HYDROSTATIC PRESSURE AND TEMPERATURE EFFECT ON THE CONFINED EXCITON BINDING ENERGY INSIDE A QUANTUM DOT

THESIS

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BY

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Abstract

The physical confinement of electrons and holes is a key feature of several last-generation quantum electronic and opto-electronic devices. Although the confinement is never complete, increasing the confinement improves the characteristics of these devices. Our thesis focused on studying the effect of hydrostatic pressure and temperature on the confined particles (electron, hole and exciton) inside the quantum dot (QD). In chapter one, we have presented brief introduction to nanotechnology, quantum dot in addition fabrication techniques of quantum dot, quantum confinement and exciton in quantum dot. In the following chapters we solved Schrödinger equation variationally with the effective mass approximation for an exciton confined inside cylindrical QD manufactured from GaAs and embedded in $Al_xGa_{1-x}As$ as barrier material. We choose a trial wave function with one variational parameter. We calculated the pressure and temperature effect on the lowest, ground-state energies and the binding energy of the exciton. . In the present work the confinement potential is finite and the exciton is confined in the three dimensions. It has bee found that the lowest and the ground-state energies are decreased by the pressure and increased by temperature effect on the QD. It has been shown that the exciton binding energy due to the presence of the hydrostatic pressure increases and the exciton becomes more stable. The exciton binding energy is significantly reduced with increasing temperature. Moreover the exciton is stable even at room temperature.

1.1-Nanotechnology

Nanotechnology involves the investigation and design of materials or devices at atomic and molecular level. The prefix nano means one-billion of something like a second or a meter. Generally nanoscience and nanotechnology refer to the world as it works on the nanometer scale, say, from one nanometer to several hundred nanometers. That's the natural spatial context for molecules and their interactions, just as a 100 yard gridiron is the relevant spatial context for football games. Naturally-occurring molecular players on the nanoscale field range from tiny three-atom water molecules to much larger protein molecules like oxygen-carrying hemoglobin with thousands of atoms to gigantic DNA molecules with millions of atoms. Whenever scientists and engineers push their understanding and control over matter to finer scales, as they now are doing on the nanoscale, they invariably discover qualitatively new phenomena and invent qualitatively new technologies [1- 3].

The down of the journey into the Nano world can be traced back to 1959, when Caltech physicist Richard Feynman painted a vision of the future of science. In a talk titled "There's plenty of room at Bottom", Feynman hypothesized that atoms and molecules could be manipulated like building blocks [4]. The first "Prof-of-principle" that atoms could be precisely positioned by a manmade tool took place in 1989 by scientists at IBM [5].

Fabrication of nanomaterials falls into one of two categories: top-down or bottom-up. The top-down method involves carving nanomaterials out of bulk materials. Approaches in this category are referred to as different forms of lithography. Lithography can be understood through the concepts of writing and replication [6]. Writing involves designing a pattern on a negative (usually a mask), and replication involves transferring the pattern on the negative to a functional material.

The bottom-up approach describes techniques for coaxing atoms and molecules to form Nano materials. One bottom up technique is chemical self-assembly. Different atoms, molecules, or nanomaterials are mixed together and, because of their unique geometries and electronic structures, spontaneously organize into stable, well-defined structures. Different categorization schemes have been used to describe the building blocks of nanotechnology. For example, some scientists categorize the building blocks into “soft” and “hard” categories. Two different, popular ways of classifying nanomaterials are described here. Nanomaterials are often classified in the literature based on dimensionality. Crucial to this classification is the concept of confinement, which may be roughly interpreted as a restriction in the ability of electrons to move in one or more spatial dimensions. 0-D nanomaterials, such as quantum dots and metal nanoparticles, are confined in all three dimensions. 1-D nanomaterials are confined in two directions and extended in only one: electrons flow almost exclusively along this extended dimension. Examples of one-dimensional nanomaterials are nanotubes and nanowires. Finally, 2-D nanomaterials, which are confined in one dimension and extended in two, include thin films, surfaces, and interfaces. Interestingly, material structures currently used as elementary semiconductor devices fall under this category. Nanomaterials can also be divided into inorganic and organic classes.

