

**Linear Spectroscopic Studies of Semiconductor Quantum
Wells**

by

M. W. Day

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This thesis entitled:
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written by M. W. Day
has been approved for the Department of Physics

Prof. Steven Cundiff

Prof. James Thompson

Prof. Judith Packer

Date _____

The final copy of this thesis has been examined by the signatories, and we find that both the content and the form meet acceptable presentation standards of scholarly work in the above mentioned discipline.

Day, M. W. (B.S.)

Linear Spectroscopic Studies of Semiconductor Quantum Wells

Thesis directed by Prof. Steven Cundiff

Manufacturing processes unintentionally introduce fluctuations in the width of semiconductor quantum wells. These fluctuations subtly modulate the optical emission energies of excitons confined within the quantum well layer. It is therefore imperative to quantify these width fluctuations so their effect on exciton confinement potentials can be accounted for in ultrafast spectroscopic studies of semiconductor quantum wells. The use of microphotoluminescence spectroscopy makes quantifying this disorder possible. I present microphotoluminescence spectroscopy work taken in pursuit of an Honors' thesis.

Dedication

To the wonderful people in my life: family and friends alike.

Acknowledgements

I'd like to sincerely thank Professor Steven Cundiff for years of tutelage. Without his guidance, I would not have been able to complete this thesis.

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Chapter 1

Introduction

Semiconductor nanostructures provide uniquely manipulable platforms for probing quantum interactions between light and matter in a highly controllable environment. These structures typically require the ability to manipulate semiconductor growth on the nanometer scale. As a consequence, manufacturing such materials is a complex and involved process. However, due to the unique physical properties of valence electrons within the crystalline lattice, describing the physics of semiconductor nanostructures is relatively simple [CITE davies]. Therefore, within the broader context of atomic and molecular optics, materials physics, and ultrafast spectroscopy, semiconductor nanostructures form the basis of deeper study into many-body physics in the quantum-mechanical regime.

Band theory, broadly construed, describes how electrons behave as atoms group together to form various materials. As this introduction is not meant to be a rigorous description of condensed matter physics, a brief sketch of band theory will be sufficient to explain the importance of semiconductor nanostructures. When a large number of atoms coalesce, the discrete energy states of electrons relative to their host atoms smear into 'bands' of allowed states electrons can occupy in the atomic superstructure. In this limit, two distinct energy bands form: the valence band (lower electron energies) and the conduction band (higher electron energies). Electrons occupying the valence band are confined to the atomic superstructure, whereas electrons within the conduction band states may roam around the superstructure. The difference between these two energies in a material is known as its

'bandgap'. In semiconductors, the bandgap is relatively small, and thus one can photo-excite an electron from the valence band to the conduction band. Electronic confinement happens when one takes a semiconductor material of lower bandgap and surrounds it with a semiconductor of higher bandgap on the length scale of the electronic wavefunction. The confinement dimensionality (generally one, but sometimes two or three), width, and energetic depth are all free parameters in this process. Because of this parametric freedom, everything from simple confinement schemes to complicated structural configurations have proven to be very useful for creating electronic and opto-electronic devices CITE NAT REVIEW. In addition, semiconductor nanostructures form the basis for photonic quantum information devices CITE DEVICE REVIEW, QI REVIEW.

For our purposes, we will be discussing semiconductor quantum wells (QWs). These nanostructures are layers of relatively low bandgap material (GaAs in our case) sandwiched between layers of higher bandgap (AlGaAs). If one excites an electron within the confined layer, that electron will be confined to move about in two dimensions. Furthermore, that electron will be weakly bound to the vacancy (or 'hole') it left in its parent atom. This electron-hole pair constitute a quasiparticle known as an exciton, the importance of which will be expanded upon later. Excitons confined within QWs can be treated theoretically using a simple particle-in-a-box quantum mechanical picture, making QWs useful devices for studying the subtleties of lights' interaction with matter, among other things.

