ELECTRONIC TRANSPORT IN ONE-DIMENSIONAL MESOSCOPIC SYSTEMS

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Chapter 1

Introduction

Figure 1.1: Physicists study the universe at many size scales, from the very large to the very small.

Mesoscopic physics, as the name suggests, is the study of the physics of the ‘in between’. (Figure 1.1.) Unlike cosmologists, who study gigantic objects such as galaxies, and particle physicists who study things so small that they cannot be seen using any microscope and have to be represented by funny (peculiar) cartoons, mesoscopic physicists study things intermediate in size between these extremes, on the order of 1nm to 1μm on a side. One might ask, of all the possible length scales between
galaxies and quarks, why 1nm to 1μm? The answer is that it is these length scales on which quantum mechanics starts to become important, on which quantum mechanical effects begin to be detectable. In other words, one might think of mesoscopics as the beginning of the realm of quantum mechanics.

Figure 1.2: Typical scheme for contacting very small objects, in this case a carbon nanotube peapod shown in the upper right in a TEM image. Metal electrodes are deposited on the molecule (lower right, false colour SEM image) which fan out into larger 100μm² contact pads. The whole substrate (in this case a silicon chip with a top layer of oxide) is glued into a chip carrier (upper left) and wires are attached between the contact pads and 'fingers' on the side of the carrier using an ultrasonic wire-bonder. The carrier can then be placed in mating sockets in various measurement setups.

In the Goldhaber-Gordon lab, we probe mesoscopic systems though electronic transport, which is to say that we study the movement of electrons (or holes) by applying voltages and measuring currents. As one might imagine, attaching wires for this purpose to objects so small is not trivial. Figure 1.2 shows a typical scheme for contacting such a system, in this instance a carbon nanotube peapod, more about
which very soon. [10]

One might be tempted to claim that mesoscopicists study things on an ‘in between’ scale because of being inherently sensible people who do all things in moderation; however, this would belie another important fact about the work of mesoscopics, which is that most of it is done at rather low temperatures — at least at 4K in our lab, and usually between 10-300 mK. (Figure 1.3.) These low temperatures are necessary for the observation of the phenomena we seek to study (as shall become clear to the reader in the course of reading this dissertation) and are achieved by the means of refrigerators operating on similar principles to the ones in most modern kitchens except that the refrigerant circulated is helium and the operating cost is thus much higher.

![Figure 1.3: It turns out that mesoscopic physicists do not practice moderation in all things. They run experiments at very low temperatures using very expensive equipment.](image)

In the field of mesoscopics, it is common to classify systems by their dimension, or (three minus) the number of directions in which they are confined, as follows. Most objects one encounters in everyday life, such as durians, are three dimensional — they
CHAPTER 1. INTRODUCTION

have extent in all three dimensions. Quantum mechanical effects can be and indeed have been studied in three-dimensional objects; however, the field of mesoscopics was given a tremendous boost by technical advances in the 1960s enabling the production of 2D electron systems (confined in one direction) first in silicon (Si) and then in the now more commonly-used gallium arsenide (GaAs). \[11\] Investigations of these novel confined systems led to the observation of unexpected phenomena such as the Quantum Hall effect, for which the Nobel Prize in physics was awarded in 1985.

It has only been recently that truly one-dimensional systems have been able to be produced. Carbon nanotubes were discovered by Sumio Iijima in 1991 \[12\] and the CEO method leading to the fabrication of the wires described in Chapter 7 was developed by Loren Pfeiffer and his colleagues in 1990. \[13\] This has opened up the prospect of the discovery of new phenomena especially as electrons and holes in 1D are expected to behave quite differently from 2D and 3D. In 2D and 3D, it is possible to think of systems of particles interacting through the Coulomb potential as ‘quasi-particles’ which behave very much like free electrons except that they have a different mass. These systems are known as Fermi liquids. \[14\] In a Coulomb-interacting 1D system (known as a Tomonaga-Luttinger Liquid), however, all excitations are collective and our intuition for the single-particle picture is not transferrable at all. (See for example References \[15\] and \[16\].) The implications of this have not been fully explored.

It is useful here to reflect on the way in which the first 1D systems were made. Carbon nanotubes were discovered serendipitously, not in the sense the experiments that led to their discovery were aimless, but that they were not directed (to my knowledge) to the production of one-dimensional systems. The fabrication of CEO wires, on the other hand, was a conceptually straightforward (though technically very difficult) extension of the methods used to produce 2D electron systems with the explicit aim of producing a 1D system. From this it would seem that science progresses by many means — some planned, some unplanned, but none entirely predictable in detail.

During my PhD I studied three different 1D systems — carbon nanotubes, peapods (carbon nanotubes with C\textsubscript{60} molecules inserted into them) and CEO hole wires. In
Chapter 2 I discuss the basic properties of carbon nanotubes and peapods, while the CEO wires are discussed in Chapter 7.

While it was the simplicity, novelty and unintuitiveness of 1D systems that first drew me to study them, many of the devices I made exhibited zero dimensional (quantum dot) effects, which can in principle happen to any small object coupled to leads whenever the temperature and tunnelling rate are smaller than the level spacing and the charging energy. \((kT, h\Gamma \ll \Delta, U\) in the language of Chapter 3, where all these terms are explained.) Therefore, in Chapter 3 I introduce 0D systems — quantum dots — which have been much studied more intensively than 1D systems and describe their basic properties.

Then, in Chapter 4, I describe in general our measurements on carbon nanotube peapods. In these experiments we discover that our devices behave like quantum dots at low temperature because of the high contact resistance between them and their leads. In addition, we found, rather unexpectedly, that the periodically intercalated C\textsubscript{60} molecules do not significantly modify the nanotube's electronic structure near the Fermi level.

In Chapter 3 I also include an description in outline of the Kondo effect, which is the focus of the middle chapters (5 and 6) of this dissertation. The Kondo effect, which occurs through the interaction of an local unpaired spin and a nearby reservoir of conduction electrons, is one of the simplest and therefore most-studied many-body problems both theoretically and experimentally; however, many details of its evolution at finite magnetic field and bias voltage (applied across the device) remain unknown. Due to a happy conjunction of materials properties as described in Chapter 5 quantum dots formed from nanotubes are particularly suited to the study of the Kondo effect at finite magnetic field. Thus, in our devices, we were able to carefully study the evolution of the Kondo effect in its most common (spin-1/2) variety in a magnetic field and compare our results in detail to several theoretical predictions. We find in the comparison qualitative agreement and at the same time quantitative disagreement.

We also observed in these devices a Kondo effect occurring when there is an unpaired spin of 1 on the nanotube quantum dot. This spin was underscreened in the regime of the experiment, which is to say that the conduction electron sea supplied a
spin in the opposite direction of only 1/2 so that the whole system had a net spin of 1/2. We were able to tune the ground state spin of the dot from 1 to 0 independently with a gate voltage as well as a magnetic field and we found that for the most part our data are explained by a simple model with a few nuances. These measurements are described in Chapter 6.

In the next chapter, I change gears to talk about the cleaved-edge-overgrowth (CEO) hole wires, describing some basic measurements from which we estimate the effective mass and subband spacings in these systems. We measured the Zeeman splitting of these subbands in two direction of magnetic field and find a large asymmetry. These experiments are ongoing and preliminary data with magnetic field oriented parallel to the wire indicate that we may directly observe a gap in the excitation spectrum due to spin-orbit coupling.

I have also measured nanotubes on and off since the beginning of my PhD, some grown in our lab and some obtained from elsewhere. In Chapter 8 I describe results from a particular experiment on devices fabricated by Xinran Wang in the Dai lab where nanotubes are suspended in air over small etched trenches.

Finally, I also in the appendices abbreviations used in this work as well as details on various technical aspects of the work.

Throughout this dissertation I have tried to write at a level comprehensible to incoming graduate students, as well as to provide suitable references and bibliographic notes for relevant topics which I do not go into in detail; I hope that this work will be of some utility to future generations of students as a description of my PhD work as well as an introduction to the field.
Chapter 2

Carbon nanotubes and peapods

In the fifteen years since their discovery, carbon nanotubes' electronic properties have generated considerable excitement among both physicists and engineers.

For those focussed on applications, they appear to hold promise for use as transistors, memory and logic elements, and field emitters. From the point of view of fundamental physics, nanotubes are among the best systems now available for the study of interacting electrons in one dimension; in addition (as described in Chapter 5), the energy scales associated with nanotube devices are particularly convenient for the study of other correlated electron phenomena such as the Kondo effect.

In this chapter I describe the crystal and band structures of nanotubes, methods of synthesis and transport measurement techniques.

2.1 Structure

Among the most well-known allotropes of carbon is graphite, which has a hexagonal lattice structure as shown in Figure 2.1. Single sheets of graphite, which are referred to as graphene, have a somewhat unusual band structure — as shown in Figure 2.3, near the Fermi level the bands look like six pairs of cones, each pair meeting apex to apex at the high symmetry K points in the Brillouin zone. The Fermi surface is thus composed of the six K points or six arcs near the K points, depending on the
Figure 2.1: Crystal structure of graphite, an allotrope of carbon, which has a staggered hexagonal structure with a two-layer period as shown. Each sphere represents a carbon atom. Blue (red) lines pass through atoms in blue (red) layers and purple lines pass through both.

Figure 2.2: Hexagonal lattice structure of graphene, showing primitive lattice vectors. Each blue dot represents a carbon atom.
Figure 2.3: Band structure of graphene. Figure reproduced from Reference [2]
position of the Fermi level relative to the Dirac point, the point (on the energy axis) where the cone apexes meet.

Figure 2.4: Examples of nanotubes with different chiralities. Figure reproduced from [3]

Rolling up a sheet of graphene and capping off the hollow ends, one arrives at a new allotrope of carbon called the carbon nanotube, several representatives of which are shown in Figure 2.4. (Note that this it not how nanotubes are actually made. See section 2.2) The way in which the graphene sheet is rolled to obtain any particular nanotube completely determines its structure.

This can be described by the ‘chiral vector’ \( \mathbf{c} \) connecting two carbon atoms on the graphene sheet, which when rolled onto or identified with each other, form the nanotube of interest. Thus, on the nanotube, \( \mathbf{c} \) is a vector wrapped about its circumference.

The chiral vector is usually defined in terms of \( \mathbf{a}_1^* \) and \( \mathbf{a}_2^* \), the primitive lattice vectors of a graphene unit cell shown in Figure 2.2: \( \mathbf{c} = n\mathbf{a}_1^* + m\mathbf{a}_2^* \). Here \( n \) and \( m \) are integers.

This rolling of the graphene sheet imposes boundary conditions on the electron wavefunction in the direction of rolling — as one might imagine, the \( k \) vector is quantised along the chiral vector resulting in lines of allowed \( k \) values in the graphene
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Figure 2.5: Brillouin zone of graphene, showing lines of allowed k imposed by boundary conditions due to rolling up of the graphene sheet to form a nanotube. (a) The allowed k lines do not cross any K points and the nanotube is semiconducting. (b) The allowed k lines cross K points, so the nanotube is metallic.

Brillouin zone as shown in Figure 2.5. The dispersions for the various subbands of the nanotube are thus the corresponding ‘allowed slices’ of the band structure in Figure 2.3. As the distance between subbands is typically on the order of a volt, for the purposes of the transport measurements described in this dissertation — which probe much lower energies — we need consider only the lowest subband, which corresponds to the two allowed k lines which pass closest to K points.

One can therefore see that the electronic properties of any particular nanotube are determined by the chiral vector and the resulting allowed k values. When the allowed k line corresponding to the lowest subband crosses the high symmetry K point, the resulting nanotube is a zero-gap semi-conductor with linear dispersion and is often called ‘metallic’. When, on the other hand, such a line does not cross the K point, the nanotube’s dispersion is a vertical slice of the cone that does not cross its apex, which is to say a hyperbola. These nanotubes are thus semiconducting with finite energy gaps depending on the particular chiral vector.

It is possible to deduce whether a given nanotube is semi-conducting or metallic by considering its chiral vector. When \( n = m \), the nanotube is metallic. \( n - m = 3j \), \( j \in \mathbb{Z} \) and \( j \neq 0 \) nanotubes should be metallic in the picture described above, but the curvature of the nanotube induces small gaps in their energy spectra. They are
thus called small band gap nanotubes. All other nanotubes are large band gap semi-conductors, which is to say that they are generally depletable at room temperature. The molecular structures of several nanotubes of different chiralities are shown in Figure 2.4.

It should be noted that it is not at present possible to control or reliably predict the chirality of nanotubes produced by any growth method.

### 2.2 Growth

The nanotubes used in the Goldhaber-Gordon lab are grown by chemical vapour deposition (CVD). In this process, which is described in much greater detail in Appendix C, nanotubes grow from iron-based catalysts when hydrogen and organic gases are flowed through the growth chamber at high temperatures.

Compared to other methods of producing nanotubes, CVD as it is done at Stanford is much more reliable at producing single-walled nanotubes as opposed to ropes or bundles of nanotubes, or multi-walled nanotubes. In addition, the amount of amorphous carbon produced simultaneously is practically negligible, thus eliminating the need for further purification steps after growth. Finally, for transport experiments CVD provides another advantage — nanotubes can be grown directly on the silicon substrates on which devices will eventually be fabricated. In other processes, where nanotubes are produced in a kind of soot, it is necessary to dissolve the nanotubes in organic solvents such as chloroform, DCE (dichloroethane), TCE (trichloroethane) or ODCB (ortho-dichlorobenzene) and then disperse them on the substrate, usually by drop-casting. Both of these operations introduce additional uncontrollable variables into the process. For all of these reasons, we decided in the end to grow the nanotubes for our devices by CVD; nevertheless, I spent some time early in my PhD learning through experience the difficulties of dispersing commercially available nanotubes grown by other methods. Typical nanotube lengths for our furnace and process conditions are 1-20 $\mu$m and typical diameters 1 - 4nm.

The growth mechanism for the CVD process — and indeed for all nanotube growth processes — is poorly understood; thus, the growth process is uncontrolled and can
be seem somewhat random. With nominally identical process parameters one can get quite different densities and chiralities of nanotubes. There is a tendency for nanotubes to align themselves with the direction of gas flow in the growth chamber but this is not strong or predictable. These unknown quantities in nanotube growth have consequences for device fabrication as described below in Section 2.4.

The CVD growth of nanotubes was pioneered by Hongjie Dai’s lab in the Chemistry department at Stanford, with whom we were collaborating at the start of my PhD. No long afterwards, this collaboration was suspended for several years. Consequently, during this time, I set up a similar CVD furnace for our own lab in 088 Moore.

The principal alternative methods of growing nanotubes are laser ablation, arc discharge and HiPCO (high pressure carbon monoxide).

In laser ablation, a laser is used to evaporate a graphite target mixed with Co or Ni as catalysts. An inert gas such as argon is flowed past the target in the direction of a collector, which is cooled and thus at much lower temperatures. Carbon evaporated from the target condenses on the collector in the form of nanotubes. This method of growth produces primarily ropes of single-walled nanotubes as well as other non-nanotube carbon structures and agglomerations of amorphous carbon.

Arc discharge was the method used to make the first nanotubes. Here a large voltage is applied between carbon electrodes in a background of helium gas. After the discharge between the electrodes, nanotubes can be found to be collected on the cathode. No catalyst is needed to obtain multi-walled nanotubes; various transition and rare earth metals can be used for the growth of single-walled nanotubes. This growth method also produces relatively large quantities of non-nanotube carbon structures.

It is also possible to produce nanotubes in other gas phase processes, where the catalyst particles are generated in situ. Perhaps the most well known of these is the high pressure carbon monoxide (HiPCO) process developed by the late Richard Smalley, who was at Rice University. In this process the decomposition of iron pentacarbonyl provides the catalyst for the growth of nanotubes from CO gas, which is flowed through at very high pressures (up to 10atm).
2.3 Synthesis of peapods

Before we discuss peapods, it is necessary first to introduce the carbon molecule buckminster fullerene, more commonly known as $C_{60}$ or a buckyball. Buckyballs look like footballs (soccer balls) with carbon atoms at every vertex — their molecular structure is shown in Figure 2.6.

![Figure 2.6: Structure of buckminsterfullerene or a buckyball, an allotrope of carbon. Each sphere represents a carbon atom. Red (yellow) lines represent single (double) bonds. Figure reproduced from Reference [4]](image)

Inserting buckyballs into a nanotube like peas into a pod, we obtain the supra-molecular structure called a peapod, a TEM image of which is shown in Figure (insert picture). The first observation of such structures was reported in 1998, but a process for their deliberate synthesis was not developed until 2000 [22]. The peas in peapod samples can be more or less tightly packed; however, in the batch of peapods we studied, most of the nanotubes were tightly-packed with $C_{60}$ as shown in the image.

The peapods in this dissertation were provided by the group of Hiromichi Kataura at AIST, Japan and were synthesised as described in Reference [23]. In the process developed in this group, carbon nanotubes grown by laser ablation with Ni/Co catalyst were purified of amorphous carbon and metal particles respectively using hydrogen peroxide ($H_2O_2$) and hydrochloric acid (HCl). They were then annealed together with buckyballs at 650 degrees Celsius in a sealed ampule for two hours. A further sonication step in toluene was performed to remove buckyballs from the surfaces of nanotubes before the peapods are filtered out of solution in the form of mats.
To fabricate our peapod devices, we sonicated these mats in chloroform or ODCB before drop-casting them on silicon substrates. ODCB-dispersed peapods we further rinsed in acetone and isopropanol to remove the ODCB, which is thought to coat nanotubes well, in order to improve contact between the electrode metal and the nanotube.

An excellent overview of the development of supra-molecular structures where $C_{60}$ and other molecules are inserted into carbon nanotubes is Ref. [22].

Brief note here about the state of theoretical literature on peapods. This is one of the questions addressed in this dissertation.

2.4 Fabricating devices and measuring transport

In all the nanotube and peapod experiments described in this dissertation, the device geometry is as shown in Figures 2.7 and 2.8. The nanotube lies atop a highly-doped silicon wafer which has a surface layer of insulating oxide. Voltages are applied to the main bulk of the wafer which acts as a gate to the device. The nanotube is contacted by source and drain electrodes to which voltages are also applied and the current through the nanotube is measured. Measurement techniques are described in more detail in Appendix B.

There are two primary methods of achieving this geometry, each with its advantages and disadvantages.

In the first, which which is geared more towards mass production, one fabricates many objects (typically about a hundred on each chip) that are lithographically likely to be devices and probes them afterwards to see which have indeed yielded suitable devices. Yields are typically between 5 and 20%. An example of the process flow for this method is shown below.

1. Obtain a highly-doped silicon wafer with a good oxide on its surface of about 100nm. Lately we have begun to grow our own oxides as commercially available wafers often have oxides of inferior quality in the sense that they have low or unpredictable breakdown voltages. It is essential to get the highest
CHAPTER 2. CARBON NANOTUBES AND PEAPODS

Figure 2.7: Side view of typical nanotube device.

Figure 2.8: Top view of typical nanotube device.
possible conductivity wafers as otherwise there will be substantial hysteresis (relaxation time of hours per volt) in the device’s response to the gate at low temperatures. Joseph Sulpizio has worked very hard on this and has found that small differences in conductivity at room temperature can translate into conductivity differences of many orders of magnitude at 4K. The reader may consult his dissertation for more details.

2. **Pattern or etch alignment marks.** We have variously used crosses with numbers next to them as well as hieroglyphs where four square pixels represent each digit. On the whole the former work better. Hieroglyphs may seem like a great idea and an elegant solution, but when one is doing AFM and twisting one’s head around to figure out which way is up one soon realises that one has been too clever for one’s own good. There is almost never such a lack of space that they become a necessity.

3. **Pattern catalyst pads.**

4. **Grow nanotubes in CVD furnace.** Note that only refractory metals such as molybdenum and tungsten will survive this step.

5. **Pattern contact electrodes with electron beam lithography.** Palladium makes best contact to larger-diameter nanotubes.

6. **Anneal chip to decrease contact resistance.** About 200 – 250°C for a few minutes in an inert gas.

As one might guess, the main problem with this method is that the growth of the nanotubes (whether they will grow from a given clump of catalyst, in which direction and how far) is almost completely unpredictable and uncontrollable. Each pair of patterned electrodes may end up with zero, one or more nanotubes spanning them; it is not always possible to tell from simple transport characteristics (conductance against gate voltage) what has happened.

This is in fact the main difficulty facing the field of nanotube transport in general; to be able to control the direction of growth, yield and chirality of nanotubes in
A growth process would represent a major technical breakthrough in this field and enable many experiments which are at present too difficult or simply impossible.

An additional and less serious problem is that probing hundreds of devices can take time especially when there is not a setup dedicated to this task.

Having said this, this is the only way to produce some types of nanotube devices such as the suspended tubes described in Chapter 8.

The second method of fabricating nanotube devices is less oriented towards mass production. Here one first disperses (either by drop-casting or CVD growth) nanotubes on a surface with pre-patterned alignment marks. One then locates them relative to these marks (usually with atomic force microscopy or AFM) and patterns contact electrodes that are appropriately aligned. A typical process flow is shown below.

1. Obtain a highly-doped silicon wafer with a good oxide on its surface of about 100nm.

2. Pattern catalyst pads.

3. Grow nanotubes in CVD furnace. Note that only refractory metals such as molybdenum will survive this step.

4. Pattern alignment marks.

5. Locate nanotubes relative to alignment marks with AFM.

6. Pattern contact electrodes with electron beam lithography.

7. Anneal chip to decrease contact resistance. About 200 – 250°C for a few minutes in an inert gas.

This method has the advantage of having a more predictable outcome; however, locating nanotubes with AFM can be very time-consuming — even an experienced microscopist will require half a day to a full day to find half a dozen likely candidates for contacting on a single chip. In addition, while one can tell quite a lot about a nanotube by its height, location and shape as determined by AFM, one still does
not know anything about its chirality (and thus expected transport characteristics) until one actually measures the device. Nevertheless, this has become our preferred method for most of our experiments.

2.5 Bibliographical Note

There has recently been published a review article on the properties of carbon nanotubes. [24] Other than that, the reader may also consult more general works such as References [25] and [26].
Chapter 3

Quantum Dots and the Kondo Effect

3.1 Quantum Dots

In introductory quantum mechanics, one of the first problems one encounters is that of a particle confined to a small region in space — the ‘particle in a box’. (Figure 3.1a.) There one learns that the allowed energies of such a particle become quantised. Here we shall call the average spacing between these quantised energy levels $\Delta$. In recent years it has become possible to fabricate through a variety of methods electrons systems small enough that this textbook problem has become a reality. ‘Quantum dot’ is the name given to any object sufficiently small that its energy levels are detectably quantised, i.e. where $\Delta \gg k_B T$.

The quantised energy levels of quantum dots may call to mind a class of naturally-occurring systems with the same property — atoms. Quantum dots have thus also been called ‘artificial atoms’ — like real atoms their energy levels are quantised; however, unlike atoms, quantum dots have the advantage that many of their properties can be designed or tuned, as shall be explicated below, thus making possible a variety of experiments not hitherto possible.