Inorganic nanomaterials describe nanostructures in which carbon is not present and combined with some other element. Examples of inorganic nanomaterials are fullerenes, carbon nanotubes, nanowires, and semiconductor nanocrystals, and nanoparticles [7-10]. Organic nanomaterials are compounds containing the element carbon. Recent advances enable researchers to create organic nanomaterials with specific atoms, geometries, and electronic arrangements [11-15].

1.2- Quantum dot

Nanoscale materials such as semiconductor quantum dots (SQDs) have electronic and optical properties between those of much large macro and micro scale bulk semiconductor crystals, and much smaller atoms and molecules. The investigation of these materials is often envisaged as the cusp of a new field-nanoscience-where heretofore unexplored interactions are possible between practitioners and components of chemistry, physics, biology, and various engineering disciplines [16].

Semiconductor quantum dots (QDs) are nanometer-sized three-dimensional structures which confine electrons and holes in all three directions giving rise to a discrete energy spectrum [17, 18]. Many types of QDs have already been developed such as electro statically and lithographically defined QDs, chemically synthesized QDs, naturally formed QDs by interface fluctuations, and epitaxially grown self-assembled QDs. Quantum dots containing electrons can be compared to atoms: both have a discrete energy spectrum and bind a small number of electrons. In contrast to atoms, the confined electrons do not move in free space, but in the semiconductor host crystal. The quantum dot host material, in particular its band structure, does therefore play an important role for all quantum dot properties. Quantum dots with a nearly spherical symmetry or flat quantum dots with nearly cylindrical symmetry can show shell filling according to the equivalent of Hund's rules for atoms [٢٠]. In contrast to atoms, the energy spectrum of a quantum dot can be engineered by controlling the geometrical size, shape, and the strength of the confinement potential.

Also in contrast to atoms it is relatively easy to connect quantum dots by tunnel barriers to conducting leads, which allows the application of the techniques of tunneling spectroscopy for their investigation. In addition, by applying small voltage to the leads one can control the flow of electron through the quantum dot and make precise measurements of the atomic-like properties.

Quantum dots are particularly significant for optical applications due to their theoretically high quantum yield. In electronic applications they have been proven to operate like a single-electron transistor and show the Coulomb blockade effect. The spin state of the electrons confined in quantum dots is one of the most promising candidates for qubits in quantum information processing.

The ability to tune size of quantum dots is advantageous for many applications. For instance, larger quantum dots, have spectra shifted towards the red compared to smaller dots, and exhibit less pronounced quantum properties. Conversely the smaller dots allow one to take advantage of quantum properties.

1.3- Fabrication of quantum dot

There are several strategies for synthesizing nanocrystals with various properties depending on the required applications [22]. Engineering of less than three dimensional semiconductor began in earnest during the early 1970s, when groups at AT&T Bell Laboratories and IBM made the first two-dimensional "quantum well" [23]. Semiconductor quantum dots (SQDs) are currently assembled or synthesized using either "dry" or "wet" methods.

Dry techniques are the most familiar to the traditional semiconductor micro fabrication community, while wet techniques are typically the domain of synthetic chemists. Though submicron nanostructures can also be formed through some what traditional paths using a top-down approach involving electron beam lithography and dry etching[24], these techniques cannot be used to make true quantum dots of Bohr exciton scale.

True dry Bohr-exciton scale quantum dots are formed using a bottom-up approach, with the use of epitaxial techniques to produce SQDs from atomic constituents and/or molecular precursors. Such SQDs are assembled using strained layer growth in ultra-high vacuum, crystal growth systems, such as molecular beam epitaxy or metal organic chemical vapor deposition. SQD growth is a consequence of the lattice mismatch between the matrix (or substrate) material and the dot (or epi-layer) material. The minimization of strain energy results in the formation of three dimensional islands, typically with a pyramidal or lens shape. For most epitaxial QDs, these three-dimensional islands occur during the Stranski-Krast- a now (S-K) growth mode, which is characterized by the formation of a two-dimensional, wetting layer, followed by the three-dimensional QD growth. The main limitations of this method are the coast of fabrication and the lack of control over positioning of individual dots.

