Application of Microphotoluminescence Spectroscopy to Study Semiconductor Quantum Well Disorder.

by

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mentioned discipline.

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Application of Microphotoluminescence Spectroscopy to Study Semiconductor Quantum Well Disorder.

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 exitons confined within the quantum well layer. It is therefore imperative to quantify these width fluctuations so their effect on exiton confinement potentials can be accounted for in ultrafast spectroscopic studies of semiconductor quantum wells. The use of micro-photoluminescence spectroscopy makes quantifying this disorder possible. I present microphotoluminesence 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.

Contents

Chapter

Figures

Figure

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 latice, 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 construction. This translates to small fluctuations in the width of the 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 experiments 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.

Chapter 2

Theory and Background

2.1   The Semiconductor Quantum Well

2.1.1   The Bandgap

In order to understand the state of electrons in a crystalline solid, one must first understand the problem of how electrons act under the effect of a periodic charge density. The Bloch theorem is a useful tool for understanding this problem quantum-mechanically, and a complete treatment can be found in CITE Iadonisi. Here, I will sketch the important physical concepts and their implications for my thesis. Consider an infinite, arbitrary crystalline atomic lattice, unbounded in each spatial direction. The crystalline structure, therefore, is homogeneous, uniform, and evidently invariant under translation along any of the crystalline axes. Thus, for each valance electron around an arbitrary lattice atom, the following is true:

$$H(\vec{r}) = H(\vec{r} + \vec{D}_n) \tag{2.1}$$

where $\vec{r}$ is an arbitrary lattice position and $\vec{D}_n$ is an arbitrary displacement from position $\vec{r}$ and $H(\vec{r})$ is the quantum-mechanical Hamiltonian for a lattice electron. We can thus define a lattice transition operator, T_n, which takes the electron wavefucntion at one lattice site and spits out an electron wave function for a chosen D_n.

$$T_n \left| \psi(\vec{r}) \right\rangle = \left| \psi(\vec{r} + \vec{D}_n) \right\rangle \tag{2.2}$$

Because our operator commutes with itself and the Hamiltonian, we can assume that T_n and H share the same eigenfunctions. Thus the following vector equation holds:

$$T_n |\psi(\vec{r})\rangle = c_n(\vec{D_n}) \left| \psi(\vec{r} + \vec{D_n}) \right\rangle \tag{2.3}$$

where the $c_n(\vec{D_n})$ coefficients are the eigenvalues of T_n. The wave function *must* remain normalized through some arbitrary displacement $\vec{D_n}$, and we can thus choose:

$$c_n(\vec{D_n}) = e^{i\vec{k} \cdot \vec{D_n}} \tag{2.4}$$

where $\vec{k}$ is the electron's wave vector (real, unless translational symmetry is broken). We can then write the Bloch theorem:

$$T_n |\psi(\vec{r})\rangle = e^{i\vec{k} \cdot \vec{D_n}} |\psi(\vec{r})\rangle \tag{2.5}$$

where $|\psi(\vec{r})\rangle = e^{i\vec{k} \cdot \vec{D_n}} |u_k(\vec{r})\rangle$ is the Bloch wave function, and $u_k((\vec{r}))$ is a periodic function of the lattice. We can then write out the full Shrödinger equation for an electron in our arbitrary lattice, after substitution of our Bloch wavefunctions:

$$\left(-\frac{\hbar^2}{2m} \bigtriangledown^2 - \frac{i\hbar^2}{m}\vec{k} \cdot \bigtriangledown + \frac{\hbar^2 k^2}{2m} + V \right) |u_k\rangle = E |u_k\rangle \tag{2.6}$$

