Experimental Quantum Transport in Carbon Nanotubes:
Josephson Quantum Dot Junctions and Double Quantum Dots

Ph.D. Thesis
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Preface

This thesis is submitted to the Faculty of Science at the University of Copenhagen in partial fulfillment of the requirements for the Ph.D. degree in Physics.

The thesis presents experimental work performed from March 2004 to June 2007 at the Niels Bohr Institute, NanoScience Center and at Hitachi Cambridge Laboratory.

The project has largely been involved in a European Union project called "ULTRA-1D" involving six universities from Europe (Jyväskylä, Karlsruhe, Lund, Copenhagen, Grenoble, and Bochum) and one from the US (California). The main aim for the Copenhagen part of this project was to make a periodic potential along a 1D-system, which chapter 5 - 7 is devoted to.

During most of the project I have had a very good and fruitful collaboration with Kasper Grove-Rasmussen. I am very grateful for this collaboration and hope it can continue.

I am also deeply grateful to my supervisor, Poul Erik Lindelof, for his excellent mix of guidance, freedom, and enthusiasm that I received during the three years, and for the trust in sending me, and bringing me along, to many good and important conferences. In the theory group a big thank also goes to Tomáš Novotný, Karsten Flensberg, Jens Paaske, and Brian Møller Andersen for help with interpreting data. For a good atmosphere at our workplace and for inspiring discussions during lunch, Journal Club, etc. I am very grateful to my colleagues Kasper Grove Rasmussen, Jonas Rahlf Hauptmann, Thomas Sand-Jespersen, Jeppe Holm, Martin Aagesen, Magdalena Utko, Ane Jensen, Anders Mathias Lunde, Claus Sørensen, Nader Payami, Søren Erfurt Andresen, Anders Eliasen, Brian Skov Sørensen, Søren Stobbe, Pawel Utko, and Inger Jensen.

Part of this work was carried out in the group of David A. Williams at Hitachi Cambridge Laboratory (chapter 7 and part of chapter 5) during a four month stay. A special thank goes to David A. Williams, Kaiyou Wang and Arthur M. Blackburn, whom I worked with at Hitachi, for four very inspiring and stimulating months.
The thesis is divided into two parts/fields: Carbon nanotube Josephson quantum dot Junctions and Carbon nanotube double quantum dots. Each part starts with an introductory chapter that describes some of the terms that will be used in the following chapters and shows how we fabricated the devices. Then follows three / two chapters, written as articles, on different subjects within the two fields. Chapter 2, 3, and 6 are (except section 3.5 and Fig. 3.5) published in basically identical forms in Refs. [1, 2, 3].

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Part I

Carbon Nanotube
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Chapter 1

Introduction

Many interesting effects can be observed in nano-scale Superconductor–Normal metal–Superconductor (SNS) junctions like Andreev reflections, Subharmonic gap structure and excess current. However, for observing the probably most interesting effect namely the Josephson effect (supercurrent) and thereby phenomena such as supercurrent reversal or its interplay with many-body effects such as the Kondo effect, a carefully designed circuit surrounding the SNS junction is needed. We will in this section show that if the surrounding circuit is not properly designed the supercurrent is suppressed due to uncontrolled phase fluctuations of the superconducting phase across the SNS junction. The maximum measurable supercurrent, called the measurable critical current (or switching current) \( I_m \), will always be lower than the (true) full magnitude of the supercurrent called the critical current \( I_C \). We show in chapter 4 that by carefully designing the surrounding circuit \( I_C \) can be extracted. We will in the following refer to the Josephson junction as the combined SNS-junction and on-chip surrounding circuit.

1.1 Designing a nano-scale Josephson junction

To design our Josephson junctions we use the so-called extended RCSJ-model [4]. The (standard) RCSJ-model is schematically shown in Fig. 1.1(a) and it models the SNS part of the Josephson junction. The extended RCSJ-model (see Fig. 1.1(b)) includes the surrounding circuit, which in our case is composed of a contact pad capacitance \( C_{CP} \) and a resistor \( R \) of the leads connecting the SNS-junction to the contact pads.

1.1.1 RCSJ-model

The basic idea in this section is to use the RCSJ-model to find dynamical properties of the phase difference across the SNS-junction \( \phi \). More specifically we want to find the oscillation frequency of \( \phi \), how strongly this oscillation is
Figure 1.1: (a) A Superconductor-Normal metal-Superconductor (SNS) junction can be modeled by a Resistor \((R_J)\), a Capacitor \((C_J)\) and a Shunt, having a certain current-phase relation \((I_S(\phi))\), by the co-called RCSJ model. (b) Extended RCSJ-model where a nearby resistor \((R)\) and capacitor \((C_{CP})\) are included. Note that \(R_J\) and \(C_J\) are not actual components but a way to model the SNS-junction, whereas \(C_{CP}\) and \(R\) are real components namely the contact pad capacitance and the resistance from the SNS junction to the contact pads.

damped, and most importantly we want to find the quality factor \(Q\), which determines whether the oscillator is over- or under-damped.

The DC and AC Josephson relations are:

\[
I_s(\phi) = I_C \sin(\phi) \\
V_J(\phi) = \frac{\hbar}{2e} \dot{\phi}
\]

(1.1)

The current through \(R_J\) (see Fig. 1.1(a)):

\[
I_{R_J} = \frac{V_J}{R_J}
\]

(1.2)

The current through \(C_J\) (see Fig. 1.1(a)):

\[
I_{C_J} = C_J \dot{V_J}
\]

(1.3)

The total current \(I_J\) is therefore:

\[
I_J = \frac{\hbar C_J}{2e} \dot{\phi} + \frac{\hbar}{2eR_J} \dot{\phi} + I_C \sin(\phi)
\]

(1.4)
1.1 Designing a nano-scale Josephson junction

We will for now assume that the current $I_J$ is a constant current that we can apply to the junction. We do this because it gives some nice simple results that we can use in the next section where we develop the correct extended-RSCJ-model.

To get insight into the dynamics of the Josephson junction Eq. (1.4) is often rewritten as (see [4] page 202 and onwards):

$$C_J \left( \frac{\hbar}{2e} \right)^2 \frac{\dot{\phi}}{\phi} = -\frac{1}{R_J} \left( \frac{\hbar}{2e} \right)^2 \dot{\phi} - \frac{\hbar}{2e} I_C \left( \sin(\phi) - \frac{I_J}{I_C} \right)$$

This equation describes the motion of a fictitious particle with mass $C_J(\hbar/2e)^2$ moving along the $\phi$-direction. The last term describes a $\phi$-dependent force acting on the particle which is analogous to the particle moving in the following potential:

$$U(\phi) = -E_J \cos(\phi) - \frac{\hbar}{2e} I_J \phi$$

Where $E_J = (\hbar/2e)I_C$ is called the Josephson energy. $U(\phi)$ is called the "tilted washboard potential", and it gives a good intuitive description of the dynamical properties of the phase in a Josephson junction. If the fictitious particle becomes trapped in one of the potential minima of the washboard potential and performs small angle oscillations ($\sin(\phi) \sim \phi$) then Eq. (1.4) can be rewritten as:

$$\ddot{\phi} = -\frac{1}{R_J C_J} \dot{\phi} - \frac{2eI_C}{\hbar C_J} \left( \phi - \frac{I_J}{I_C} \right)$$

In this form it is equivalent to a damped harmonic oscillator with oscillation frequency $\omega$, and damping coefficient $\gamma$ given as:

$$\omega = \sqrt{\frac{2eI_C}{\hbar C_J}}, \quad \gamma = \frac{1}{R_J C_J}, \quad Q \equiv \frac{\omega}{\gamma} = \sqrt{\frac{2eI_C}{\hbar C_J}} R_J C_J$$

where $Q$ is the quality factor determining whether the oscillation is over-damped $Q \ll 1$ or under-damped $Q \gg 1$.

1.1.2 Extended RCSJ-model

We are unfortunately not in control of the current $I_J$. The current we control $I_{sd}$ is not applied directly to the SNS-junction but to the contact pads as schematically shown in Fig. 1.1(b). We therefore need to extend the model to include the contact pad capacitance and resistance from the SNS-junction to the contacts pads.

The current we apply can be given as:

$$I_{sd} = I_J + I_{C_{CP}}$$

where $I_{C_{CP}}$ is the current through the contact pad capacitor, given by:

$$I_{C_{CP}} = C_{CP} V_{sd}$$
The source-drain bias $V_{sd}$ is a dynamical parameter when the device is current biased, given by:

$$V_{sd} - V_J = RI_J \quad \Rightarrow \quad \dot{V}_{sd} = RI_J + \dot{V}_J$$  \hspace{1cm} (1.11)

We can now rewrite Eq. (1.9):

$$I_{sd} = I_J + C_{CP}(RI_J + \dot{V}_J)$$  \hspace{1cm} (1.12)

By inserting Eq. (1.4) and (1.1) into this equation we get:

$$I_{sd} = \frac{\hbar C_J C_{CP} R}{2 e} \phi + \frac{h}{2 e} \left( C_J + \frac{C_{CP} R}{R_J} + C_{CP} \right) \dot{\phi} + \left( \frac{h}{2 e R_J} + RC_{CP} I_C \right) \cos(\phi) + I_C \sin(\phi)$$  \hspace{1cm} (1.13)

This equation can have a solution of the form $e^{\omega t}$ where $\omega$ is the oscillation frequency. By inserting values of the components ($R = 1.5 \text{k}\Omega$, $R_J \sim 20 \text{k}\Omega$, $C_{CP} \sim 2 \text{pF}$, and $C_J \sim 1 \text{fF}$, see chapter 4) we find that the third order term (third derivative term) is much smaller than the other terms. We therefore neglect the third order term and rewrite Eq. (1.13) in a form analog to Eq. (1.5):

$$m \ddot{\phi} = -b \dot{\phi} - F(\phi)$$  \hspace{1cm} (1.14)

with

$$m = \left( C_J + C_{CP} \left( \frac{R}{R_J} + 1 \right) \right) \left( \frac{h}{2 e} \right)^2$$

$$b = \frac{h}{2 e} \left( \frac{h}{2 e R_J} + RC_{CP} I_C \right)$$

$$F(\phi) = \frac{h}{2 e} I_C \left( \sin(\phi) - \frac{I_{sd}}{I_C} \right)$$  \hspace{1cm} (1.15)

We see that the tilted washboard model can be used again, where $m$ is the mass of the particle, $b$ is friction, and $F(\phi)$ is the $\phi$-dependent force acting on the particle. The tilted washboard potential ($\int F(\phi) d\phi$) becomes almost identical to what we found in Eq. (1.6):

$$U(\phi) = -E_J \cos(\phi) - \frac{\hbar I_{sd}}{2 e} \phi$$  \hspace{1cm} (1.16)

If the fictitious particle gets trapped in a potential minimum of this potential and makes small angle oscillations we again get the harmonic oscillator equation:

$$\ddot{\phi} \simeq -\gamma \dot{\phi} - \omega^2 \left( \phi - \frac{I_{sd}}{I_C} \right)$$  \hspace{1cm} (1.17)

now with

$$\omega = \sqrt{\frac{2 e I_C}{h (C_J + C_{CP} \left( \frac{R}{R_J} + 1 \right))}}$$

$$\gamma = \frac{h + 2 e I_C RR_J C_{CP}}{h R_J (C_J + C_{CP} \left( \frac{R}{R_J} + 1 \right))}$$  \hspace{1cm} (1.18)

and quality factor given by [5]:

$$Q \equiv \frac{\omega}{\gamma} = \sqrt{\frac{h [C_{CP} (1 + R/R_J) + C_J] / (2 e I_C)}{R C_{CP} + h / (2 e I_C R_J)}}$$  \hspace{1cm} (1.19)

This quality factor is used in chapter 4 and is identical to Eq. (4.1).
1.1 Designing a nano-scale Josephson junction

1.1.3 Quantized supercurrent

To use Eq. (1.19) we also need to determine the critical current. The critical current and Josephson energy in an open QD (i.e. without Coulomb blockade) with $\Gamma = \Gamma_L + \Gamma_R \gg \Delta$ is given by [6, 5]:

$$I_C = \frac{e\Delta}{\hbar} N \left(1 - \sqrt{1 - T_{BW}}\right)$$  \hspace{1cm} (1.20)

$$E_J = \frac{\Delta}{2} N \left(1 - \sqrt{1 - T_{BW}}\right)$$  \hspace{1cm} (1.21)

where $N$ is the number of spin degenerate channels, and $T_{BW}$ is the Breit-Wigner transmission probability given by [7]:

$$T_{BW}(\varepsilon) = \frac{\Gamma_L \Gamma_R}{(\frac{1}{2}(\Gamma_L + \Gamma_R))^2 + \varepsilon^2}$$  \hspace{1cm} (1.22)

where $\Gamma_L / \hbar$ and $\Gamma_R / \hbar$ are the tunnel rates through the left and right barrier.

Note that the Josephson energy for a nanotube device in the Fabry-Perot regime with e.g. $G_N \sim 3e^2/h$ at resonance, as in chapter 2, is $E_J \sim \Delta/2 = 50\mu eV$ corresponding to 580 mK. Therefore, to have any chance of observing the supercurrent in a nanotube device we need to cool it down to very low temperatures.

