a low density of defects.

Quantum dots have facilitated large advances in the performance of semiconductor lasers. The pursuit of lower threshold values and increased power output is continually being challenged as researchers push the envelope of understanding further. For example, a very recent publication by Bimberg has reported the most impressive quantum dot laser performance to date [26].

It is clear that advances have been made in the performance of quantum dot lasers. However, many materials and device design issues have still to be explored before QD lasers meet their potential. In particular for quantum dot lasers, the problems have been in attaining the theoretical expectations of such devices. In the ideal case, the quantum dots are sized, shaped, and distributed as desired to attain the best theoretical performance. However, more realistic models, such as finite barriers and non equilibrium carrier distribution, have led to the understanding that the size, shape, and number of electron and hole levels can greatly affect the resulting threshold current densities measured. The area of research for quantum dot laser systems is vast, and there is much left to discover as fabrication methods are improved and hence material quality, and device structures are optimized.

4.3. Photovoltaic Devices

Ever, since the inception of the solar cell in 1954 by Chapin, Fuller, and Pearson, much research has been conducted in search of improved efficiency and performance [27]. In addition to studying many different semiconductor materials and structures, including single-crystal, polycrystal, and amorphous, researchers have become very clever in device configuration in an attempt to yield high conversion efficiency [28]. Despite all the research and advances made, the Si p-n homojunction has remained as the solar cell material for applications.
Perhaps the single most important issue in solar cell performance is ideal conversion efficiency.

The ideal conversion efficiency is defined as the ratio of the maximum power output to the input power. So the more incident light i.e. absorbed and turned into carriers, the better the conversion efficiency. However, basic materials properties dictate the absorption characteristics of a material. The conventional p-n junction solar cell has only a single band gap, $\boldsymbol{E_g}$. Once the cell is exposed to the solar spectrum, any photon with energy less than Eg will not be absorbed by the device and thus contributes nothing to the cell output. This is known as spectral mismatch, in which light composed of a spectrum of energies has only one band gap for absorption. The device is a low pass filter, absorbing light of greater energy than the band gap. However, the excess energy is lost as heat and only the energy equal to $\boldsymbol{E_g}$ will contribute.

The application of quantum dots or low-dimensional structures to the area of photovoltaic has been previously theorized [27]. Although the experimental evidence to support such claims is still in its infancy; potential device designs have been suggested. Das and colleagues are currently developing photovoltaic cells using an electrochemical fabrication technique [29].

In order for quantum dots to successfully be incorporated into working photovoltaic devices, much research will need to be done. Of primary importance is the development of fabrication techniques and conditions that can consistently provide control over the quantum dot size distribution and periodicity. In addition, ideal materials systems need to be developed, including the best semiconductor material for the quantum dot nanostructures, as well as investigating which material would best serve as a matrix for a p-n junction with the dots. Despite the clear issues that will need to be overcome to make the quantum dot solar cell a viable device, quantum dots have the potential to be the next leap forward in advancing the conversion efficiency of solar cells.

5. CONCLUSIONS AND FUTURE PROSPECTS

Over the last decade nanoscience has been a very popular topic of research. This review has provided a glimpse into the science behind the semiconductor quantum dot, including a few of the ways it can be experimentally observed and some of the more exciting areas for which it is being considered for application.

The ability of the quantum dot to vary its band gap energy with size, and the quantized energy states that result, are what give such nanoscale device structures their potential. Much of the published data in this field reflects this fact; however, nanoscale research is still a relatively new topic and, as such, has much to overcome. Materials issues, fabrication control, and device design are all issues in need of further investigation and understanding. From this effort advances have already been made in the control of quantum dot fabrication and properties and have led to collaborations between many different scientific disciplines. This merging of ideas has opened up many new areas for possible quantum dot device application.

The quantum dot has already proved a unique and useful structure for both the understanding at nanoscale and for improving device design and performance. As research continues, our ability to control the properties of the quantum dot will allow advances in science and industry. Finally quantum dot application will some day play a revolutionary role in advancing technologies such as microelectronics and as cross-disciplinary research continues the quantum dot will likely shape the ideas of future device applications.

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