Much recent work is also in the area of bottom-up synthesis of wet colloidal SQDs, grown in solution in a chemical reaction vessel. Because SQD particle size, and hence the peak luminescence emission wavelength, can be controlled with great precision in wet colloidal synthesis, and because the surface chemistry of SQDs can be modified to have compatibility with a variety of solvents including water, the first major commercial application of SQD is in luminescent tags for biomedical applications. Wet SQDs can be also assembled into quantum dot solids through various deposition techniques such as drop-casting, spin-casting, and Langmuir-Blodgett methods.

1.4- Quantum confinement

The inherent size dependence of these macroscopic nanocrystal properties is the result of two primary mechanisms: free energy variation and quantum confinement. Since nanocrystals are typically made up of tens to thousands of atoms; geometric considerations promote a very high surface-to-volume ratio, compared to the bulk version of the same nanocrystal. Since for any material the surface atoms contribute proportionately to the total free energy, compared to their bulk counter parts, nanocrystals exhibit significantly differentiated thermodynamic properties. In general, size reduction to the nano regime causes a melting temperature depression.

Semiconductor nanocrystals exhibit distinct variations in their electrical and optical properties with size variation. The core reason for these changes lies in the systematic transformation of the crystal's density of states as a function of energy, which varies as a function of the nanoparticle interior. These changes are known as quantum size effects. From a density of states perspective, nano crystals lie between the discrete atomic and bulk 'continuous band' limit (figure1.1). In any material there will be a size below which one observes a fundamental shift in electronic and optical properties as a function of size; this quantum size effect regime begins when the energy level spacing exceeds the kT temperature. For a particular temperature, this effect occurs at a relatively large size in semiconductors (in comparison to small molecular crystals, metals, and insulators).

The energy required to add successive charges onto an extended (bulk) crystal does not vary. In a nanocrystal, however, the presence of one charge acts to prevent the addition of another. Thus, in metals or semiconductors, the current-voltage curves of individual crystallites resemble an additive step function(a stair case). This effect is often termed a "Coulomb blockade".

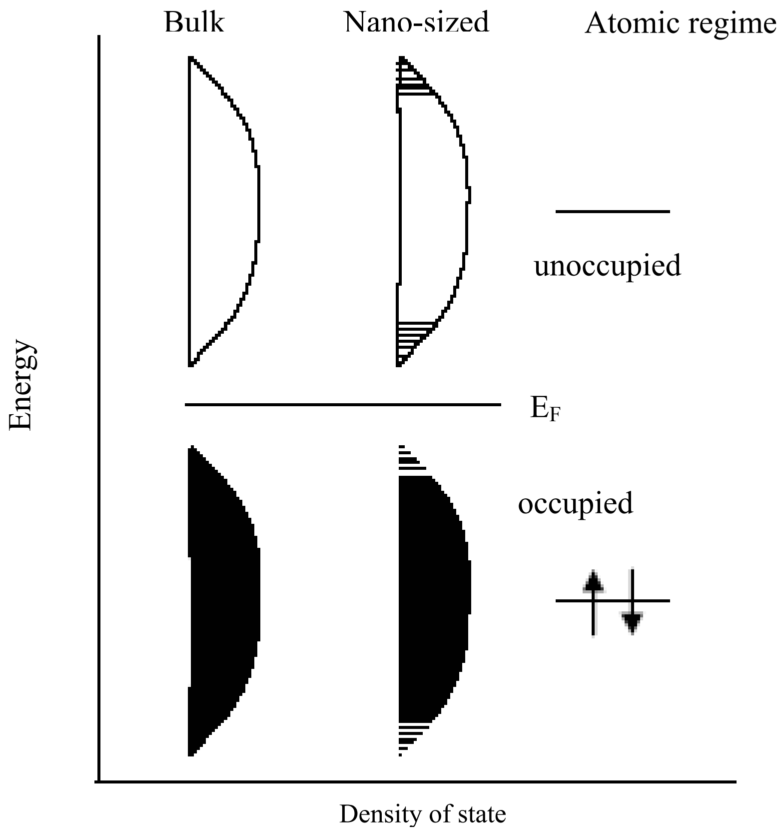

Figure 1.1- Density of States in semiconductor nano-crystals.
The nanocrystal density of states is discrete at the band edges.