1.1.4 The illusive resistor “$R$”

In order to observe a supercurrent flowing through a Josephson junction the AC Josephson relation states that the phase has to be maintained at a constant value. To increase the probability for having a constant phase we need to damp oscillations of $\phi$, which is done by decreasing the Q-factor $Q < 1$ (over-damped junction). Eq. (1.19) therefore describes how to increase the measurable critical current ($I_m$) in a nano-scale Josephson junction by decreasing $Q$. There are four parameters entering Eq. (1.19) ($R$, $R_J$, $C_{CP}$, and $C_J$) that we can control when designing the Josephson junction. With parameter values normally obtained when we are making devices ($R \sim 10\Omega$, $R_J \sim 6.5-500\ k\Omega$, $C_{CP} \sim 2\ pF$, and $C_J \sim 1-100\ fF$) we get an under-damped ($Q > 1$) Josephson junction. The most important parameter to change in order to get an over-damped junction ($Q < 1$) is the resistor $R$. By increasing $R$ to the kilo-ohm range an over-damped junction can be obtained, provided that the contact resistance is not too high ($R_J \lesssim \hbar/e^2$).

Numerical solutions to the full equation (1.13) (i.e. the time-dependence of the $\phi$) is shown in Fig. 1.2(a) and (b). The blue curves in (a) and (b) are calculated with $R = 10\ \Omega$, $R_J = 8\ k\Omega$, $C_{CP} = 2\ pF$, $C_J = 1\ fF$, and $I_C = 24\ nA$ (found using Eq. (1.20)) and the red curves are for the exact same parameters except $R = 1.5\ k\Omega$. This gives a quality factor $Q = 0.06$ (over-damped) for the red curves and $Q = 8$ (under-damped) for the blue curves. In (a) the applied current is $I_{sd} = 0.1\ nA$ and in (b) it is $I_{sd} = 5\ nA$. We see that both of the curves in Fig. 1.2(a) has a constant solution after some time, i.e., they can both carry a
Figure 1.2: **Over- and under-damped Josephson junctions.** (a) Numerical solutions to Eq. (1.13). Blue curve is with $R = 10 \Omega$, $R_j = 8 \k \Omega$, $C_{CP} = 2 \pF$, $C_J = 1 \fF$, $I_C = 24 \nA$, and $I_{sd} = 0.1 \nA$, and red curve is with the same parameters except $R = 1.5 \k \Omega$. (b) Same as (a) but with $I_{sd} = 5 \nA$. (c) Experimental curves at 0.3 K of the differential conductance $(dI/dV)$ as function of $V_{sd}$ for two devices fabricated with the same parameters as used in the theoretical calculations in (a) and (b). (d) Same measurement as in (c) but integrated to get the current-voltage curves.

supercurrent of 0.1 nA. The under-damped (blue) curve oscillates a lot before it settles at a constant value, which can be viewed as a weakly damped oscillation of the fictitious particle in a potential minimum of the washboard potential. The over-damped (red) curve is strongly damped and does not oscillate. In (b) a much larger current is applied ($I_{sd} = 5 \nA$), and in the under-damped case the particle is no longer trapped in the washboard potential. For the over-damped case we see the red curve has a few step like patterns before settling at a constant value, i.e., the particle is trapped. The steps indicate that the particle is skipping from one minimum to the next a few times before being
Note also that the steps are successively getting longer indicating that the particle is loosing momentum in each skip. These calculations clearly show that increasing $R$ from the normally obtained value of $\sim 10 \Omega$ to $\sim 1.5 \text{k}\Omega$ increased the measurable critical current ($I_m$) considerably. We compare this to experiments in Fig. 1.2(c) and (d) on two devices where the only difference is the value of $R$. Device 1 (the blue curves) and device 2 (the red curves) has approximately the same parameters as the theoretical blue and red curves in (a) and (b). Both devices are in the Fabry-Perot regime with $G_N \sim \frac{3e^2}{h}$ at resonance and measured in the same cryostat at 0.3 K. In (c) we plot the differential conductance as function of source-drain voltage at resonance for the two devices, and in (d) we plot the same measurement integrated up to get a current-voltage (I-V) plot instead. Good qualitative agreement between theory and experiment is seen. The over-damped device has a much larger measurable critical current that the under-damped device.

### 1.2 Fabrication

We focus on two different types of Josephson quantum dot junctions in this thesis. Under-damped devices are analyzed in chapter 2 and 3 (except in Section 3.5), and over-damped devices are analyzed in chapter 4. The over-damped devices are made the following way (the recipe is given in appendix D). We start by making a set of alignment markers by optical lithography for aligning all of the following sets of lithography accurately on top of each other. Islands of catalyst material are then made at specific positions relative to the alignment marks (see Fig 1.3(c)). Carbon nanotubes are catalytically grown from these islands at high temperatures ($850-950^\circ\text{C}$) in a tube furnace with a controlled flow of Methane (0.5 L/min) Hydrogen (0.1 L/min) and Argon (1.1 L/min). The catalyst is a suspension of Iron nitrate and Molybdenum acetate in methanol with suspended Aluminum-oxide nano particles (see appendix C for details). By inspecting the catalyst islands with scanning electron microscope after growth we typically see that there are many nanotubes close to the island and that they often bundle together into robes within 1-2 $\mu$m from the island. The superconducting source and drain electrodes, made by standard electron beam lithography, are therefore placed a few microns away from the islands in hope of contacting just one single nanotube (see Fig. 1.3(b)). The area of the superconducting electrodes are kept small to reduce the junction capacitance and thereby decrease the quality factor from Eq. (1.19). The superconductor is made of 5 nm Titanium, 40-60 nm of Aluminum, and 5 nm Titanium, which gives a superconducting alloy that makes good contact to the nanotube and has a measured transition temperature $T_C \sim 0.75\text{K}$ and superconducting energy gap $\Delta \sim 100\mu\text{eV}$. The superconducting source and drain electrodes are each contacted by two normal metal leads to facilitate four probe measurements. Since these normal metal leads constitutes the resistance $R$ in the extended RCSJ-model we want their resistance to be around $\sim 1\text{k}\Omega$. They are therefore made very long and thin, as can be seen Fig. 1.3(c). The last step is to connect
Figure 1.3: **Nanotube Josephson quantum dot junction design.** (a) Schematic figure of a Single wall carbon nanotube contacted by superconducting electrodes. (b) Scanning electron microscope image of a real device. (c) Colored optical image of the central parts of the Josephson junction with blue superconducting electrodes, yellow normal metal leads with resistance $R$. (d) Leads made by optical lithography connects the electrodes out to contact pads (outside image).

The leads to contact pads by optical lithography (see Fig. 1.3(d)) so the device can be bonded onto a chip carrier and cooled down to very low temperatures.

The under-damped devices are fabricated very similarly. The only difference is that the normal metal leads seen in Fig. 1.3(c) are not made, and only two-probe measurements can be made. The superconducting electrodes are extended to overlap with the optical lithography as seen in Fig. 1.3(d). The resistance $R$ in these devices is therefore the resistance of the optical lithography leads, which is estimated to $R \sim 10 \Omega$. 
Chapter 2

Electronic transport in single wall carbon nanotube weak links in the Fabry-Perot regime

Abstract:\textsuperscript{1}
We fabricated reproducible high transparency superconducting contacts consisting of superconducting Ti/Al/Ti trilayers to gated single-walled carbon nanotubes (SWCNTs). The reported semiconducting SWCNT have normal state differential conductance up to $3e^2/h$ and exhibit clear Fabry-Perot interference patterns in the bias spectroscopy plot. We observed subharmonic gap structure in the differential conductance and a distinct peak in the conductance at zero bias which is interpreted as a manifestation of the supercurrent. The gate dependence of this supercurrent as well as the excess current are examined and compared to the coherent theory of superconducting point contacts with good agreement.

\textsuperscript{1}This chapter has been published in Physical Review Letters 96, 207003 (2006)
Electron transport through a SWCNT bridging two metal electrodes has been studied intensively over the last years. At low temperatures different transport regimes depending on the transparency of the metal-SWCNT interfaces have been identified. With low transparency contacts a quantum dot (QD) will be defined in the SWCNT [8, 9, 10, 11], and with intermediate transparency contacts Kondo resonances around zero bias are observed [12, 13]. High transparency contacts have in recent years also been reported, where the SWCNT constitutes an electron waveguide with Fabry-Perot (FP) interferences [14, 15].

Changing the metal electrodes to superconductors (SC) dramatically changes the transport characteristics. In such junctions the carbon nanotube (CNT) forms a weak link between the two superconductors and several interesting effects can be observed. At zero bias a supercurrent can flow through the weak link [5, 16, 17] due to the Josephson effect [4], while at low biases current will be carried by Multiple Andreev Reflections (MAR) at the two CNT-S interfaces [18, 19, 20]. For large bias, these effects will give rise to an excess current. In fact, only very recently these effects have been seen in SWCNT devices [5] similar to the one presented here. In this chapter, we present transport measurements on a gated S-SWCNT-S Josephson junction at low temperatures, with high transparency contacts. We focus on the gate dependence of the excess current and zero-bias conductance peak in the FP regime.

2.1 Experimental details

The SWCNTs are grown by chemical vapor deposition (CVD) from catalyst islands made by electron beam lithography and positioned relative to predefined alignment marks. The details of the CVD-growth procedure are described in chapter 1. After growth, source and drain electrodes consisting of superconducting trilayers are positioned next to the catalyst islands to contact the SWCNT. The gap between the source and drain trilayer films is approximately 500 nm. The superconducting trilayers consist of 5 nm titanium to make good contact to the SWCNT, then of 40 nm aluminum to raise the transition temperature, and finally 5 nm titanium to stop oxidation of the aluminum.

Our devices are made on a highly doped silicon wafer with a 0.5 μm thermally oxidized SiO$_2$ layer on top. We use the silicon substrate as a back gate. To be able to measure the transition temperature $T_C$ and the critical field $B_C$ of the trilayer films at low temperatures we furthermore define a four probe device of the superconducting trilayer. For the device in this chapter we find $T_C = 750$ mK, $B_C = 75$ mT and from BCS theory we calculate a superconducting energy gap of $2\Delta \simeq 3.5k_BT_C = 230$ μeV. However, the actual effective value of $\Delta$ for the CNT weak link might differ from this measured value due to interface effects and, indeed, we found from the fit of the excess current measurements (see below) that the effective gap is reduced by about a factor of $\sim 0.7$. All measurements are performed at 300 mK in a sorption pumped $^3$He cryostat (Oxford Instruments Heliox). The measurements are made with standard DAQ cards, lock-in amplifiers (excitation 5 μV), and opto-couplers to reduce noise.
2.2 Opening a closed quantum dot by gate tuning

Fig. 2.1 shows a gatesweep from -10 V to 0 V with $V_{sd} = 1 \text{ mV} (> 2\Delta/e)$. It displays strong gate dependence: High conductance at high negative gate voltages and low conductance at small gate voltages, which indicates that the SWCNT is semiconducting. The SWCNT defines a QD with gate depended Schottky barriers at each interface. The gate thus tunes both the energy levels of the QD and the strength of the Schottky barriers.

From $V_{\text{gate}} \sim -4 \text{ V}$ to $V_{\text{gate}} \sim -2 \text{ V}$ the Schottky barriers are large and the SWCNT constitutes a closed QD, i.e., the charging energy $U_C$ is larger than the broadening of the energy levels $\Gamma$. Transport is here dominated by charging effects and Coulomb blockade peaks are clearly visible. Some of the peaks are spaced into periods of four due the four-fold degeneracy (spin and orbital) of each energy level, also confirmed by bias spectroscopy plots (not shown). Such characteristics is a sign of a high quality SWCNT. As the gate voltage is decreased to more negative values the Schottky barriers are decreased, leading to an increase of $\Gamma$. Below $V_{\text{gate}} \sim -5 \text{ V}$ the dot opens, $\Gamma$ becomes larger than $U_C$, and charging effects of the QD are no longer dominant. Instead, FP interference of the electron waves being reflected at the SWCNT-electrode interfaces dominates transport.

Fig. 2.2(a) shows a bias spectroscopy plot in this gate region with a small magnetic field applied ($B = 100 \text{ mT}$) to suppress the superconducting state of the electrodes. The average differential conductance in this gate region is around $\sim 2.5e^2/h$ with maximums of about $\sim 3e^2/h$, approaching the theoretical limit.
Figure 2.2: (a) Bias spectroscopy plot in the high transparency gate region, with a small magnetic field ($B = 100$ mT) to suppress the superconducting state of the electrodes. (b) Analogous to (a) but without magnetic field, i.e., with superconducting electrodes. (c) Close-up on two of the resonances from which the excess current and supercurrent of Fig. 2.4 are extracted. Arrows are pointing to the gate voltages where the graphs in Figs. 2.3(a-b) are measured.
2.3 Excess current

of $4e^2/h = (6.5 \text{k}\Omega)^{-1}$. The maximum value of $\sim 3e^2/h$ of the conductance implies a rather large asymmetry $\Gamma_L/\Gamma_R = 3$ of the CNT couplings to the contacts (found from a resonant level model representing the system well around a conductance peak), in contrast to fairly symmetric couplings reported elsewhere [19, 5]. As $V_{\text{gate}}$ and $V_{\text{sd}}$ are changed the dips in conductance evolve into straight lines, forming a mesh of crossing dark lines. These pronounced oscillations in differential conductance versus $V_{\text{gate}}$ and $V_{\text{sd}}$ are clear signs of FP interferences [14]. In the FP region we estimate $\Gamma = \Gamma_L + \Gamma_R \sim 2 \text{meV}$ by fitting the resonances in Fig. 2.2(a) with a Lorentzian line shape, and the level spacing $\Delta E \sim 4 - 7 \text{meV}$ as half of the distance in source-drain voltage between two resonances.