In the transport measurements done in the Goldhaber-Gordon lab, source and drain electrodes are attached to the quantum dot and usually there are also nearby
Figure 3.1: (a) Box with five electrons occupying single-particle energy levels. Interactions between particles have been ignored. \( \Delta \) is the average energy spacing between levels. (b) Transport measurement of a quantum dot. Levels are filled up to the Fermi energy of the leads (‘source’ and ‘drain’) and a capacitive charging energy \( U \) has to be taken into account. The walls of the box also have to be at least slightly leaky in order for current to be measured. In this chapter, ‘source’ and ‘drain’ are just names for particular leads — the source is not necessarily at a higher potential than the drain and we could equally have called them ‘A’ and ‘B’. This is a common convention in the literature.
electrodes called gates which couple capacitively to the dot. (Figure 3.1b.) The interactions between electrons on the dot and of the quantum dot with its environment and practical considerations of the measurement now introduce other relevant parameters which immediately make themselves apparent.

For instance, when one tries to add an electron onto the dot, one finds that one requires not only the level spacing $\Delta$ but also an additional energy $U$ due to the capacitance of the quantum dot. As one might naïvely expect, at least to within some factors of 2 and $\pi$, $U = \frac{e^2}{C}$ where $C = \sum_i C_i$ is the sum of the capacitances of all electrodes to the quantum dot. In many experimental systems, $U$ is on the same order of or larger than $\Delta$.

In addition, in order to be able to measure anything at all, the barriers of the quantum dot have to be at least slightly leaky — electrons have to be able to tunnel on and off the dot at some small rate. The leakiness of the barriers are characterised by transmission coefficients $t_{\alpha}$, where $\alpha = s, d$. These coefficients can of course in principle be dependent on any number of things, such as temperature, the state of the dot being tunnelled into and so on, but it is not necessary at the moment for us to pay attention to these things.

The considerations above allow us to surmise that the Hamiltonian for the system at this point looks something like the following.

$$H = \sum_\nu \epsilon_\nu c_\nu^\dagger c_\nu + \frac{e^2}{2C} \left( \sum_\nu n_\nu \right)^2 + \sum_{\nu k} \left( t_{sk} c_\nu^\dagger c_k + t_{sk}^* c_k^\dagger c_\nu \right) + \sum_{\nu l} \left( t_{dl} c_\nu^\dagger c_l + t_{dl}^* c_l^\dagger c_\nu \right)$$

(3.1)

Here $c$ and $c^\dagger$ are annihilation and creation operators; and $\nu$, $k$ and $l$ label states respectively on the dot, on the lead known as the 'source' and on the 'drain'. The first term is thus the sum of all the single particle energies in the dot and the last two represent tunnelling between the dot and the two leads. The second term is the energy of a classical capacitor — $q^2/2C \equiv U/2$. It is non-trivial to see how we get from this term in the Hamiltonian to the statement above that one adds a 'charging energy $U$' for each electron. For a detailed description, the reader may consult Reference [27] where this is laid out in more detail. The third term describes processes where an
electron hops on (off) the dot from (to) the source while the fourth term describes similar processes involving the dot and the drain.

The reader should note that there are now at least four important energy scales in the problem that we are aware of: (1) $U$, the charging energy; (2) $\Delta$, the average level spacing; (3) $kT$, the temperature, which will affect the distribution of occupied states and (4) much less obviously, energies related to the leakage rates through the barriers — $\hbar \Gamma_s$ and $\hbar \Gamma_d$, where $\hbar$ is Planck’s constant and roughly speaking $\Gamma_s \propto |t_s|^2$ and $\Gamma_d \propto |t_d|^2$. (The reader may consult Chapter 10 of Reference [14] for details such as sums over states and factors of $\pi$.)

3.2 Transport at Zero Bias: Coulomb Blockade

Now consider a transport measurement through a quantum dot which has source and drain electrodes (leads) attached and a gate electrode (or several) nearby. (Figure 3.2.) An infinitesimally small AC excitation voltage is applied between source and drain and the resulting current measured. (‘Small’ here means that $eV < k_B T$.) The quantity measured here — $dI/dV$ at zero $V$ — is called the ‘linear conductance’ as opposed to the ‘differential conductance’ which is the same quantity measured with a finite DC component of $V$.

If we assume that both the temperature $kT$ and the leakage rates $\hbar \Gamma_s, \hbar \Gamma_d$ are very much less than the level spacing $\Delta$, in most situations the configuration of the dot looks something like Figure 3.2a. There is a well-defined number of electrons on the dot and current does not flow through it as there are only inaccessible virtual states by which electrons can cross from source to drain through the dot. By changing the voltage on a gate, the levels in the dot can be capacitively tuned relative to the source and drain electrodes as described in the previous section. At particular values of gate voltage, when there is an alignment of a level in the dot with the leads as in Figure 3.2b, the number of electrons in the ground state of the system can fluctuate between some integer $n$ and $n + 1$ — current flows through the dot.

Therefore, when one measures the linear conductance of the dot against gate voltage one should see that current flows only at specific and relatively periodic points
CHAPTER 3. QUANTUM DOTS AND THE KONDO EFFECT

Figure 3.2: (a) A quantum dot in a stable configuration with $n$ electrons on it. Source and drain leads are between allowed energy levels and no current flows. (b) A change in gate voltage changes the energy levels of the dot relative to source and drain. Here the leads are aligned with an allowed energy state and current flows through the dot. (c) A trace of conductance against gate voltage taken in a nanotube device on 22 April 2003. Conductance is finite only in the region of certain gate voltage for which the situation in (b) is true. This phenomenon is known as the Coulomb blockade.
in gate voltage — and this is indeed what is observed, as shown for example in Figure 3.2c. this phenomenon is referred to as the Coulomb blockade (CB).

What can we learn about a quantum dot from its Coulomb blockade peaks?

Firstly, the spacing between peaks tells us something about the energy levels in the dot. Note, however, that this is not in units of energy as we should like but scaled by the unknown capacitance of the gate to the dot. To convert this to energy, further measurements must be made at finite bias as described in the following section.

Secondly, the height of the CB peaks depend on the overlap of the wave function of the aligned state with the leads. A review of work on this can be found for example in Reference [28].

Thirdly, the lineshape of the peaks tells us about the relative sizes of the different energy scales in the problem. In what follows, we assume that transport occurs primarily though one level in the dot at once, i.e. that \( k_B T \ll U, \Delta \), which is the case for all of the measurements in this dissertation. (Other regimes of \( k_B T \) with respect to \( U \) and \( \Delta \), where transport occurs through many levels in the dot, are discussed for example in Ref [28].) We shall then consider different regimes of the barrier transmissions \( \Gamma_s \) and \( \Gamma_d \) with respect to temperature, assuming for the moment also that \( \Gamma_s \approx \Gamma_d \), which is to say that there is no great asymmetry between the barriers.

In the case where \( h\Gamma_s, h\Gamma_d \ll k_B T \), the CB peaks are broadened by temperature and the lineshape is given by

\[
G = \frac{\overline{G}\beta\Delta}{4} \cosh^{-2}\left(\frac{\beta\delta}{2}\right) \tag{3.2}
\]

where \( \beta = 1/k_B T \); \( \delta \) is the distance from the resonance peak in units of energy; and \( \overline{G} \) is the conductance in the limit of high temperature — \( k_B T \gg U \).

When \( h\Gamma_s, h\Gamma_d \gg k_B T \), the CB peak lineshape is instead of the form

\[
G = \frac{e^2}{2h} \frac{(h\Gamma/2)^2}{(h\Gamma/2)^2 + \delta^2}. \tag{3.3}
\]

where \( \Gamma = \Gamma_s + \Gamma_d \).

This equation is one that shows up in many places in physics and has many names
— it is characteristic of a resonance broadened primarily by a finite lifetime. In optics, it is the transmission of a Fabry-Perot cavity and is called a Lorentzian. In atomic physics it appears as the Breit-Wigner formula describing nuclear scattering.

A simple way to distinguish between these two lineshapes (and to tell whether the peaks in transport through one's quantum dot is temperature- or $\Gamma$-broadened) is to plot the CB peaks with a logarithmic scale for the conductance and a linear scale for the gate voltage (which is linearly proportional to $\delta$). With these axes, temperature broadened peaks have straight tails, whereas the Lorentzian will appear to have concave tails. (To see this, the reader should plot the mathematical formulae for both line shapes in Matlab or else do a series expansion far away from the peak.)

The case of very strong coupling, where the Kondo effect is observed, will be discussed later in this chapter.

### 3.3 Transport at Finite Bias: Coulomb Diamonds

Now consider the case where a finite bias voltage is applied to the quantum dot. We assume here that a symmetric bias is applied which is to say that $V_L = -V_R = V/2$ and also that the source and drain electrodes are symmetrically capacitively coupled to the dot. This ensures that we can neglect any capacitive effects from the source and drain as we apply a bias voltage. In real life this is often not true of course but again this is another detail that can be added in later — it is not necessary for understanding the main features of the data.

Imagine that the source and drain are between two energy levels with the assumptions above. No conduction is allowed as we are in a blockaded region, but as the bias voltage is increased, eventually we reach the situation shown in Figure 3.3a where the source is aligned with the lowest unoccupied state (LUDS) in the dot and the drain with the highest occupied state (HODS). Then current can flow. If we now consider starting not exactly in between two blockade peaks but closer to one of them than the other, we can see that a smaller bias voltage is required to achieve conductance. (Figure 3.3b.) Only one lead needs to be aligned with either the LUDS or HODS in order for current to flow.
Figure 3.3: (a) Starting with leads right in between two allowed energy states (dotted lines, arrow to (c)) a voltage can be applied so that current flows. A symmetrically applied bias and symmetric couplings to source and drain are assumed. (b) Starting closer to one allowed state than another (dotted lines, arrow to (c)) a smaller voltage is needed. (c) Mapping out all of the parameter space in gate and bias voltages, regions of no conductance are blue and red means that conductance is allowed. These features are known as Coulomb diamonds.
If, in a 2D plot of conductance against bias and gate voltages, we colour in the regions where conductance is expected, we find a diamond-like structure as shown in Figure 3.3c. The slope of the sides of the diamond give the very useful conversion between energy and gate voltage for the particular gate being swept.

Figure 3.4: A closer look at a Coulomb diamond where the dot has asymmetric couplings to its leads. Red regions are where there are conductance and blue where there is no conductance. U measures the charging energy.

Now let us spend a little more time identifying the events marked by the edges of the diamonds — which lead lines up with each level. By thinking about things for a bit the reader can ascertain that the identification of the edges as in Figure 3.4 is true and also that the height of the diamond gives the charging energy $U$, or strictly speaking the charging energy plus the level spacing. Note that in the figure we have arbitrarily identified one lead as ‘source’ and the other as ‘drain’ and defined the bias voltage as $V_s - V_d$. This is a common convention in the literature.

We have also added the complication of slightly asymmetric couplings to the two leads. The effect of this is to distort the diamonds so that they are no longer four-fold
symmetric but appear slightly slanted as shown in the figure. From the distortion of the diamonds, one can obtain the relative capacitive couplings of the two leads using the following relation, which I also encourage the reader to think about.

\[
\frac{\gamma}{\beta} = \frac{\text{capacitance of dot drain}}{\text{capacitance of dot to source}}.
\]  

Thus, assuming that there are only two leads and one gate to a quantum dot, in principle, its Coulomb diamonds allow us to determine the capacitance of each electrode to the dot.

Additional structure is often observed inside and outside the Coulomb diamonds which gives us more information about the dot. Some of these are relevant to the work in this dissertation and will be described in what follows. For everything else, I refer the reader to Andreas Fuhrer’s dissertation done under Klaus Ensslin at ETH Zürich.

### 3.4 The Kondo Effect

The Kondo effect – the screening of a local moment by a reservoir of conduction electrons – is perhaps the simplest many-body effect and hence holds a special place in the study of correlated electron systems [29]. In bulk metals, in which the Kondo effect was first studied, it manifests itself as an increase in resistance at low temperatures. (Figure 3.5, Reference [5].) This anomalous resistance increase was first observed in the 1930s but for a long time could not be explained. It was expected that the resistance of metals should decrease with decreasing temperature as phonon modes were frozen out and that the resistance should saturate to a constant value at low temperatures due to defects and impurities in the metals. Instead, what was observed was a rise in resistance below what is now known as the Kondo temperature.

In the 1960s experimental work was done — primarily by Myriam Sarachik — which associated this puzzling resistance increase with the presence of magnetic impurities. (Figure 3.6, References [30, 6].) About the same time a theoretical explanation was also proposed by Jun Kondo for this phenomenon [31].
Figure 3.5: Figure reproduced from Reference [5] showing resistance minimum in temperature in two different gold samples.
Figure 3.6: Figure reproduced from Reference [6].
CHAPTER 3. QUANTUM DOTS AND THE KONDO EFFECT

Before discussing Kondo’s solution of the resistance minimum problem, let us first consider the Philip Anderson’s earlier work describing the formation of local magnetic moments in bulk metals. [32] The Anderson Hamiltonian is

$$H = \sum_{\nu} \epsilon \nu c_{\nu}^\dagger c_{\nu} + \sum_{\nu k} \left( t_{k,\nu} c_{\nu}^\dagger c_{\nu}^{\dagger k} + t_{k,\nu}^* c_{\nu}^{\dagger k} c_{\nu} \right) + U n_{d\downarrow} n_{d\uparrow}$$ (3.5)

This is essentially the same as the Hamiltonian we had earlier for the quantum dot except that now (1) instead of the ‘dot’ we have an impurity atom, (2) instead of the leads we have the bulk metal as the reservoir of electrons, (3) we consider only the subspace of states with ‘0’, ‘1’ or ‘2’ electrons on the impurity and (4) $\epsilon_{\nu}$ is the same for all states, so we drop the subscript and take it outside the sum.

The first term gives the energies of the electrons on the impurity, the second describes tunnelling between the impurity and the conduction sea and the third describes interactions on the impurity.

There are two conditions for the formation of a local moment. (1) $n = 1$ must be the ground state — $U/2 > |\epsilon + U/2|$. (2) Compared with their repulsion due to the intra-impurity interaction $U$, the states on the impurity cannot be mixed too much by interactions with the conduction sea — $U > U_c \sim \pi \Gamma_b$. Here $\Gamma_b$ is the broadening of the level due to the second term and is given by Fermi’s Golden Rule: $\Gamma_b(E) = \pi \sum_{k} |t_k|^2 \delta(E_k - E)$. A very readable account of the calculations leading up to these conditions can be found in Reference [33].

The Kondo model is a simplification of the Anderson model and has the following Hamiltonian.

$$H = \epsilon \sum_{\nu} c_{\nu}^\dagger c_{\nu} + \sum_{\nu\nu'\alpha\beta} J_{\nu\nu'} c_{\nu\alpha}^\dagger s_{\alpha\beta} c_{\nu'\beta} \cdot \hat{S}$$ (3.6)

Here $\hat{S}$ is the spin on the impurity, $\vec{\sigma}$ are the Pauli spin matrices and

$$J_{\nu\nu'} = t_{\nu\nu'}^* \left( \frac{1}{U + \epsilon + E_F} + \frac{1}{E_F - \epsilon} \right)$$ (3.7)

if the temperature is sufficiently low so that only electrons in the conduction sea close
to the Fermi level participate in interactions with the impurity. Another way to put it is that the spread in energies in the conduction sea is small compared to the other terms in the denominators above i.e. $kT \ll U$. This, as noted earlier, is true for all the measurements considered in this dissertation.

The Kondo Hamiltonian is derived from the Anderson by performing a unitary transformation which eliminates terms which linear in $t$ and neglecting higher order terms. This was first done by Schrieffer and Wolff [34]. The original paper is quite readable, but the transformation is also described perhaps more pedagogically and in some detail in Chapter 10 of Reference [14] and also in Reference [33].

In the Kondo Hamiltonian one can more transparently see that magnetic impurities at low temperatures will tend to anti-align with the spins of conduction electrons in their vicinity, forming a singlet state. The increased scattering due to these bound states leads to a resistance higher than that expected for simple non-magnetic impurities.

Kondo's original perturbative solution of the Hamiltonian named after him was able to predict the $\ln(T)$ dependence of the resistance, but failed as $T \to 0$ where the measured resistivity does not continue to diverge logarithmically. The full solution of the Kondo Hamiltonian at all temperatures, even in equilibrium and at zero magnetic field, had to await the numerical application of Wilson’s ‘renormalisation group’ technique and an exact analytical solution was later found using the method of the Bethe ansatz. [35]

### 3.5 The Kondo Effect in Quantum Dots

As can be seen from the reverse exercise above, the Kondo and Anderson Hamiltonians can be quite straightforwardly mapped from the ‘impurities in a bulk metal’ problem onto the problem of a quantum dot coupled to leads.

A recent resurgence of interest in the Kondo effect was spurred by the ability to measure transport through a single magnetic site: a quantum dot with an unpaired spin [36]. Here, analogous to the situation in bulk metals, the unpaired spin on the quantum dot forms a spin singlet with conduction electrons in the leads. Unlike bulk
metals, however, the increased scattering now gives rise to an increase in conductance near zero bias.

In the Coulomb diamond measurements shown earlier, the Kondo effect thus manifests itself as a stripe of increased conductance about zero bias in regions where there is an unpaired spin — usually diamonds in which there is an odd number of electrons on the dot.

The tunability of this novel Kondo system — the quantum dot — has enabled tests of long-standing theoretical predictions, for example, finite magnetic field and finite bias behaviour as explicated the following two chapters.

3.6 Bibliographical Note

Ethan Bradley Foxman’s dissertation (under Marc Kastner at MIT) contains an excellent description of early experiments on GaAs quantum dots. Reference [28] is a good review article on quantum dots focusing mostly on experiments while Reference [37] is more theoretical.

In writing the above description of the Kondo effect I drew most heavily from References [33], [14] and [38]. In the lectures by Coleman [33] the Kondo effect is discussed primarily in the context of local moment physics, whereas the book by Bruus and Flensberg places the discussion in a chapter on quantum dots. Altland and Simons’s book [38] has many illuminating problems relating to the Kondo effect and outlines of solutions. The book by Hewson [35] is considered the standard reference on the subject, and as a reference it is indeed valuable, but I would not recommend it as an introduction as the reader can be easily overwhelmed by details that are not immediately relevant.

The introduction to the Hewson book [35] nevertheless provides a good historical overview of the theoretical methods applied to the Kondo problem as, to a lesser extent, do the Coleman lectures [33]. For an introduction to the RG in a condensed matter context, the reader is encouraged to consult References [39] and [40]. Piers Coleman has also written an excellent short article on the development of the field of many-body physics, putting both the Kondo problem and the RG in context. [41]
Chapter 4

Transport Properties of C\textsubscript{60} Peapods

4.1 Motivation

In recent years, it has become possible to synthesize supra-molecular structures by inserting smaller molecules such as C\textsubscript{60} fullerenes into nanotubes to form ‘peapods’. Early experiments have shown that the inclusion of fullerenes modifies the electronic structure of a nanotube at energies far from the Fermi level, and that a peapod's conductance can depend on the choice of encapsulated species, raising the prospect of novel transport phenomena in these molecules.

In studying peapods, we were motivated primarily by the question: what does transport through a 1D system with a periodic potential look like, especially as interactions are taken into account?

Consider first a 1D system with a periodic potential imposed. Here we see the formation of allowed bands of energies analogous to the bands in 3D crystals that the reader might be more familiar with. In nanotubes, we expect the formation of gaps in the spectrum whenever the density $\rho$ is equal to $4m/\lambda$ where $m$ is an integer and $\lambda$ is the spatial period of the potential.

In the presence of interactions but no periodic potential, a 1D system can form a Wigner crystal where electrons arrange themselves as shown in Figure 4.1b.
Finally, putting a periodic potential and interactions together (Figure 4.1c), it has been predicted that the picture should be not so different from that in Figure 4.1a except that \( m \) can be a fraction rather than an integer. \([7, 45]\) In transport, these gaps should appear as dips in the conductance at zero bias voltage and at gate voltages corresponding to the appropriate densities; however, as described below we do not see these.

Figure 4.1: (a) Formation of ‘mini-bands’ in 1D periodic potential with no interactions. (b) Formation of Wigner crystal in 1D system with interactions. (c) Formation of incompressible states in 1D system with a periodic potential and interactions. Figure reproduced from Reference [7].

### 4.2 Measurements

In this chapter we report measurements of the conductance of carbon nanotube peapods at temperatures from 250mK to room temperature. We were surprised to find that despite the addition of C\(_{60}\) molecules our devices exhibit a range of low-energy transport behavior similar to that previously seen in empty nanotubes. At room temperature, the nanotubes are semi-conducting or metallic; at low temperature, we observe Coulomb blockade, and both spin-1/2 and spin-1 Kondo effects. Here
we discuss the overall behavior of our ensemble of devices and make some statistical statements about them. The Kondo effects are described in detail in the following chapters.

Our devices are carbon nanotube C$_{60}$ peapods contacted by palladium source and drain electrodes, 150nm to 500nm apart. The peapods lie on a 500nm or 1$\mu$m thick thermal oxide atop a highly-doped silicon substrate, which acts as the gate. The peapods are synthesized by the sublimation technique described in Reference [23]. They are then dispersed by sonication in chloroform or ortho-dicholorobenzene. The dispersion is deposited on the substrate and allowed to dry. We locate the peapods relative to pre-existing alignment marks using atomic force microscopy (AFM), and fabricate the electrodes using standard electron-beam lithography techniques. All nanotubes studied are 1-4nm in diameter according to AFM measurements.

Figure 4.2: (a) TEM image of bundles of carbon nanotubes deposited from our chloroform suspension, most filled with C$_{60}$. Arrows point to unobscured single nanotubes representative of those counted in our analysis — black arrows to filled tubes, white ones to unfilled. The scalebar is 30nm long. (b) A single nanotube filled with C$_{60}$ peas. The scalebar is 5nm long. (c) A bundle of nanotubes viewed at an angle, showing the C$_{60}$ molecules inside. The scalebar is 5nm long.
Figure 4.2 shows representative transmission electron microscopy (TEM) images taken of nanotubes deposited from our dispersion. We deposited electrodes on 20 different nanotubes, of which 7 were found to be conductive at room temperature. The other 13 nanotubes are discounted from the analysis that follows as they are likely not connected due to handling or alignment problems. [46]

Next, we perform a statistical analysis of our group of 7 nanotubes in light of TEM images of many other nanotubes deposited from the same ensemble. Such a statistical analysis is crucial, as no synthesis method yields 100% filled peapods, and it is impractical to verify directly that a given nanotube in transport studies is filled with fullerenes — TEM is the only established method for differentiating between filled and unfilled carbon nanotubes, and the specimen requirements for TEM imaging are incompatible with the standard geometry of nanotube transistors. From images such as those in Figure 4.2, we identify 109 single nanotubes, which were unobscured and in focus. Of these, 92 nanotubes are filled, and 17 empty. We observe no partially-filled tubes. We also note that these tubes were deposited in the same way as those in the devices, i.e. by drop-casting from solution.

![Figure 4.3](image_url)

Figure 4.3: Calculated probabilities, using Bayes's Theorem, for the number of filled nanotubes in our sample of 7, taking into account the proportion of filled nanotubes in our TEM images (92 out of 109).

