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Electronic Raman Spectroscopy on Semiconductor Quantum Dots

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Abstract

We present Raman and photoluminescence experiments on low-dimensional electron systems based on the compound III-V semiconductors GaAs, AlAs and InAs. A detailed analysis of electronic Raman spectra measured on an ensemble of self-assembled InAs quantum dots on GaAs, excited resonantly at the $E_0 + \Delta$ gap is shown. The highly asymmetric double quantum well system consisting of the backgate and the wetting layer is also studied by resonant Raman spectroscopy. Apart from the single-well single-particle, charge and spin density excitations the measurements show electronic Raman signals that can be explained by a weak tunnel coupling between the two quantum wells. A comparison of the single particle excitations to the results of self-consistent calculations of the Schrödinger and Poisson equations of the system shows a good agreement. Capacitance and photoluminescence spectroscopy was performed on samples with two close layers of self-assembled InAs quantum dots on GaAs. We further present results of photoluminescence excitation (PLE) spectroscopy on InAs/GaAs quantum dots measured at the $E_0 + \Delta$ gap illustrating the possibilities and limitations of the PLE method when applied on a large ensemble.

Raman and photoluminescence spectroscopy was performed exciting resonantly at the E_0 gap transitions on InAs dots grown on AlAs, grown on GaAs and grown on GaAs with the so-called In-flush technique. Macroscopic ensemble measurements on the InAs/AlAs dots did not exhibit any distinguishable electronic Raman signals but featured what in retrospective may be assumed to be photoluminescence peaks of few resonantly excited quantum dots as well as excitations that shift with half the laser energy, for which a physical model is proposed. Measurements on the InAs/GaAs systems are shown ranging from macroscopic ensemble measurements to measurements on single quantum dots. We examine the resonance behavior of the dots and establish and apply methods to distinguish between single dot Raman and single dot photoluminescence signals.

Inhaltsangabe

In dieser Arbeit präsentieren wir Raman- und Photolumineszenz-Experimente an niedrig-dimensionalen Elektronensystemen basierend auf den III-V-Verbindungshalbleitern GaAs, AlAs und InAs. Wir zeigen eine detaillierte Analyse elektronischer Raman-Messungen an einem Ensemble von selbstorganisiert gewachsenen InAs-Quantenpunkten auf GaAs, die resonant am $E_0 + \Delta$-Gap angeregt wurden. Ebenso untersuchen wir ein hoch asymmetrisches Doppelquantumwell-System, das durch das Backgate und die so genannte Wetting Layer in unseren Proben entsteht, mittles resonanter Raman-Streuung. Neben den Einteilchen-, Ladungs- und Spindichteanregungen aus einzelnen Quantum Wells konnten wir auch Ramansignale beobachten, die durch eine schwache Kopplung der beiden Systeme erklärt werden können. Ein Vergleich zwischen den gemessenen Einteilchenanregungen und selbstkonsistenten Berechnungen der Schrödinger- und Poisson-Gleichungen zeigt eine gute Übereinstimmung. Weiterhin führten wir Kapazitäts- und Photolumineszenz-Spektroskopie an einer Probe mit zwei Lagen von Quantenpunkten durch. Ferner zeigen wir Ergebnisse von Photolumineszenz-Anregungs-Spektroskopie am $E_0 + \Delta$-Gap von InAs/GaAs-Quantenpunkten um die Möglichkeiten und Grenzen der Methode, angewandt auf ein großes Ensemble, aufzuzeigen.

Raman- und Photolumineszenz-Spektrokopie, resonant angeregt am E_0-Gap, wurden durchgeführt an Proben, die auf herkömmliche Art auf AlAs oder GaAs, bzw. mit der In-Flush-Technik auf GaAs gewachsen wurden. Makroskopische Ensemble-Messungen an den InAs/AlAs-Quantenpunkten zeigten keine eindeutigen elektronischen Raman-Signale, enthielten aber schmale Anregungen, die im Nachhinein als Photolumineszenz-Signale weniger Quantenpunkte gedeutet werden könnten sowie Anregungen, die mit halber Laserenergie auf absoluter und relativer Energieskala schieben. Messungen an den InAs/GaAs-System werden gezeigt, die von makroskopischen Ensemble-Messungen bis zu Messungen an einzelnen Quantenpunkten reichen. Wir untersuchen das Resonanz-Verhalten der Quantenpunkte und etablieren Methoden um zwischen Einzelpunkt-Raman- und Photolumineszenzsignalen zu unterscheiden.