2.3 Excess current

As we turn off the magnetic field, i.e., turn on the superconducting state of the electrodes, an overall increase in differential conductance between $V_{\text{sd}} \sim \pm 2\Delta/e$ is observed (Fig. 2.2(b)). A more detailed bias spectroscopy plot of this overall increase through two successive resonances is shown in Fig. 2.2(c). Detailed measurements with lock-in amplifier of differential conductance versus $V_{\text{sd}}$ at gate voltages indicated in Fig. 2.2(c) are shown in Figs. 2.3(a-b), where (a) is a cut through a resonance and (b) is a cut through an antiresonance. A characteristic conductance variation between $V_{\text{sd}} \sim \pm 2\Delta/e$ is seen for all gate voltages. Close to $|V_{\text{sd}}| \sim 2\Delta/e$ the conductance starts to increase while at smaller source drain voltages a dip centered around zero bias also develops. In Fig. 2.3(b) this dip can be seen between $V_{\text{sd}} \sim \pm 80 \mu\text{V}$ and less strongly in Fig. 2.3(a). The change in conductance between $V_{\text{sd}} \sim \pm 2\Delta/e$ to typically higher, but sometimes also lower values, than the normal state differential conductance $G_N$ is because superconductivity induced transport mechanisms occur.

Between $V_{\text{sd}} \sim \pm 2\Delta/e$ transport is governed by Andreev Reflections (ARs) [21] and normal reflections. An electron with energy $|\epsilon| < \Delta$ relative to the Fermi energy in the normal region has (depending on the barrier strength) a probability for being AR on the superconductor as a hole effectively transferring two electrons (one Cooper pair) through the NS interface [22, 23]. For $|\epsilon| > \Delta$ ARs are still possible but fall off rapidly. MAR between the two superconducting leads at finite bias give rise to a sub gap structure (SGS) [24, 25, 26, 27], while at zero bias a dissipationless supercurrent can flow provided that the interfaces are sufficiently transparent. In Fig. 2.3(b) we observe features for $|V_{\text{sd}}| \leq 2\Delta/e$ and a distinct peak around zero bias, which is a general trend in our S-SWCNT-S junctions with high transparency. At $|V_{\text{sd}}| \gg 2\Delta/e$ transport is mostly due to quasiparticle transport and the FP pattern is seen in Fig. 2.2(b). As we approach $|V_{\text{sd}}| \sim 2\Delta/e$ from above the quasiparticle transport is enhanced due to the modified density of states of the superconductors and below this point a subharmonic gap structure (SGS) is expected to appear. We observe a complex pattern in the subgap region for the lower transparency case (Fig. 2.3(b)) while the higher transparency one (Fig. 2.3(a)) doesn’t show much structure.
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Figure 2.3: Differential conductance versus source drain voltage measured with lock-in amplifier (5 µV excitation) at different gate voltages as indicated in Fig. 2.2(c). Upper curve (black) is for the superconducting electrodes and lower curve (red) is with the magnetic field of 100 mT to suppress the superconductivity of the electrodes. (a) is measured at a resonance of the Fabry-Perot pattern while (b) is measured at an antiresonance. Blue arrows point to the supercurrent peak.

in qualitative agreement with theoretical predictions [28, 27]. The structure in Fig. 2.3(b) seems too smeared to allow for quantitative comparison with theory, yet it is an interesting subject for further studies.

Instead of studying in detail the SGS, we focused on its integral effect in the form of the excess current, $I_{\text{exc}}$, which is defined as the difference in current between having the electrodes in the superconducting state and the normal state at $V_{sd} \gg \Delta/e$. It can therefore be found as half of the difference in area between the two set of data points in Figs. 2.3(a-b). In Fig. 2.4(a), we have extracted the excess currents from the bias spectroscopy plot in Fig. 2.2(c) and plotted them as functions of the normal state differential conductance. Since the level width is much larger than the SC gap, $\Gamma_{L,R} \gg \Delta$ (see above), we can use for the interpretation of the results the well-established theory of SC point contacts [29, 28] and fit the excess current with the function

$$I_{\text{exc}}(g) = \frac{e\tilde{\Delta}g^2}{h(4-g)} \left[ 1 - \frac{g^2}{4\sqrt{4-g}(8-g)} \log \frac{2 + \sqrt{4-g}}{2 - \sqrt{4-g}} \right]$$

(2.1)

where $g$ is the conductance measured in units of $e^2/h$ and where $\tilde{\Delta}$ is the gap parameter at the superconductor-SWCNT interface. Allowing for renormalization of $\tilde{\Delta}$ and performing a least-square fit to the measured data, we get $\tilde{\Delta} \sim 0.7\Delta$. Using this value in Eq. (2.1) yields the curve in Fig. 2.4(a) and also the data points in Fig. 2.4(d), showing good agreement between experiments and the theoretically extracted excess current.
2.3 Excess current

![Graphs showing measurements and calculations](image)

Figure 2.4: Comparison between measured and theoretical data. (a) Measured (dots) and fitted (line) excess current as a function of conductance ($G = g e^2/h$). The fitted line is calculated from Eq. (2.1) using $\Delta$ as a fitting parameter. (b) Measured (dots) and fitted (lines) supercurrent. The fits are to $a(I_C)^{3/2}$ (full line) and $aI_C$ (dashed line), using $a$ as a fitting parameter, see text for discussion. (c) Measured conductance in the Fabry-Perot regime as a function of gate voltage. (d) Measured (dots) and calculated (crosses) excess current. (e) The zero bias anomaly area measured (dots) and calculated (crosses). The theoretical points (crosses) in (d) and (e) are calculated using the measured conductance in (c) and Eqs. (2.1) and (2.2), respectively.
2.4 Supercurrent

Next, we discuss the zero bias anomaly that we observe for all gate voltages in the FP-region. The peak has a full width of only $\sim 25 \mu V$ (see Fig. 2.3). Such a peak in conductance at zero bias was observed earlier [19, 20] and was attributed to a dissipative quasiparticle current [30]. In this work, however, we pursue an alternative interpretation viewing the peak as a manifestation of the supercurrent. We interpret half of the area of the zero-bias peak (not the whole area under the peak) as a measure of the supercurrent. At the first glance, this interpretation seems inconsistent since the measured peak area is of the order of 0.2 nA, while the expected magnitude of the supercurrent is on the order of $2e\tilde{\Delta}/\hbar \sim 35$ nA, i.e., more than two orders of magnitude higher. However, a similar discrepancy between the measured and expected values of the supercurrent has been observed previously [5, 31]. In particular, in a very recent study [5] on a similar device to ours where the supercurrent was measured as zero bias current a discrepancy of the order of 15 between the measured and expected value was found. The suppression can be understood in terms of the dynamics of the SC phase of a resistively and capacitively shunted Josephson junction with the environment, resulting in a measurable critical current $I_m$ scaling as $I_m \propto I_C^{3/2}$ with $I_C$ being the bare critical value of the supercurrent $I_C$, see Refs. [5, 31] for details.

Adopting this idea, we perform a fit to the measured zero-bias-peak area as a function of the normal state conductance, Fig. 2.4(b). The supercurrent is determined as [27, 29]

$$I_C(g) = \frac{e\tilde{\Delta} g \sin \varphi_{\text{max}}}{4h \sqrt{1 - \frac{2}{\pi} \sin^2 \left(\frac{\varphi_{\text{max}}}{2}\right)}} \tanh \frac{\tilde{\Delta} \sqrt{1 - \frac{2}{\pi} \sin^2 \left(\frac{\varphi_{\text{max}}}{2}\right)}}{2k_B T} \tag{2.2}$$

where $\varphi_{\text{max}}$ is the phase at which the supercurrent in Eq. 2.2 is maximal. Using the renormalized value of the gap, we have fitted the data to both $aI_C(g)$ and $aI_C^{3/2}(g)$ with $a$ being a fitting parameter. We clearly see that the conductance dependence of $I_C^{3/2}(g)$ fits the measured data very well. On the other hand, the dependence of $I_C(g)$ doesn’t fit the data at all, comparable with analogous results of Ref. [5]. The resulting $V_{\text{gate}}$-dependence of the peak area is plotted in Fig. 2.4(e) using the fitted values of $I_C^{3/2}(g)$ with $\tilde{\Delta}$. Thus, we conclude that the zero-bias-peak results are fully consistent with the theoretical predictions based on the supercurrent interpretation.

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The important parameter for the validity of this description is the quality factor $Q > 1$ which is estimated following Ref. [5] to be larger than 2 in our case, with $C = 4 \mu F$, $C_J = 1 \mu F$, $R = 60 \Omega$, $R_J = 10 \mu \Omega$, and $I_C = 35$ nA.
2.5 Conclusion

In conclusion, we have successfully fabricated gated S-SWCNT-S Josephson junctions with high transparency contacts. In the Fabry-Perot regime of the semiconducting SWCNT reported here we observed quasiparticle tunneling at \(|V_{sd}| = 2\Delta/e\), enhanced current due to MARs for \(|V_{sd}| < 2\Delta/e\), and a conductance peak around zero bias. We interpret the zero bias conduction peak as a not-fully-developed supercurrent. The excess current, which has not been analyzed before for such junctions, fits very well to the theory of coherent SNS junctions.

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Chapter 3

Kondo resonance enhanced supercurrent in single wall carbon nanotube Josephson junctions

Abstract\(^1\).

We have contacted single wall carbon nanotubes grown by chemical vapor deposition to superconducting Ti/Al/Ti electrodes. The device, we here report on is in the Kondo regime exhibiting a four-fold shell structure, where a clear signature of the superconducting electrodes is observed below the critical temperature. Multiple Andreev reflections are revealed by sub-gap structure and a narrow peak in the differential conductance around zero bias is seen depending on the shell filling. We interpret the peak as a proximity induced supercurrent and examine its interplay with Kondo resonances.

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3.1 Introduction

Single wall carbon nanotubes (SWCNTs) have been under intense investigation for more than a decade due to their unique mechanical and electrical properties. They are one-dimensional conductors with two conducting modes and when contacted to electrodes they behave as quantum dots, where phenomena as Fabry-Perot interference [14], Kondo effect [12] and Coulomb blockade [8, 9] have been observed. The possibility of contacting carbon nanotubes to superconducting leads [17, 18, 32, 33] opens up for the interesting study of effects related to superconductivity such as supercurrent and multiple Andreev reflections [19] together with the above mentioned phenomena. In the Fabry-Perot regime recent experiments have confirmed that the supercurrent is modulated by the quantized nature of the energy spectrum of the SWCNT, i.e., a Josephson field effect transistor with only two modes (chapter 2, and [5, 1]). Experimental access to the less transparent regimes [34, 35, 36, 37] is even more interesting due to the possibility of probing the competition between effects related to Coulomb blockade and superconductivity. Coulomb blockade generally suppresses the supercurrent and interesting phenomena such as π-junction behavior has been observed in the closed quantum dot regime in nanowires [36]. For more transparent devices the supercurrent can be enhanced due to Kondo physics [38]. Both supercurrent and Kondo physics are two extensively studied many-body effects in condensed matter physics and SWCNT Josephson junctions thus give a unique possibility to examine their interplay. The supercurrent is predicted to coexist with Kondo resonances provided that the Kondo related energy scale $k_B T_K$ is bigger than the superconducting energy gap $\Delta$ [38, 39, 40]. The importance of the ratio between these parameters has recently been addressed in different measurements [20]. In this chapter we extend this investigation to SWCNTs contacted to superconducting leads in order to experimentally probe the interplay between supercurrent and Kondo physics. We investigate the gate dependence of a narrow zero bias conductance peak interpreted as a proximity induced supercurrent and show that the observation of a supercurrent in Kondo resonances depends on the ratio between the two energy scales ($k_B T_K/\Delta$) with a crossover close to 1 qualitatively consistent with theory.

3.2 Experimental Details

SWCNTs are grown from catalyst islands consisting of Fe-oxide and Mo-oxide supported by aluminum nano-particles [41]. Growth is performed by chemical vapor deposition at 850 °C with a controlled flow of gases Ar: 1 L/min, H$_2$: 0.1 L/min, CH$_4$: 0.5 L/min. During heating the furnace is kept under an Ar and H$_2$ flow, whereas cooling is done in Ar. We reduce cooling time by air-cooling the furnace. The substrate is a doped silicon wafer (used as back gate) with a 500 nm SiO$_2$ layer on top. Pairs of superconducting electrodes of Ti/Al/Ti (5/40/5 nm) are defined directly on top of the SWCNT by electron beam lithography followed by optical lithography to define the Cr/Au contact
Figure 3.1: (a) Bias spectroscopy plot of a SWCNT device at 75 mK showing a four-fold Coulomb blockade shell structure. The numbers indicate the additional hole filling, where big diamonds correspond to filled shells. Four Kondo resonances K1-K4 are identified. A magnetic field of 180 mT is applied to suppress superconductivity in the leads. (b) Bias spectroscopy plot for the same gate range as (a) but with the leads in the superconducting state. A sub-gap structure emerges, most clearly visible at $\pm \Delta/e$ and $\pm 2\Delta/e$ (horizontal green dashed lines). The green dashed rectangles are the gate voltage regions shown in Fig. 3.2(c) and 3.2(d).

pads.