Taking into account the frequency of filling in the nanotubes examined by TEM and assuming no prior knowledge of the fraction of filled tubes, we use Bayes's Theorem for continuous probability distributions [47, 48] to evaluate the probability that
our 7 measured nanotubes included any specific number of filled tubes from 0 to 7. This information is presented in Figure 4.3. The expected number of filled tubes is found by this method to be 5.86. Details of these calculations are included in the Appendix; however, we would like to emphasize here that our approach is more conservative than simply taking 92/109 as the fraction of filled nanotubes, yielding a higher calculated probability that many of our nanotubes are unfilled.

![Figure 4.3](image)

**Figure 4.3:** Our devices, which include some peapods (see Figure 4.3) show a range of room-temperature transport properties indistinguishable from those of unfilled nanotubes. (a) Room temperature linear conductance traces for devices exhibiting some (blue line, right axis) and no (black line, left axis) change as the gate voltage is swept. (b) Room temperature linear conductance of a completely depletable semiconducting device.

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Returning to the transport properties, Figures 4.4 and 4.5 show representative measurements of the conductance of our devices as a function of gate voltage at room temperature and at 250mK.

In the room temperature measurements, we observe devices with conductances significantly modified by the gate as well as ones that are unaffected by it (Figure 4.4a):
'semiconducting' and 'metallic', respectively, in the conventional description of carbon nanotubes. [49, 50] Only a few devices with rather low overall conductances are completely depletable (Figure 4.4b).

Figure 4.5: (a) At 250mK, device showing Coulomb Blockade. (b) Also at 250mK, device with higher conductance showing Coulomb Blockade. (Inset) Detail showing regularity of peaks, which continues over the whole range. (c) Conductance versus bias and gate voltage of device in (b). The color scale is blue (low) to red (high conductance). Regular diamonds indicate that this is a single quantum dot.

At 250mK-350mK, two of the seven nanotubes in our ensemble have undetectably low conductance. The question naturally arises as to whether these, and only these, are peapods. As seen in Figure 4.3, we find that the probability that three or more of
CHAPTER 4. TRANSPORT PROPERTIES OF $C_{60}$ PEAPODS

<table>
<thead>
<tr>
<th>Device Label</th>
<th>Tube length ($\mu$m)</th>
<th>Intercontact distance (nm)</th>
<th>Oxide thickness ($\mu$m)</th>
<th>$U$, charging energy (meV)</th>
<th>Predicted $U$ (meV)</th>
<th>Predicted $U$ for entire tube (meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>2</td>
<td>500</td>
<td>0.5</td>
<td>3-5</td>
<td>9-11</td>
<td><strong>2.3-2.8</strong></td>
</tr>
<tr>
<td>B*</td>
<td>5.5</td>
<td>500</td>
<td>0.5</td>
<td>5-10</td>
<td><strong>9-11</strong></td>
<td>1.7-2</td>
</tr>
<tr>
<td>C</td>
<td>4</td>
<td>250</td>
<td>0.5</td>
<td>15-40</td>
<td><strong>18-23</strong></td>
<td>1.1-1.4</td>
</tr>
<tr>
<td>D*</td>
<td>2</td>
<td>250</td>
<td>0.5</td>
<td>1-5</td>
<td>18-23</td>
<td><strong>4.6-5.6</strong></td>
</tr>
<tr>
<td>E1</td>
<td>4</td>
<td>250</td>
<td>1</td>
<td>3-4.5</td>
<td>20-24</td>
<td>2.2-2.4</td>
</tr>
<tr>
<td>E2</td>
<td>4</td>
<td>150</td>
<td>1</td>
<td>1-3</td>
<td>34-41</td>
<td><strong>3-4.1</strong></td>
</tr>
</tbody>
</table>

Table 4.1: Correlation of energy scales with device geometry. Each letter corresponds to a different tube. For tube E, results are reported for transport between two different pairs of contacts (E1, E2). Charging energies are experimentally determined from intersection of lines with opposite slopes in Coulomb diamond plots such as Fig. 4c. For comparison, charging energies are predicted for electrons confined to a stretch of nanotube between two neighboring metal contacts, assuming a tube diameter of 1-4 nm [1]. These predictions are dramatically off for some devices – in those cases charging energy is instead consistent with electrons delocalized over the entire tube. Whichever prediction is consistent with our measured value is shown in bold. Note: when we observe double dot behavior (tubes B and D), each of the two predictions assumes that the relevant tube is divided into two equal segments, each acting as one dot. See text for details and discussion. * These devices act as double dots.

our tubes in this sample of seven are filled is 99.75%. (See Appendix for details.) It is therefore practically a certainty that one or more of our quantum dot devices are formed on peapods.

All devices measurable at low temperature show Coulomb Blockade behavior, but with widely varying peak conductances. Representative data are shown in Figure 4.5. Devices on all five tubes show Coulomb diamonds in measurements of conductance versus gate and bias voltages (Figure 4.5c is representative), indicating that each device acts as a single or double quantum dot.

In Table 1 we summarize results from devices on the five tubes which conduct at low temperature. We compare measure charging energies to estimates obtained by using the classical expression for capacitance between a metallic wire and a metallic plane, $C = 2\pi \varepsilon L / \ln(4h/d)$, and $U = e^2 / C$. [1] Here $d$ is the tube diameter, $L$ the distance between contacts, $h$ is the oxide thickness and $\varepsilon = 3.9\varepsilon_0$ the dielectric.
CHAPTER 4. TRANSPORT PROPERTIES OF $C_{60}$ PEAPODS

permittivity for SiO$_2$.

For two of the five tubes our measured and calculated values agree; however, in three tubes (four devices) the measured charging energies are lower than would be expected if the quantum dot or double dot were delimited by the contacts. The most likely explanation is that in those cases the contacts do not break the tube into electrically-separate regions – rather, the quantum dot(s) extend beyond the contacts, as was observed in one previous experiment in which a tube was draped across multiple contacts [51]. Assuming for tubes A and E that electrons are delocalized over the entire length of the tube, and for tube D that the entire length of the tube acts as a double dot, yields predicted charging energies consistent with our measured values (Table 1). (See section 4.3 for details.) As delocalization of electrons in a nanotube beyond a junction with a metal contact deposited on top of the tube is previously unknown, we speculate that intercalation of $C_{60}$ makes nanotubes more radially rigid and more difficult to crush.

This possibility of a structural effect of $C_{60}$ intercalation is intriguing and deserves further study, but it does not impact the main conclusions of our work on the electrical effects. If, as argued above, some of the devices in Table 1 are formed from peapods, the observation of Coulomb diamonds leads to the conclusion that the encapsulated $C_{60}$ does not introduce substantial backscattering of electrons passing through a nanotube: electrons are delocalized over hundreds of peas. This somewhat surprising result is consistent with data from recent photoemission studies. [52] The absence of backscattering in peapods may be due to the long wavelength of the perturbation introduced by the encapsulated $C_{60}$. Due to an unusual bandstructure, backscattering in single-walled carbon nanotubes is expected to require a very large momentum transfer, which can only be produced by a nearly atomically sharp perturbation or a perturbation so large that it locally depletes the tube.

To our knowledge, there have been only two previous or contemporaneous reports of transport measurements on nanotubes believed to contain $C_{60}$ molecules (though several studies have been published on metallofullerene peapods). (1) Yu et al. found modulation of conductance by gate and bias voltages on large energy scales, and weak conductance at zero bias, suggesting formation of multiple dots in series within
the nanotube [53]. In contrast, we always see conductance at zero bias and see Coulomb blockade behavior consistent with formation of single or double dots. These differences between our results and theirs may be due to the irregular spacing of C60 peas in their nanotube, in contrast to the regular spacing in our tubes (Fig. 1). (2) Since the first submission of this manuscript, Utko et al. have reported measurements which substantially agree with ours. They find single dot behavior in several tubes from an ensemble of peapods. Their observed charging energies are consistent with electrons in a dot delimited by metal contacts – unlike in our study, in which contacts are narrow (250 nm) palladium strips that cross the tubes, Utko et al. cover each tube entirely with palladium metal except for a short portion through which they measure transport. [54]

4.3 Analysis of charging energies

![Figure 4.6: Common device geometry for transport measurement through carbon nanotubes, top view.](image)

There is a large body of experimental literature on transport through nanotubes. In most cases the device geometry is as shown in Figure 4.6, with metal contacts either above or below the nanotube. When the coupling between the tube and the contacts is not very good (which is to say in most cases) the nanotube acts as a quantum dot.

In most cases, the quantum dot is found to be delimited by the contacts. This is thought to be because the nanotube is crushed by the contacts (if it is beneath them...
as shown above) or because kinks are formed in the nanotube as it is draped across the contacts (as shown in Figure 4.7).

![Figure 4.7: Common device geometry for transport measurement through carbon nanotubes, side view.](image)

**Nanotube A**

While the scenario described above is true for the majority of cases, it can sometimes happen that the quantum dot is delimited by the length of the nanotube rather than by the contacts (see picture below and Reference [55]). This leads to lower charging energies than would be expected if the quantum dot were defined by the contacts.

![Figure 4.8: The quantum dot can be formed over the whole nanotube instead of being delimited by the contacts.](image)

For example, assuming that the whole 2μm length of nanotube A above acts as a
single dot, the charging energy $U$ is $2.3 - 2.8\text{meV}$ according to $C = 2\pi e L / \ln(4h/d)$ and $U = e^2/C$. (Reference [1])

(Here $L = 2\mu m$, $h = 0.5\mu m$, $d = 1 - 4nm$.)

This is much closer to the observed value ($U = 3 - 5\text{meV}$) than the calculation in Table 1 which assumes that the dot is formed between the contacts.

We find that this explanation allows us to account for all of the unexpectedly low charging energies as shown in the following pages. We speculate that the addition of C$_6$0 makes nanotubes more radially rigid and difficult to crush.

**Nanotube E**

![Diagram of Nanotube E](image)

Figure 4.9: Nanotube E.

If we assume that the whole $4\mu m$ length of tube acts as a single dot, the charging energy $U$ is $1.3 - 1.5\text{meV}$ according to $C = 2\pi e L / \ln(4h/d)$ and $U = e^2/C$.

In this case we would have $L = 4\mu m$, $h = 1\mu m$, $d = 1 - 4nm$.

If we assume that there is one defect that splits the tube up into two parts of lengths $1.5\mu m$ and $2.5\mu m$, then we get charging energies
CHAPTER 4. TRANSPORT PROPERTIES OF C\textsubscript{60} PEAPODS

\[ U_1 = 3.4 - 4.1 \text{meV} \]  
\[ U_2 = 2 - 2.4 \text{meV} \]

which agree with what is observed in the two devices on this tube: \( U_1 = 3 - 4.5 \text{meV} \) and \( U_2 = 1 - 3 \text{meV} \).

Nanotube D

\[ \text{quantum dot} \quad \text{defect/barrier} \quad \text{quantum dot} \]

Here we appear to have a defect between the contacts as the device looks like a double dot.

If we assume that the 2\( \mu \text{m} \) length of tube is split into two equal parts, we have \( L = 1 \mu \text{m}, h = 0.5 \mu \text{m}, d = 1 - 4 \text{nm} \).

The charging energy \( U \) is then 4.6 - 5.6meV according to \( C = 2\pi eL/\ln(4h/d) \) and \( U = e^2/C \) which is close to the observed value, \( U = 1 - 5 \text{meV} \).

4.4 Conclusions

We have measured the transport properties of carbon nanotube samples including some C\textsubscript{60} peapods at room temperature and at 250-350mK, and have done a careful statistical analysis of such an assembly of devices. Our results indicate that C\textsubscript{60}
peapods do not differ collectively from nanotubes in their electronic transport characteristics. We note that this complements earlier STM work, [42] where C$_{60}$ peas were found to induce significant perturbations in electronic structure of a nanotube only at much higher energies than are accessed in our present measurements.

Photoemission studies nevertheless suggest that other peapod species may yield more exotic behavior in transport — for example, a Tomonaga-Luttinger- to Fermi-liquid transition with increased potassium doping. [56] A more detailed picture of the range of transport properties of peapods may emerge when transport measurements can be combined with \textit{in situ} structural characterization. Meyer et al. [57] have commenced work in this direction.
We introduced the Kondo effect in Chapter 3. There it was mentioned that there has been a resurgence of interest in this well-studied effect with the advent of quantum dots, which afford greater tunability of various parameters in the problem. A great deal of work has been done but behavior at finite magnetic field remains controversial and ripe for quantitative experimental study. Magnetic field is a particularly important tool for studying the Kondo effect: as in many correlated-electron systems, the many-body physics in this system stems from local spin fluctuations, to which magnetic field couples strongly.

Quantum dots formed from carbon nanotubes are particularly well-suited to studying the Kondo effect in magnetic field. Advantages of nanotube-based dots relative to the gated GaAs-based dots in which Kondo effect has been more thoroughly studied include: (1) High Kondo temperature. The strength of the Kondo effect is characterized by the Kondo temperature $T_K$. Within a simple model, $T_K \sim \sqrt{U} e^{\pi \epsilon_0 (e_0 + U)/\Gamma U}$ where $\epsilon_0$ is the energy of a localized state relative to the Fermi level, $U$ the charging energy and $\Gamma$ the coupling to the leads [58]. The underlying energy scales, and hence the maximum achievable $T_K$, grow with decreasing dot size, with $T_K$ reaching several Kelvin for nanotube dots [59], an order of magnitude higher (in the middle of the Kondo valley) than for gate-defined GaAs dots [60, 61]. (2) Large $g$-factor. The
Landé g-factor – the strength of magnetic field coupling to the spin of the local site – is large and well-defined in nanotubes \((g \approx 2)\) \([62, 51]\) compared to GaAs-based dots \((|g| = 0.1 \text{ to } 0.44)\) \([63]\). Thus, the Kondo temperature can be much larger than accessible temperatures but still smaller than Zeeman splitting at accessible magnetic fields. (3) Pure Zeeman coupling. The large g-factor and small area of nanotube quantum dots render orbital effects of magnetic field insignificant for magnetic field perpendicular to the tube axis. Magnetic field parallel to the tube axis does strongly affect orbital states \([64]\), so we apply field perpendicular rather than parallel to the tube axis to ensure that only the Zeeman effect is important.

In this chapter, I present data on the magnetic field evolution of the spin-1/2 Kondo effect. I summarise the extensive existing theoretical literature and compare our data to representative models, showing that they agree qualitatively but not quantitatively. I also note previous experimental work which foreshadow our results.

### 5.1 Measurement and Results

Our device is one of those measured in Chapter 4 and the device geometry is described there. For this particular device, the distance between source and drain electrodes is 250nm and the nanotube was 1nm in diameter according to AFM measurements. The differential conductance \((G)\) across the device was measured as a function of the bias voltage \((V_b)\), gate voltage \((V_g)\) and magnetic field \((B)\) as described in Appendix B. All of the measurements in the Oxford top-loading \(^3\)He refrigerator in our lab, whereas those in Chapter 6 were done in a nominally identical system at Bell Labs.

In measurements of \(G\) against \(V_g\) and \(V_b\), we see Coulomb diamonds with striking horizontal features — peaks in \(G\) at \(V_b = 0\) of voltage \(V_g\) — which signal the presence of the Kondo effect (Figure 5.1a) \([65]\) as discussed in Chapter 3. These zero-bias anomalies are more prominent at lower temperatures (Figure 5.1b, insets), as expected, and from their widths we estimate the Kondo temperature to be 1.7K, 1.8K and 1.5K in the middle of valleys (i), (ii) and (iii) \([66, 67]\).

Upon application of a magnetic field, a Kondo zero-bias conductance peak with
CHAPTER 5. SPIN-1/2 KONDO EFFECT

Figure 5.1: (a) Differential conductance $G$ as a function of bias voltage $V_b$, showing zero bias features for odd electron occupancy. Data to the right of the location marked ‘x’ have been shifted to account for a random charging event. (b) Linear conductance as a function of voltage $V_g$ on a back gate, at $T \approx 317$ mK (blue), 1.8K (green) and 6K (red). (Insets) Zero bias features in valleys ii and iii at the same temperatures.

$T_K = 2K$ splits into two peaks at energies $\pm \delta$ corresponding to peaks in the densities of spin-up and spin-down states (Figure 5.2), as predicted [68] and previously observed [69, 63, 62]. Naively, one might expect these peaks to occur at the Zeeman energy $\pm g\mu_B B = \pm \Delta$ and this indeed what we observe at high fields. Between $B = 4$–7 T ($g\mu_B B = 2.9$ to $5.1kT_K$), we find linear behavior $2\delta|\mu V| = 240B[T] - 5 \pm 14$, corresponding to $g = 2.07 \pm 0.02$ (95% confidence) (Figure 5.3b) [70]. This linearity at high fields has been predicted theoretically [68, 71] and our value for the $g$-factor agrees well with previous work on nanotubes [62, 51].

At low fields, however, we find that the extracted peak energy falls below linear (Figure 5.3b, c). This is in qualitative agreement with the theoretical literature which predicts $\delta < \Delta$ [71, 72, 73], with $\delta$ reduced to $2/3\Delta$ as $B \to 0$ [74] due to the attraction of the Kondo resonance to the Fermi level [73].

The field at which this reduction disappears is therefore another measure of the strength of the Kondo interaction. Despite broad theoretical agreement on the high and low field limits, differing descriptions exist of this crossover region. Here
Figure 5.2: (a) Kondo diamond at zero magnetic field. \( T \approx 352 \text{mK} \). Black is low conductance and white high. (b) The same diamond at 4T. (c) Evolution of the features in the middle of the valley in 2a-b with magnetic field. (d) Slices of the data in 2a in 0.5T steps. (Our data are twenty times denser.) Successive curves are offset vertically by \( 0.02 \times 2e^2/h \). Peak locations for the central Kondo peaks were obtained by fitting the data from (c) with two Lorentzians plus a field-independent background (black crosses). A slightly different background is assumed for a similar fit at intermediate field points (blue crosses). Between 4 and 6T the two versions of our fitting procedure produce nearly-indistinguishable results (see also Fig. 3a-d).
we consider two representative theories due to Moore and Wen (MW) [71] and Costi [75, 73]. MW's Bethe-ansatz calculation converges to the high-field result extremely slowly (logarithmically), predicting for example $\delta \approx 0.9\Delta$ for $g\mu_B B = 1000kT_K$ [71]. Costi, using a density matrix renormalization group approach, predicts a more rapid crossover, with convergence to the high-field result around $g\mu_B B = 20kT_K$ [73]. In contrast, our data have crossed over to linearity by $g\mu_B B = 2.8kT_K$, a much lower field than predicted by either theory [76, 77] but also a more intuitive result.

This can be seen in Figure 5.3c where we compare our data to the Moore-Wen and Costi predictions: our data have already saturated to the linear splitting value at $4T$, yet both theories predict this saturation only at much higher fields off the scale of the plot (assuming $g = 2$). Another way to look at the contrast between theory and experiment is to normalise the predicted splittings to our data. (This is equivalent to assuming, unrealistically, $g = 2.9$ and 2.35 for MW and Costi respectively.) One then sees that the theories predict a more gradual evolution to high field behavior than we observe (Figure 5.3d).

The splitting of the spin-1/2 Kondo peak has been measured in some detail on two previous occasions. Cronenwett et al. [69] measure $\delta \sim g\mu_B B$ at all fields but with rather large variations around this value. In measurements by Kogan et al. [63], we note that there appears to be some suppression of $\delta$ starting around $g\mu_B B = 1.1T_K$ in their data. In both these sets of measurements, $T \sim T_K/3$ — whereas $T \sim T_K/6$ for us — and the higher temperature may have obscured the low-field suppression of $\delta$ that we observe. [71]

Another question of interest concerns the field at which splitting between the two spin-resolved resonances first becomes visible: although these resonances in principle move away from zero energy even at very low fields, they initially overlap so that the measured Kondo peak does not visibly split into two until a critical field, $B_c$. Costi has predicted [75] that at low temperatures ($T < T_K/2$, satisfied in our case) $g\mu_B B_c \approx kT_K$. We observe this splitting first at $B = 2.3T$, modestly higher than the predicted $B_c = 1.5T$, and roughly where we can start to robustly fit peak positions. Previous work in GaAs also found the splitting occurring roughly at $B_c$ [61].
Figure 5.3: (a) Peak positions in (c) obtained as described in 2d. Black (blue) dots mark results from the high (intermediate) field fit. (b) Energy difference between peaks from 3a. The green line is a fit to the high field points. (c) Splitting from 3a normalized by the naïve Zeeman energy — $\delta/2\mu_B B$ (black and blue dots). The red line is the Moore-Wen prediction, the grey line is the Costi prediction, the flat green line is the predicted high-field limit, and the red dot marks the Logan-Dickens low-field prediction, all for $g = 2.07$. (d) The data from 3b are reproduced (black and blue dots). The red line is the Moore-Wen prediction and the grey line is the Costi prediction, now using unrealistically large Zeeman coupling $g = 2.9$ and 2.35, respectively, to attempt to fit the data.
5.2 Note on our fitting procedure

For both high and intermediate field regimes, we did several sets of fits to our data, starting with four completely unconstrained Lorentzians at each field (representing two low-bias Kondo peaks and two higher-bias inelastic cotunnelling peaks) on various (parabolic, V-shaped, flat) backgrounds. After each set, we looked at the goodness of the fits through numerical measures as well as by comparing fit results slice-by-slice (field-by-field) to the actual data. Through this procedure we determined that our background is more V-shaped than parabolic or flat and is very constant through the field range.

We then started pruning the number of free parameters and studying the results as before.

For the high field regime (4-7T) we found that we could obtain very good fits to our data while holding the V-shaped background and the two side peaks fixed — the free parameters here are then the weight, width and location of the central Kondo peaks. We found that adding further constraints — such as requiring the central peaks to be symmetrically spaced about zero bias — worsened the quality of the fits considerably, so we stopped here.

For the lower fields around 3.5T, we also obtained very good fits for fixed V-shaped background and side peaks (with slightly different parameter values for the side peaks as noted in the paper). We next also constrained the central peaks to have equal widths and be equidistant from zero bias. Note that the free parameters here are thus two fewer than before — the weights of the central Kondo peaks, plus the width and distance from zero bias which are the same for both.

The quality of fits remained excellent and these additional constraints did not change the results over a wide field range: 2.3-6T. This is the range for which we report results for this second 'intermediate field' fit.

When we observed the sublinear splitting described in the paper, we considered that temperature-broadening might have the effect of making peaks appear to be closer together than they actually were, especially in the crucial intermediate field regime. We therefore included temperature-broadening of 350mK (which was the
measurement temperature) in our fits but found that it had a negligible effect, so we ended up leaving this out.

As can be seen in the figures, our two fitting procedures produce nearly-indistinguishable results over a large overlap region of 4-6T, another indication of the robustness of the fits.

5.3 Note on comparing existing calculations to transport experiments

We wish to note some issues in comparing existing calculations to any transport experiment and in particular to ours.