Despite immense progress in the growth of quantum wells, manufacturing processes still unintentionally introduce inhomogeneities to the layer thickness during crystal deposition. This translates to uneven interface between two different materials on the scale of a few crystal monolayers. What this means for a quantum well structure is that small fluctuations in interface flatness translate to small fluctuations in well width as these happen at both interfaces of the quantum well and barrier layers. These small width fluctuations are known as disorder.

Semiconductor quantum wells are studied optically by our lab. In most of our exper-

iments involving these structures, excitons are generated in the wells, and their dynamic behavior can be monitored by several different spectroscopic techniques, notably Multidimensional Coherent Spectroscopy (MDCS). As will be shown later, small changes in well width cause changes in confinement potentials for excitons and thus affect the energies at which excitonic states exist in the wells. Therefore, disorder is the main cause of inhomogeneous broadening within QW samples. This is known to be true CITE BRISTOW, but quantifying the spatial distribution of disorder is important because complete information concerning inhomogeneous contributions to the overall spectral response of QWs can help us enrich our understanding of many-body physics of excitons within quantum wells. In this thesis, I present my development of a method known as micro-photoluminescence spectroscopy to obtain a spatial map of emission energy fluctuations. Furthermore, I translate this spatial picture into a quantitative measurement of disorder on three different types of QW structures: a periodic ten-quantum well structure, a periodic four-quantum well structure, and an interfacial quantum dot ensemble.

In addition to studying quantum well disorder, linear spectroscopy is useful for characterizing the energy profile of electron states in matter. For example, it is useful to study exciton states the asymmetric double quantum well, as these systems are great testbeds for various quantum coupling phenomena CITE AQW Review. Understanding incoherent coupling between exciton states in asymmetric double quantum wells allows us to fulfill two important experimental goals: we obtain a better understanding of the exact absorption energies of the various exciton states, and we can explore incoherent coupling between states in each of the wells.

Chapter 2

Theory and Background

In this chapter, I will provide the theoretical background necessary to understand the work presented in this thesis. The first section will deal with the definition and importance of the bandgap of a material, the second will introduce the effects of confinement on electrons within semiconductors, leading to a useful physical model, the semiconductor quantum well. The third will introduce the concept of semiconductor quantum well disorder, and the fourth will motivate the study of semiconductor quantum well disorder with micro photoluminescence spectroscopy.

2.1 The Semiconductor Quantum Well

INTRO

2.1.1 Bandgap

In order to understand the state of electrons in a crystalline solid, one must first understand how electrons act under the effect of a periodic potential. When we bring electron orbitals together in a molecule, the wave functions perturb each other. In the case of semiconductors, the Bloch theorem is a useful tool for understanding this problem quantum-mechanically, and a complete treatment can be found in CITE (Iadonisi and Siradesh). Fundamentally, if one assumes that valance electrons exist in an infinite, periodic, homogeneous lattice, then electron wave functions are invariant under translation from one lattice

point to the next. The Bloch theorem states that under translation from lattice site to lattice site, the electron wavefunction will pick up a phase:

$$\vec{k} \cdot \vec{R}_n \quad (2.1)$$

where \vec{k} is the displacement wavevector and \vec{R}_n is the lattice vector for the n th lattice site to which the wavefunction is translated CITE Iandonesi. Thus, the electron wavefunction becomes

$$\psi(\vec{r}) = e^{i\vec{k} \cdot \vec{r}} u_k(\vec{r}) \quad (2.2)$$

where \vec{r} is the lattice coordinate and $u_k(\vec{r})$ is a periodic function for each primitive cell in the crystal. In the limit that many primitive cells are brought together in a crystal, two things happen: each n -fold degenerate electronic energy level will split into n components, and these levels will become so close that they will smear into allowed and disallowed energy bands CITE Indonesi, Siradesh, Grif, Davies. If electrons have a high enough energy, they can occupy the ‘conduction band’ states where they are free to move about the lattice structure. They are otherwise confined to their host atoms and occupy the ‘valance band’ states. The energy difference between the valance band and conduction band is called the bandgap, and is illustrated in the example dispersion curve, 2.1.