The first titanium layer of the metallic trilayer ensures good contact to the SWCNT, whereas the thicker middle aluminum layer is the actual superconductor in the device. Finally, the top layer of Ti is intended to stop oxidation of the aluminum. The gap between the source and drain electrode is typically around 0.5 $\mu$m. In the same evaporation process a four-probe device is made next to the S-SWCNT-S devices, which is used to measure the transition temperature $T_C = 760$ mK and the critical field around $B_C = 100$ mT of the superconductor. From BCS theory we deduce the superconducting energy gap $\Delta = 1.75 k_B T_C = 115 \mu eV$. The devices are cooled in a $^3$He-$^4$He dilution fridge with a base electron temperature around 75 mK and we use standard lock-in techniques.
3.3 Sub-gap structure and supercurrent

At room temperature the current through the device is gate dependent, which reveals that the SWCNT is semiconducting. Figure 3.1(a) shows a bias spectroscopy plot at 75 mK of the SWCNT device in the Kondo regime. It is measured at negative gate voltages and thus transport takes place through the valence band. The superconductivity in the leads is suppressed by a relative weak magnetic field of 180 mT ($>B_C$). The plot shows Coulomb diamonds, four of which are indicated with white dashed lines. Due to the high transparency contacts significant cotunneling is allowed which tends to smear the features. The Coulomb blockade diamond structure exhibits a four-fold degenerate shell structure due to spin and orbital degrees of freedom, where each shell contains two spin degenerate orbitals $[42]$. The filled shells corresponding to the big Coulomb blockade diamonds are marked in Fig. 3.1(a) by the additional number of holes on the SWCNT quantum dot. In the gate region shown 14 holes are added to the SWCNT quantum dot. An estimate of the level spacing between the shells can be found by the horizontal onset of conductance (white horizontal lines) at finite bias in the big diamonds giving $\Delta E \sim 4$ meV $[43]$. The charging energy is estimated as half the source-drain height of the very faintly visible small diamonds giving $U_C \sim 4$ meV (indicated by white lines for hole filling 13-15). A rough estimate of the line-width broadening ($\Gamma \sim 1$ meV) is extracted from the current plateau through ground state. Conductance ridges at low finite bias for hole filling 13-15 are seen, which are attributed to inelastic cotunneling through the two slightly split orbitals in that shell. We note that these lines are not related to superconductivity. Furthermore, four Kondo resonances K1-K4 are identified (green arrows) and labeled in ascending order based on their Kondo temperatures. The Kondo temperatures are estimated by fitting the normal state conductance versus $V_{sd}$ (solid line in Fig. 3.3a-b) to a Lorentzian line shape. The half width at half maximum of each Lorentzian fit yields the estimated Kondo temperature giving $T_K \sim 2$ K, 4.5 K, 5 K and 6 K for K1-K4.

Figure 3.1(b) shows a bias spectroscopy plot at low bias voltages for the same gate range as Fig. 3.1(a) with the leads in the superconducting state. A sub-gap structure clearly appears. The peaks in differential conductance at $V_{sd} = \pm 2\Delta/e \sim \pm 230 \mu V$ are attributed to the onset of quasi-particle tunneling. At lower bias the transport is governed by Andreev reflections due to the relative high transparency between the SWCNT and the superconducting leads $[21]$. Features at biases $V_n = \pm 2\Delta/(en)$, $n = 2, 3, ...$ are expected due to the opening of higher order multiple Andreev reflection processes as the bias is lowered $[44, 27]$. Peaks at $V_{sd} = \pm \Delta/e \sim \pm 115 \mu V$ are clearly seen consistent with the energy gap found from BCS theory $[19]$. Furthermore, for some ranges in gate voltage a narrow zero bias conductance peak is seen. We note, that this zero bias peak is visible in most of the Kondo resonances and is the subject to analysis below.

These above mentioned effects due to superconductivity are more clearly revealed in high resolution data shown in Fig. 3.2(c) and 3.2(d) corresponding
Figure 3.2: (a-b) Bias spectroscopy plot at 75 mK with the leads in the normal state ($B = 180 \text{ mT}$) for Kondo resonance K4 and K1, respectively. (c-d) High resolution data with the leads superconducting for the same gate and bias voltages as in the normal state above. The corresponding gate regions are shown by the dashed green rectangles in Fig. 3.1(b). A sub-gap structure appears at bias voltage $\pm 2\Delta/e$ and $\pm \Delta/e$ indicated by the green dashed lines. This is particular clear in the Coulomb blockade region (d) and also seen in the green traces, which are $dI/dV$ versus bias curves extracted from the plots. A zero bias conductance peak (red arrow) is also visible throughout Kondo resonance K4 as shown by the red trace in (d) and for some gate voltages in (c).

to the green dashed rectangles in Fig. 3.1(b). Figure 3.2(a) and 3.2(b) show the same gate and bias regions as (c) and (d) with the leads in the normal state for comparison. Clearly the sub-gap structure is due to the proximity of the superconducting leads. The peaks at $\pm 2\Delta/e$ and $\pm \Delta/e$ are most pronounced in the Coulomb blockade region (d) while being more smeared in the high conducting region of K4 (c). Green curves show bias cuts, where higher order multiple Andreev reflections features are faintly visible in (c). The turning of the sub-gap structure to lower bias values close to resonance is also observed on both side of K1 in agreement with previous observations [19]. The zero bias conductance peak indicated by the red arrow is clearly visible in Kondo resonance K4 (red
Figure 3.3: $dI/dV$ versus $V_{sd}$ for Kondo resonances K1-K4 shown in Fig. 3.1(a) illustrating the effect of superconducting leads. (a-b) Solid lines and circles are with the leads in the normal and superconducting state, respectively. (c) The four Kondo resonances K1-K4 for small bias voltages with the leads in the superconducting state. The measurable critical current ($I_m$) given by the zero bias peak area is zero for the lowest value of the ratio $k_B T_K / \Delta$, while it increases as this ratio is increased.

trace in (c)) and also present for some gate voltages in (d). Similar behavior of a zero bias peak in Kondo resonances have been observed in other devices [35]. We interpret the zero bias conductance peak as being due to a proximity induced supercurrent running through the SWCNT. The magnitude of the supercurrent in these measurements, called the measurable critical current $I_m$, is estimated by the area of the peak giving a typical value in the order of $I_m \sim 0.2 \text{nA}$. A similar analysis of a zero bias conductance peak as supercurrent was carried out for an open quantum dot in chapter 2 (Ref. [1]), i.e., without Coulomb blockade effects. We note that an alternative interpretation of the origin of the zero bias peak as quasiparticle current has also been suggested [30], but the supercurrent interpretation will be pursued here. The value of $I_m$ is highly suppressed compared to the full magnitude of the supercurrent, called the critical current $I_C$. For one spin-degenerate level at resonance $I_C$ is theoretically expected to be $I_C = e \Delta / h \sim 28 \text{nA}$ in the so-called short regime $\Gamma > \Delta$ [45]. Despite having two spin-degenerate levels in the SWCNT, only one level is available due to Coulomb blockade. The suppression is partly explained within an extended resistively and capacitively shunted junction model due to interaction of the S-SWCNT-S junction with its electrical environment. The important part of the electrical environment for this device is the low resistance between the superconducting electrodes and the contact pads, which plays a crucial role making the
device an under-damped Josephson junction [46, 5]. However, in contrast to the reported Fabry-Perot regime (chapter 2, [5, 1]) the effect of Coulomb blockade and single hole tunneling also contribute to the suppression of the supercurrent because Cooper pair transport is a two particle tunneling process.

3.4 Kondo vs. Josephson

We now return to the Kondo resonances K1-K4 and analyze the interplay between Kondo physics and supercurrent. The solid curves in Fig. 3.3(a) and 3.3(b) show bias cuts with the leads in the normal state through the center of Kondo resonance K2 and K4, respectively (see Fig. 3.1(a)). The circles correspond to the behavior when the leads are superconducting. In both cases an enhancement of the differential conductance is observed for bias voltages below $\pm 2\Delta/e$ and a zero bias conductance peak is present due to supercurrent. The supercurrent is largest for the Kondo resonance with the highest Kondo temperature (K4), i.e., broadest Kondo resonance. In Fig. 3.3(c) bias sweeps at the center of all four Kondo resonances are shown with the leads in the superconducting state. The Kondo temperature normalized by the superconducting

Figure 3.4: (a) The measurable critical current ($I_m$) is zero for K1 and shown to increase as a function of $k_B T_K / \Delta$ (K1-K4). An exponential fit to the data points is given by the solid curve as guideline, where the Kondo temperature is based on the width of the resonance (black diamonds). The red squares are the Kondo temperature obtained from the temperature dependence (K1, K3 and K4). (b) $I_m$ versus gate voltage in the range with the Kondo resonances K1-K4, where the additional hole number is given for filled shells as in Fig. 3.1(a). A finite supercurrent is seen in the center of the broader Kondo resonance K2, while being zero in the center of narrowest Kondo resonance K1. This behavior does not reflect the normal state conductance $G_N$ shown for the same gate range below in (c).
energy gap is the important parameter and is given for each resonance in the figure. It is seen that the supercurrent vanishes for the lowest ratio (blue circles) while it emerges and increases as the ratio is increased. The crossover is close to $k_B T_K / \Delta \sim 1$.

To illustrate this point more clearly, we plot $I_m$ versus $k_B T_K / \Delta$ in Fig. 3.4(a) for the four Kondo resonances analyzed above (black diamonds). The solid curve shows a guideline to the eye based on an exponential fit to the available data points. The overall trend is qualitative consistent with existing theory, which predicts a suppression of the supercurrent in the so-called weak coupling regime ($k_B T_K \ll \Delta$), while the supercurrent coexists with Kondo resonances in the strong coupling regime ($k_B T_K \gg \Delta$) [40, 39]. The red squares show the measured supercurrent where the Kondo temperature is extracted from the temperature dependence for completeness (only data available for $K_1, K_3$ and $K_4$) [47].

Finally, Fig. 3.4(b) shows $I_m$ as function of gate voltage in the range including the four Kondo resonances. It is strongly gate dependent illustrating effects of the four Kondo resonances. Figure 3.4(c) shows the zero bias conductance $G_N$ with the leads in the normal state for the same gate range. The supercurrent does not reflect the normal state conductance $G_N$ as in the case of an open quantum dot [5, 1]. This is most clearly seen by comparing Kondo resonance $K_1$ and $K_2$. No supercurrent is present in the center of the Kondo resonance $K_1$ despite the high normal state conductance, while a finite supercurrent is present in Kondo resonance $K_2$ with equally high normal state conductance. Similarly, the Kondo resonances $K_3$ and $K_4$ have almost the same normal state conductances, but very different supercurrent. These observations support the above analysis that the interplay between Kondo physics and superconductivity has the fraction $k_B T_K / \Delta$ as the important parameter for the observation of supercurrent in Kondo resonances. We also note that the supercurrent in diamond 4, 8 and 12 is zero due to Coulomb blockade. The supercurrent in the high $T_K$ Kondo resonances can thus be view as being enhanced from the suppressed values by Coulomb blockade due to the formation of Kondo resonances. The gate behavior is quite similar to the observed behavior of Ref. [37], but the authors do not show the magnitude of the supercurrent as a function of Kondo temperature.

### 3.5 Increasing the measurable critical current

We have made a second device where we have designed the circuit to increase $I_m$, as explained in chapter 1. The device is in the same contact regime as the device above showing regular Kondo resonances for every second electron added to the QD, see Fig. 3.5(a). The device exhibits odd-even shell filling pattern compared to the four-fold pattern seen above. The zero-bias conductance peak again emerges, see Fig. 3.5(b). $I_m$ is larger for this device as can be seen in Fig. 3.5(d). The black areas in (d) in every second diamond (indicated with green dashed lines for two diamonds) is a low-resistance state due to the su-
3.6 Conclusion

In conclusion SWCNTs have been contacted to superconducting Ti/Al/Ti leads creating SWCNT Josephson junctions. We observe sub-gap structure due to multiple Andreev reflections and a narrow zero bias conductance peak. The zero bias peak is interpreted as a proximity induced supercurrent with a suppressed magnitude due to the under-damped nature of the Josephson junction and Coulomb blockade. We examine interplay between the supercurrent and the Kondo resonances, and the measurable critical current is shown to coexist with Kondo resonances which have high Kondo temperatures compared to the percurrent flowing through the QD. The edge of the black areas (green dashed lines) therefore plots the magnitude of $I_m$ versus gate-voltage, going from zero in the even numbered diamonds into a peak (perhaps cusp-like peak) in the odd numbered diamonds. This is consistent with the analysis above.

Figure 3.5: Second device with designed circuit to increase the measurable critical current. (a) Differential conductance ($dI/dV$) versus gate voltage ($V_{\text{gate}}$). Clear odd-even behavior with a thick Kondo conductance ridge around zero bias in the odd numbered diamonds. (b) Same as (a) but at low bias voltage where the sub-gap structure and zero bias conductance peak (supercurrent) are seen. (c) Same as (a) but in the normal state (0.9 K). (d) Differential resistance ($dV/dI$) versus $V_{\text{gate}}$. The black areas in the odd numbered diamonds plots out the magnitude of measurable critical current ($I_m$) versus $V_{\text{gate}}$, as indicated with green dashed lines in two diamonds to the left. $I_m$ is seen to peak (perhaps in a cusp) in every odd diamond.
superconducting energy gap, while being suppressed when the Kondo temper-
atures becomes comparable with the superconducting energy gap, qualitative
consistent with existing theory.