1. All the theories calculate spin-resolved density of states, whereas experiments measure the sum of the two spin-resolved densities of states. We address this by fitting our measurements with two Lorentzian peaks and interpreting the extracted peak positions as the positions of the resonances in the spin-resolved density of states. We find that the results are insensitive to details of the fitting procedure for sufficiently high fields, $g\mu_B B > 2.3T(1.6kT_K)$. At lower fields we do not report peak positions, as details of the fitting procedure influence the extracted peak positions when the peaks strongly overlap. Information on the actual non-Lorentzian (and non-analytic) Kondo density of states at small but finite B would likely be necessary to extract reliable (self-consistent) splittings at these low fields.

2. Lineshape asymmetries at high magnetic field predicted by Rosch et al. and Logan-Dickens may also influence our extracted peak positions [78, 74] but the fact that we extract splitting that is highly linear with field at high field, extrapolating to zero at zero field, suggests that these issues of lineshape do not have a strong effect on our extracted splittings.

3. Most theories calculate an equilibrium density of states. In transport experiments a finite bias is applied across the quantum dot, changing the density
of states of the system. Konik et al. extended the Moore-Wen Bethe-ansatz analysis to take into account non-equilibrium effects, and achieved very similar predictions. [72] We expect non-equilibrium effects to be small in our experiments since our dot is coupled much more strongly to one lead than to the other: the ratio of couplings is roughly 8 as judged from our maximum observed conductance.

5.4 Conclusions

In conclusion, we have studied the magnetic field evolution of the spin-1/2 Kondo effect in a quantum dot formed from a carbon nanotube. Carefully tracking the Kondo peak as it splits in magnetic field, we find that its spin-resolved constituents is linear with field at high field and sublinear at low field, qualitatively matching predictions. however, the crossover from sublinear to linear splitting is shaper than predicted by any exiting theory and occurs at lower-than-expected field.
Chapter 6

Spin-1 Kondo Effects

In this chapter we turn our attention to Kondo effects when there are an even number of electrons on the dot (Figure 6.1).

Empirically, in quantum dots with even electron occupancy the ground state is usually a singlet ($S = 0$), particularly in the presence of strong tunnel coupling to leads [79]. However, when the exchange energy gained from aligning spins is larger than the energy difference between singlet and triplet states the ground state will be a triplet ($S = 1$).

6.1 Singlet-triplet transition at zero field

In our dot we observe for one particular even occupancy a zero bias conductance peak which splits into peaks at finite bias as a function of gate voltage (Fig. 4a-b). These features can be understood as a transition between triplet and singlet ground states [80], respectively, as follows.

On the lefthand side of the Coulomb diamond in question, the triplet is the ground state and correlated interactions with the leads arise from the degeneracy of the three triplet states. As the total spin on the dot is 1, interactions with two channels in the leads are required to fully screen this spin [81]. (See also Section 6.4.) When couplings to the two leads are comparable, previous experiment and theory have shown two peaks in conductance at finite bias, flanking a zero-bias conductance dip:
Figure 6.1: (a) Zero bias feature in an even Coulomb diamond (parity determined by careful study of ten diamonds on either side). $T \lesssim 250$ mK. (b) Higher resolution scan of the region in 4a bounded by the rectangular box. The data indicate a gate-induced transition between singlet and triplet ground states for the dot. (c) Magnetic field dependence of the conductance at the point marked by the white triangle in 4a.
the ‘two-stage’ Kondo effect [82, 83, 84, 85]. Our contrasting observation of a simple zero bias peak indicates strong coupling to only one channel so that the spin is underscreened [86, 87, 88, 80].

On the righthand side of the Coulomb diamond, the ground state is a non-degenerate singlet and so conductance is low at zero bias; however, peaks appear at finite bias corresponding to the energy difference between singlet and triplet states. This sort of gate-driven Kondo singlet-triplet transition at zero magnetic field has been reported only once before, in a GaAs dot [80]. The mechanism proposed there was the Stark effect, which is small for quantum dots in nanotubes. A different gate-driven spin transition has been observed in nanotubes [55] and was thought to be due to electron-electron correlations, which could play a role here too.

6.2 Magnetic field evolution

As the transition is gate-driven and as the orbital effects of magnetic field are negligible in nanotubes as noted above, we can study the magnetic field evolution of the Kondo features on both sides of the transition in a simple manner.

Magnetic field-driven transitions of ground state spin in a quantum dot are more common than gate-driven transitions (cf. [88, 89]), but they usually stem from orbital physics. In contrast, we can apply a pure Zeeman field, allowing us to study the magnetic field evolution of the Kondo features on both sides of the gate-driven transition in a simple manner.

On the triplet (lefthand) side of the diamond, a magnetic field linearly splits the zero-bias peak into two finite-bias peaks (Fig. 5a, e). The voltage of these peaks corresponds to the energy difference between the two lowest triplet states (Fig. 5c). As with the spin-1/2 Kondo peak, the splitting here first occurs at slightly larger-than-expected field $B = 1.8T > B^\text{Costi}_c = 1.1T$ [75, 90].

On the singlet (right hand) side, the two finite-bias peaks each split with increasing field, with two of the resulting features meeting at zero bias at finite field and then splitting apart again. (Figure 6.2b).

This field evolution can be understood by considering finite-energy transitions
Figure 6.2: (a,b) Magnetic field evolution of conductance versus bias voltage on the left (triplet) and right (singlet) sides of the diamond in 4a. The gate voltages chosen have been marked with dashed lines in 4a. Black is low conductance and white high. (c,d) Schematic of singlet (s) and triplet (t) energy levels as a function of magnetic field in the configurations corresponding to 5a and 5b respectively. Double-headed arrows indicate transitions seen in our data. (e) Peak positions in 5a obtained by fitting two Lorentzians, corresponding to the transition shown in 5c. The lines are \( \pm g\mu_B B \), with \( g = 2.07 \). (f) Analysis of data in 5b. The black circles are peaks obtained using a simple peak-finding function. The grey triangles are the locations of steepest slope for the step. The lines are guides to the eye and have slope \( \pm g\mu_B B \), with \( g = 2.07 \) and mark the transitions shown in 5d.
between the singlet (s) and the three Zeeman-split triplet states t(-1), t(0) and t(1), labeled according to their spin in the direction of the field (Fig. 5d, cf. [91] Fig. 2b.)

(i) s ↔ t(-1) The features moving toward zero bias with increasing field correspond to this transition, so the zero-bias crossing is when t(-1) becomes degenerate with s. Beyond this crossing field, t(-1) is the ground state and we continue to see transitions between t(-1) and s, but not the those between t(-1) and t(0) or t(1). The latter is forbidden at lowest order because of the large change in spin; the former should be possible, but would occur at very high bias. (ii) s ↔ t(1) The features starting at zero field that move to higher bias with increasing field mark this transition. These features are less pronounced, probably because of stronger decoherence at higher bias, and disappear around when t(-1) becomes the ground state. (iii) s ↔ t(0) We do not clearly discern this transitions, likely because the three overlapping peaks associated with singlet-triplet transitions form a plateau, in which only the edges of the plateau, associated with t(-1) and t(1), can be easily identified, while the middle peak remains hidden.

The transformation in Figure 6.2b of the upper peak into a step after the crossing point can be understood as being caused by asymmetric coupling to the leads [91].

6.3 Mysteries

We observe several additional strongly gate-sensitive features in our diamond plots (Figure 6.1a), which we investigate in less detail. At least one of them appears to be Kondo-related — the conductance decays logarithmically with B at high field, and saturates to a constant value at low field Figure 6.1c).

The question arises as to whether the strongly gate-dependent finite-bias feature in Figure 6.2 is due somehow to the likely encapsulation of buckyballs in our nanotube. We believe that this is not the case for the following reasons.

1. Our results in Chapter 4 and the work of other researchers has shown that as a whole C_60 peapods look similar to unfilled tubes in transport at low biases. [92]

2. Similar gate-dependent finite-bias Kondo features have previously been observed
in unfilled nanotubes. See for example Ref. [91] As in our case, the mechanism for the gate-dependence is not identified in this work.

3. The energy of the unexplained feature (\(\sim 1 - 12\) meV) is too low to match vibrational states (\(> 30\) meV) of \(C_{60}\), so it is not clear what \(C_{60}\)-associated mechanism would produce the feature. [93]

We propose a simpler explanation: This feature may mark the transition between the lowest \(m_z = 0\) triplet state and the first excited singlet state. This candidate transition does not change \(m_z\), and hence does not move with magnetic field, in agreement with observation. We do not know the mechanism for gate-tuning of this feature nor whether the fact that it appears to pass through one corner of the diamond is anything more than an accidental alignment of energy levels at that point.

6.4 More on screening the spin-1 Kondo effect

When the net spin on a quantum dot is 1, interactions with two channels in the leads are required to fully screen this spin. Each channel will have a coupling strength \(\Gamma_{\alpha}\) to the dot and an associated Kondo temperature, \(T_{K\alpha}\).

In the case where \(T_{K1} \neq T_{K2}\) and \(T_{K1} \sim T_{K2}\), a two-stage Kondo effect can be observed as reported in Ref.s [83] and [82]. In this case a dip with width corresponding to the smaller of the \(T_{K\alpha}\)s is superimposed on the main zero bias peak of width corresponding to the larger of the \(T_{K\alpha}\)s.

Of course one must have \(T \ll T_{K1}, T_{K2}\) as well in order for this to the case. As in the spin-1/2 case, if the temperature is too high, the 'bond' between the dot and one or both of the leads will be 'broken' and this effect will not be observed.

For example in the case where \(T \ll T_{K1}\) and \(T, T_{K1} \ll T_{K2}\), it is possible to have an underscreened Kondo effect — only one screening channel in the leads is well-coupled to the dot and so the total spin of 'dot and leads' is reduced to 1/2 rather than to 0. There will then be only a simple peak at zero bias instead of a peak with a super-imposed dip. The observation of such a peak in a spin-1 diamond as in our work and others' is suggestive of such a scenario. [87, 88, 80]
The observation of a simple peak in a spin-1 diamond as in our work and others’\[87, 88, 80\] is suggestive of such a scenario; however, recently, Posazhennikova and Coleman [86] have proposed further, more striking signatures of the underscreened Kondo effect — according to their noncrossing approximation (NCA) calculations there should be a singularity at low temperature and voltage such that \( d^2I/dVdE \) diverges with increasing 1/\( E \), where \( E \) is either \( V \) or \( T \).

While in principle we should be able to compare our data to these predictions, we unfortunately find that we do not have enough data points at low voltage (i.e. high 1/\( V \)) to say conclusively that we observe singular (rather than say Lorentzian or Gaussian) behaviour.

6.5 Conclusions

In conclusion, we have studied the magnetic field evolution of the underscreened spin-1 Kondo effect in a quantum dot. We also demonstrate the independent gate and magnetic field tuning of a spin-1 Kondo effect, and find good quantitative agreement with a simple model using the g-factor previously extracted in chapter 5.
Chapter 7

Cleaved-Edge-Overgrowth Hole Wires

Cleaved-edge-overgrowth (CEO) is a Molecular Beam Epitaxy (MBE) process by which very long and clean one-dimensional electron and hole systems have been fabricated in gallium arsenide (GaAs). It is at first glance a simple conceptual extension of the process by which two-dimensional electron gases (2DEGs) are produced, but it is technically extremely challenging, so much so that to my knowledge only two MBE groups in the world have carried it out with any success — those of Loren Pfeiffer at Bell Labs and Werner Wegscheider (who was Pfeiffer's postdoc) at Regensburg. (A subsequent literature search yielded Reference [94]. I have no personal knowledge of this work, though Hidefumi Akiyama presently collaborates with the group at Bell Labs.)

The cleaved-edge-overgrowth method was developed by Loren Pfeiffer and his colleagues in 1990 [13] and subsequently used to produce one-dimensional wires. These wires had electrons as their carriers and were first measured optically [95] and subsequently also in transport. [96] Since then, these wires have been very well characterised in transport, primarily by Amir Yacoby and Rafael de Picciotto.

Other than QPCs made from conventional semiconductor 2DEGs, CEO wires are the only one-dimensional system measured in transport where the number of populated subbands can be tuned and studied in a reliable way. Thus CEO wires
have provided an independent realm for the exploration of the physics of such systems and have allowed us to develop a understanding of them which is not tied as much to the particularities of either QPCs or CEO wires. Measurements on CEO wires have shed light, for instance, on the 0.7 structure, the origin of which is a long-standing puzzle in the physics of tunable quasi-one-dimensional systems. [97] In addition, their unique geometry has made possible tunnelling experiments giving evidence of spin-charge separation in 1D. This work on tunnelling has been done primarily by Amir Yacoby, Ophir Auslaender and Hadar Steinberg. [98, 99, 100]

During my Ph.D., I had the privilege of spending eight months at Bell Labs working on the fabrication and measurement of the first CEO hole wires with Rafael de Picciotto, Loren Pfeiffer, Kenneth West and Kirk Baldwin. In this chapter I shall describe some initial transport measurements on these wires. At the time of writing (the end of 2007) measurements are being continued at Stanford on these devices after a hiatus of almost two years due to problems with our dilution refrigerator. (See Appendix E.) These will be completed by Joseph Sulpizio after my departure, so it is unfortunately not yet possible to give the reader a complete picture of these experiments.

N.B. In writing this chapter I have run into problems akin to those faced generally by humanists due to the gendered-ness of pronouns in the English language. In what follows, the reader should note that ‘electron’, ‘hole’ and ‘carrier’ have been used more or less interchangeably except where I refer to my specific devices where the carriers are always holes. It is not my intention to imply that all carriers are electrons any more than I wish to imply that all physicists are women.

7.1 The cleaved-edge-overgrowth process

The idea behind CEO is quite simple and requires knowledge of two facts stemming from semiconductor band structures: (a) In a AlGaAs-GaAs-AlGaAs heterostructure, electrons/holes (from donor/acceptor dopants) will tend to accumulate in the GaAs ‘quantum well’ layer and (b) Electrons/holes (again from dopants) tend to accumulate at GaAs-AlGaAs interfaces. (The reader may refer to the book by Davies for further
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Figure 7.1: Simplified cross-sectional schematic of a GaAs wafer containing a 1D wire on its edge. The figure is not to scale. In the main part of the wafer, electrons (or holes) accumulate in a GaAs well with AlGaAs barriers. On the edge, growth of additional AlGaAs leads to accumulation of carriers in a 1D line (into the page) at the interface of the new layer with the existing well indicated by the black dot. Dopant atoms are embedded in the crystal in short pulses at the locations shown. Si (C) atoms act as donors (acceptors), yielding electrons (holes) as carriers.

details. [101]) As shown in Figure 7.1, to grow a CEO wire, one starts with a conventional 2DE/HG formed in a GaAs quantum well and by growing additional AlGaAs on the edge of the substrate, causes electrons to accumulate in a one-dimensional region at the interface between the existing 2DEG and the new layer.

As mentioned above, very few MBE groups have been able to grow CEO wires. Partly because of this, many other processing tricks surrounding the fabrication of these wires are also not particularly obvious and not perhaps well-known to most groups studying mesoscopic physics. I therefore describe these below in some detail so that this knowledge is not lost.

Growing the 2DHG

The first step of the CEO process involves the growth of a conventional quantum well 2DE/HG on the (100) surface of a GaAs substrate. The 2DE/HG is formed in a GaAs well with AlGaAs barriers as shown in Figure 7.1. The dopants (C for holes and Si for electrons) are in a ‘delta-doped’ layer outside the GaAs well.

All 2DE/HGs grown by Loren Pfeiffer and Kenneth West at the time I was at
Bell were characterised by Kirk Baldwin — at 4K and sometimes also in a $^3$He fridge — using Quantum Hall measurements to obtain their mobility and density.

The use of carbon as a dopant for 2DHGs on (100) surfaces had at the time just been developed Michael Manfra, Loren Pfeiffer and their colleagues [102] as well as two other groups independently [103, 104]. Our substrates typically have mobilities of about $1 - 2 \times 10^5 \text{cm}^2/\text{Vs}$ and densities of about $1 - 2 \times 10^{11} \text{cm}^{-2}$ at 4K. In a small number of samples cooled down further to 300mK, the mobility increased 5-10 times, while the density stayed roughly constant.

In most of the substrates, the Al content was 32%. In two instances 50% substrates were grown, but the carrier density in our CEO wires grown on the edges of these substrates was found to be extremely unstable over time upon cooling down — the densities would start very high and decrease over some hours to untenable values.

Figure 7.2: Optical image of the gates patterned by photolithography on a portion of a 10mm by 12mm piece of substrate after the first MBE growth. The 13 gates intersecting the future cleave are each 2$\mu$m wide and 2$\mu$m apart. A scribe mark for cleaving will be made at the location of one of the horizontal tick marks on either side of the chip.
Patterning the gates by photolithography

Gates are then patterned on the wafer as shown in Figure 7.2 with standard photolithography techniques. Each of the gates in Figure 7.2 is 2\(\mu\)m wide and the spaces between them are also 2\(\mu\)m wide. The metal evaporated is usually tungsten, which as a refractory metal is able to withstand the elevated temperatures in the MBE chamber.

Thinning the substrate

The substrate is then thinned from the standard wafer thickness of about 500\(\mu\)m to about 80\(\mu\)m with bromine methanol (9 or 10 to 1 and pour the bromine in last) in a chemical-mechanical etching process. Before the sample (usually a quarter of a 2” wafer) is thinned, chips are defined on the front side by scribing as this will be next to impossible to do at the end. (The wafer is not broken after scribing.) The sample is then attached to a quartz disc with wax by melting the wax onto the disc at 55°C on a hotplate and placing the sample face down in the molten wax. One should also place a weight on the sample to ensure that it is flat on the disc.

Using a round wafer box as a holder for the quartz disc, the sample is thinned by rubbing it against a glass plate. The disc will probably sit somewhat loosely in the wafer box, but this is not a big problem. The glass plate is prepared by taping a sheet of paper to it and then pouring a small amount of bromine methanol solution onto the plate, enough to soak the sheet. One then abrades the sample against the plate in a circular motion, taking care to change the orientation of the sample from time to time to avoid unintentionally creating a non-uniform thickness.

After about a minute, one washes the etching solution off the sample with water, blows it dry and measures its thickness at several points using a gauge such as the Peacock R5C. (It is helpful to leave a tap running throughout this whole process.) This will tell one the rate of thinning and also if the thinning is non-uniform across the sample. If it is the case the one side of the sample is higher than another, one can adjust the etching motion to try to correct for this.
The thinning intervals get shorter and shorter as one approaches the desired thickness. As the solution evaporates, it has to be replenished — one puts on fresh sheets of paper (which do not have to be taped on) and soaks them in the etching solution for each thinning step. About 100mL of bromine methanol is usually consumed in the course of thinning down one wafer.

Finally, when the wafer is at the desired thickness of 80-90μm, the wax is dissolved so that the chips can be removed from the quartz disc. This is done by immersing the disc in acetone which is in a beaker on a hotplate set to 155°C. It is essential that the beaker be quite tall so that the boiling acetone does not overflow and ignite on the hotplate. At the same time the beaker cannot be completely covered so that the acetone vapour does not have a means of escaping. A 500mL beaker with a spout and a suitably sized watch glass works quite well.

As the chips are very fragile, it is helpful to place a mesh basket in the beaker so that they can be easily removed from the acetone solution. It will also make one’s life easier if the disc is held with a contraption hung over the edge of the beaker so that the chips can fall into the basket and do not have to be pushed off the disc. (Figure 7.3.) After removing the chips, they should be gently rinsed with isopropanol and blown dry.

**Cleave and overgrowth**

An additional short scribe mark is then made on each chip at the intended position of the cleave. (See Figure 7.2 and caption.) In the MBE chamber, the cleave is made by hitting the chips with a mechanical arm to expose a fresh (110) surface on which AlGaAs is grown. (Figure 7.4.) The edge is also doped with a ‘delta’ pulse of C. (Figure 7.1.) This step was performed by Kenneth West and Loren Pfeiffer. A test wafer is grown at the same time as the overgrowth. Typical densities from the 2DHGs in these test wafers at 4K are $1 - 3 \times 10^{11}/\text{cm}^2$ and typical mobilities $1 - 4 \times 10^5 \text{cm}^2/\text{Vs}$. Additional MBE technical details on the cleave step may be found in Reference [105]. Additional parameters for our samples can also be found in Appendix F.
Figure 7.3: Schematic of apparatus for detaching a quarter wafer from a quartz disc, to which it had been attached with wax, after thinning. The wafer will fall from the disc in individual chips along the lines demarcated earlier with a scribe. Each chip should contain one device as shown in Figure 7.2.
Figure 7.4: The cleave and overgrowth process. Thinned chips containing 2DEGs are mounted in such a way that they can be hit with a mechanical arm in the MBE chamber, cleaving them in two. Additional material is then grown on the edge as shown.
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Ohmics

An InZn eutectic is used to make ohmic contact to the wires. The eutectic was made by melting the appropriate ratios of In and Zn together in a petri dish on a hotplate. (For electron wires, InSn is used instead.) Blobs of the eutectic are placed on the chip with a soldering iron and the chip is then annealed in nitrogen or argon at between 400-440°C for about two minutes in a rapid thermal annealer (RTA).

Wiring

![Image of CEO hole wire chip wired to a chip carrier](image)

Figure 7.5: Optical image of a fully-processed CEO hole wire chip wired to a chip carrier and ready for measurement. The chip carrier is about 2cm in its horizontal dimension.

The chips are glued to GaAs pieces of standard thickness with photoresist to make them easier to handle. (During this process take care not to put down too much photoresist as it can flow over the chip, making it impossible to wire.) In general, the thinned chips by themselves are handled as little as possible and the whole process described here has been optimised in this direction. The pieces are heated slightly on a hot plate to dry to photoresist and then stuck on to 16 pin chip carriers with Dow Corning high vacuum grease. Gold wires are attached between the ohmic contacts and gates on the chip and the pins of the chip carrier using indium solder. Figure 7.5 shows a CEO wire that has been wired to a chip carrier in this fashion and is ready
for measurement.

7.2 Transport through a quasi-1D system: rough sketch

In this section we shall explore what is naively expected for conductance through a tunable quasi-1D system to give the reader a feel for the problem. In the following sections, making certain assumptions, a more mathematical model shall be presented.

Consider a one-dimensional channel with a small number of occupied sub-bands connected to two contacts as shown in Figure 7.6(a). Similar to the allowed energy levels in quantum dots, a nearby gate electrode can shift these bands up and down with respect to the contacts. When the sub-bands are all higher in energy than the electrochemical potential of the contacts, the wire is entirely depleted of carriers and no current can flow through it. As the gate voltage is varied such that one after another of the sub-bands cross the Fermi level of the contacts, current flows through the wire and one expects a sharp increase in conductance as each sub-band crosses the contact Fermi energy as in Figure 7.6(c). At finite temperature, of course, these steps will look like Fermi-Dirac distributions rather than sharp steps.

What is the contribution of each sub-band to the conductance? It turns out that for perfectly transmitting contacts one can expect a contribution of one quantum of conductance $2e^2/h$ per sub-band. This is the famous Landauer formula. (See Reference [15] for instance for a derivation and explanation.) In the situation in Figure 7.6(a) for instance, one expects a conductance of $4e^2/h$ as two sub-bands are below the Fermi energy of the contacts. In real measurements, the steps are often suppressed from this ideal value because of a series resistance due e.g. to filters in the wiring of the fridge or the way in which the wires are contacted. The plateaus are also often not very smooth, especially in CEO wires, due to disorder. Both of these issues and others will be addressed below in the presentation of our data.