Contents

Chapter 1

Introduction

Many of today's everyday appliances would not be conceivable were it not for the rapid advances that the semiconductor industry has made in the last decades. Mobile phones with the computing power of a ten year old personal computer are just one example of what the massive increase in component density on computer chips has made possible. This increase in density does of course go along with a size reduction of the basic elements that the electronic components consist of — the next generation of processors (to go into serial production as of 2007/2008) has been announced to contain structures as small as 45 nm. As the structures reach the limit where the number of atoms to a structure becomes countable it is not only the realization of these «nanoscopic» structures that poses a challenge — the physics of electrons and holes on this scale is quite different. Therefore, a detailed analysis and knowledge of low-dimensional systems, where electrons are contained in one or even all three dimensions within a few nanometers, is crucial for the development of future electronic components. Whether these will be similar to those that we know (but smaller), or whether at some point the new properties of the low-dimensional systems will be used to create the famed quantum computer, only time will show.

The focus of this work are the electronic properties of quantum dots, i.e. structures where the electrons are contained within few nanometers in all three dimensions by a potential barrier. Because of the three-dimensional enclosure the optical properties of quantum dots are not unlike those of atoms — although on a different energy scale — which is why they are also referred to as *artificial atoms*. Changing the size and material composition of the quantum dots enables us to change the energies of the electrons within the dots. A lot of research has been done on quantum dots in recent years due to the fundamental interest in adjustable atoms as well as possible new applications such as single photon sources or quantum bits.

There is a variety of different quantum dot structures that have received varying attention over the years. This work concentrates on different self-assembled quantum dots. Other notable quantum dot systems include deep-mesa etched systems, gate-defined systems and chemically synthesized nano-structures. Similarly, the material used for structure growth can be anything from Si and Ge to III-V compound semiconductors such as GaAs or II-VI semiconductors such as CdTe and ZnS. We used structures based on the semiconductors InAs, GaAs and AlAs because of their favorable optical properties and the well understood growth mechanisms.

Electronic Raman spectroscopy has been used to study the many-particle excitations of low-dimensional electron systems as early as the late 1970's [Pin79, Abs79]. First Raman measurements on quantum dots were performed on etched GaAs/AlGaAs quantum dots in 1994 by Strenz *et al.* [Str94]. The self-assembled quantum dots have the advantage of containing only very few electrons as compared to the hundreds of electrons that etched quantum dots usually contain (although recently García *et al.* have shown Raman experiments on etched GaAs/AlGaAs quantum dots with only few electrons [Gar05]). The first publication on Raman scattering on self-assembled InAs quantum dots on GaAs was by Chu *et al.* [Chu00] and reported on measurements on a sample with fifteen layers of quantum dots separated by GaAs and doping layers. By using a highly sensitive *CCD* camera and exciting resonantly we have succeeded in enhancing the Raman signal enough to be able to measure Raman signals of a single layer of quantum dots that we could charge with single electrons [Bro03a].

The ultimate goal would be the detection of the Raman signals of a single quantum dot, thus eliminating the inhomogeneous broadening effects of an ensemble measurement. For photoluminescence experiments this has been accomplished several years ago, see e.g. [Bay00]. To separate the single dot Raman signal from any other signal that the sample may exhibit, a profound understanding of the quantum dot luminescence properties as well as the Raman properties of all electronic systems in the sample is necessary.

This thesis tries to contribute to this understanding and establishes methods for the distinction of the different optical signals as well as showing first Raman measurements on few self-assembled quantum dots. It is organized as follows.