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Chapter 4

Critical current 0-\(\pi\) transition in designed Josephson quantum dot junctions

Formation of a quantum dot (QD) between two superconductors, called a Josephson QD junction, enables study of supercurrent through a single discrete energy level of an artificial atom (the QD) occupied by a gate-controllable number of electrons. Supercurrent [17, 48, 49] through QD structures in carbon nanotubes [5, 1, 37, 2] and nanowires [36] has been observed to be suppressed below theoretical predictions. Since supercurrent only flows when a constant phase difference across the QD is maintained, observation of the full magnitude of the supercurrent (critical current) depends crucially on the ability to damp fluctuations in the phase [50, 51]. Here we analyze Josephson QD junctions, created in a single wall carbon nanotube, with a carefully designed on-chip circuit that damps phase fluctuations and thereby allows extraction of the critical current. The analysis reveals reversals of the critical current for every electron added to the QD, due to the so-called 0-\(\pi\) transition [52, 53, 54, 38, 55, 56, 57, 58, 59]. Both the observed magnitude of the critical current as well as the observed shift of the 0-\(\pi\) transition points agree well with a simple model. The realization of a well controlled nano-scale Josephson junction opens new routes toward superconducting quantum bits [60, 61, 62] \(^1\).

\(^1\)This chapter has been accepted for publication in NanoLetters
When a Josephson QD junction is placed in an electromagnetic environment, the phase difference across the QD (\(\phi\)) becomes a dynamical variable, which determines the time-averaged current, \(I\), and voltage drop, \(V_J = \langle (\hbar/2e) \dot{\phi}/dt \rangle\), across it. The \(I-V_J\) characteristics of the junction thus depend in a non-linear fashion on the phase dynamics induced by the electromagnetic environment. Inspired by Refs. [51, 5] we utilize a designed external circuit in order to control the phase fluctuations which enables us to infer the true magnitude of the critical current, \(I_C\), from the measurable critical current/switching current, \(I_m\), by a theoretical fitting procedure. \(I_m\) can significantly differ from \(I_C\) as demonstrated previously for single wall carbon nanotube (SWCNT) based Josephson junctions [5, 1, 2].

### 4.1 Designing a nano-scale Josephson junction

To be able to design the external circuit we model our sample (Fig. 4.1(a) and (b)) by an extended resistive and capacitively shunted junction (RCSJ) model [5], yielding the schematic circuit diagram shown in Fig. 4.1(c). The real Josephson QD junction is represented by an ideal Josephson junction with current-phase relation \(I_J(\phi)\), in parallel with a junction capacitor \(C_J\), and junction resistor \(R_J\) accounting for current carried by multiple Andreev reflections. The resistor \(R_J\) generally depends on \(V_J\) and gate voltage \((V_{\text{gate}})\) but for small enough voltage as used in our analysis \(V_J \ll \Delta/e\), where \(\Delta \sim 0.1\,\text{meV}\) is the superconducting energy gap, we approximate \(R_1\) to depend only on \(V_{\text{gate}}\). Assuming a sinusoidal current-phase relation \((I_J(\phi) = I_C \sin \phi)\) the dynamics of \(\phi\) in the circuit of Fig. 4.1(c) becomes equivalent to the damped motion of a fictitious particle in the so-called tilted washboard potential [4]. The damping of the motion of this particle is characterized by the quality factor [51, 5] (see chapter 1 for details).

\[
Q = \frac{\sqrt{\hbar[C(1 + R/R_J) + C_J]}/(2eI_C)}{RC + \hbar/(2eI_CR_J)} \tag{4.1}
\]

given by the ratio of the local minimum oscillation frequency to the friction coefficient. Low \(Q\) implies higher probability of trapping the particle in a potential minimum resulting in a constant phase difference and, thus, the observation of the supercurrent.

We have used Eq. (4.1) to design overdamped samples shown in Fig. 4.1(b). The superconducting electrodes (blue leads in Fig. 4.1(b)) are made with a small area to reduce the junction capacitance, \(C_J \sim 1\,\text{fF}\). Yellow leads, connecting the superconducting electrodes to contact pads, are long thin normal metal wires designed to have a large resistance, measured to be \(R \sim 1.5\,\text{k}\Omega\). The contact pads are fabricated with a large area to increase their capacitance, \(C \sim 2\,\text{pF}\). By inserting the above mentioned values and \(R_J \geq h/e^2\) (see Fig. 4.3(a)), into Eq. (4.1) we get a strong damping \(Q < 1\) for \(I_C \geq 0.1\,\text{nA}\) which is also the smallest critical current we have been able to measure (see Fig. 4.4(b)).
4.2 The diffusive supercurrent branch

The measurements are performed in a $^3\text{He}-^4\text{He}$ dilution fridge at a base electron temperature of 75 mK. In Fig. 4.1(d) we show a current biased four-probe measurement of an $I$-$V_J$ curve at very low bias voltage $V_{sd} \ll \Delta/e$. In this measurement, $I_m$ is the point where the curve has a large change in slope (as indicated in Fig. 4.1(d)), giving $I_m \sim 0.2\,\text{nA}$. For $|I| < I_m (< I_C)$ the particle in the tilted washboard potential has a high probability, after thermal activation out of one potential minimum, to be subsequently retrapped in the next potential minimum. The motion of the particle is therefore diffusive, leading

![Figure 4.1: Sample design and measurement scheme.](image)

(a) Schematic illustration of the Superconductor(S)-Quantum dot(QD)-Superconductor(S) part of the device, made of a single wall carbon nanotube and a superconducting trilayer of 5 nm Ti, 60 nm Al, and 5 nm Ti, with transition temperature $T_C = 0.75\,\text{K}$ and energy gap $\Delta = 0.1\,\text{meV}$ [1]. (b) Colored optical image of the device. Blue leads are the superconducting electrodes with a spacing of 300 nm, contacting a nanotube (represented by a dashed line). Yellow leads are long thin Cr/Au (15 nm/15 nm) leads connecting the superconducting leads to large-area contact pads, outside the image. (c) Schematic circuit diagram of the device with four-probe current-controlled measurement setup. Blue square: S-QD-S junction as depicted in (a). Yellow region: The resistors $R$ are the yellow leads in (b), and the capacitors $C$ are the large-area contact pads. (d) Current ($I$) versus junction voltage ($V_J$) close to a resonance (see the corresponding arrow in Fig. 4.4(b)). Measurement (diamonds) are fitted with Eq. (4.2) (red line) yielding a critical current $I_C = 1.3\,\text{nA}$ and a junction resistance $R_J = 90\,\text{k}\Omega$. 
Critical current 0-π transition in designed Josephson quantum dot junctions

to a small average phase velocity, i.e., a low but finite junction voltage. The branch in the I-VJ curve at |I| < Im, called the diffusive supercurrent branch, therefore has finite resistance. As the current is ramped further up (I > Im) the friction in the tilted washboard potential is no longer large enough to retrap the particle once it is activated out of a minimum. This leads to a high phase velocity and hence the supercurrent is averaged out.

4.3 Extraction of the full critical current

In order to find Ic, we fit the measured I-VJ curves to an overdamped (Q < 1) extended RCSJ model. For the overdamped Josephson junction we need to consider the classical dynamics only. In Ref. [64] the overdamped RCSJ model (without RJ) was calculated for sinusoidal current-phase relation and was extended to general current-phase relations in Ref. [63]. High-resistance

---

Figure 4.2: Fitting procedure for extracting the critical current. (a) Theoretical current-phase relations calculated using a phenomenological “external exchange” model [57], with Eex ≡ U/2 = 15Δ, Γ = ΓL + ΓR = 11Δ, ΓL = ΓR, T = Δ/10, I0 = eΔ/ℏ ≃ 25 nA. The three curves are calculated very close to resonance at Δε = U/30, −U/60, −U/15 from top to bottom (see arrows in Fig. 4.4(a)), where Δε is the potential on the quantum dot measured from resonance. Even though this model is phenomenological it seems to capture correctly all qualitative features of the 0-π transition known from the numerical renormalization group [40] or quantum Monte Carlo [39] calculations. Diamonds in (b) are I-VJ curves calculated using the current-phase relations from (a) in the full theory [63], valid for a general current-phase relation and in the presence of thermal fluctuations. Eq. (4.2) is fitted to the diamonds (solid lines) yielding very good fits in the whole gate voltage range apart from the closest vicinity of the 0-π transitions, which is below the experimental resolution.
4.3 Extraction of the full critical current

Figure 4.3: **Supercurrent versus odd and even electron occupation.** (a) Surface plot of the differential conductance $(dI/dV_{sd})$ versus bias voltage $(V_{sd})$ and gate voltage $(V_{gate})$. Coulomb blockade diamonds, indicated with black dotted lines, are alternating in size between large and small with a corresponding even and odd number of electrons localized on the QD. (b) Surface plot of differential resistance $(dV_j/dI)$ versus applied current $(I)$ and $V_{gate}$, in the same gate-voltage range as in (a). (c) Three $I$-$V_j$ curves from (b) at indicated positions, the right and left graphs are shifted by 20 µV for clarity. Circles are measured with odd occupation on the dot in the $\pi$ junction regime, squares at the 0-π transition point, and triangles with even occupation on the dot in the 0 junction regime. The solid lines are fits using Eq. (4.2) yielding critical currents of 0.65 nA(0.35 nA) for even(odd) electron occupation, and zero at the transition point.
Critical current \(0-\pi\) transition in designed Josephson quantum dot junctions

tunnel junctions have sinusoidal current-phase relations, but QD junctions with resistances comparable to the resistance quantum \(h/e^2\) may have non-sinusoidal current-phase relation (see, e.g., Eq. 2.2 in chapter 2). Nevertheless, we approximate the current-phase relation by the simple sinusoidal form parameterized by \(I_C\), and we justify this approximation in Fig. 4.2 by comparing with a theoretical calculation where the full non-sinusoidal current-phase relation is included. We generalize the theory of Ref. [64] to include current carried via multiple Andreev reflections by the resistor \(R_J\) which is assumed to be much larger than the lead resistance \(R\). The subsequent fitted values of \(R_J\) are consistent with this assumption, see Fig. 4.4(c). Under these assumptions \((I_J(\phi) = I_C \sin \phi\) and \(R_J \gg R\)) the \(I-V\) curve is given parametrically via \(V_{sd}\) by the relations

\[
I(V_{sd}) = I_C \text{Im} \left[ \frac{I_{1-i\eta(V_{sd})} (I_C h/2 e k_B T)}{I_{-i\eta(V_{sd})} (I_C h/2 e k_B T)} \right] + \frac{V_J(V_{sd})}{R_J} \tag{4.2}
\]

where \(I_{\alpha}(x)\) is the modified Bessel function of the complex order \(\alpha\), and \(\eta(V_{sd}) = h V_{sd} / 2 e R k_B T\). Since all the parameters entering Eq. (4.2) apart from \(I_C\) and \(R_J\) are experimentally known we can use \(I-V_J\) curves to determine both \(I_C\) and \(R_J\). The solid curve in Fig. 4.1(c) is a fit of Eq. (4.2) to the measured data, yielding \(I_C = 1.3\,\text{nA}\) and \(R_J = 90\,\text{k\Omega}\). The theory seems to capture the experimental measurement very well, which shows that the suppression of \(I_C = 1.3\,\text{nA}\) into the diffusive supercurrent branch with \(I_m = 0.2\,\text{nA}\) indeed is caused by thermal fluctuations. A total of five samples were fabricated with different contact resistances. They all showed \(I-V_J\) curves with qualitatively the same behavior as the sample in Fig. 4.1(c) and showed excellent fits to Eq. (4.2) (see Fig. 4.5).

### 4.4 Controlled reversal of the critical current

In Fig. 4.3(a) we show a color-scale plot of the differential conductance \((dI/dV_{sd})\) versus \(V_{sd}\) and \(V_{gate}\). Coulomb blockade diamonds, indicated with black dotted lines, alternate in size between large and small as \(V_{gate}\) is increased. This indicates that the QD has two-fold spin degeneracy of each discrete energy level, with an odd (even) number of electrons on the QD in the small (large) diamond. Coulomb repulsion energy \(U \sim 3\,\text{meV}\) and level spacing \(\Delta E \sim 2.5\,\text{meV}\) are extracted from this plot. The edges of the Coulomb diamonds are somewhat blurred due to the tunnel coupling between QD and leads. The tunnel coupling can vary from device to device and in Fig. 4.5(c), we show similar measurements on another device (fabricated the same way) exhibiting sharer edges of the diamonds due to a slightly lower tunnel coupling. At low bias voltage two parallel conductance ridges (indicated with black arrows) are seen, reflecting the peak in density of states of the superconductors at \(V_{sd} = \pm 2\Delta/e\), yielding \(\Delta \sim 0.1\,\text{meV}\). The energy broadening \(\Gamma = \Gamma_L + \Gamma_R\), where \(\Gamma_L(R)\) is the coupling to the left (right) lead, and the asymmetry \(\Gamma_L/\Gamma_R\) of each Coulomb oscillation
4.4 Controlled reversal of the critical current

Figure 4.4: Critical current $0$-$\pi$ transition compared to theory. (a) Solid line is $|I_C|$ versus potential on the quantum dot obtained as $|I_J(\pi/2)|$ from [57] with the same parameters as in Fig. 4.2. Crosses are the critical currents obtained by fitting Eq. (4.2) to the full theory as described in Fig. 4.2(b). Apart from very close to the transition point, the match between the two approaches is very good. (b) and (c) Experimental critical current $|I_C|$ and junction conductance $1/R_J$ as extracted from fitting measured $I$-$V_J$ curves to Eq. (4.2), versus the gate voltage $V_{gate}$. The error-bars in (b) at the plateau around $V_{gate} = -2.56\, \text{V}$ show the estimated precision of the fit. (d) Normal state differential conductance ($B = 150\, \text{mT}$) at zero bias, in the same gate-voltage range as in (b) and (c). The dotted vertical lines indicate the positions of charge-degeneracy resonances. Note the good correspondence between the phenomenological model in (a) and the measurement in (b), both the observed magnitude of the critical current as well as the shift toward odd occupation of all the $0$-$\pi$ transition points are in agreement with the theory.
are determined in the normal state \((B = 150 \text{ mT})\) by fitting the even valley part of the Coulomb oscillation peaks, Fig. 4(d), to a Lorentzian (see Fig. 4.6). This gives approximately constant values of \(\Gamma \sim 1.1 \text{ meV}\) and asymmetry parameters \(\Gamma_L/\Gamma_R\) in the range 1 to 4. These values are used in the theoretical plots in Fig. 4.2 and Fig. 4.4(a).