Now consider the case of a finite bias across the contacts. Consistent with the analysis below, we shall call the higher electrochemical potential $\mu_+$ and the lower $\mu_-$.
Figure 7.6: Rough picture of main features of transport through a tunable 1D channel. (a) Zero bias. The conductance is $2e^2/h$ times the number of sub-bands (two in this case) with their edges below the Fermi energy. (b) As the bias is increased, eventually either $\mu_-$ reaches the bottom of the sub-band or else (as in this case) $\mu_+$ reaches the next sub-band. Here the conductance is $5e^2/h$. Each 'half sub-band' contributes $e^2/h$. (c) As the gate voltage is swept at zero bias, the conductance is increased by $2e^2/h$ every time the Fermi energy hits the bottom of a new sub-band. (d) A 'two-dimensional' sketch of conductance against bias and gate voltages in units of $e^2/h$. 
regardless of which contact is which. (Note that this is in contrast to the quantum dot analysis in Chapter 3 where we labelled one contact the ‘source’ and the other the ‘drain’ regardless of the relative Fermi energy difference between them.)

Assume now that we are sitting on a plateau in conductance, as in Figure 7.6(a), but are closer to the bottom of the third sub-band than to that of the second. Without assuming too much about the capacitances of the channel to the various electrodes, one can imagine that at a certain bias, $\mu_+$ will hit the next sub-band while $\mu_-$ has not as in Figure 7.6(b). This represents a contribution of half a quantum of conductance, so that in the situation in the figure, one expects a conductance of $3e^2/h$.

Mapping out the conductance with respect to all gate and bias voltages, one can see that something like what is shown in Figure 7.6(d) will result. The straight lines marking the ‘diamonds’ in the Figure are not a priori to be taken literally. In QPCs, a model is often used which maps the bottom of the sub-bands to the allowed states in the quantum dot problem, ignoring the details of the density of states in the 1D sub-bands. In this case, the diamonds are indeed straight-edged as shown, as are Coulomb diamonds in quantum dots; however, in CEO wires, this picture breaks down — the 1D density of states has to be accounted for and the diamonds have curved borders, as explicated in Section 7.5.

### 7.3 Why hole wires?

We embarked on this experiment mostly because holes in GaAs are expected to experience a strong SO interaction which can in principle have quite dramatic effects on transport in a 1D system, around which several schemes for the development of spin filters have been proposed.

It must be emphasised here, especially to those used to dealing with the much more well-studied electron gases in GaAs, that hole gases in general and in particular those grown on (100) surfaces are much newer systems about which much is unknown — theoretically as well as experimentally — including many basic materials properties that are taken for granted in 2DEGs. Partly because of this and partly because we started measuring the CEO wires only in 2005 [105], our own thinking about and
understanding of the CEO hole wires is at present limited. It is already clear that there are many differences between hole and electron wires, but their origin is not at present always clear.

In this section I try to the extent possible to convey some of the reasons which spurred us to perform this experiment as well as to record what is known about 2D hole gases and our thoughts to date on the quite striking data we have been getting (and still are) from the hole wires. While I have confidence that the data in this chapter are presented in a systematic and clear way, I apologise to the reader for the many speculative statements and interpretive errors which must of necessity be present here; we hope to present a more coherent and correct picture in a subsequent publication upon completion of the data-taking and analysis.

![Figure 7.7: The spin-orbit coupling gives rise to a momentum (k) dependent spin-splitting for each sub-band.](image)

**Spin-orbit coupling in one dimension**

A result from special relativity is that electrons moving transverse to an electric field see an effective magnetic field in their rest frame — see for example, Chapter 12 of Reference [106]— which modifies their energy depending on their spin. This is the essence of the mechanism for the spin-orbit interaction.

Starting with the expansion of the Dirac equation — which describes relativistic
spin-1/2 particles [107]— in powers of $mc^2$ the lowest order contribution of the spin orbit interaction to the Hamiltonian is [108]

$$H_{SO} = \beta \mathbf{\sigma} \cdot \mathbf{k} \times \nabla V \equiv \beta \mathbf{\sigma} \cdot \mathbf{B}_{SO}$$  \hspace{1cm} (7.1)$$

where $\mathbf{\sigma}$ are the Pauli spin matrices, $\mathbf{k}$ is the momentum of the particle and $V$ the electrostatic potential it experiences and we have included a material-dependent constant, $\beta$, motivated by Reference [9].

If we make the further simplification that the electric field is constant and perpendicular to the direction of the wire, we can write for a one-dimensional system

$$H = \frac{\hbar^2 k^2}{2m} \pm \beta kE_{SO}$$  \hspace{1cm} (7.2)$$

where $k$ is in the direction of the wire and $E_{SO} \equiv |\nabla V|$. $k$ and $E_{SO}$ are now only numbers, and the $\pm$ is for spin up and spin down in the direction of $B_{SO}$.

We can rewrite Equation 7.2 as follows.

$$H = \frac{\hbar^2 k^2}{2m} \pm \beta kE_{SO}$$  \hspace{1cm} (7.3)$$

$$= \frac{\hbar^2}{2m} \left[ k^2 \pm \frac{2\beta kE_{SO}m}{\hbar^2} + \left( \frac{\beta mE_{SO}}{\hbar^2} \right)^2 - \left( \frac{\beta mE_{SO}}{\hbar^2} \right)^2 \right]$$  \hspace{1cm} (7.4)$$

$$= \frac{\hbar^2}{2m} \left[ k \pm \frac{\beta E_{SO}m}{\hbar^2} \right]^2 - \frac{\beta^2 E_{SO}^2 m}{2\hbar^2}$$  \hspace{1cm} (7.5)$$

$$\equiv \frac{\hbar^2 (k \pm k_{SO})^2}{2m} + \text{constant}$$  \hspace{1cm} (7.6)$$

We can see from this last equation that the initially spin-degenerate parabolic band has been split into two non-degenerate bands with centres shifted by $\pm k_{SO} = \pm \beta E_{SO}m/\hbar^2$ as in Figure 7.2. There is also a constant shift in the overall energy which can be ignored. Note, however, that this k-dependent spin-splitting cannot be detected in the transport properties of the system at zero magnetic field, which is rather disappointing.

Can we detect this shifting of the spin-split bands by applying a magnetic field?
Figure 7.8: Dispersions and conductance traces against gate voltage for a quasi-one-dimensional system with spin-orbit coupling for different directions of magnetic field. Figure reproduced and modified from Reference [8].
Let us now qualitatively explore this possibility, following closely the analysis of References [8] and [109].

In the direction of $B_{SO}$, spin is still a good quantum number, so applying a magnetic field in this direction also should simply raise (lower) the spin up (down) band by the Zeeman energy without mixing the two bands. The result in transport is thus not any different from the case where there is no spin-orbit coupling — one observes that each plateau splits into two corresponding to spin up and spin down. (Figure 7.8a and b.)

If, however, a magnetic field is applied in a direction perpendicular to $B_{SO}$, this represents a off-diagonal matrix element in the original spin basis — parallel and anti-parallel to $B_{SO}$. The two spin branches are thus mixed at their crossing, resulting in a gap or anti-crossing at that point as shown in Figure 7.8(e). This can be seen in a striking manner in the transport through the wire. Focussing our attention on the first spin up and down sub-bands, when the Fermi energy is tuned by the gate voltage to be in the gap, in the Landauer-Büttiker picture, there is now only one ‘equivalent’ sub-band and so the conductance drops from the normal plateau value of $2e^2/h$ to $e^2/h$. (Figure 7.8(f).) The size of this drop or dip in gate voltage depends on the size of the gap. Similar dips should be seen on all the conductance plateaus. These features will of course be blurred at finite temperature as shown in Figure 7.8.

Note that when the Fermi energy is in the gap, the wavefunction of the left-movers will be dominated by the spin-down component, while the right-movers will be mostly spin-up. This has been the basis for several proposals for spin filtering and spin accumulation applications, such as that described in Reference [109].

Finally, note that when the angle of the applied field is something other than zero or $\pi/2$ with respect to $B_{SO}$, both of these effects — Zeeman spin-splitting and a dip in the plateau due to the SO gap — can be observed simultaneously. (Figure 7.8(c) and (d).)
From the abstract to the concrete: grappling with materials properties

Bulk GaAs is one of the most well-studied semiconductors and 2DEGs in GaAs heterostructures are the bread-and-butter system of semiconductor mesoscopic physics. Comparatively little work has been done, however, on 2DHGs in GaAs especially those grown in the (100) plane; and almost nothing is known experimentally about (quasi-)one-dimensional hole systems in GaAs. In this section I shall discuss the state of the field with respect to the materials properties of GaAs which are relevant to our work, in particular the strength and nature of the spin-orbit coupling and the values of quantities such as the mass and $g$ factor of holes.

We first note that in engineered semiconductor structures such as our CEO wires, the electric field described in the previous section as the source of spin-orbit coupling can arise in two ways. First, inversion asymmetry in the bulk material. This is often referred to in the literature as bulk inversion asymmetry (BIA) and leads to what are called the 'Dresselhaus terms' in the SO Hamiltonian. Second, inversion asymmetry in the externally imposed confining potential or structural inversion asymmetry (SIA) gives the 'Rashba terms'. In the CEO wires, the sources of SIA will include the dopants, the triangular well on the edge of the wafer as well as the gates. The effect of spin-orbit coupling is seen most directly in the band structure of GaAs (described below) which is caused by the Dresselhaus terms. The Rashba terms are a little more straightforward to think about as they are in principle simply proportional to the electric field due to the confining potential; however, their prefactors contain constants of the band structure, so the Rashba terms are indirectly related to the strength of the Dresselhaus terms [9].

Figure 7.9 shows the band structure of GaAs. The lowest conduction band arises from s-like atomic orbitals so that the orbital angular momentum $l$ is zero, while the highest valence band arises from p-like ones so that $l = 1$. Thus, the total angular momentum $j$ of the holes in the valence band can be either $1/2$ or $3/2$, giving rise to six possible bands. These bands are degenerate at the $\Gamma$ point in the absence of SO coupling, but the presence of (Dresselhaus) SO coupling due to the inversion
The asymmetry of the crystal causes the $j = 1/2$ and $j = 3/2$ bands to have different symmetries and thus be non-degenerate. [9] The energy difference between them is known as the spin-orbit gap and the $j = 1/2$ band is known as split-off band. The $j = 3/2$ states are further split at finite $k$ depending on their value of $j_z$. The $j = 3/2, j_z = 3/2$ band is known as the heavy hole (HH) band because of its smaller curvature and hence larger mass, while the $j = 3/2, j_z = 1/2$ one is known as the light hole (LH) band.

In 2DHGs, the degeneracy between heavy and light hole bands at $k = 0$ is lifted because of the confinement potential. Additionally, the confinement splits each 3D hole band into a number of 2D sub-bands. Mixing between the two sets of 2D sub-bands leads to the non-parabolicity of both sets of sub-bands. (See Reference [9], Section 4.5). I do not know of any theoretical work for 1D; however, one would expect that the HH-LH mixing would be reduced with the increasing confinement energy such that the 1D sub-bands arising from the lowest lying 2D sub-band would have mostly heavy hole character and the bands more parabolic than in 2D.

In bulk GaAs, there is extensive literature on the effective masses of heavy holes,
a summary of which can be found in Reference [110]. Values for the effective mass appear to vary widely — the author of the article recommends $0.34m_0$ for the $<100>$ direction and $0.79m_0$ for the $<111>$ direction. These are much larger than the equivalent (isotropic) value for the electrons of $0.067m_0$. Measurements on 2DHGs in the (100) plane with Be-doping indicate that the mass of the first sub-band, which is the one relevant for us, is relatively isotropic [111]. Another set of measurements on substrates similar to the ones from which we fabricate our CEO wires show the hole effective mass depends strongly on both the well width as well as the carrier density due to the non-parabolicity of the 2D sub-bands; values between 0.2 and $0.5m_0$ were reported [112]. For the reasons mentioned above, we might expect these dependences to be much weaker for our wires, with values of the mass perhaps on the higher end of what is reported. Note that the larger effective mass of holes compared to electrons should give rise to stronger hole-hole interactions (compared to kinetic energy), but we do not at present consider the effects of this.

In addition to the effects noted above, spin-orbit coupling is also expected to affect the Landé $g$ factor of semiconductors. In bulk materials, $g$ can be significantly modified from the free particle $g$ factor of two [113]. In 2D, not only can $g$ deviate from 2, it is also predicted theoretically to be highly anisotropic, more so for holes than for electrons [9]. Most experiments on 2DHGs in magnetic field have focussed on an apparent field-induced metal-insulator transition — see for example References [114, 115, 116, 117] — but there is one recent work that shows that $g$ in the [332] direction is much lower than $g$ in the [110] direction [118]; however, due to the limitations of the experiment accurate quantitative analysis could not be done on the data and so values for $g$ were not reported. In 1D, there is as far as I know no theoretical work on how further confinement might affect the $g$ factor; however, measurements have been made of quantum point contacts defined on a 2DHG grown in the (311) direction which show $g$ to be between 0.7 and 1 in the [233] direction (parallel to the QPC channel) while in the [011] direction (perpendicular to the QPC channel) no splitting of the conductance plateaus was observed up to 9T [119]. In both cases the magnetic field was in the plane of the 2DHG.
7.4 The first CEO hole wires

The first hole wire devices were made shortly after I arrived at Bell and we measured them in a $^3$He fridge. I believe this was the first attempt and Loren Pfeiffer was a little surprised that it worked as he'd 'guessed' at growth parameters based on what worked from electron wires.

![Figure 7.10: Measurement of a cleaved edge overgrowth wire. The wire is defined by a gate extending all the way across the chip which, when a voltage is applied to it, first depletes the 2DE/HG underneath it and then depletes each subband in the wire. Ohmic contacts are attached on both sides of the gate. Note that the wire is not measured 'directly' but by contacting the 2DE/HG, which is coupled to the wire.](image)

CEO wires are measured in the configuration shown in Figure 7.10. Conductance is measured between two ohmic contacts on each side of a gate electrode, which extends across the whole chip. As a positive voltage is applied to the gates, the 2DHG beneath it is first depleted, then each of the each of the 1D modes in the wire in succession. This gives rise to conductance plateaus as a function of gate voltage reminiscent of those in quantum point contacts as described in the previous section.
and sketched in Figure 7.6 (c). Typical voltages for which both wire and 2DHG are completely pinched off are between 2.7 and 6V.

Figure 7.11: (a) Conductance steps in gate voltage measured in a CEO quantum wire at different temperatures. A series resistance corresponding to the measured resistance with no voltage on the gate has been subtracted. Temperatures are as measured with a thermometer attached to the cold finger and may not represent the electron temperature in the device. (b) The data in (a) with successive curves offset by $0.2e^2/h$.

Figure 7.11 shows these conductance steps measured in a particular wire at different temperatures. One sees immediately that these data differ from the ideal sketched in Figure 7.6 (c) in a number of ways, each of which helps us understand something about the physics of the system beyond the naïve picture.

First, the plateau heights are suppressed from $2e^2/h$ even after the subtraction of a contact resistance; this suppression does not occur in QPCs. This is most likely due to
a fundamental difference in how these systems are formed and contacted. A QPC repre-
sents a saddle point in the potential of a 2DEG, with the 2DEG smoothly evolving
spatially into a quasi-1D constriction. CEO wires in contrast are long, atomically-
smooth 1D channels which are contacted through the overlap between 2D and 1D
states in regions where the 2D is not pinched off as shown in Figure 7.10 and as
mentioned previously. Thus, the suppression of the plateaus in this system from the
ideal conductance is most likely due to this 2D-1D contact resistance [120].

Second, the plateaus are not entirely smooth — there are reproducible corruga-
tions on each step. These features are most likely resonances due to disorder in the
wire potential leading to some amount of localisation of the charge carriers and are
particular to each wire, i.e. they are not reproducible from wire to wire.

As the temperature is increased, there is an improvement of the ‘noise’ due to
disorder (as opposed to inherent noise in the circuit due for example to ground
loops). One might at this point be tempted to invoke electron-electron interactions
as described in Tomonaga-Luttinger liquid theories for the temperature dependence
as well as the initial suppression of the plateaus at the base temperature of the
fridge [121, 122, 15]; however, it must be pointed out that the temperature depen-
dence of the 2D-1D coupling, which is poorly understood (and could in principle also
incorporate some TLL effects), cannot be ruled out as a factor and must at this point
be considered the far more likely explanation.

7.5 Finite bias

The main analytical paradigm for thinking about CEO wires at finite bias is due to
Rafael de Picciotto and is described primarily in Reference [123]. Here I present the
model in some detail and compare it to the data in hole wires. (A similar analysis
for QPCs will I am sure appear in Lindsay Moore or Chou Hung Tao’s dissertations
and I encourage the reader to compare the two.)

First, we assume a parabolic dispersion so that

\[ \mu = \frac{\hbar^2 k_F^2}{2m} \]  

(7.7)
where $\mu$ is the chemical potential (Fermi energy), $k_F$ the Fermi wavevector and $m$ the effective mass of the carriers.

We then relate the Fermi wavevector to the gate voltage with the following.

$$k_F = \frac{n\pi}{2} = \frac{\pi c V_g}{2 e}, \quad (7.8)$$

where $n$ is the density of carriers, $c$ the capacitance of the (sub-band in) the wire to the gate and $V_g$ the gate voltage. The first equality can be got by counting allowed states in momentum space up to the Fermi energy in 1D in the usual way. The second equality assumes that the gate is the only object that has significant capacitance to the wire. Those more familiar with QPCs will note that this is a significant departure from the normal QPC model where capacitances to the source and drain are very important; however, due to the geometry of the CEO system, this is a reasonable assumption here.

Finally, we write down the definition of the bias voltage $u$, which is the difference between the electrochemical potentials in the leads

$$\mu_+ - \mu_- = eu \quad (7.9)$$

and impose conservation of charge.

$$\sqrt{\mu_+} + \sqrt{\mu_-} = 2\sqrt{\mu} \quad (7.10)$$

Here $\mu_+$ is the higher of the electrochemical potentials in the leads and $\mu_-$ is the lower of the two. From Equations 7.7 and 7.8, one can see that Equation 7.10 says that the number of charges moving in one direction plus the number moving in the other direction must equal the total charge.

After a bit of algebra, Equations 7.7 to 7.10 above give

$$\mu_\pm = \mu \left(1 \pm \frac{eu}{4\mu}\right)^2 \quad (7.11)$$

Now assume that we are sitting on the first plateau, i.e. that $\mu$ is between the
CHAPTER 7. CLEAVED-EDGE-OVERGROWTH HOLE WIRES

bottom of the first subband and the bottom of the second subband as shown in Figure 7.6(a). Let us call the energy difference between the bottoms of the first and second subbands $\Delta_1$.

Then imagine increasing the bias voltage between the leads. When $\mu_+ = \Delta$, the electrochemical potential of the ‘higher’ lead has hit the bottom of the second subband as shown in Figure 7.6(b) and this is the condition for finding the shape of the upper curve of the first diamond, yielding the equation $u = u_\Delta(V_g)$. To find the lower curve of the first diamond, set $\mu_- = 0$ above, giving $u = u_0(V_g)$.

At the ‘tip’ of the diamond, $(u = u^*, V_g = V_g^*)$, $\mu_+ = \Delta_1$ and $\mu_- = 0$, so that $u^* = \Delta_1/e$ — the sub-band spacing $\Delta_1$ can be read directly off this point if it is distinct enough. To find $V_g = V_g^*$ at this point, we note that from Equations 7.7 and 7.8, $\mu = \xi V_g^2$, where $\xi$ is a constant. At the bottom of the second sub-band $\mu = \Delta_1$, so $V_{\Delta 1} = \sqrt{\Delta_1/\xi}$. At the tip of the diamond, from Equation 7.9, we see that $\sqrt{\Delta_1} - \sqrt{\theta} = 2\sqrt{\mu}$ so that $\mu = \Delta_1/4 = V_g^*2$. Thus $(V_g^*/V_{\Delta 1})^2 = 4$ and $V_g^* = V_{\Delta 1}/2$ — the tip of the diamond occurs in gate voltage exactly halfway between pinch-off and the rise to the second plateau.

Note here (and in what follows) that all energies are measured from the bottom of the first subband, which is assumed to be at a constant gate voltage for all biases. This is a key assumption in this model and the question immediately arises as to whether it breaks down, for instance, in finite magnetic field.

Note also that this picture cannot predict the shape of the pinch-off line.

In Figure 7.12, we show data from a measurement of conductance against gate and bias voltages in a CEO hole wire in the region of the first diamond, together with an attempted fit. The upper lines, corresponding to $u_\Delta(V_g)$ are fit to the data with one fitting parameter — $\alpha \equiv cl^2/m$. Inputs to the fit are the threshold gate voltage (4.17V) and the gate voltage at the rise to the second plateau (3.52). $\Delta 1$ can be read off the plot or calculated from $\alpha$. Here it has been found to be about 2.9meV. The value of $\alpha$ that best fits the upper curves of the diamond was found to be $(43pF/metre)^2/m_0$ where $m_0$ is the bare electron mass.

The bottom curves are completely determined by the fixing of $\alpha$, but unlike the electron wires, we note no feature in the data corresponding to these curves. There is
Figure 7.12: Conductance against bias and gate voltages in a CEO hole wire showing the region around the first diamond. A series resistance has been subtracted corresponding to the resistance measured for the 2DHG with no voltage on the gate. The upper black lines are fits to the rise from the plateau with one fitting parameter. The lower black lines are entirely determined from the fit to the upper lines.
also a persistent dip at zero bias which is not seen in the electron wires, possibly due
to disorder-induced localisation. On the second plateau, this dip evolves into a dip
flanked by two peaks as described below, a surprising divergence from the electron
wire data, for which we have no explanation at the moment.

A third difference with electron wires can be seen in the pinch-off line, i.e. the gate
voltage (as a function of bias voltage) at which there is no longer any conductance.
Below the pinch-off line, as the band structure of the wire is no longer relevant, one
might think that the QPC model would again be applicable. In this case, one expects
the pinch-off line to be a straight line on this sort of 2D plot. In electron wires, this is
roughly true, except for a small cusp close to zero bias. [123] This cusp is thought to
be due to localisation at low densities and according to Rafael de Picciotto has been
seen to get smaller with increasing temperature, though these data are not published.
In our hole wires, however, the pinch-off line is far from linear and is not understood.