The electrical characteristics of individual semiconductor nanocrystals, in which Coulomb blockade effects and discrete of energy levels are both expected to occur, are a topic of much current research. It is in fact the actual dependence of the optical and electrical properties of semiconductor nanocrystals on their size which forms the basis of the brood interest they have garned in recent years.

The change in the density of available electronic states (the quantized density of states near the band offset shifts to a higher energy) is understood by initially considering Heisenberg's uncertainty relationship between momentum and spatial positioning of both free and confined particles.

$$\Delta P_x \quad \Delta x \quad \geq \hbar \qquad (1.1)$$

For either a free particle or one which is confined to periodic potential, the energy and the momentum $\hbar k$, k is the wave vector, may be well-defined, but simultaneously the particle position is not defined. Localization of the particle to within a particular restricted range results in a decrease in the momentum certainty but a simultaneous increase in the position certainty. From the well-known Schrödinger equation, energy eigenfunctions are produced through the Hamiltonian operating on the wave function. These energy eigenfunctions of the localized particle can be viewed as super positions of k state associated with the bulk. For the case of an extended solid (electron not localized), there is a definite relationship between energy and momentum. The change in energy as a function of size can be estimated through the fact that the energy of the confined particle arises via superposition of bulk k states of varying energy. For a free particle, the dependence of energy E on the wave-vector k is quadratic.

$$E = \frac{\hbar^2 k^2}{2m^*} \tag{1.2}$$

In the effective mass approximation, the relationship of equation (1.2) is assumed to hold for an electron or hole in a semiconductor periodic potential, with the substitution of a reduced mass inversely proportional to the curvature of the band. We are given that momentum uncertainty (i.e. the superposition of bulk states) and spatial localization resulting from confinement are inversely proportional in the Heisenberg relation. Coupling the uncertainty with the energy dependence on the square of the wave-vector leads directly to the approximate dependence of energy on size as the inverse of a^2. This corroborates the result obtained through the particle in a box analysis, where energy is also proportional to the inverse of a^2 i.e.

$$E = \frac{n^2 \pi^2 \hbar^2}{2m^* a^2} \tag{1.3}$$

1.5- Exciton in quantum dot

A free exciton is a bound electron-hole (e-h) pair that has a binding energy of a few meV. If optical excitations are of sufficiently high frequency, they impart enough energy to ionize an electron previously in the valance band of the crystal. A vacancy (hole) is left behind in the position where the electron once resided. As long as the optical excitation continues, the excited electron acts as an ion in a perfect crystal.

In a perfect crystal all electrons in the highest occupied molecular orbital are capable of being excited. Since the coupling between the ions outer shells is strong, the excitation energy can readily be transferred from ion to ion. Thus the e-h pair behaves as a single hydrogenic particle and can move through the crystal without requiring that the individual ions switch spatial position. As such, excitons which are not spatially bound to a region smaller than their natural separation distance (the Bohr Radius) are free to move about the crystal. For the free exciton scenario, it is more appropriate to think of the e-h pair as not being localized at all [25]. Rather, it is more relevant to consider the exciton to be a feature of the electronic crystal, a superposition of states among all crystal atoms.

The significance of the excitonic Bohr radius is that it provides a threshold value below which the confinement regime becomes relevant. That is, an exciton in a nanoparticle will not experience confinement if it is restricted to a spatial dimension larger than that of its own preferred electron-hole spatial separation. Confinement energy will not be imparted to energy levels available to the exciton unless the e-h spatial separation is reduced below the excitonic Bohr radius. Below this threshold value begins the exhibitions of quantum sizing features, such as distinct variation of states with nanocrystal size.

In the present theses we study the effect of hydrostatic pressure and temperature on the exciton binding energy confined in cylindrical GaAs quantum dot. The GaAs material and its