In Fig. 4.3(b) we plot the differential resistance \(dV_J/dI\) as a function of \(I\) and \(V_{\text{gate}}\). Three representative \(I-V_J\) curves selected from Fig. 4.3(b) at indicated positions are shown in Fig. 4.3(c). The diffusive supercurrent branch is observed at most gate-voltages also at the resonances, but not in a narrow gate-region close to each resonance (open square in Fig. 4.3(c) and Fig. 4.4(b)). Full gate-voltage dependence of \(I_C\), and \(R_J\) obtained by fitting the \(I-V_J\) curves from Fig. 4.3(b) to Eq. (4.2) is shown in Fig. 4.4(b) and (c), together with the normal-state \((B = 150 \text{ mT})\) zero-bias conductance in Fig. 4.4(d). We observe that \(I_C\) oscillates in accordance with the number of electrons (odd or even) localized on the QD. \(I_C\) furthermore exhibits sharp dips to zero for every electron added to the QD, signifying a reversal of the sign of \(I_C\) due to a 0-\(\pi\) transition of the current-phase relation (see Fig. 4.2). All the transition points are seen to systematically shift toward odd occupation on the QD. In Fig. 4.4(a) we have used the phenomenological model of Ref. [57] with the same parameters as used in Fig. 4.2 to calculate the full gate-dependence of the critical current. Even though the QD in reality has an unpolarized magnetic moment in the odd valleys, the theory, which is based on a polarized magnetization, does capture two important consequences of the magnetic moment: (i) The magnitude of the critical current and thus its strong suppression as compared to a non-interacting resonant level \((I_0 = e\Delta/h \simeq 25 \text{ nA})\), which is a result of the transport of Cooper-pairs through a strongly correlated electronic system. (ii) The slight shift of the transition points toward odd occupation.

In fig. 4.5 we present measurements on another device exhibiting the same 0-\(\pi\) behavior versus gate-controllable odd or even electron occupation, as shown above.

### 4.5 Outlook

We end by noting that Josephson junctions fabricated in Al-AlO\(_x\)-Al are already used in superconducting quantum bits (qubits) [60, 61, 62]. The realization of a well-controlled Josephson QD junction in a SWCNT, could open the route towards new superconducting qubits where better control of the Josephson junctions can be achieved.
Figure 4.5: **Critical current 0-π transition for another device.** (a) Critical current $|I_C|$ extracted by fitting Eq. (4.2) to the measured data in (b). (b) Color scale plot of differential resistance ($dV_J/dI$) versus applied current ($I$) and gate voltage ($V_{gate}$). (c) Color scale plot of differential conductance ($dI/dV_{sd}$) versus bias voltage ($V_{sd}$) and $V_{gate}$ with the electrodes driven normal by a small magnetic field, $B = 150$ mT. Coulomb blockade diamonds, indicated with black dotted lines, are alternating in size between large and small with an even (odd) number of electrons on the QD in the large (small) diamond. The vertical green dotted lines indicate the position of the charge degeneracy resonances.
Critical current 0-π transition in designed Josephson quantum dot junctions

Figure 4.6: Extraction of level broadening and asymmetry. Differential conductance ($dI/dV$) at zero bias versus gate voltage ($V_{\text{gate}}$) or level energy ($\varepsilon$). Measurement (circles) are identical to Fig. 4(d), and solid lines are Lorentzian fits. The $\Gamma$ and $\alpha$ hereby obtained are 1.2 (1), 1.3 (2.5), 1.1 (3.8), 1.0 (4.0), from left to right.

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Part II

Carbon Nanotube Double Quantum Dots
Chapter 5

Introduction

In this chapter we examine electron transport through two QDs in series, called a double quantum dot (DQD). We start with a phenomenological description of the so-called honeycomb diagram, and introduces some general parameters and quantities used in analysing DQDs. We describe how the capacitive coupling parameter ($U_{Cm}$) and the tunnel coupling parameter ($t$) qualitatively are observed in the honeycomb diagram. In the second section we give a theoretical analysis of the honeycomb diagram and model it theoretically. We show that by fitting the theoretical model to measurements a quantitative estimate of $U_{Cm}$ and $t$ can be made. Formulas used in chapter 7 are also derived here.

In the third section three different designs (side-gate, top-gate, and trench-gate) to fabricate DQD devices with carbon nanotube (CNTs) are described. In this project we started out with the side-gate design, but we never managed to make a device that worked as a DQD with that design. Next we tried the top-gate design, which is the design we have had the most success with, both in chapter 6 and 7 the top-gate design is used. The trench-gate design are more recently developed, therefore, we do not have that many measurements on those devices yet. However, measurements so far are promising.

5.1 The honeycomb diagram phenomenologically

When two QDs are connected in series and contacted with metal electrodes electrons can tunnel through at certain electrostatic potentials in each dot named $E_1$ for dot 1 and $E_2$ for dot 2. $E_1$ and $E_2$ are controlled by the voltages applied to gate 1 ($V_{G1}$) and gate 2 ($V_{G2}$). A schematic figure of a DQD is shown in Fig. 5.1(a).

We will now plot the regions in gate space (gate 1 and gate 2 in Fig. 5.1(a)) where transport can occur. Assuming two-fold (spin) degenerate energy levels and that the device is in the Coulomb blockade regime, each dot has a charging energy $U_{C1}$, $U_{C2}$ for filling in an extra electron and a level-spacing between the spin degenerate levels $\Delta E_1$, $\Delta E_2$, as shown in Fig. 5.1(b). For each dot an odd-
Figure 5.1: (a) Schematic representation of a double quantum dot (DQD) connected to metal electrodes with gate-voltages tuning each quantum dot as well as the inter-dot tunnel barrier. (b) Schematic energy diagram of the chemical potentials in each dot, where $\Delta E_1$ and $\Delta E_2$ are level spacings, and $U_{C1}$ and $U_{C2}$ are Coulomb energies. (c) Schematic honeycomb diagram without capacitive and tunnel coupling ($U_{Cm} = 0$, and $t = 0$) as described in the text. $E_1$ and $E_2$ are electrostatic potentials in each dot controlled by gate voltages.
5.1 The honeycomb diagram phenomenologically

Even shell filling pattern will therefore emerge as the corresponding gate-voltage is increased. Fig. 5.1(c) shows this shell filling pattern, where the potential in dot 1 ($E_1$) and dot 2 ($E_2$) is tilted 45° with respect to vertical and horizontal. The lines perpendicular to the $E_1$ ($E_2$) axis corresponds to a chemical potential in QD1 (QD2) being aligned within the bias window.

When capacitive coupling between the QDs is introduced (still neglecting tunnel coupling) a Coulomb repulsion between electrons in the two dots will emerge, which changes the filling pattern just described. By going vertically up through any two intersecting lines (crosses) in Fig. 5.1(c), two electrons are filled into the DQD. Due to coulomb repulsion these two electrons are now added to the DQD at different energies, splitting the intersection points in Fig. 5.1(c) into two new points called triple points. The splitting, called the mutual charging energy ($U_{C_m}$), is the change in energy of QD1 (QD2) when an electron is added to QD2 (QD1). The new filling pattern is schematically shown in Fig. 5.2(a), where the black solid lines (almost) perpendicular to the $E_1$ ($E_2$) axis corresponds to having a chemical potential in QD1 (QD2) within the bias window.

The notation (N,M) will be used for the electron occupation in the two dots (for one shell), where N (M) is the electron occupation in dot 1 (dot 2). Transport takes place at each triple point via three degenerate charge states. Between (1,0) and (0,1) two triple points exist, with transport of electrons at the low-energy triple point: (0,0) ⇒ (1,0) ⇒ (0,1) ⇒ (0,0) and transport of holes at the high-energy triple point: (1,1) ⇒ (1,0) ⇒ (0,1) ⇒ (1,1). The pattern emerging in this plot is hexagons of different sizes, and it is therefore called a honeycomb diagram.

When a finite tunnel coupling between the two dots is included the wave-function in each dot starts to overlap and new hybridized (molecular) states are formed, changing the way transport takes place. There are no longer localized states in each dot for the electrons to tunnel through, instead there are one hybridized DQD-state, and transport is governed by sequential tunneling from source to the hybridized state, and then to drain. The state in dot 1 ($|\phi_1\rangle$) mixes with the state in dot 2 ($|\phi_2\rangle$) to form two new states called bonding ($|\phi_B\rangle = \alpha|\phi_1\rangle + \beta|\phi_2\rangle$), and antibonding ($|\phi_A\rangle = \alpha'|\phi_1\rangle - \beta'|\phi_2\rangle$), where the bonding state is the ground state. The energy-separation between $|\phi_B\rangle$ and $|\phi_A\rangle$ depends on the so-called detuning parameter ($\varepsilon$) with a minimum of $2t$, where $t$ is the tunnel coupling parameter. Detuning is the difference in electrostatic potential in the two dots defined as $\varepsilon = E_2 - E_1$, with zero-point where the separation between $|\phi_B\rangle$ and $|\phi_A\rangle$ has its minimum, i.e., the separation between $|\phi_B\rangle$ and $|\phi_A\rangle$ is $2t$ at $\varepsilon = 0$. The tunnel coupling effectively rounds the corners in each hexagon as schematically illustrated in Fig. 5.2(b). Therefore, where we before (without tunnel coupling, $t = 0$) had triple points in the honeycomb diagram, an anti-crossing is now seen with a lower and upper wing (schematically shown in Fig. 5.2(b)).
Figure 5.2: This figure extend Fig. 5.1(c) to the case of capacitive coupling ($U_{Cm}$) and tunnel coupling ($t$). (a) Schematic honeycomb diagram with finite capacitive coupling and no tunnel coupling. Note that the coordinate system is changed compared with Fig. 5.1(c), we now use total energy ($E = E_2 + E_1$), and detuning ($\varepsilon = E_2 - E_1$). (b) Both capacitive and tunnel coupling are included. Light gray lines are identical to the black lines in (a). Separation between upper and lower wing at each anti-crossing is $2(2t + U_{Cm})$, where the factor 2 stems from the coordinate system.
5.2 The honeycomb diagram theoretically

After this phenomenological description of the honeycomb diagram we will in this section give a theoretical analysis. We find analytical expressions for the shape of the two wings at each anti-crossing point in gate-voltage space, i.e., in \( V_{G1}, V_{G2} \) space. We then use the expression to extract the two coupling parameters \( U_{Cm} \) and \( t \) by fitting it to measured data.

5.2.1 Region between (1,0) and (0,1)

We start with the two wings in the region around charge state (0,0), (1,0), (0,1), and (1,1). In this region we have a 0-particle state in the (0,0)-region, a 1-particle state in the (1,0) and (0,1)-region, and a 2-particle state in the (1,1)-region. We find the eigenenergy of the ground state in each of these three regions. The condition for transport through the lower wing is that the chemical potential for adding the first electron (eigenenergy for the 1-particle state minus the eigenenergy of the 0-particle state) is equal to the chemical potential of the source and drain. While the condition for transport through the upper wing is that the chemical potential for adding the second electron (eigenenergy for the 2-particle state minus the eigenenergy of the 1-particle state) is equal to the chemical potential of the source and drain.