For the second diamond, things get a little more complicated. Assume that we
can are sitting on the second plateau. Then Equations 7.7 to 7.10 are modified as
follows.

\[
\mu = \frac{\hbar^2 k_{F1}^2}{2m} \quad \text{and} \quad \mu - \Delta_1 = \frac{\hbar^2 k_{F2}^2}{2m} \tag{7.12}
\]

\[
n_1 + n_2 = \frac{(c_1 + c_2)V_g}{e} = \frac{2}{\pi} (k_{F1} + k_{F2}) \tag{7.13}
\]

\[
\mu_+ - \mu_- = \epsilon u \tag{7.14}
\]

\[
\sqrt{\mu_+ + \Delta_1} + \sqrt{\mu_- + \Delta_1} = 2(\sqrt{\mu} + \sqrt{\mu - \Delta_1}) \tag{7.15}
\]

From these equations we can get that

\[
\frac{(c_1 + c_2)V_g}{e} = \frac{2}{\pi} (k_{F1} + k_{F2}) \tag{7.16}
\]

\[
= \frac{2}{\pi} \sqrt{\frac{2m}{\hbar}} \left( \sqrt{\mu} + \sqrt{\mu - \Delta_1} \right) \tag{7.17}
\]
\[
\Rightarrow \sqrt{\mu_+ + \mu_+ - \Delta_1} + \sqrt{\mu_- + \mu_- - \Delta_1} = 2(\sqrt{\mu + \mu - \Delta}) = \frac{(c_1 + c_2)V_g}{e} \sqrt{\frac{\hbar}{2m}} \quad (7.19)
\]

Let us call the energy spacing between the second and third subbands \(\Delta_2\). To find the upper and lower curves for the second diamond, we insert \(\mu_+ = \Delta_2\) and \(\mu_- = \Delta_1\) into Equation 7.14 and then Equation 7.14 into Equation 7.19.

These calculations can be generalised in a straightforward manner to the nth diamond, though the algebra becomes rapidly more complicated. Note that fitting to data for the nth diamond requires all the prior subband spacings, which can only be obtained by looking at all the prior diamonds.

### 7.6 Finite magnetic field

This model can be easily extended to finite magnetic field, where each sub-band is split into two bands with opposite spin with energy difference \(\delta = g\mu_B B\), the Zeeman energy. For convenience in speaking of them later, we shall call the lower energy sub-band spin-down and the higher energy one spin-up. The goal is the analysis of data such as those shown in Figure 7.14, i.e. we desire to have a relation between magnetic field and the location in gate voltage of the rise to each spin-split step.

As before, we label as \(\Delta_1, \Delta_2\ldots\) the energy differences between the sub-bands, which are now the energy differences between the spin-down bands. The analysis is much simpler than for the finite bias data above. For each spin-split band (labelled by \(\alpha\)), we need only the equations

\[
\mu_\alpha = \frac{\hbar^2 k_{F\alpha}^2}{2m} \quad (7.20)
\]

and

\[
k_{F\alpha} = n\pi = \frac{\pi eV_g}{e}. \quad (7.21)
\]
Figure 7.13: (a) The first two plateaus in a CEO wire in magnetic field from 0T (blue) to 9T (red). The field is applied in the plane of the 2DHG and perpendicular to the wire. A series resistance was subtracted at each field corresponding to the measured resistance at that field with no voltage on the gate. The temperature is about 300mK. (b) The same data as in (a) with successive curves offset by $0.05 \frac{e^2}{h}$. 
What we seek is a curve $V_g(B)$ for the edge of each spin-split band. For the first spin-down band, we can relate $V_g$ to $\mu$ simply:

$$\frac{\pi c_1 V_g}{e} = k_F = \frac{\sqrt{\mu^2 m}}{h}. \quad (7.22)$$

If we assume that the bottom of the first (spin-down) sub-band does not move with magnetic field (an assumption worth examining) and again measure all energies from that point, we can see that we get the step to the next ($G = e^2/h$) plateau when $\mu = \delta$, so that the curve $V_g(B)$ we seek is

$$V_g = \frac{e\sqrt{2m\delta}}{c_1 \pi h}. \quad (7.23)$$

Following a similar calculation, the curve for the edge of the second spin-down band is

$$V_g = \frac{e\sqrt{2m}}{c_1 \pi h} \left( \sqrt{\mu} + \sqrt{(\mu - \delta)} \right) \quad (7.24)$$

and that for the second spin-up band is

$$V_g = \frac{e\sqrt{2m}}{(c_1 + c_2) \pi h} \left( \sqrt{\mu} + \sqrt{(\mu - \delta)} + \sqrt{(\mu - \Delta_1)} \right). \quad (7.25)$$

Note that to plot these curves we require the quantities $\alpha_n \equiv c_n^2/m$ which we obtain fits to the finite bias data. The free parameter is $g$.

This model ignores any orbital effects of the magnetic field, which are the dominant effect in electron wires due to their small $g$ factor and which could also be quite large in the hole wires. [96] One way to eliminate these effects from the analysis is to look at energy differences between two spin-split bands from the same orbital state, but this analysis has not yet been done.

### 7.7 Perpendicular Field

Figure 7.14 shows the evolution of the first two plateaus for a particular CEO wire in magnetic field applied in the plane of the 2DHG but perpendicular to the wire.
Figure 7.14: The second plateau from Figure 7.13 with the conductance of the first plateau subtracted.
The first plateau shows no clear dependence on magnetic field, but any such dependence could easily have been obscured by the large amount of switching close to pinch-off. The second plateau on the other hand is clearly split by the magnetic field; in principle we can obtain a $g$ factor from this, but the data have not yet been analysed quantitatively in this fashion.

Another qualitative observation can be made, about the heights of the plateaus. From Figure 7.14 we can see that the first plateau is suppressed with magnetic field even after the resistance of the 2DHG is subtracted, whereas the incremental height of the second plateau is relatively constant with magnetic field. (Figure 7.14.)

### 7.8 Parallel Field: Direct Observation of SO Coupling?

Quite recently we have measured the conductance plateaus in magnetic field roughly parallel to the wire. The results of these measurements are shown below in Figure 7.15.

On this cooldown, the first plateau is much less noisy, most likely because Joseph Sulpizio cooled down the fridge very slowly. (It has been found empirically that cooling down slowly between room temperature and about 100K does wonders for reducing switching in GaAs samples.) This allows us to see more clearly that there is almost no change in the first plateau in magnetic field, which is quite surprising as the second plateau is clearly spin-split. In addition, we note that both plateau heights hardly change at all with magnetic field in this direction. We expect parallel magnetic field also to have minimal effect on the energy of the orbitals, which means that the location of the rises to the plateaus should be fixed in gate voltage. This is true for the second plateau, but in the first plateau we observe a slight shift.

The second plateau also shows a clear and striking dip that increases in depth and width with increasing magnetic field. It is possible that this is a direct observation of the gap in the bands induced by spin-orbit coupling as described in Section 7.3. At the time of writing measurements are being made to confirm this and also to check for this feature on other plateaus and devices.
Figure 7.15: Conductance plateaus in parallel magnetic field from 0T (blue) to 9T (red). The temperature is 300mK.
The dip is not observed on the first plateau. One possible reason for this is that we have simply not reached high enough fields — there is no reason that dips should appear on all plateaus at the same field. An alternative explanation is that the dip on the second plateau is caused not by mixing between spin up and down branches of the same 1D sub-band but between two branches coming from different sub-bands as discussed to some extent in References [124] and [125]. We do not understand why the first plateau does not appear to be spin-split even at 9T.

One surprising thing about these data is the coexistence of the dip and the spin-splitting of the plateaus. If the dip is indeed caused by spin-orbit coupling as described in the model in Section 7.3, this indicates a significant spin-orbit effective magnetic field along the axis of the wire, perhaps due to the Dresselhaus spin-orbit effect. This idea has to be explored in greater detail.

The reader might think that we should apply magnetic field in the third direction (perpendicular to both wire and 2DHG) to get a complete picture. It is a priori unclear whether we will get meaningful data in this way because the Quantum Hall effect in the 2DHG may overwhelm the signal we are looking for; however, this may be worth grappling with.

7.9 Unexplained Resonances

In measurements of conductance against gate and bias voltages around the region of the second plateau, the usual diamond shape was observed an in addition two symmetric peaks at finite bias across part of the diamond. (Figure 7.16.) It is at present date still a complete mystery as to what this is; however, we have several clues.

At zero magnetic field, this resonance appears to extend over a range of gate voltages but to be fixed in bias voltage.

In magnetic field, the location of the feature in bias voltage, as well as its magnitude, hardly change, if at all. The most striking change in magnetic field is a change in the extent of this feature in gate voltage. At zero magnetic field, the feature starts at the side of the diamond closer to the rise to the third plateau and ends abruptly
Figure 7.16: Resonance flanking second plateau (top) and its evolution with magnetic field. The box in the top plot marks the boundaries of the other plots. The temperature is 300mK.
before reaching the other side. (Top of Figure 7.16.) With increasing magnetic field, the location of this abrupt termination shifts to the left, closer to the third plateau, diminishing the extent of the feature in gate voltage. (Rest of Figure 7.16.) The question may be asked: what is this point of termination? Comparing Figure 7.16 to Figure 7.14, one can see that it corresponds to the point in gate voltage at the end of the rise to the second plateau, i.e. where the conductance has reached its plateau value.

![Graph](image)

**Figure 7.17:** Temperature dependence of the resonance in Figure 7.16.

Yet another clue is the dependence of this feature on temperature. Increasing temperature suppresses this feature, though the temperature for complete suppression would appear to be well above 300mK. (Figure 7.17.)

This resonance was also observed in another device on the same chip and is persistent over thermal cycling.

As mentioned previously, we have at present no explanation for this feature.
7.10 Conclusions

We have measured the transport properties of the first GaAs CEO hole wires, which are quasi-one-dimensional systems. As expected, the subband spacing in these systems is smaller than the corresponding electron system, likely due to their higher effective mass. Measuring conductance plateaus with magnetic field parallel and perpendicular to the wire, we observe that the $g$ factor is highly anisotropic. Preliminary data from measurements in parallel magnetic field suggest a direct observation of gap in the spectrum induced by spin-orbit coupling. We also note an unexplained finite bias resonance on the second conductance plateau.

7.11 Bibliographical Note

As the reader will have noted by now, Roland Winkler’s book [9] is the canonical reference on spin-orbit in reduced (mostly two) dimensions. There is no review article or book that I am aware of on spin-orbit in one dimension. It is instructive to compare the relative degree of development of this problem to that of the Kondo problem.
Chapter 8

Measurements on Nanotubes

For the first two years of my PhD and intermittently thereafter I worked on mea­
surements of carbon nanotubes with the primary goal of studying in these ideal one-
dimensional systems (resonant) tunnelling behaviour in Tomonaga-Luttinger liquids
as a function of directly tunable tunnel barriers. Unbeknownst to me at the time, a
prerequisite for the fabrication of these devices was the solution of several very dif­
ficult processing problems. Now, several years later, we believe that we have finally
overcome all of these obstacles and Joseph Sulpizio, into whose capable hands these
experiments have been transferred, will describe them in his dissertation.

In this chapter I described a separate but related set of measurements done in late
2006 on suspended nanotube devices fabricated by Xinran Wang in Hongjie Dai's lab.
The data analysis is not complete and may be continued at a later date.

8.1 Device geometry

The device is similar to those described in Chapter 2 except that the nanotube is
suspended over a etched trench and the nanotube growth is the last step.
8.2 Peak height alternation

At low temperatures, we observe Coulomb blockade, as in the devices described in Chapter 4. Here, however, we also observe a pairing pattern in the peak heights for some ranges of gate voltage: two high, two low etc. as shown in Figure 8.2. A preliminary analysis of the peak spacings shows that a periodic pattern of $A$, $B$, $C$, $B$ where $A > C > B$ may hold. Further scrutiny may allow us to compare our results to predictions e.g. from random matrix theory or from nanotube shell-filling models; however analysis is complicated by switching in the gate as described in Section 8.4.

8.3 Magnetic field dependence

Applying a magnetic field and tracking the motion of peaks allows us to identify the states corresponding to each peak as in Figure 8.3. For the most part, adjacent peaks move in opposite directions indicating that they have opposite spins; however, after a certain magnetic field some peaks change direction indicating an internal spin flip as described in Reference [126].
CHAPTER 8. MEASUREMENTS ON NANOTUBES

Figure 8.2: Alternating pattern of Coulomb blockade peak heights (two high, two low...) seen in suspended nanotube devices. The temperature is the base temperature of the Leiden dil fridge, i.e. lattice temperature ~20 mK and electron temperature probably about 75 mK. (Inset) Focus on a quartet of peaks. ± in the labels mark orbital states, whereas ↑ and ↓ mark spin states. These are determined from the magnetic field dependence of the peak positions described in section 8.3.

Figure 8.3: Tracing the positions of seven pairs of Coulomb blockade peaks from -9 to -8V. Blank space has been removed between all adjacent pairs of peak; each section demarcated with vertical black lines is 20 mV wide in gate voltage.
8.4 Finite bias

Figure 8.4: Conductance against gate and bias voltages for a suspended nanotube device.

Finite bias measurements of Coulomb diamonds, an example of which is shown in Figure 8.4, show occasional gate switching, which may prove to be problematic for analysis of peak spacings at zero bias. A key question, which may be elucidated by further analysis, is whether these switches occur randomly in time or whether they are induced at particular gate voltages. From Figure 8.3 it would appear that the former is probably the case.
# Appendix A

## List of abbreviations

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
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<tbody>
<tr>
<td>AFM</td>
<td>Atomic Force Microscopy</td>
</tr>
<tr>
<td>ALD</td>
<td>Atomic Layer Deposition</td>
</tr>
<tr>
<td>BIA</td>
<td>Bulk Inversion Asymmetry</td>
</tr>
<tr>
<td>CAS</td>
<td>Chemical Abstracts Service</td>
</tr>
<tr>
<td>CEO</td>
<td>Cleaved-Edge-Overgrowth</td>
</tr>
<tr>
<td>CVD</td>
<td>Chemical Vapour Deposition</td>
</tr>
<tr>
<td>DMM</td>
<td>Digital Multimeter</td>
</tr>
<tr>
<td>IVC</td>
<td>Inner Vacuum Chamber (of cryostat/refrigerator)</td>
</tr>
<tr>
<td>GLAM</td>
<td>Geballe Laboratory for Advanced Materials</td>
</tr>
<tr>
<td>MBE</td>
<td>Molecular Beam Epitaxy</td>
</tr>
<tr>
<td>NPGS</td>
<td>Nanometer Pattern Generation System</td>
</tr>
<tr>
<td>OFHC</td>
<td>Oxygen Free High Conductivity</td>
</tr>
<tr>
<td>PMMA</td>
<td>what does it stand for</td>
</tr>
<tr>
<td>RTA</td>
<td>Rapid Thermal Annealer</td>
</tr>
<tr>
<td>SEM</td>
<td>Scanning Electron Microscopy</td>
</tr>
<tr>
<td>SIA</td>
<td>Structural Inversion Asymmetry</td>
</tr>
<tr>
<td>SMU</td>
<td>Source-measure unit</td>
</tr>
<tr>
<td>SNF</td>
<td>Stanford Nanofabrication Facility</td>
</tr>
<tr>
<td>SO(C)</td>
<td>Spin-orbit (coupling)</td>
</tr>
<tr>
<td>STM</td>
<td>Scanning Tunnelling Microscopy</td>
</tr>
<tr>
<td>TEM</td>
<td>Transmission Electron Microscopy</td>
</tr>
</tbody>
</table>
Appendix B

Experimental Miscellany

B.1 Measurement setup

The measurements in this dissertation were performed in four cryostats: (1) an Oxford Instruments HelioxTL single shot $^3$He fridge at Stanford, (2) an Oxford Instruments HelioxTL single shot $^3$He fridge at Bell Labs, (3) an Oxford Instruments sample-in-vacuum dil fridge at Bell Labs and (4) a Leiden Cryogenics MCK-50-100 dil fridge. The principle of operation of these systems will not be discussed here. The reader may consult Reference [127], for example.

Figure B.1 shows a rough schematic of a typical measurement setup and Table B.1 lists typical components of the measurement setup. Matlab code for most of the programmable instruments can be found in the folder \kopenhagen\analysis\Charis\Matlab'.

At present my preferred DC voltage source is the Yokogawa 7651. It is an extremely quiet DC source requiring practically no filtering, but at $3000 per channel it is an expensive solution for experiments requiring a large number of sources. The Keithleys are almost as expensive and considerably noisier; I would not recommend their use except for the crudest measurements at 4K. The multi-channel boards from Harvard and National Instruments are much more cost-efficient. With Joshua Folk’s filtering, the NI board worked reasonably well for me during the short time when I was using our $^3$He fridge. I also used an early version of James MacArthur’s BIASDAC
but found that it was quite noisy and also had a tendency to rail. After substantial effort put into filtering its output, the BIASDAC has been used with considerable success by several members of our lab and initial tests on the new version (which does not have a switching power supply) seem to indicate that it has fewer problems.

For the temperature controllers, the Oxford Instruments ITC was found to introduce substantial noise in our 3He setup and was later replaced by John Cumings and Lindsay Moore with a Lakeshore controller. An identical 3He fridge at Bell Labs with an ITC did not have these problems. The difference might be in the wiring of the thermometers on the two systems as we have observed a general lack of predictability in the quality of the wiring on the Oxford systems.

### B.2 Device fabrication

Many of the less well-known device fabrication details have been discussed in the main part of this dissertation. Photolithography and electron beam lithography processes routinely used in this lab will be discussed in Lindsay Moore’s dissertation. In Table B.2 I list the apparatus used in the fabrication of the samples measured for this work.
APPENDIX B. EXPERIMENTAL MISCELLANY

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Model/Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lock-in amplifier</td>
<td>PAR 124A (also AC voltage source)</td>
</tr>
<tr>
<td>Current pre-amplifier</td>
<td>DL 1211</td>
</tr>
<tr>
<td>Voltage sources</td>
<td>Yokogawa 7651, Programmable DC source</td>
</tr>
<tr>
<td></td>
<td>NI PCI-6703, Analog voltage output board</td>
</tr>
<tr>
<td></td>
<td>Keithley 2400, General purpose source meter</td>
</tr>
<tr>
<td></td>
<td>Keithley 236, Source measure unit</td>
</tr>
<tr>
<td></td>
<td>James MacArthur of Harvard’s BIASDAC</td>
</tr>
<tr>
<td>Voltage readout</td>
<td>HP/Agilent 34401A</td>
</tr>
<tr>
<td>Temperature controller</td>
<td>Oxford Instruments ITC</td>
</tr>
<tr>
<td></td>
<td>Lakeshore 370, AC resistance bridge</td>
</tr>
<tr>
<td>Magnet power supplies</td>
<td>Oxford Instruments IPS</td>
</tr>
<tr>
<td></td>
<td>American Magnetics, Inc. 420</td>
</tr>
<tr>
<td></td>
<td>Lakeshore 625</td>
</tr>
</tbody>
</table>

Table B.1: Instruments used in measurement setup.

It is not advisable to use an AFM without closed-loop feedback (for the lateral position of the tip) for images which one intends to use for alignment purposes. (I did not have any choice in this at the beginning of my Ph.D.) E-beam without closed-loop feedback (for the stage position) on the other hand can work very well for many patterns provided they are carefully designed so that all features requiring fine alignment are written in the same ‘writefield’, without any mechanical movement of the SEM stage.

One of the things that one should learn during one’s Ph.D. is how to judge the robustness of any given device fabrication process or construction project. By a robust process I mean one that, with enough practice, an experimentalist with reasonably ‘good hands’ can get to work reliably most of the time. A fragile process is in contrast one where due to the inherent limitations of the instruments one is using, even someone with an experimental ‘green thumb’ will succeed 10-20% of the time depending on the direction of the wind and the phase of the moon. ‘Robust’ and ‘fragile’ are not the same as ‘difficult’ and ‘easy’. Fabricating CEO wires using the process developed at Bell Labs is difficult and robust; if one pays attention to what one is doing, it is possible almost never to break a sample. Fabricating devices where crucial alignment images are taken on a non-linearly drifty, no-feedback tube scanner AFM is easy and fragile (and very foolish). It is best to plan experiments with as few non-robust steps
## APPENDIX B. EXPERIMENTAL MISCELLANY

<table>
<thead>
<tr>
<th>Equipment</th>
<th>Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electron beam lithography</td>
<td>Raith 150 (SNF)</td>
</tr>
<tr>
<td></td>
<td>Hitachi HL-700F (SNF)</td>
</tr>
<tr>
<td></td>
<td>JEOL SEM with Nabyte NPGS (Ginzton Lab)</td>
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<tr>
<td></td>
<td>FEI Sirion SEM with NPGS</td>
</tr>
<tr>
<td></td>
<td>(Shared by GG, Mellosh and Cui labs)</td>
</tr>
<tr>
<td>Photolithography</td>
<td>Karl Suss mask aligner (Bell Labs)</td>
</tr>
<tr>
<td>Atomic force microscopy</td>
<td>Digital Instruments Multimode</td>
</tr>
<tr>
<td></td>
<td>(Various models: GLAM, Dai lab, Quate lab)</td>
</tr>
<tr>
<td></td>
<td>Digital Instruments Nanoscope 3000 (SNF)</td>
</tr>
<tr>
<td></td>
<td>Asylum Research MFP-3D-SA (GG lab)</td>
</tr>
<tr>
<td>Electron beam evaporation</td>
<td>Innotec ES26C (SNF)</td>
</tr>
<tr>
<td></td>
<td>GG lab e-beam evaporator</td>
</tr>
<tr>
<td></td>
<td>(Parts from Kurt J. Lesker, assembly by Lindsay Moore)</td>
</tr>
<tr>
<td>Thermal evaporation</td>
<td>Evaporator of doubtful provenance in 089 Moore</td>
</tr>
<tr>
<td></td>
<td>(GG lab)</td>
</tr>
</tbody>
</table>

Table B.2: Equipment used for device fabrication.

As possible; however, it should be noted that it is often possible to turn a fragile process into a robust one. For instance, soldering a delicate connector in a dark corner of the lab with an enormous soldering iron tip at an unknown temperature after an over-night e-beam write is fragile; soldering the same connector held in a vice with adequate sleep and lighting, the right solder and a tip chosen carefully for size and temperature is remarkably robust and can even, with practice, be easy and enjoyable. Similarly, the many fabrication tricks described in Section 7.1 make what has the potential be a very frustrating process relatively pain-free.

### B.3 Adder box

As the reader will have noted by now, in many of our measurements it is necessary to add a DC voltage and an AC voltage. I have often used a passive adder circuit such as that shown in Figure B.2. (If the device were drawn in the circuit it would look like a resistor in parallel with $R_4$.)

Assuming typical component values of $R_1 = 4 M\Omega$, $R_2 = R_3 = 2 k\Omega$, $R_4 = 40\Omega$ and $C = 4.7 \mu F$, this circuit will (a) divide an AC voltage by about $10^5$, (b) divide
APPENDIX B. EXPERIMENTAL MISCELLANY

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Figure B.2: Passive circuit for adding AC and DC voltages.

a DC voltage by about $10^2$, (c) filter out high frequencies coming from the DC side with a cutoff of about 30Hz and (d) add AC and DC together.

The rationale behind the values of the components is as follows. $R_4$ is chosen to be very small compared to the typical resistance of the device so that the device itself does not affect the output voltage. $R_1$ is chosen to divide the AC side by $10^5$. $R_2 + R_3$ is chosen to divide the DC side by $10^2$. $C$ was chosen so that together with $R_3$ it is a low pass filter with a cutoff frequency somewhat lower than 60Hz but at the same time not so low as to affect the speed of the measurement. It is placed between $R_2$ and $R_3$ (instead of in parallel with $R_4$) so that the AC voltage is not divided across it resulting in a frequency-dependent AC side voltage division.