The full functional shape of dispersion curves is dependent on the physical properties of solids. Thus, solids can be broadly construed into three categories: metals, insulators, and semiconductors. In metals, the conduction band energies are below the Fermi level and thus some electrons are free to move about and conduct charge. In insulators, the bandgap is relatively large, and therefore electrons are confined to their host atoms. By contrast in semiconductors, the bandgap is fairly small and therefore only a small amount of energy is required to promote an electron to the conduction band from the valance band. Importantly, semiconductor electrons can be photo-excited into the conduction band CITE Iandonesi. This fact forms the basis of experimental studies of semiconductor nanostructures

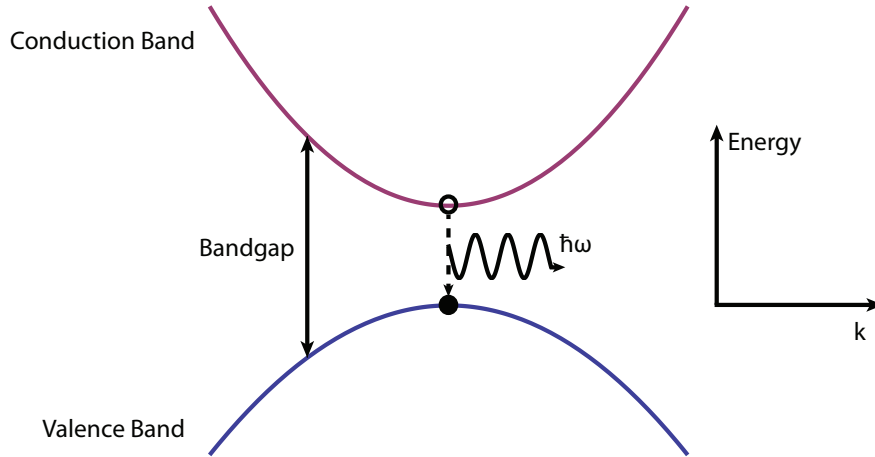


Figure 2.1: A typical dispersion curve minima for a direct-gap semiconductor. An optical transition is illustrated at $\vec{k} = 0$, where an electron is emitting a photon as the result of a transition from the conduction band to the valence band.

(CITE Steve review), photonic devices, and certain types of theoretical quantum information processing schemes (CITE Nature Review of QI).

(Discussion of band structure in GaAs arising from p-orbital smearing, figure highlighting the direct-gap nature of GaAs).

2.1.2 Confinement and The Exciton

It is well known that nanometer scale confinement of particles results in quantized energy states CITE Griff. In the previous section, I briefly introduced the bandgap, and its important physical properties. In this section, I'll illustrate an interesting application of band theory: the semiconductor quantum well. First, it is important to understand what we mean by confinement, and how quantized energy levels arise for confined particles. I will then construct a physical picture of the semiconductor quantum well (QW), and then I will discuss the formation of excitons and a simple physical model of their behavior, sufficient for understanding the spectroscopy conducted in this thesis.

Perhaps the simplest problem in quantum mechanics is the of confinement of a particle in a finite, one dimensional potential well. I will sketch a derivation of the wave function

of a particle trapped in such a well, and use this derivation as the basis for exploring the physics of the QW exciton. We will begin by considering an arbitrary particle confined in a one dimensional infinite potential well. The potential that our arbitrary particle feels is:

$$V(x) = \begin{cases} 0 & 0 < x < L \\ \infty & |x| > 0 \end{cases}$$

where L is the length of the potential well. Graphically, the potential the particle feels looks like 2.2.