Chapter 2 outlines the basic theoretical concepts necessary for the understanding of this work. In Chapter 3 and 4 we show details of the sample preparation and the experimental setups. In Chapter 5 we discuss the experimental results, which are divided into several parts. Starting with results on self-assembled InAs/GaAs quantum dots obtained by exciting resonantly at the $E_0+\Delta$ gap, we move on to Raman measurements on the double quantum

well system backgate-wetting layer. The third section shows results of capacitance and photoluminescence spectroscopy on double quantum dots and the fourth section evaluates the possibilities of photoluminescence excitation spectroscopy at the $E_0 + \Delta$ gap on large ensembles of quantum dots. The following sections show the results of resonant Raman spectroscopy at the E_0 gap on InAs/AlAs dots, InAs/GaAs dots and InAs/GaAs dots grown with the In-flush technique before we gradually decrease the number of observed dots in the last part by using a microscope Raman setup and nanoapertures in an Al mask on top of the sample. Chapter 6 summarizes this work.

Chapter 2

Theory

2.1 Low-dimensional electron systems

2.1.1 Two-dimensional electron systems

In a bulk semiconductor, electrons in the conduction band can move freely through the crystal in every dimension. They can occupy a quasi-continuum of energies and momentums, experiencing a quadratic energy dispersion near the Γ-point in the reciprocal lattice which can be written as

$$E = \frac{\hbar^2 |\vec{k}|^2}{2m^*}, \tag{2.1}$$

where m^* is the effective mass of the electron and $\vec{k}$ is the wave vector. Confining the electrons in one direction leads to a so-called two-dimensional electron system (*2DES*) or quantum well. We will call the directions where the electrons can still move freely x and y while z will be the direction of the confinement. In our experimental systems this is also the growth direction. While the focus of this work will be on zero-dimensional systems, our samples contain at least one quantum well (the so-called wetting layer) and often a second one which serves as a back contact. We will use these systems as examples to further discuss the physics and the possible collective excitations of electrons in *2DES*s.

The wetting layer is a thin layer of InAs that forms during the growth of the self-assembled quantum dots used in this work (see Chapter 3.2 for details). This layer is surrounded by GaAs or AlAs, which have a much larger band gap. The resulting band structure is depicted for GaAs in Fig. 2.1 (a). The shape of the confinement potential is in first approximation rectangular and symmetric, unless doping layers are introduced. Fig. 2.1 (b) shows a

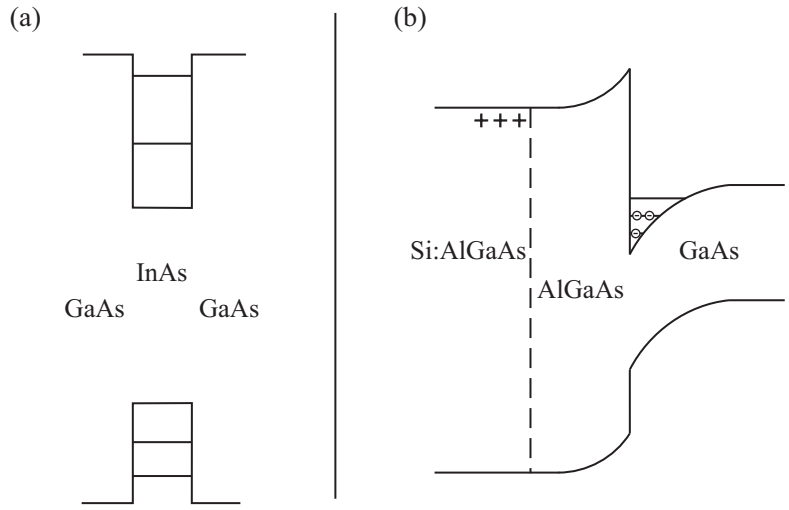

Figure 2.1: Band structures of (a) an InAs wetting layer surrounded by GaAs and (b) a *2DES* created by modulation doping as used in this work as a back contact.

2DES grown with a different approach. An AlGaAs layer highly n-doped with Si is overgrown by a thin layer of undoped AlGaAs — called the spacer layer — and a thick layer of GaAs. The excess electrons from the doping layer travel through the spacer layer and into the GaAs, losing energy mainly to phonons in the process. This leaves the Si donor atoms with a positive charge, attracting the free electrons that are now in the GaAs layer. Therefore, the electrons accumulate near the AlGaAs/GaAs heterojunction, changing the band structure. The potential for the electrons can be calculated self-consistently by solving the Poisson and Schrödinger equations alternately (see Chapter 2.2). The resulting quantum well structure is rather triangular than rectangular in shape. Usually, the structure is grown with the doping layer on top and applied in High Electron Mobility Transistors (*HEMT*s). The way the structure is grown here, it is called an *inverted HEMT* structure.