As the reader can see, passive circuits such as this must be designed with care and ‘solved self-consistently’ to make sure that the different parts of the circuit do not interfere with each other. Do not forget to think about the output impedances of your voltage/current sources as well as the input impedances of your current/voltage measurement equipment.

B.4 Subtracting series resistance

It frequently happens that one wants to subtract a (known or estimated) constant series resistance in the lines leading up to one’s device. This method of subtracting
series resistance was developed in conversation with David Goldhaber-Gordon and Hung-Tao Chou.

1. The data are in the form of two vectors for the bias applied to the whole system of 'device plus series resistance' (D+SR) and the conductance measured across the same.

2. Doing a numerical integration using, for example, Simpson's Rule [128] one obtains the current through D+SR against bias voltage applied to D+SR. By conservation of charge, the current has to be the same through both the device and the series resistance, so we now have the current through the device.

3. To obtain the voltage across the device alone, start with the voltage across D+SR and subtract the current multiplied by the series resistance.

4. Finally, to obtain the conductance through the device, numerically integrate the current through the device by the voltage across it.

It is tempting to simply convert conductance into resistance in the data above by saying resistance = 1/conductance at each point of the bias voltage; subtracting off a series resistance and converting back to conductance, but this is not rigorous. This procedure may however be OK for a first pass at data analysis or if one is not trying to get very numerically accurate results.

**B.5 Minimising noise**

In order to make good transport measurements at low temperatures, it is necessary to minimise the noise in your measurement system. Noise is something that is very difficult to get a handle on as one does not have control over and often cannot even identify all of its sources. Here I record the conventional wisdom in our lab regarding noise and some general guidelines for reducing it.

The first thing that one does upon noticing that the noise in one's experiment is drowning out the signal (which will inevitably happen if one takes no precautions due
to the second law of thermodynamics) is to measure the noise, which will (a) give one a clue as to where it is coming from and (b) help one to know quantitatively whether ones successful in the attempts to reduce it.

Generally I do this in two ways: (1) I connect the output of my lock-in (or sometimes of my current pre-amp) to a spectrum analyser to ascertain the frequency distribution of the noise. Nine times out of ten, you will find an enormous peak at 60Hz, more about which later. (2) I often also switch the PAR 124A lock-in over to AC voltmeter mode, turn off any filtering I have on the lock-in and also of course also the excitation to the device. This way I am measuring the ‘noise’ at all frequencies coming from my device.

Now that one has a direct measure of how bad the noise is, one can try to reduce it in various ways. Some of the major causes of noise are:

- **Ground loops.** A ground loop is a connected path by which current can flow in the ground of your measurement setup. Stray radiation induces currents in the loop which couple to your signal. A common ground loop in the setup in Figure B.1 is from the ground supplied out of the wall to the shield of the lock-in output (AC voltage source) to the breakout box of your fridge to the shield of the current pre-amp back to the wall. There are two philosophies for dealing with ground loops. The one which I generally prefer is to ground all my instruments at a single point. For instance, the loop described above can be broken by running the current pre-amp off car batteries, a common practice in our lab. Another approach, favoured by Rafael de Picciotto, it to create as many ground loops as possible as they will tend to cancel out as $1/\sqrt{N}$ where $N$ is the number of loops.

- **Floating (ungrounded) shields and chassis** can pick up noise which then couples into your signal, so each shield and chassis should be grounded exactly once. If a shield/chassis is grounded more than once, you have a ground loop. Do not forget to ground your instrument rack.

- **Ground loops and ungrounded shields** of course have to pick up radiation from somewhere. Major culprits are digital instruments especially the Agilent 34401A
DMMs and instruments containing switching power supplies. Mechanical vibrations, e.g. from pumps, can also induce noise in your measurement. It helps to have a shielded room with as few instruments as possible in it and lines to pumps that are well mechanically anchored. If you are not in a shielded room, try turning off other equipment in the room one by one especially those connected to the same power circuit.

- Intermittent connections in lines caused by cold solder joints, damaged cables (that have been stepped on) etc.

Noise can and will change from week to week or even day to day as people turn on and off pumps, ventilations systems, elevators and other equipment in one's building or the building next door. It is good practice to quantitatively check the noise level in your measurement every so often, especially if you notice that the quality of your signal has significantly degraded.

B.6 Bibliographical Note

Almost everything I know about running fridges and making low-temperature measurements I learned from people in lab and I think this is really primarily how one should learn how to be an experimental physicist. Sometimes, however, it's useful to be able to look up thermal contraction coefficients or read about the experiences of others. For general low-temperature experimental techniques some good references are [129] and [130]. This term (Autumn 2007) a course is being offered at Stanford by Aharon Kapitulnik and Kathryn Moler using Reference [131] as the text. And finally, Reference [132] is a reasonably good book on noise reduction and grounding.
Appendix C

Growth of carbon nanotubes by chemical vapour deposition

C.1 CVD furnace

Figure C.1 shows a photo of our CVD furnace. It is modelled after the one in the lab of Hongjie Dai in the Chemistry Department here at Stanford. The Dai lab had done pioneering work on nanotube growth by CVD. I set up most of the furnace during my first year and Joey Sulpizio added the argon and air lines. The furnace in itself is not a very complicated object (see Figure something for schematic), but safety regulations particular to the Moore building slowed its construction considerably.

The main components in our furnace are as follows.

Lindberg/Blue M Mini-Mite tube furnace
MKS 647C mass flow programmer
MKS 1179A mass flow controller (one for each gas)

C.2 Nanotube catalyst recipe

This recipe was provided to us by the Dai lab in chemistry.
Figure C.1: Photo of our CVD furnace in 088 Moore. I also did the bulk of the construction of the (more complicated) nanowire furnace to the right (not shown), but never used it for an experiment.
C.2 Schematic of a CVD furnace for nanotube growth. The mass flow controllers are controlled by a mass flow programmer connected electrically.

**Ingredients**
- 20mg iron nonahydrate, CAS number 7782-61-8
- 15mg aluminium oxide, CAS number 1344-28-1
- 1mg bis(acetylaceto)-dioxomolybdenum (VI), CAS number 17524-05-9
- 15mL methanol

Mix all ingredients and sonicate for about fifteen minutes. Sonicate briefly before each subsequent use to homogenise suspension.

Note: It is not easy to get aluminium oxide in reasonable quantities, as minimum order quantities are enormous. So ask for a sample say of ‘Aluminium Oxide C’ from Degussa and you will get a lifetime supply.

C.3 Growing Nanotubes: Preparing the chip

The following are instructions for preparing a silicon chip for growth of nanotubes.

1. Start with a chip with the desired pattern of catalyst pads developed in PMMA.
2. If necessary, turn on the oven and let it heat to 160°C.

3. Sonicate catalyst for one or two minutes. Drop catalyst on the chip with a pipette, about half as much as you would drop PMMA on a chip to spin.

4. Leave under a petri dish to evaporate, propping up the dish slightly so that it doesn’t take a couple aeons. Wait about five minutes.

5. Place the chip on a glass slide or glass petri dish and put it in the oven for five minutes.

6. While the chip is still hot, drop it into acetone to liftoff the PMMA. Wait about five minutes.

7. Transfer the chip to a new vial of acetone. Sonicate briefly at a low power if necessary.

8. Rinse chip with acetone and isopropanol as usual. Blow dry.

**C.4 Growing Nanotubes: Running the furnace**

The following are instructions for growing nanotubes in the furnace once the chip has been prepared as described above.

1. Put the chip in the furnace tube. Make sure all seals are good. Conflat connections should be made in a ‘star formation’. Wear gloves. Do not lay the fool; this is a flammable gas furnace.

2. If necessary, turn all gases on at the cylinders. (DO NOT TOUCH THE VENT VALVE in the gas cabinets. As they say, do that ‘and you will surely die’. I’m not kidding. Just don’t do it.)

3. Let all gases flow at the desired rates for about five minutes to purge the lines. At this point, the furnace is OFF.
4. Now turn off all gases except hydrogen — turn them off at the controller and also close the manual valves in the rack. You may or may not want to increase the flow rate of hydrogen.

5. Turn on the furnace and let it heat to the desired temperature. All this while, only hydrogen is flowing.

6. When the temperature stabilises (five to ten minutes), turn the flow rate of hydrogen back to normal, and turn on the ethylene and methane. Flow all gases for three to ten minutes depending on what has worked in the recent past.

7. Turn off ethylene and methane and lower the furnace setpoint to room temperature. Raise the lid of the furnace and turn on the fan, directed away from you and other people.

8. When the furnace reaches about 100 – 150°C, TURN OFF THE HYDROGEN. Wait a few more minutes. Then you can open the tube and take your sample our. Record all data in the furnace logbook. Make notes of any unusual events.

9. Turn off all gases if you’re the last person that day.
Appendix D

Leiden Dilution Refrigerator: Operating Instructions

This document is intended as a list of guidelines/reminders to a trained user on how to run the Leiden dilution refrigerator (dil fridge for short) in the Goldhaber-Gordon lab, not as a substitute for training. This is also not a tutorial on how dilution fridges work. For that, the reader is advised to consult Reference [127].

A general rule of thumb is that one doesn’t learn to run fridges well by following recipes, but by keeping one’s eyes open and getting a feel for the system. It is also a good idea to, in the words of Mark Topinka, ‘engage brain before pressing button’.

In addition to this fridge, I’ve also used one made by Oxford Instruments at Bell Labs. In what follows I occasionally compare the two to give the reader some idea of the variety of ways in which a fridge like this can be designed as it is often helpful to learn the same thing from different angles, even (or perhaps especially) technical things such as running a dil fridge.

It is worth noting here that both dil fridges I used are ‘non-top-loading’, which is to say that the sample is in a vacuum can and it is necessary to warm the whole fridge up change samples. The Oxford dil fridge in our lab (and one of the ³He fridges is ‘top-loading’ which is to say that the sample is immersed directly in liquid helium and can be removed without warming up the fridge. I shall not discuss the differences between these two classes of fridges here.
D.1 Cooling down the dewar

- If the dewar is already cold, skip this section. The dewar temperature can be estimated by measuring the magnet resistance.

- Make sure you have all the fittings you need before you begin.

- Pump out the dewar with a scroll pump, then vent and over-pressure with nitrogen gas.

- Repeat the previous step as often as you like. Be careful to have the dewar over-pressured whenever you exchange or remove fittings.

- Fill the dewar with liquid nitrogen. Leave for a few hours, preferably overnight, so that the magnet equilibrates to 77K.

- Over-pressure with nitrogen gas to force out the liquid into a storage dewar. When no more liquid is coming out, blow nitrogen gas into the bottom of the dewar for about half an hour to ensure that the remaining nitrogen has evaporated. Make sure that the magnet resistance has started to rise above its 77K value.

- Pump and purge again, now with helium gas.

- Over-pressure with helium gas and fill with liquid helium. Note that as the dewar is relatively warm, you do not need to wait for the transfer tube to cool.

- Note that 60L of liquid helium should get you to about 80Ω. If you find that you are using much more helium than usual, you may have left some liquid nitrogen in the dewar. Another sign of frozen nitrogen is a quenching magnet.

D.2 Loading your sample

- Put your chip into the chip carrier

- Attach the radiation seal.
• T off the IVC. One side goes to the 3He cylinder, the other to a pump. It is a good idea to have a valve on the T on the pump side.

• Coat the IVC cone seal with Dow Corning 111 ‘valve lubricant and sealant’ (in the green tubes). Attach the can and pump on the IVC for a few hours.

The other common method of making a low temperature seal is with an indium O-ring. Here the joint consists of flanges on each side much like conflat or KF connections, except that the O-ring is indium and has to be remade out of indium wire with every cooldown. If one has to make such a seal it is advisable to coat the indium wire with a little vacuum grease to make it easier to remove later (with plastic tweezers, one does not want to scratch the flange or it will cease to seal). Also make sure that the wire overlaps itself by about 1cm as it goes around the flange; otherwise there will be no seal. It is really a toss-up as to which sort of seal is more finicky — indium or cone. The grass just seems greener on the other side and with practice one can learn to make both kinds reliably.

• One may also pump on the fridge at this time.

• When you are satisfied that there are no leaks and the IVC has got to a low pressure (low $10^{-3}$ mbar), close off and disconnect the pump, and let a few mbar of 3He into the IVC. The valve mentioned before ensures that most of the 3He goes into the IVC rather than into the bellows connecting the T to the pump.

• You might want at this point to check, to the extent that this is possible, that your sample is alive at this point at room temperature so that you don’t cool down the fridge for nothing.

• Note that we have removed the sorb from the IVC. See Appendix E.
D.3 Cooling down to 4K

- There are four thermometers on this fridge: a) a RuO₂ on the 1K pot, b) a Dale 10K on the still, c) one from Dr. Frossati on the mixing chamber and d) a nominally identical one attached to the cold finger. Calibration curves for (b) and (c) have been provided by Giorgio Frossati, for (a) from Oxford. [Note that this information is no longer correct since the fridge’s last trip to Leiden in late 2007. Check the lab notebook or talk to Joseph Sulpizio for up-to-date information.]

- Lower the fridge into the dewar slowly, making sure that the helium blowoff from the vent port is not excessive. With GaAs samples one might want to cool even more slowly as this has been shown to decrease ‘switching’ in devices later.

- With the 1K pot input above the compression seal at the top of the dewar, attach all the bellows.

Note that it is the normal practice to over-pressure the 1K pot with helium throughout this process of cooling to 4K rather than to pump on it in this fashion. In this case, the pumping is necessary because of the absence of a needle valve at the input to the 1K pot which can be opened during the over-pressureing. Instead, there is a fixed impedance in the form of a very small capillary. This is partly because of space constraints (this is a very small fridge) and also partly because, as far as I understand, Giorgio Frossati believes that needle valves are not necessary.

- Lower the fridge a bit more until the 1K pot input is about 5cm below the compression seal. Start pumping on the 1K pot: start S4, open A7 and A1. Make sure that the Speedivalve before the over-pressure valve is closed.

- When P1 has dropped to nothing, close A7 and watch for increasing pressure on P1 as helium gas enters the 1K pot. In two minutes, the pressure should rise
to 4–6mbar. If it doesn’t, you have a frozen 1K pot pickup port. You will want to warm it up to room temperature and try again.

- Lower the fridge all the way and leave it there while it cools to 4K. Usually the fridge will get caught on the magnet. When this happens, make sure that the rope is not too slack and rotate the fridge gently while supporting it as much as possible. Within a turn it will find its way into the magnet bore.

- Remember to open the Speedivalve when P1 is above atmosphere.

- As the fridge is cooling to 4K, pump on the lines and the cold trap for a few hours. Open A8, A9, 5, 8, 2 and 0.

- Turn on the turbo pump after P4 has dropped below 20mbar.

**D.4 Cooling down to 1K**

- When the fridge is at 4K, pump on the IVC until pressure is off-scale ($<5\times10^{-4}$). Wait at least another half hour to an hour.

- Close off the IVC and remove the pump.

- Start pumping on the 1K pot and watch its temperature drop to below 2K.

**D.5 Cooling down to base**

- The main indicator of how well the fridge is cooling is the 1K pot temperature; however, also keep an eye on the still pressure, P5, the flow rate and other temperatures.

- Close 5, put nitrogen in the cold trap dewar and the cold trap in it.

---

Note that most fridges also have a helium cold trap. This is not necessary in this case because all our pumps are dry, i.e. there is no oil in them.
• Check that all valves are closed except for those associated with the 1K pot. Open valve 4 to begin letting helium into the cold trap.

• Open the circulation path. Open valves Aux-1, 0, 2, 3, 5 and 6.

• Start the pumps. Begin with pump 3 and then when P4 drops below 20, turn on the turbo.

• Start letting out '3He' behind the scroll pump. Open M1, 9 and 10. Release the gas bit by bit at first, watching the 1K pot temperature and all indicators especially those mentioned above. Do not let the 1K pot temperature rise above 1.8-2K. When the fridge is working well, it should be possible to let gas out continuously.

In most fridges, one does not start circulating until all the mixture is out. One opens the needle valve for increased cooling power and lets the mixture out on both sides (still and condensing) of the fridge, relying on the cryopumping to get the mixture out of the dumps. In this scenario, one can check that the main impedance is not blocked by first letting mixture out on one side and watching the pressure rise on the other side. (After a few cooldowns, one should have some canonical rate of increase to compare to.)

Note that the gas condensing on the still side will not pass through the cold trap(s) — the thinking is that any impurities should freeze to the walls of the still and not affect the main impedance of the fridge.

• When the pressure behind the pump rises to 300 or so and it becomes impossible to let any more out that way, use the pump to push the rest of the gas out. Close valve 9. Open 12 and close 2. In addition to monitoring everything else, do not let the pressure behind the turbo pump rise above 20. You may hear it complaining even before that.
• Do the same thing for $^4$He but leave 200-250mbar in the dump. This is because
the amount of helium supplied to us was sufficient to cool the fridge without a
cold finger attached. As the cold finger takes up space in the mixing chamber,
less $^4$He is now required. Monitor the capacitance of the still. It should rise
about 2pF over the course of the emptying of the dumps.

• When all the mixture is out of the dumps, close all valves except M1, M2 and
the circulation path.

• You may apply heat to the still to increase the cooling power and decrease the
base temperature. We have found that something in the range of 3-8% usually
works quite well, but this will depend on your heat load etc.

• To increase the sample temperature, apply heat to the sample. We have sta-
bilised the temperature in this fridge up to 500mK, but did not push this to its
limit, so it should be possible to go higher than that.

• Keep an eye on the fridge at all times. Feed it nitrogen twice a day and put in
60L of helium whenever you can. i.e. when the level has got to 100Ω. (The 1K
pot pick up is at about 130Ω, but I try not to live dangerously.)

D.6 Warming up from base to 4K

• Close the circulation path by closing valve 4.

• Immediately open a path to the 3He dump by opening valves 9 and 10. M1
should already be open.

• Open valve 8 to pump on both sides of the fridge.

• Stop pumping on the 1K pot by closing valve A7. This is to allow the 1K pot
temperature to rise to 4K and stop its providing cooling to the fridge.

• Heat may be applied judiciously to the mixing chamber and still thermometers
to expedite fridge evacuation.
• Monitor P1. When the pressure rises above atmosphere - this will take tens of minutes to a couple of hours - open the Speedivalve in the gas handling cabinet that is located before the blow-off valve on the 1K pot pumping line.

• When the pressure in the 3He dump reaches the value on the label, stop pumping gas into it and start putting gas into the 4He dump instead. Do this by closing valve 10 and opening valves 14 and 11. Close M1 as well.

• When the \(^4\)He dump is full, close all valves and turn off all pumps in a sensible order. Remember to close M2.

D.7 Warming up to room temperature

• Double-check that all valves are closed.

• Disconnect all hoses and measurement cables. The 1K pot will get blocked but that is OK as you are going to warm it up soon.

• Raise the fridge out of the dewar and wait for it to warm up. You may put some exchange gas into the IVC to speed up the process.

• Hang the fridge from the Genie lift.

• To take off the IVC, vent it to helium and over-pressure slightly (1-2 psi). It is usually necessary to hit the IVC gently at the end with a medium-sized screwdriver or small spanner/wrench while holding on to it. It is a good idea to have the fridge close to, but not touching, the floor at this time.

• Don’t forget to pump out the nitrogen cold trap. Take it out of its dewar, make sure S4 is on, and open valves A8, A9 and 5. Never warm up a trap without also pumping on it. When the trap is at room temperature, you can close all valves and then turn off the pump.
D.8 Other Notes

- The helium level meter should be run at 70mA which can be supplied from the triple current source in the gas handling cabinet and its resistance is 4.5Ω per centimeter. The 1K pot pickup port is at about 140Ω, but you should put in a 60L dewar of helium whenever you can. Don’t live dangerously.

- Monitor your helium consumption. If there is an increase without any clear reason, it may be because the vacuum jacket on your dewar has gone ‘soft’, which is to say that air has got into it and the pressure has increased.
Appendix E

Leiden Dilution Refrigerator: Adventures, Trials and Tribulations

We received a dil fridge from Leiden Cryogenics in September 2005, just before I returned from Bell Labs. The thinking was that I would wire and set up the fridge in order to measure wires from Bell as well as carbon nanotube samples together with Joseph Sulpizio. Many things happened to prevent this from happening in a timely fashion and it was only in these past few months that the fridge has been working in a somewhat reliable fashion.

In this appendix I describe the process of setting up the fridge in hopes that it will be instructive and perhaps edifying to future users of the fridge as well as others who are setting up dil fridges.

E.1 Preliminaries

To begin with as the fridge came from the Netherlands, its electrical plug did not fit any of the sockets in the lab. This problem was solved by the simple expedient of cutting off the plug and soldering on one of the appropriate form. Mercifully, we did have the right sort of power supplied to some of the outlets in the lab. It was also
fortunate that there was only one plug, which powered a strip that everything else was plugged into.

Next, cooling water had to be supplied to the turbo pump. As Lindsay Moore had recently dealt with the problem of making sure that cooling water was supplied to the lab in the course of setting up our electron beam evaporator, this was just a matter of finding the right fittings.

Finally, as the dewar for the fridge was not in a pit, we could not use a Genie Lift forklift to raise and lower the fridge out of and into the dewar. Thus, we had to purchase a hoist/winch from McMaster-Carr and attach it to an eye bolt in the ceiling. In this and in much of the initial testing of the fridge I had the invaluable help of Ileana Rau and Hung-Tao Chou. The Manoharan lab also kindly lent us some sort of special drill for making a hole in the ceiling. Upon ascertaining that we could swing from the winch chain without causing it to detach from the ceiling, we concluded that it would support the fridge which weighs about 40lbs.

The winch was also useful for another purpose: the magnet and dewar came in several pieces. We soldered the magnet to its leads and lowered the magnet support into the dewar with the new winch. These were again not very heavy, though I forget the exact weight — perhaps around the same weight as the fridge.

E.2 Superfluid Leak

Early into our first attempt to cool down the fridge for the first time, we concluded that we had a superfluid helium leak from the fridge circulation path to the IVC. Following the procedure outlined in the preceding appendix, after letting most of the $^3$He'out of the dumps, the following occurred as documented in an e-mail to Giorgio Frossati of Leiden Cryogenics.