Solving this equation is far beyond the scope of the thesis, but the upshot is that in a periodic lattice (i.e. a periodic charge distribution establishes periodic boundary conditions), allowed energy states smear into allowed 'bands'. Crudely, electrons 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 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
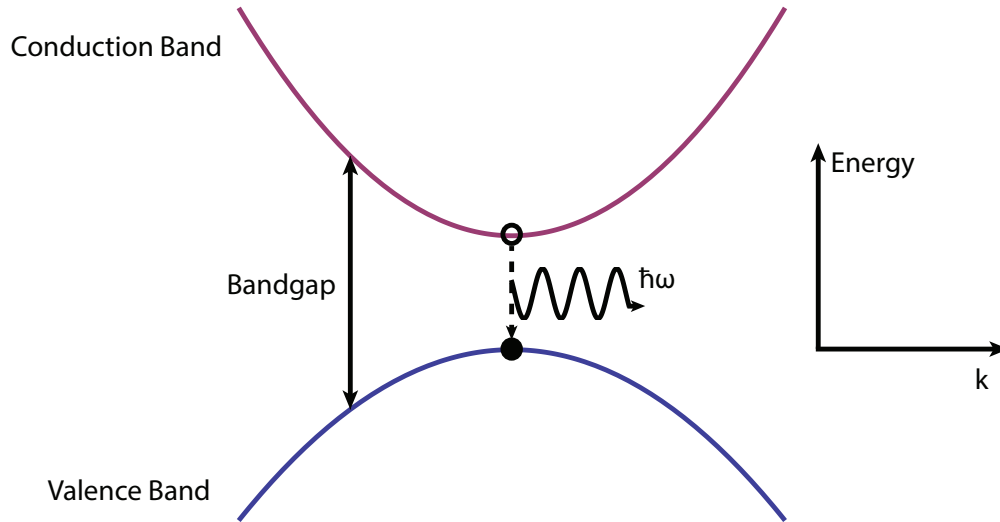
Figure 2.1: A typical dispersion curve minima for a direct-gap semiconductor. An optical transition is illustrated at $\vec{k} = 0$, where an electron is emitting a photon as the result of a transition from the conduction band to the valance band.

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 optically photo-excited into the conduction band. This fact forms the basis of experimental studies of semiconductor nanostructure CITE Steve, photonic devices, and certain types of theoretical quantum information processing schemes CITE Nature Review of QI.

2.1.2   Confinement and The Exciton

It is well known that nanometer scale confinement of particles results in quantized energy states CITE someone. I'll illustrate the important physical effects of confinement, and in the next section, I will introduce the semiconductor quantum well. Initially, I will sketch the derivation of the finite potential well that confined particles feel. This derivation will be sufficient to explore the physical effects of confinement, but the subtleties of the semiconductor electron potential will be addressed in the next section. The most illustrative theoretical model of a semiconductor quantum

well is the canonical particle in a finite well, and the goal here is to derive the energy of the first

quantized energy state in the limit that the well is fairly deep. For a more complete treatment of

the finite potential well, see CITE Griffiths.

$$V(x) = \begin{cases} 0 & x < -L \\ -V_0 & -L \leq x \leq L \\ 0 & L < x \end{cases}$$

The most illustrative approach is to solve the Shrödinger Equation for the stationary states,

and find the first excited state energy for the finite well in the limit of a tall well. Within the well,

the Time Independent Schrödinger equation reads:

$$-i\hbar \frac{d}{dt} |\psi\rangle = E |\psi\rangle \tag{2.7}$$

2.1.3   Physical Model of a Semiconductor Quantum Well

A semiconductor quantum well is a grown structure with two barrier layers 'sandwiching'

the quantum well layer CITE Miller. The barrier layers are manufactured from a higher band-gap
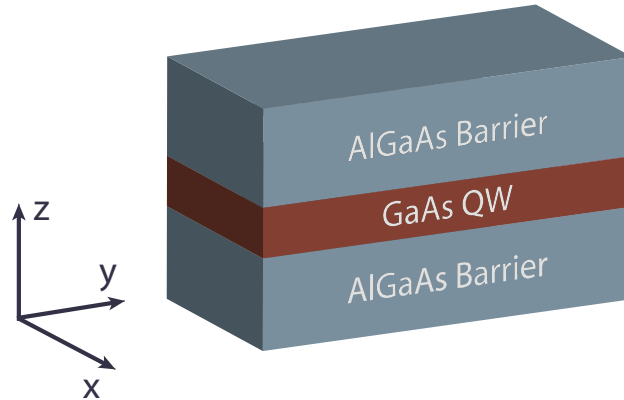
material than the

Figure 2.2: sports

2.2   **Quantum Well Disorder**

2.3   **Microphotoluminescence Spectroscopy**