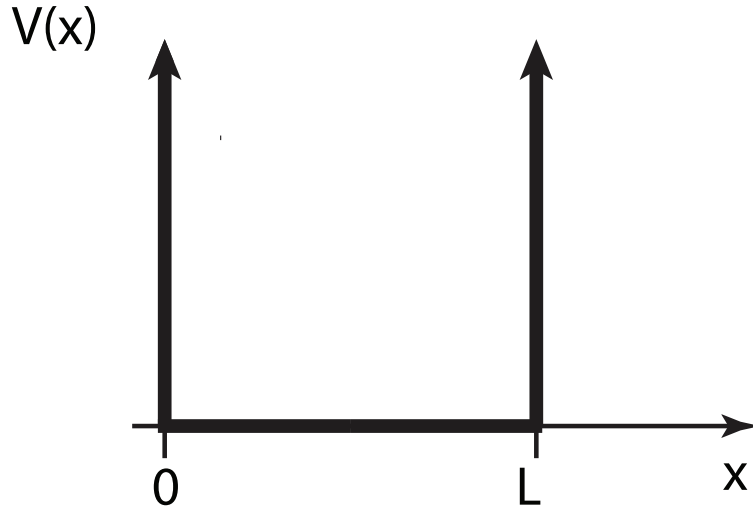


Figure 2.2: A graphical representation of the one-dimensional infinite potential well of width L .

Our task is to solve the time independent Schrödinger equation to show how quantized bound states arise for one-dimensional confinement. The time independent Schrödinger equation reads:

$$\hat{H}\psi = E\psi \tag{2.3}$$

where E is the energy of the particle, and $|\psi\rangle$ is the particle's wavefunction. The particle will evidently be confined to the well, so our Hamiltonian inside the well is just

$$\hat{H} = -\frac{\hbar^2}{2m} \frac{\partial^2}{\partial x^2} \quad (2.4)$$

where m is the particle's mass, and E is the particle's total energy. The time independent Schrödinger equation now reads:

$$\frac{\partial^2}{\partial x^2} \psi = -\alpha \psi \quad (2.5)$$

where we define

$$\alpha = \frac{2mE}{\hbar^2}. \quad (2.6)$$

Now, eq. 2.4 looks like the familiar simple harmonic oscillator equation from classical mechanics. Because the wave function must be continuous at $x = L$ and $x = 0$, i.e. it vanishes at those locations, and the potential is odd about the origin, solutions to eq. 2.5 have the form:

$$\psi(x) = A \sin(kx) \quad (2.7)$$

where k contains E and is determined by our boundary conditions. Now we want $\psi(L)$ to vanish, but we can't have $A = 0$, because that is the trivial solution to eq. 2.5. Therefore, because we want $\psi(L) = A \sin(kL) = 0$, we must have $kL = \pm n\pi$ where $n \in \mathbb{N}$. Now, we can absorb all of the negative combinations of kL into our normalization constant, A . We have, then, that

$$k_n L = n\pi \quad (2.8)$$

where the subscript denotes the fact that we now have infinitely many, *discrete* solutions to eq. 2.5. Evidently

$$k_n = \frac{n\pi}{L} \quad (2.9)$$

and therefore

$$\psi(x) = A \sin\left(\frac{n\pi x}{L}\right). \quad (2.10)$$

Now, if we let $k_n = \alpha$ and solve for E, we obtain

$$E = \frac{n^2 \pi^2 \hbar^2}{2mL^2}. \quad (2.11)$$

It will do us no good to normalize the wavefunctions we found, as I am not interested in the exact behavior of each state for our particle. The important part is that confinement in one dimension resulted in our particle occupying *discrete* energy levels.

Using layers of semiconductors, one can generate similar confinement of electrons in one dimension. A simple way of confining particles is to sandwich a low-bandgap semiconductor in between two layers of higher bandgap materials (CITE Davies). One period of this structure is shown in figure ???. If the well material is a direct-gap semiconductor, then simple vertical optical transitions across the bandgap can be made, as the transition illustrated in 2.1. Figure 2.4 is the band structure for GaAs, the chosen well material for the studies of growth disorder. Annotated on the figure is the direct-gap transition zone of interest. The potential well created by the semiconductor sandwich leads to quantization of electron states within the well layer CITE Miller, Davies, Steve Review. We can access these transitions optically, making the QW a great testbed for exploring electron dynamics within a simple and well-known potential CITE Steve Review.