7. When the $^3$He' dumps were at 100mbar, something strange happened:
   a. The 1K pot went from 28-29kOhm to 26-27kOhm.
   b. The still pressure rose from mid-2 to 1.6-1.7e-1mbar
APPENDIX E. FUN WITH THE LEIDEN DF

8. Within the next five minutes, all hell broke loose:

a. The 1K pot went to 28-29kOhm
b. The still pressure went to 2.5e-1mbar
c. The turbo pump quieted down
d. The still went to 58kOhm
e. The MC went to 3.5kOhm

and then...

f. The still pressure shot up
g. The IVC pressure increased to high-4 to low-3.

and we went into the same state we were in yesterday where:

h. The still and IVC pressures are inversely proportional.
i. The 1K pot is stuck at 23-25kOhm.
j. The still is slightly colder than the IVC
k. The MC is around 3.1kOhm
l. Nothing gets any colder
m. The process is not reversible (we tried to put things back into the dumps to see what would happen)

8. We also let a small amount of gas out of the IVC into the leak detector. It was all 4He with a partial pressure of 10e-9. No 3He was detectable.
APPENDIX E. FUN WITH THE LEIDEN DF

On the suggestion of Dr. Frossati, we tried to leak check by immersing the fridge (without the IVC can) in liquid nitrogen and then raising it up slowly while spraying helium gas around the level of the liquid and leak testing on the main volume of the fridge. If there is a detectable leak at 77K, one might expect to see an increase in the helium signal when the fridge is raised so that the position of the leak is just above the liquid nitrogen level. For cold leaks, this is not a bad idea as most leaks that appear at 4K can be detected also at 77K. In our case, however, we did not see a signal and eventually shipped the fridge back to Leiden in late October 2005 and received it back a week before Christmas. This was the point at which we removed the sorb from the IVC in order to determine unambiguously that the helium we were seeing in the 'leak' was coming from the fridge circulation path rather than the sorb. We never put the sorb back and have not found it necessary. (If we were using $^4\text{He}$ as exchange gas this might not be true, but I'm not sure as I've only used $^3\text{He}$.)

E.3 Touch

In early 2006 we were able to cool down the fridge with the cold finger stub supplied by Leiden, though the cool down time was on the long side (6-8 hours as I recall) as the 1K pot impedance was quite high.

Early in the summer of 2006, after construction of the cold finger and radiation shield (the revised version of which is described in Section E.6 below), and assembly/construction of all the necessary electronic components, we attempted to cool down with a sample. During this time, we were able to perform some measurements on nanotubes (described in Chapter 8); however, we soon concluded that we needed to re-design our cold finger and radiation shield as the radiation shield would occasionally touch the IVC can, causing a thermal short from the fridge to 4K.

Two factors contributed to this. Firstly, the cold finger stub protruding from the mixing chamber has a long aspect ratio and is made of copper. This means that it is very flexible. In fact, we found that if we lay the fridge flat on a table with our cold finger attached, the stub would deform over time. This was quite a frightening discovery, as you might imagine, after which we started hanging the fridge on the
Genie Lift instead. Secondly, the initial radiation shield was designed to be rather larger in diameter (42mm) than it needed to be as I had (mistakenly) counted on the whole assembly being rather rigid. (The OD of the IVC is 50mm and its wall thickness is less than 1mm.)

After attempting to salvage the original radiation shield by attaching plastic spacers to it (we learned from Giorgio Frossati that a certain kind of Vespel with 40% graphite is very thermally non-conductive) finally I gave up and re-designed the cold finger. (See drawings in Section E.6.) This meant re-wiring the whole thing as well.

### E.4 Blocked 1K Pot

Starting in August 2006, we also ran into the problem of the 1K pot impedance blocking. The symptoms are obvious — the 1K pot pressure drops to zero and its temperature rises to 4K. The first time this happened, the block cleared on cycling to room temperature, leading us to suspect impurities in the helium.

Soon afterwards, in early October, we experienced a block which did not go away at room temperature. Here again is an excerpt from my correspondence at the time about tests done at room temperature.

1. We over-pressured the 1K pot with helium gas to about 1300 mbar, then valved off the helium cylinder and waited for the pressure to drop. It didn’t drop noticeably.

2. We pumped on the 1K pot, then stopped pumping and waited for the pressure to rise. It rose from ‘-2’ to 3[mbar] in two minutes, which in my experience is very low.

We received new capillaries and filters from Leiden and together with Joseph Sulpizio and Benjamin Huard, I changed out the impedance. Initially in November 2006 this seemed to have worked in the sense that the capillary was not blocked at
room temperature though its impedance was still rather high. On trying to cool down
the fridge, we discovered that it took three days to condense compared to about two
hours in the normal mode of operation (at the time of writing).

There was a hiatus after this as I was first at home and then (somewhat prematu­
rely, I thought at the time) gallivanting around Europe for job interviews. One
must bear in mind also that in parallel to all this the cold finger re-wiring and such
were going on. Also, Joseph Sulpizio and I were perfecting processing techniques for
nanotube devices, which I hope will be measured very soon as part of his dissertation.

In March 2007 I replaced the impedance, again with the help of Joseph Sulpizio
and now found that it was completely blocked at room temperature.

We sent it back to Leiden in April and received it back in May. Since then, we
have not have too many issues, except that four sample pins are now useless due to
re-wiring at Leiden that did not correspond to our wiring here. (Pin drawings can be
found in the fridge notebook.)

E.5 Damaged IVC cone seal

In August 2007, in a heroic combined effort, Joseph Sulpizio and I damaged the cone
seal on the IVC can — I propped it up against the Genie lift and he kicked it over. We
could no longer achieve vacuum in the IVC. After some consultation with Paul Brink
and Walter Ogburn in the Cabrera group as well as Giorgio Frossati, we decided
to have the machine shop lathe out the IVC can to the 2° half angle specified by
Giorgio Frossati. The shop could not measure this angle directly from the IVC can
as, according to Kalheinz Merkle, ‘it is egg-shaped’.

After this, there was still a slight leak. Taking the advice of Giorgio Frossati, we
abraded the can against the fridge. As we could not lay the fridge flat and clamp
it to a table as he suggested, we formed a vacuum seal as usual except that instead
of vacuum grease, we used a paste mixed from vacuum grease and abrasive powder
containing particles about 10\(\mu\)m in size. (We found this powder on a long-forgotten
shelf in the machine shop, but similar things can be obtained from companies such
as McMaster-Call.) After two rounds of this, we cleaned the seal very carefully and
found that the IVC now held vacuum. A couple of weeks after the catastrophe, we were back in business.

We did not take the other piece of advice that Dr. Frossati gave us, which was to make a $2^\circ$ half angle cone out of stainless steel and pound it into the can to reshape the seal.

Figure E.1: Drawing of Part A of a cold finger/radiation shield. This attaches to a stub protruding from the mixing chamber. The material is OFHC copper and the units millimetres.
E.6 Cold Finger Construction

Two cold fingers were made for two different chip carriers. The machine shop drawings for the various pieces (A, B, C and D in Figures E.1 to E.4) of one cold finger and the associated radiation shield are shown on the following pages. In this section I describe the cold finger in general and also some things to be aware of when designing such an object. Please note that the drawings below were 'almost final' drafts. Changes have been made subsequently in consultation with the Physics department machine shop. The reader may use these drawings as a template for his/her own work but should not just get them made as drawn. All original SolidWorks files can be found in ‘\kopenhagen\analysis\Charis\DilFridge’.

Part A is designed to be bolted to a stub of similar aspect ratio protruding out of the mixing chamber. It is made of OFHC copper (subsequently gold-plated) for good thermal conductivity. As mentioned above these copper stubs bend very easily and must be handled with care. A design suggestion I made to Giorgio Frossati was to have the stub on the mixing chamber (and for us on this piece) rotated by 90° and also be made fatter so that it is less likely to bend.

Part B is the main part of the cold finger. It is gold-plated to improve thermal contact to Part A. The central section is designed for mounting PCB filters as shown in Figure E.5. Wires are wrapped around the cylindrical sections above and below the PCBs to improve thermal contact between them and the cold finger. After wrapping, the wires are ‘epoxied’ in place with GE varnish. The 32 pin chip carrier (not shown) is attached to the bottom of this piece with a set screw which is screwed and epoxied into it (the chip carrier). As much as possible the corners of this piece were rounded to avoid cutting wires wrapped around it. A small rod was press fit at the top of the upper cylindrical section and at the end of the bottom one similar to Figure E.5 to make wrapping the wires easier. In addition, at the suggestion of Lindsay Moore, small holes (not shown) were made in the cylindrical sections in order to prevent the creation of a semi-sealed space. This could have been done also by drilling the bores of these sections straight through the ‘bars’ marking the beginning of the PCB section.
Figure E.2: Drawing of Part B of a cold finger/radiation shield. This is the main part of the cold finger. The material is brass and the units millimetres.
Figure E.3: Drawing of Part C of a cold finger/radiation shield. This is the radiation shield. It attaches to Part A. The material is brass and the units millimetres.
Part C is the radiation shield. It shields the sample from black body radiation at 4K coming from the wall of the IVC. It is attached to Part A with screws.

Figure E.4: Drawing of part D of a cold finger/radiation shield. This is the cap at the bottom of the radiation shield. It attaches to Part C. The protruding tube is soldered in and subsequently bent. The material is brass and the units millimetres.

Part D completes the radiation shield. It is screwed on to Part C.

Figure E.5 shows the assembled and wired cold finger with a 16 pin chip carrier, which is not what is drawn in Figures E.1 to E.4 but is also not too different. Note here that the wires coming out of the tube at the top have been encased in heat shrink tubing in order to protect the insulation as they are passed through the tube. They are soldered into a circular ring, which plug into a similar ring at the bottom of the mixing chamber. The plugging/unplugging process can be quite difficult ('fragile'
Thermal Anchoring

As mentioned above, Parts A and B were gold-plated (without nickel, which is magnetic) to improve thermal contact between them. Ideally they should also be press-fit together, but I erred on the side of caution and allowed a little more wiggle room.

An oxygen-free high-conductivity (OFHC) copper braid soldered to a wire running through the mixing chamber is wrapped around the cold finger over its whole length as shown in Figure E.5.

There was some concern about solder going superconducting (and thus thermally insulating) at low temperatures, so I painted over most of the connections on both the chip carrier side as well as the connector ring side with silver paint. I am not sure if this was necessary, but it is a quite difficult and not very robust process, in the sense that even after a lot of practice, there is much room for error because of geometrical and mechanical constraints. (I forgot to do this for the socket side of the 16 pin cold finger, which were used for the hole wire measurements in Chapter 7.)
Filtering

The filtering in this fridge is the same as on the Oxford dil fridge (see Ronald Potok's dissertation) except that we do not have coax. We have not as yet been able to determine the electron temperature precisely, but based on the data in Chapter 7 we expect that it is about 75mK, which was really all we were hoping for given that the fridge is in a particularly electrically noisy room.

Other Notes

Pay attention to the thermal contraction coefficients of your materials and ask whether joints will become tighter or looser at low temperatures.

There is also an inherent trade-off to be aware of, especially if one plans to ramp magnetic field frequently, due to the Wiedeman-Franz law connecting electrical and thermal conductivity. High conductivity metals (such as OFHC copper) will cool down quickly, but will also support large eddy currents in changing magnetic field, leading to greater heating of the sample. The obverse is true for low conductivity materials.

Be aware of the possible unintentional creation of nearly-isolated volumes of gas, which can cause problems with 'out-gassing' — gas will be pumped out more slowly from these spaces than from the rest of the chamber (usually the IVC) giving the appearance a slow leak. In Part B, for instance, one will notice a small hole in the cylinders to prevent this. Similarly, in Part D, one will notice a bent tube serving the same purpose for the volume inside the radiation shield.

Why a bent tube? One should never have a hole in one's radiation shield — that would defeat the whole purpose of having a radiation shield; however, one does need openings in order to get wires etc. in and out. Following advice by Ronald Potok, which I have not verified independently, radiation can be greatly attenuated by tubes (instead of direct opening) with aspect ratios of three or more. If possible, one can also insert copper gauze into the various tubes.

Obviously the installation of a fridge is a complex process with lots of room for error. I pass on to the reader some wise words from John Cumings: 'When you can
get orders of operations right, you are ready to graduate.'
Appendix F

Catalogue of Hole Substrates and Devices

The tables in this appendix catalogue all 2DHG substrates grown for the CEO hole wire project as well as all overgrowths (cleaves) done. All these were done by Loren Pfeiffer and Kenneth West at Bell Labs. A brief explanation of terms that may be confusing precede them. Further details on the MBE growths can be found in my lab notebook.

- The setback is the distance of the first pulse of carbon after the AlGaAs well.
- The numbers listed under the column heading ‘C (s)’ are the durations in seconds of the two pulses of carbon, one near the well for doping and the other near the surface to prevent all the carriers from the first pulse from migrating to the surface.
- The density of mobility for the cleaves were measured on a 2DHG grown simultaneously in the MBE chamber.
- A maximum of five chips can be processed during a single overgrowth step.
Table F.1: 2D substrates used in fabrication of CEO hole wires

<table>
<thead>
<tr>
<th>Label</th>
<th>Mobility (10^5 cm^2/Vs)</th>
<th>Density (10^{11} cm^{-2})</th>
<th>Well width (Å)</th>
<th>% Al</th>
<th>Setback (Å)</th>
<th>C (s)</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>8-19-03.1</td>
<td>49 (0.3K)</td>
<td>1.8</td>
<td>150?</td>
<td>32</td>
<td>400?</td>
<td>11/20</td>
<td>Never used for anything.</td>
</tr>
<tr>
<td>11.6.03.1</td>
<td>1.4 (4K)</td>
<td>1.7</td>
<td>220</td>
<td>32</td>
<td>400</td>
<td>11/20</td>
<td>Used up?</td>
</tr>
<tr>
<td>1-10-05.1</td>
<td>2.1 (4K)</td>
<td>2.6</td>
<td>150</td>
<td>32</td>
<td>400</td>
<td>11/20</td>
<td>Flash of light at 0.3K.</td>
</tr>
<tr>
<td>1-10-05.1</td>
<td>15 (0.3K)</td>
<td>1.9</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3-24-05.2</td>
<td>1.1 (4K)</td>
<td>2.4</td>
<td>220</td>
<td>32</td>
<td>400</td>
<td>11/20</td>
<td></td>
</tr>
<tr>
<td>5-17-05.2</td>
<td>1.1 (4K)</td>
<td>1.6</td>
<td>220</td>
<td>32</td>
<td>400</td>
<td>11/20</td>
<td></td>
</tr>
<tr>
<td>5-20-05.1</td>
<td>1.5 (4K)</td>
<td>2.1</td>
<td>220</td>
<td>32</td>
<td>400</td>
<td>11/20</td>
<td>Used up.</td>
</tr>
<tr>
<td>6-2-05.1</td>
<td>1.3 (4K)</td>
<td>2.3</td>
<td>220</td>
<td>50</td>
<td>500</td>
<td>11/20</td>
<td>50% Al bad.</td>
</tr>
<tr>
<td>6-9-05.1</td>
<td>1.5 (4K)</td>
<td>2.1</td>
<td>220</td>
<td>32</td>
<td>400</td>
<td>17/30</td>
<td>3/4 to be processed.</td>
</tr>
<tr>
<td>6-9-05.2</td>
<td>1.4 (4K)</td>
<td>2.8</td>
<td>220</td>
<td>50</td>
<td>500</td>
<td>17/30</td>
<td>3/4 left, some pieces ready to load, but 50% Al is not good.</td>
</tr>
<tr>
<td>7-13-05.2</td>
<td>1.5 (4K)</td>
<td>1.1</td>
<td>220</td>
<td>32</td>
<td>400</td>
<td>11/20</td>
<td>Copy of 5-20-05.1.</td>
</tr>
<tr>
<td>Label</td>
<td>Substrate</td>
<td>Mobility $(10^5 \text{cm}^2/\text{Vs})$</td>
<td>Density $(10^{11} \text{cm}^{-2})$</td>
<td>Comments (overgrowth)</td>
<td>Comments (devices)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>----------</td>
<td>-----------</td>
<td>--------------------------------------</td>
<td>----------------------------------</td>
<td>-----------------------</td>
<td>--------------------</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2-17-05.1</td>
<td>1-10-05.1</td>
<td>1.9 (4K)</td>
<td>2.6</td>
<td>The data in the APL are from #1.</td>
<td>Only two pieces. Both broke. #1 Average depletion (AD) 2.7V. #2 AD 2.4V.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3-29-05.1</td>
<td>1-10-05.1</td>
<td>2.6 (4K)</td>
<td>1.9</td>
<td>Setback 32nm.</td>
<td>#1 Used for ohmic test, could be rewired. #2 Severe leakage.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4-1-05.1</td>
<td>3-24-05.2</td>
<td>2 (4K)</td>
<td>1.4</td>
<td>Same as 2-17-05.1.</td>
<td>#1 Severe leakage. #2 Severe leakage. #3 Sacrificed for studying leakage. #4 Sacrificed for studying leakage. #5 Five nd, seven leak, one works.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5-19-05.1</td>
<td>3-24-05.2</td>
<td>2.9 (4K)</td>
<td>2.1</td>
<td>Same as 2-17-05.1.</td>
<td>Only two pieces. #1 Severe leakage. #2 Sacrificed for studying leakage.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5-26-05.1</td>
<td>1-10-05.1</td>
<td>0.99 (4K)</td>
<td>3.3</td>
<td>Setback 31.5nm.</td>
<td>#1 AD 4V, seven leak. #2 AD 4.5V, four leak, two nd/funny. #3 Bad leakage.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5-17-05.1</td>
<td>0.99 (4K)</td>
<td>3.3</td>
<td></td>
<td>#4 Bad leakage.</td>
<td>#5 AD 5.5V, two leak, one nd.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5-31-05.1</td>
<td>5-20-05.1</td>
<td>2.6 (4K)</td>
<td>2.7</td>
<td>Same as 5-26-05.1.</td>
<td>Three pieces broken. #1 Bad to severe leakage in all but one gate. #2 Severe leakage. #3 Perhaps still a possibility, not wired up.</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table F.2: Catalogue of cleaved-edge-overgrowth processes and devices
<table>
<thead>
<tr>
<th>Label</th>
<th>Substrate</th>
<th>Mobility ((10^5 \text{cm}^2/\text{Vs}))</th>
<th>Density ((10^{11} \text{cm}^{-2}))</th>
<th>Comments (overgrowth)</th>
<th>Comments (devices)</th>
</tr>
</thead>
<tbody>
<tr>
<td>6-3-05.1</td>
<td>5-20-05.1</td>
<td>2.5 (4K)</td>
<td>2.4</td>
<td>Setback 50nm, Al 50%</td>
<td>33% Al in substrate problematic? #1 - Bad leakage and looks like no wire. #2 Severe leakage. #3-5 Not wired up.</td>
</tr>
<tr>
<td>6-13-05.1</td>
<td>6-2-05.1</td>
<td>1.8 (4K)</td>
<td>2.6</td>
<td>Same as 6-3-05.1</td>
<td>Density for 50% substrates is not stable, goes down with time. #1 No ohmics, temperature too low. #2 AD 5.5V, one gate leaked, five nd. #3 No wire, some leakage. Piece broken? #4 AD 6V, one leaky gate, 2 nd. #5 AD 3.5V, no gate leakage, unstable at low temperature, density decreases a lot.</td>
</tr>
<tr>
<td>7-12-05.1</td>
<td>5-20-05.1</td>
<td></td>
<td>Setback 28.5nm</td>
<td>#1 Very bad leakage. #2 Very bad leakage. #3 Severe leakage. #4 Scrap piece. #5 Severe leakage.</td>
<td></td>
</tr>
<tr>
<td>7-27-05.1</td>
<td>5-20-05.1</td>
<td>2.7 (4K)</td>
<td>2.6</td>
<td>Same as 5-26-05.1</td>
<td>#1 Generally bad leakage, but two possibilities, depleting at 4.2V and 6V.</td>
</tr>
<tr>
<td>6-9-05.1</td>
<td></td>
<td></td>
<td></td>
<td>#2 Severe leakage. #3 Bad leakage. #4 AD 3.9V, nine possible, four leak.</td>
<td></td>
</tr>
</tbody>
</table>

Table F.3: Catalogue of cleaved-edge-overgrowth processes and devices, continued
<table>
<thead>
<tr>
<th>Label</th>
<th>Substrate</th>
<th>Mobility (10^5 cm^2/Vs)</th>
<th>Density (10^{11} cm^-2)</th>
<th>Comments (overgrowth)</th>
<th>Comments (devices)</th>
</tr>
</thead>
<tbody>
<tr>
<td>8-1-05.1</td>
<td>7-13-05.2</td>
<td>0.52 (4K)</td>
<td>3.7</td>
<td>Setback 33nm</td>
<td>#1 AD 3.5V, seven possible, six leak.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Very, very ugly at low temperature.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>#2 AD 4.5V, eight possible, five leak.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Quite nice at low temperature.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>#3 Only two gates leak, but only four</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>possibilities, many other problems.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Piece cracked. And now become unstuck.</td>
</tr>
<tr>
<td>8-1-05.2</td>
<td>7-13-05.2</td>
<td>1.9 (4K)</td>
<td>1.9</td>
<td>Same as 5-26-05.1.</td>
<td>#1 AD 4V, six possibilities, seven leak.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>#2 AuBe contacts, bad leakage.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>#3 AS 3.2V, ten possible, three leak.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>#4 No wire, as expected.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Also no leakage at all.</td>
</tr>
</tbody>
</table>

Table F.4: Catalogue of cleaved-edge-overgrowth processes and devices, continued
Bibliography

the quantum capacitance of interacting electrons in carbon nanotubes. *Nature


alloys of iron in Mo$_{0.8}$Nb$_{0.2}$. *Physical Review*, 137(2A):659–663, 1965.


[8] Y. V. Pershin, J. A. Nesteroff, and V. Privman. Effect of spin-orbit interac-
tion and in-plane magnetic field on the conductance of a quasi-one-dimensional

Systems*, volume 191 of *Springer Tracts in Modern Physics*. Springer, Berlin,
2003.
[10] In the interests of full disclosure, none of the four parts of this figure are from the same chip and only the image in the upper right really contains a peapods. The other images are of nanotube devices. So the figure as a whole is a little misleading, but it works for the purposes of illustration.


[46] We deposited multiple electrodes on each of 20 nanotubes. All but one of the electrically-inactive nanotubes were from a batch with known handling issues. Even at 0.5V bias and ±20V on the gate at room temperature, every adjacent electrode pair on these nanotubes showed no discernible current above a ~ 4pA noise floor, implying a resistance above 100GΩ. In contrast, even the most resistive of the 'conductive' nanotubes passed several nA of current under similar conditions, corresponding to a nanotube resistance of $\lesssim 200M\Omega$.


[67] We define \( T_K \) and \( V_K \) according to \( G(T_K, V = 0) \equiv G(0, 0)/2 \) and \( G(T = 0, V_K) \equiv G(0, 0)/2 \). According to NRG calculations, \( T_K \) and \( V_K \) are related by \( eV_K = 2.10k_B T_K \). (T. A. Costi, personal communication (2006) and [75].) Hence, \( T_K \) can be estimated from the HWHM \( V_K \) of the zero-bias anomaly at low \( T \).
BIBLIOGRAPHY


[70] A series resistance of 650Ω in the leads was taken into account.


[76] Conductance peaks at larger bias, which come from inelastic cotunneling processes involving orbital rather than spin transitions, are observed within the same Coulomb diamond (Figure 2a-b) [133]. In our fits to the data in Figure 2, we assume that these cotunneling peaks do not change with field. Thus the only fitting parameters at each magnetic field are associated with the peaks of interest. Note: the device’s configuration (and hence $T_K$) changed slightly between earlier measurements and this measurement.

[77] Konik et al. build on the Moore-Wen calculation, and agree with its predicted density of states. Logan and Dickens predict high and low-field limits but not a detailed crossover.


[90] We determine $T_K$ here from the peak width in the same way as for spin-1/2 as there is no explicit calculation for spin-1.