The Hamiltonian for the DQD system is given by an electrostatic part \( H_{el} \), and a tunnel coupling part \( H_t \):

\[
H_{el} = n_1 n_2 U_{Cm} + (n_{1\uparrow} n_{1\downarrow} + n_{2\uparrow} n_{2\downarrow}) U_C - n_1 E_1 - n_2 E_2
\]

where \( E_1 \) and \( E_2 \) are the electrostatic potential in dot 1 and dot 2 imposed by the gates. We have for simplicity assumed identical dots i.e. \( U_{C1} = U_{C2} \equiv U_C \).

\[
H_t = -t \sum_{\sigma} (c_{1\sigma}^\dagger c_{2\sigma} + c_{2\sigma}^\dagger c_{1\sigma})
\]

where \( c^\dagger \) and \( c \) are creation and annihilation operators, and \( \sigma = \uparrow, \downarrow \) is the electron spin. The Hamiltonian \( H = H_{el} + H_t \) is only designed to handle one shell i.e. one spin degenerate level in each dot. The eigenenergy of the 0-particle state \( E_{00} \) is not so hard to find:

\[
E_{00} = 0
\]

The eigenenergy of the 1-particle state is more difficult! The first electron added to the shell is either in dot 1, dot 2, or in a super position of the two dots. In general the state can be written as a Bonding state or as an Antibonding state:

\[
|B_{\sigma}\rangle = \sin(\theta/2)|\sigma_1\rangle + \cos(\theta/2)|\sigma_2\rangle
\]
\[
|A_{\sigma}\rangle = \sin(\theta/2)|\sigma_1\rangle - \cos(\theta/2)|\sigma_2\rangle, \quad \sigma = \uparrow \text{ or } \downarrow
\]

where \( \theta \in [0, \pi] \) is a parameter determining the spatial weight of the wave function, strongly dependent on detuning. It is found that the bonding state
always has a lower energy than the antibonding state, and we therefore neglect
the antibonding state (excited state) and concentrate about the bonding state
(ground state). Since we do not include the magnetic field dependence, the spin
up and spin down states are degenerate, and we find the eigenenergy to be given
by:

\[ E_B(E, \varepsilon) = -t \sin(\theta) - \frac{1}{2} E - \frac{1}{2} \varepsilon \cos(\theta) \]  

(5.5)

where we have defined the total energy \( E = E_2 + E_1 \) and detuning \( \varepsilon = E_2 - E_1 \).
Minimizing this energy with respect to \( \theta \) \((dE_B/d\theta = 0)\) yields:

\[ \tan(\theta) = \frac{2t}{\varepsilon} \]  

(5.6)

By inserting this back into the above equation we get:

\[ E_B(E, \varepsilon) = -\frac{1}{2} (\sqrt{(2t)^2 + \varepsilon^2} + E) \]  

(5.7)

We now have the eigenenergy of the 0-particle state and the 1-particle ground
state which is enough for finding the shape of the lower wing. But for finding
the shape of the upper wing we also need the eigenenergy of the 2-particle ground
state in the (11) region. The available states in region (11) are:

\[
\begin{align*}
|S(11)\rangle &= \frac{1}{\sqrt{2}} (|\uparrow_1 \downarrow_2\rangle - |\downarrow_1 \uparrow_2\rangle) \\
|T_+(11)\rangle &= |\uparrow_1 \uparrow_2\rangle \\
|T_0(11)\rangle &= \frac{1}{\sqrt{2}} (|\uparrow_1 \downarrow_2\rangle + |\downarrow_1 \uparrow_2\rangle) \\
|T_-(11)\rangle &= |\downarrow_1 \downarrow_2\rangle
\end{align*}
\]

(5.8)

a singlet and three triplets. We find that the states are degenerate with eigen-
energy:

\[ E_{S(11)} = U_{CM} - E \]  

(5.9)

The S(11) state has in fact a lower energy than the three T(11) states (ap-
proximately \((2t)^2/U_C\) called the exchange energy) due to the tunnel coupling.
Since we here approximate the state in region (11) to be one of the pure (un-
hybridized) states in Eq. (5.8) we do not see this small contribution.
The condition for transport through the lower wing is that the chemical po-
tential for adding the first electron to the bonding state \((\mu_{00-B} = E_B - E_{00})\)
has to be between the chemical potential of source \((\mu_S)\) and drain \((\mu_D)\) (the
bias window) which we approximate to be zero \((\mu \equiv \mu_S = \mu_D = 0)\), i.e., zero
bias voltage. The condition for transport through the lower wing is therefore
\( E_B = E_{00} \), yielding (total energy \( E \) versus detuning \( \varepsilon \)):

\[ E_{LW1001}(\varepsilon) = -\sqrt{(2t)^2 + \varepsilon^2} \]  

(5.10)

where the subscript "LW1001" means the Lower Wing in the region between
(10) and (01). Transport through the upper wing occurs when the chemical
potential for adding the second electron into state \(|S(11)\rangle\), given that the first
5.2 The honeycomb diagram theoretically

electron is in state |B⟩, \( \mu_{B \rightarrow S(11)} = E_{S(11)} - E_B \) is in the bias window. We therefore have that \( E_{S,11} = E_B \), yielding:

\[
E_{UW1001}(\epsilon) = \sqrt{(2t)^2 + \epsilon^2} + 2U_{Cm} \quad (5.11)
\]

where the subscript "UW1001" means the Upper Wing in the region between (10) and (01). The separation between the upper and lower wing is:

\[
E_{\Delta 1001}(\epsilon) = 2\sqrt{(2t)^2 + \epsilon^2} + 2U_{Cm} \quad (5.12)
\]

These three equations can be transformed to be given in terms of the gate voltages, \( V_{G1} \) and \( V_{G2} \), by using the following relations:

\[
E_1 = \alpha_{11}V_{G1} + \alpha_{21}V_{G2} \\
E_2 = \alpha_{12}V_{G1} + \alpha_{22}V_{G2} \quad (5.13)
\]

where the four \( \alpha \) factors are gate couplings. For identical dots we have \( \alpha_{11} = \alpha_{22} \) and \( \alpha_{12} = \alpha_{21} \). We also define a new coordinate system (45° rotated), \( V = (V_{G2} + V_{G1})/\sqrt{2} \), and \( \Delta V = (V_{G2} - V_{G1})/\sqrt{2} \). Eq. (5.10) can now be written as:

\[
V_{LW1001}(\Delta V) = -\sqrt{\left(\frac{2t}{\sqrt{2}(\alpha_{11} + \alpha_{12})}\right)^2 + \left(\frac{\alpha_{11} - \alpha_{12}}{\alpha_{11} + \alpha_{12}}\Delta V\right)^2} \quad (5.14)
\]

The gate couplings are given by:

\[
\alpha_{11} = \frac{U_C}{\Delta V_G}, \quad \alpha_{12} = \frac{U_C\Delta V_G^m}{\Delta V_G} \quad (5.15)
\]

where \( \Delta V_G \) is the width of hexagon (11) (since we assume identical dots the height and width are the same i.e. \( \Delta V_{G1} = \Delta V_{G2} \equiv \Delta V_G \)). By using \( U_{Cm}/U_C = \Delta V_G^m/\Delta V_G \) (Eq. (6.1) without cross capacitance term) it can be shown that \( (\alpha_{11} - \alpha_{12})/(\alpha_{11} + \alpha_{12}) = (U_C - U_{Cm})/(U_C + U_{Cm}), \) and \( \sqrt{2}(\alpha_{11} + \alpha_{12}) = \sqrt{2}(U_C + U_{Cm})/\Delta V_G \equiv \alpha \). The parameter \( \alpha \) can be interpreted as a gate coupling factor. We can now rewrite Eq. (5.10) as:

\[
V_{LW1001}(\Delta V) = -\sqrt{\left(\frac{2t}{\alpha}\right)^2 + \left(\frac{U_C - U_{Cm}}{U_C + U_{Cm}}\Delta V\right)^2} \quad (5.16)
\]

By similar method we can rewrite Eq. (5.11) and (5.12):

\[
V_{UW1001}(\Delta V) = \sqrt{\left(\frac{2t}{\alpha}\right)^2 + \left(\frac{U_C - U_{Cm}}{U_C + U_{Cm}}\Delta V\right)^2} + \frac{2U_{Cm}}{\alpha} \quad (5.17)
\]

\[
V_{\Delta 1001}(\Delta V) = 2\sqrt{\left(\frac{2t}{\alpha}\right)^2 + \left(\frac{U_C - U_{Cm}}{U_C + U_{Cm}}\Delta V\right)^2} + \frac{2U_{Cm}}{\alpha} \quad (5.18)
\]

Eq. (5.16) and Eq. (5.17) are plotted in Fig. 5.3 as the two red wings between (0,1) and (1,0).
5.2.2 Region between (2,0), (1,1), and (0,2)

We now want to analyze the region around charge state (01), (11), (02), and (12), and find the shape of the wings in that region. We have in this region a 1-particle state in the (01)-region, a 2-particle state in the (11)- and (02)-region, and a three particle state in the (12)-region. To find the shape of the two wings we again need to find the eigenenergies of the ground states in each of these three regions.

The 1-particle states in the (01)-region are:

\[ | \uparrow_2 \rangle, \quad | \downarrow_2 \rangle \]

The eigenenergy with the same Hamiltonian as above (Eq. (5.1), and (5.2)) is:

\[ E_{01}(\varepsilon) = -E_2 = -\frac{1}{2}(E + \varepsilon) \quad (5.19) \]

We could also use the bonding state from above Eq. (5.7) at large positive detuning, which gives the same result.

The available 2-particle states in the region (11) are given in Eq. (5.8) and the available states in region (02) are:

\begin{align*}
| S(02) \rangle &= \frac{1}{\sqrt{2}} (| \uparrow_{1} \downarrow_{2} \rangle - | \downarrow_{1} \uparrow_{2} \rangle) \\
| T_{+}(02) \rangle &= | \uparrow_{2} \downarrow_{2} \rangle \\
| T_{0}(02) \rangle &= \frac{1}{\sqrt{2}} (| \uparrow_{2} \downarrow_{2} \rangle + | \downarrow_{2} \uparrow_{2} \rangle) \\
| T_{-}(02) \rangle &= | \downarrow_{2} \downarrow_{2} \rangle \\
\end{align*}

(5.20)

As we detune between the (11) and (02) region, states with the same total spin and same spin in the z-direction \( S_z \) couple together into bonding and antibonding states:

\begin{align*}
| S_B \rangle &= \frac{1}{\sqrt{2}} (| S(11) \rangle + | S(02) \rangle) \\
| S_A \rangle &= \frac{1}{\sqrt{2}} (| S(11) \rangle - | S(02) \rangle) \\
\end{align*}

(5.21)

The \( T_{S_z}(11) \) and \( T_{S_z}(02) \) states with the same spin in z-direction also hybridize. But, since the three \( T_{S_z}(02) \) states need two energy levels to exist and therefore has a higher energy than the \( S(02) \) (approximately one level spacing), we will exclude those states for now. At elevated magnetic fields the hybridized triplet with \( S_z = -1 \) can become ground state and we therefore return to the triplet states in section 5.2.4. The magnetic field dependence of the honeycomb diagram will be analyzed in chapter 7.

We find that the 2-particle ground state in the (11) to (02) region (at low magnetic fields) is the molecular singlet bonding, which in general form can be written:

\[ | S_B \rangle = \sin(\theta/2)| S(11) \rangle + \cos(\theta/2)| S(02) \rangle \]

\[ = \frac{1}{\sqrt{2}} \left[ \sin(\theta/2)(| \uparrow_{1} \downarrow_{2} \rangle - | \downarrow_{1} \uparrow_{2} \rangle) + 2 \cos(\theta/2)| \uparrow_{2} \downarrow_{2} \rangle \right] \quad (5.22) \]
5.2 The honeycomb diagram theoretically

We find that its eigenenergy can be written as:

\[ E_{S_n} = U_{Cm} - E - \cos^2(\theta/2)(\varepsilon + U_{Cm} - U_C) - t \sin(\theta/2) \cos(\theta/2) \]
\[ = U_{Cm} - E - \frac{1}{2} \varepsilon^*(\cos(\theta) + 1) - t \sin(\theta) \]  
(5.23)

where we define a new (shifted) detuning parameter, \( \varepsilon^* = \varepsilon + U_{Cm} - U_C \).

Minimizing its energy with respect to \( \theta \) yields:

\[ \tan(\theta) = \frac{2t}{\varepsilon^*}, \quad \text{where} \quad \varepsilon^* = \varepsilon + U_{Cm} - U_C \]  
(5.24)

Inserting this back into the equation above yields:

\[ E_{S_n}(E, \varepsilon) = U_{Cm} - E - \frac{1}{2} (\sqrt{t^2 + \varepsilon^*2} + \varepsilon^*) \]  
(5.25)

So, this is the \( E \) and \( \varepsilon \) dependent eigenenergy of the ground state in the (11)- and (02)-region.

The 3-particle states in the (12)-region are:

\[ |\uparrow\downarrow\downarrow\rangle, \quad |\downarrow\uparrow\downarrow\rangle, \quad |\uparrow\downarrow\uparrow\rangle, \quad |\downarrow\downarrow\uparrow\rangle \]

They all have the same eigenenergy:

\[ E_{12}(E, \varepsilon) = 2U_{Cm} + U_C - \frac{3}{2} E - \frac{1}{2} \varepsilon \]  
(5.26)

To have transport through the lower wing in the (11)- and (02)-region the chemical potential for adding an electron into the \( |S_B\rangle \) state has to be at the chemical potential of source and drain, which we defined to be at zero, i.e., \( \mu_{S_n 
leftrightarrow 01} = E_{S_n} - E_{01} = 0 \). This gives:

\[ E_{LW1102}(\varepsilon^*) = -\sqrt{(2t)^2 + \varepsilon^*2} + U_C + U_{Cm} \]  
(5.27)

Similarly we get for the upper wing (\( \mu_{12 
leftrightarrow S_n} = E_{12} - E_{S_n} = 0 \)):

\[ E_{UW1102}(\varepsilon^*) = \sqrt{(2t)^2 + \varepsilon^*2} + U_C + 3U_{Cm} \]  
(5.28)

And the separation:

\[ E_{\Delta1102}(\varepsilon^*) = 2\sqrt{(2t)^2 + \varepsilon^*2} + 2U_{Cm} \]  
(5.29)

These three equations can, analogously to above, be transformed to be given in terms of \( V \) and \( \Delta V \):

\[ V_{LW1102}(\Delta V) = -\sqrt{\left(\frac{2t}{\alpha}\right)^2 + \left(\frac{U_C + U_{Cm}}{U_C - U_{Cm}}\left(\Delta V - \frac{\Delta V_G}{\sqrt{2}}\right)\right)^2} + \frac{U_C + U_{Cm}}{\alpha} \]  
(5.30)
Figure 5.3: Theoretical honeycomb stability diagram with $t = 0.2\,\text{meV}$, $U_{\text{Cm}} = 0.2\,\text{meV}$, $U_C = 3\,\text{meV}$, $\Delta V_G = 1\,\text{V}$, and $B=0\,\text{T}$, using Eq. (5.16) and (5.17) between (10) and (01), and Eq. (5.30) and (5.31) between (20) and (11) and (02), and Eq. (5.33) and (5.34) between (21) and (12).