The simple picture presented above doesn't quite adequately represent the physics of an electron within a quantum well, however. After an excited electron moves from the valence band to the conduction band, it will leave behind a vacancy, or 'hole', around its parent atom CITE Miller, Davies. The electron feels a *screened Coulomb potential* from this vacancy, as the vacancy is positively charged. I'll briefly sketch why this potential arises and then introduce the key concept of this section: the excitation. Imagine a large number of electrons

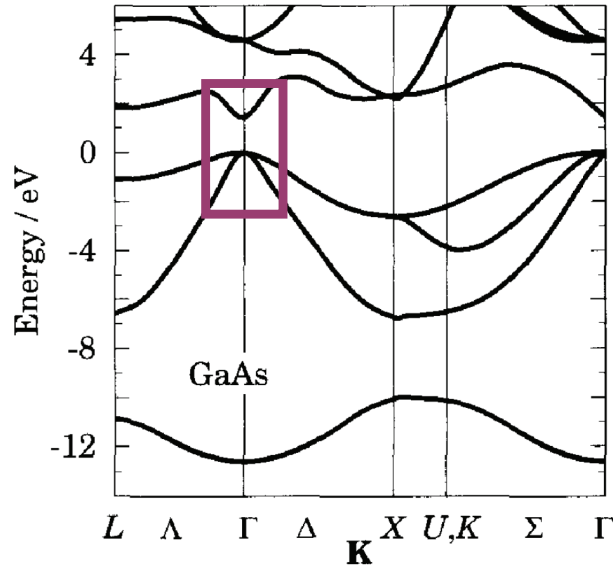


Figure 2.3: The band structure of GaAs, the allowed states are the thick horizontal curves, and the boxed region is the direct-gap region, in which electrons can be photo-excited across the gap. Note, the minima of this region look like the dispersion curve in 2.4.

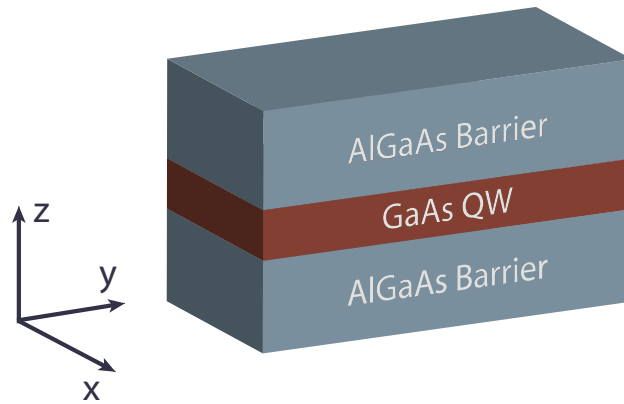


Figure 2.4: An example of the semiconductor quantum well. These layers can be repeated arbitrarily many times with arbitrary dimension and thickness for many types of application.

have been excited to their first excited state within the quantum well. Now, spatially, one will have a quasi-neutral distribution of electrons and holes in the well layer. Imagine inserting into this situation a positive charge. Adjacent electrons will immediately surround the positive charge, modifying the total Coulomb potential due to that charge, *screening* its effects from charges far away. This phenomenon, known as Debye shielding CITE Chen,

modifies the potential electrons feel from adjacent holes. If the potential electrons feel was exactly Coulombic in nature, then any bound state between an electron and a hole would be Hydrogenic CITE Griffiths. This potential, however, is *not* exactly Culombic in nature, so the bound states between the electrons and holes don't exhibit exactly Hydrogenic behavior. The subtleties of the functional form of the wave function are treated in CITE Rogers, but a sketch of their form is in figure ???. The photoexcited electron and an hole can form a bound pair, known as an exciton. The energy levels of this quasiparticle are discretized by its confinement in the quantum well. Additionally, coherent coupling of exciton states can be explored using multidimensional coherent spectroscopy CITE Steve Review, Bristow's PRL.

2.2 Quantum Well Disorder

2.3 Microphotoluminescence Spectroscopy

2.4 Asymmetric Double Quantum Wells

2.5 Photoluminescence Excitation Spectroscopy