Both systems still have parabolic energy dispersions in k_x and k_y with a series of subbands because of the confinement. They can be written as

$$E_i = E_i^z + \hbar^2 \frac{(k_x^2 + k_y^2)}{2m^*}, \tag{2.2}$$

where i is the index of the subbands. E_i^z depends strongly on the confinement potential.

In a simple single particle picture, an excitation of an electron is represented by lifting the electron to a higher state which can be either on another subband (intersubband excitation) or, with the need of a momentum

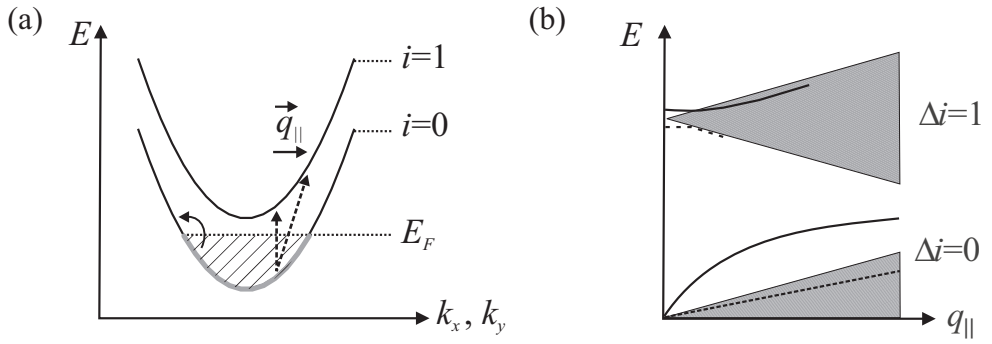

Figure 2.2: (a) Possible intersubband (dashed lines) and intrasubband (solid line) excitations in a quantum well with one subband filled with electrons. The horizontal line marks the Fermi energy E_F. (b) Dispersions of different intersubband ($\Delta i = 1$) and intrasubband ($\Delta i = 0$) excitations. The gray areas show the single-particle continua, the solid lines the *CDE*s and the dashed lines the *SDE*s.

transfer, the same subband (intrasubband excitation). These excitations are depicted in Fig. 2.2 (a). Without momentum transfer only intersubband excitations are possible. Due to the Coulomb and exchange interactions between the electrons all excitations in the quantum wells are *collective* excitations. There are, however, some excitations which have all the characteristics of a single particle excitation (*SPE*) and are only very slightly shifted in energy which shall therefore be called *SPE*s. The other collective excitations are divided into charge density excitations (*CDE*) and spin density excitations (*SDE*) depending on whether the center of charge oscillates (*CDE*s) or not (*SDE*s). *SDE*s are usually shifted to lower energies because of the exchange interaction. Since the center of charge moves in the *CDE*s, the Coulomb interaction becomes more important and the *CDE*s are shifted to higher energies with respect to the corresponding *SPE* and *SDE*. The difference between the energies of the *CDE* and the *SDE* is called depolarization shift. Pinczuk *et al.* have calculated the energies of the collective excitations in a modulation doped quantum well to be [Pin89]:

$$\omega_{SDE}^2 = E_{ij}^2 - 2n_i E_{ij}\beta_{ij} \tag{2.3}$$

$$\omega_{CDE}^2 = E_{ij}^2 + 2n_i E_{ij}\left(\frac{\alpha_{ij}}{\epsilon(\omega_{CDE})} - \beta_{ij}\right), \tag{2.4}$$

where E_{ij} is the difference between the two subbands i and j and n_i is the two-dimensional electron density. α_{ij} and β_{ij} are Coulomb matrix elements, where α_{ij} describes the depolarization shift. The dielectric function $\epsilon(\omega_{CDE})$ also takes into account the interaction of *CDE*s with optical phonons. GaAs is a