These three equations can also be used in the region between (20) and (11) just by replacing $\Delta V$ with $-\Delta V$. Eq. (5.30) and Eq. (5.31) and their counterparts with $\Delta V$ replaced with $-\Delta V$ are plotted in Fig. 5.3 as green lines.

5.2.3 Region between (2,1) and (1,2)

Since the derivation for shape of the wings in the region between (21) and (12) are very similar to the derivation in the region between (10) and (01), we
5.2 The honeycomb diagram theoretically

Figure 5.4: The black circles are the position of the wings at anti-crossing points from Fig. 7.1(b). Charge states are written between the graphs indicating where the data points are extracted. The separation between the red lines, and the green lines are fitted with Eq. (5.18). Least square fitting to the measurement yields the mutual charging energy $U_{Cn}$ and tunnel coupling $t$ in units of meV as written on the graphs.

only state the results:

$$V_{\Delta_{2112}}(\Delta V) = -\sqrt{\left(\frac{2t}{\alpha}\right)^2 + \left(\frac{U_C - U_{Cn}}{U_C + U_{Cn}}\Delta V\right)^2} + \frac{2U_C + 2U_{Cn}}{\alpha}$$ (5.33)

$$V_{\Delta_{1101}}(\Delta V) = \sqrt{\left(\frac{2t}{\alpha}\right)^2 + \left(\frac{U_C - U_{Cn}}{U_C + U_{Cn}}\Delta V\right)^2} + \frac{2U_C + 4U_{Cn}}{\alpha}$$ (5.34)

And the separation is identical to Eq. (5.18): $V_{\Delta_{2112}}(\Delta V) = V_{\Delta_{1101}}(\Delta V)$. Eq. (5.33) and Eq. (5.34) are plotted in Fig. 5.3. The theoretical honeycomb diagram for one shell (Fig. 5.3) is now finished and seems to correspond well with measurements, see Fig. 7.1. To extract $U_{Cn}$ and $t$ we have fitted Eq. (5.18) to the measured anti-crossings from Fig. 7.1 in Fig. 5.4.
5.2.4 Return of the Triplet

We now analyze the region between (11) and (02) with one energy level in dot 1, and two energy levels in dot 2 separated by a level spacing $\Delta E$. We do this to be able to include the triplets states from Eq. (5.20) (which we excluded in the analysis above). The Hamiltonian therefore has to be changed. The electrostatic part adds the level spacing if two of the same spin is placed in dot 2.

$$H_{el} = U_{Cm} n_1 n_2 + \frac{1}{2} U_C \left( n_1 (n_1 - 1) + n_2 (n_2 - 1) \right) + \frac{1}{2} \Delta E \sum_\sigma n_{2\sigma} (n_{2\sigma} - 1) - n_1 E_1 - n_2 E_2$$  \hfill (5.35)

The tunnel part has to include two levels in dot 2:

$$H_t = -t \sum_{i,\sigma} (c_{1\sigma}^{\dagger} c_{2i\sigma} + c_{2i\sigma}^{\dagger} c_{1\sigma})$$  \hfill (5.36)

where $i = 1, 2$ is the level number in dot 2 (we still only consider one level in dot 1). The magnetic field contribution to the Hamiltonian is:

$$H_B = b (n_{1\uparrow} + n_{2\uparrow} - n_{1\downarrow} - n_{2\downarrow})$$  \hfill (5.37)

The available triplet states in the region between (11) and (02) are:

$$|T_{B S_z}\rangle = \frac{1}{\sqrt{2}} \left( |T_{S_z}(11)\rangle + |T_{S_z}(02)\rangle \right)$$

$$|T_{A S_z}\rangle = \frac{1}{\sqrt{2}} \left( |T_{S_z}(11)\rangle - |T_{S_z}(02)\rangle \right), \quad S_z = -1, 0, +$$  \hfill (5.38)

The ground state at finite magnetic field is $|T_{B-}\rangle$, which in general form is written:

$$|T_{B-}\rangle = \sin(\theta/2) |T_{-}(11)\rangle + \cos(\theta/2) |T_{-}(02)\rangle$$

$$= \sin(\theta/2) |_{1\downarrow_{1\downarrow}} + \cos(\theta/2) |_{1\downarrow_{2\downarrow}}$$  \hfill (5.39)

It has eigenenergy:

$$E_{T_{B-}} = U_{Cm} - E - \frac{1}{2} \epsilon^* - \Delta E \left( \cos(\theta) + 1 \right) - t \sin(\theta)$$  \hfill (5.40)

Minimizing with respect to $\theta$ gives:

$$\tan(\theta) = \frac{2t}{\epsilon^* - \Delta E}$$  \hfill (5.41)

By inserting this back into the equation above we get:

$$E_{T_{B-}} = U_{Cm} - E - \frac{1}{2} \sqrt{(2t)^2 + (\epsilon^* - \Delta E)^2 + \epsilon^* - \Delta E} - 2b$$  \hfill (5.42)

In chapter 7 we will need the chemical potential for adding an electron into the singlet bonding state ($\mu_{S_{B\rightarrow 01}}$) and triplet bonding state with $S_z = -1$ ($\mu_{T_{B-\rightarrow 01}}$). They are given by:

$$\mu_{S_{B\rightarrow 01}}(E, \epsilon) = E_0 - E_{S_{B\rightarrow 01}} = \frac{1}{2} U_C + \frac{1}{2} U_{Cm} - \frac{1}{2} E - \frac{1}{2} \sqrt{(2t)^2 + \epsilon^*} + b$$  \hfill (5.43)
5.3 The hitchhiker’s guide to the nanotube double dot

where we have included the magnetic field dependence by using the Hamiltonian in Eq. (5.37).

\[
\mu_{T_{B_{-01}}}(E, \varepsilon) = E_{T_{B_{-01}}} - E_{01} = \frac{1}{2} U_C + \frac{1}{2} \Delta E + \frac{1}{2} U_{Cm} - \frac{1}{2} E - \frac{1}{2} \sqrt{(2\varepsilon)^2 + (\varepsilon^* - \Delta E)^2} - b
\]

In chapter 7 we subtract the term \(\frac{1}{2} U_C + \frac{1}{2} U_{Cm}\) (i.e. we change the energy zero-point) for simplicity, and we only analyze Eq. (5.43) and (5.43) along the black dashed line in Fig. 7.4(a), i.e., for constant total energy \(E = 0\).

5.3 The hitchhiker’s guide to the nanotube double dot

The DQDs studied in this project are made in single nanotube molecules. The nanotubes are grown by chemical vapor deposition as explained in section 1.2. The two QDs are defined and independently tuned by local gates. Several different methods to make the local gates have been pursued during the project. We will in this section, in chronological order, explain some of the many attempts toward making a DQD in a SWCNT. As you will see there were also many failed attempts!

5.3.1 Side-gate and AFM manipulation

A device with the side-gate design for making a DQD in a nanotube is shown in Figs. 5.5 and 5.6. The structure is made the following way: First carbon nanotubes are grown in an in-house chemical vapor deposition system (see section 1.2), followed by electron beam lithography (EBL) of alignment marks (two squares with the corners facing, see Fig. 5.5(c)). Then an atomic force microscope (AFM) is used to image the nanotubes and alignment marks. One specific nanotube is chosen from this image based on its height profile i.e diameter, preferably with a diameter \(\leq 1\) nm since that would indicate that it is a SWCNT. The position of the chosen SWCNT is then measured in a coordinate system made from the alignment marks. With the position of the SWCNT known, an EBL mask is made in a software program (Elphy) for contacting the SWCNT with source and drain, and gating it with five side gates on both sides, see Fig. 5.5(d) and (e). Finally optical lithography is used to connect the electrodes and gates to macroscopic contact pads, see Fig. 5.5(a). Several devices similar to the one shown in figure 5.6 were fabricated, but they unfortunately all had a bad quality CNT that did not show regular CB oscillations, or there was no electrical contact between source and drain (probably due to defects in the CNT). So no useful measurements were obtained on this devise geometry.

The device shown in Fig. 5.7(a) is made in much the same way as described above. In Fig. 5.7(b) we subsequently made a kink in the SWCNT with an AFM. The kink is intended to induce a local defect in the SWCNT which would act as a tunnel barrier for the electrons dividing the SWCNT into two QDs,
Figure 5.5: A series of optical images and atomic force micrographs of the side-gate geometry. The series is a close-up all the way from macroscopic UV-lithography in (a) to the nanolithography in (d) and (e).

Each tunable with the outer side gates (SG1 and SG2 in Fig. 5.7(b)). The middle side gate is intended to tune the tunnel barrier, but since it was leaking we could not try this. In Fig. 5.7(d) we show the transconductance (dI/dSG1) as function of voltage applied to SG1 and SG2 with $V_{sd} = 1 \text{ mV}$, and since the middle side gate was leaking it is kept floating. Transconductance lines with a small curvature are observed. Each line is continuous through the plot with a negative slope and can thus be described as being due to transport through discreet energy levels of one QD. In a second device shown in Fig. 5.7(c) we tried to make the barrier in the middle of the SWCNT by placing a narrow electrode (50nm wide) on top of the SWCNT. Unfortunately we were only able to measure on the left-hand QD since there was no contact to the right-hand QD. In Fig. 5.7(e) we show current through the left-hand QD as function of voltage applied to the back gate and voltage applied between the upper and middle electrode marked "S" and "D" in Fig. 5.7(c). Clear diamond like structures with no current due to CB are observed. In each of these diamonds there is a fixed number of electrons on the QD. To add an electron to the QD an addition energy ($E_{add}$) is needed. The addition energy is given by a charging energy $U_C$ for every electron added to the QD plus a level spacing energy $\Delta E$ for every fourth electron added to QD (assuming no orbital splitting). The
Figure 5.6: Raith Micrograph Award 2004. A single wall carbon nanotube is contacted by palladium wires, and gated by five side-gates. The side-gates are made by very small nano-lithography (linewidth down to 50 nm) and aligned with high precision (±50 nm) to the nanotube. This micrograph won the first price in ”Raith Micrograph Award 2004” .
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insert of Fig.5.7(e) shows $E_{\text{add}}$ as function of electrons added to the QD. The addition energy is estimated from the (source-drain) height of each diamonds. From that plot we estimate $U_C \sim 10-11$ meV and $\Delta E \sim 3$ meV. This gives an estimated length of the QD of $L = \hbar v_F / (4\Delta E) \sim 275$ nm, which fits well with the measured length of the QD from the AFM micrograph $\sim 300$ nm.

As is probably clear by now the side-gate design for making a DQD newer worked for us. The fabrication process is very time consuming especially because, for every single device, the position of a SWCNT has to be measured with an AFM and a specific EBL mask has to be drawn. Furthermore, since the yield of devices with contact to the SWCNT was low, we decided that we needed a new fabrication scheme. In the following two sections we show two new fabrication schemes based on top-gates and trench-gates, which produce a much greater number of devices in each batch and also is much faster. The only downside of these two methods is that, before measuring at low temperature, it is difficult to know if there are more than one nanotube bridging the source

Figure 5.7: (a) A SWCNT (diameter $\sim 1$ nm) contacted with source and drain, and gated with three side-gates. (b) Same device as in (a) but after a kink is made in the nanotube with an AFM. (c) A device fabricated similar to (a), where a narrow electrode (50 nm wide, and marked “D”) is placed on top of the SWCNT in an attempt to make a local tunnel barrier. (d) Transconductance ($dI/dV_{SG1}$) versus voltage applied to the two side gates (SG1 and SG2) of the device in (b). (e) Current through the left-hand dot in (c) i.e. from “S” to “D” as function of voltage applied to the back-gate.
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and drain electrodes.

5.3.2 Top-gate

The top-gate design is the design we have used most, and it will be explained in chapter 6 and 7. The recipe is given in appendix E. When we started making these devices we were worried that making gates directly on top of the nanotube by EBL would damage the tube. For the center top-gate, which is intended to make a tunnel barrier this is fine, but for the two outer gates that should tune a QD situated below the gate this could become a problem. We therefore also started working on another fabrication scheme that could eliminate this problem by making the gates first and then subsequently grow the nanotube across the gates. These gates are etched into the substrate in narrow trenches and therefore called trench-gates.

5.3.3 Trench-gate

In the trench-gate design the gates are made first and the nanotubes are subsequently grown across. We experience that a smooth surface is crucial for making the nanotubes grow across the gates. Therefore, to smoothen the surface over the gate-electrodes we place them in narrow etched trenches and cover them with SiO$_X$. To ensure the gate electrodes are still metallic after growth of the nanotubes at 900°C they are made of platinum. To increase the number of devices made in each batch and decrease processing time, optical masks for alignment marks, gate oxide, catalyst islands and contact pads are made, so that only two steps of the more time consuming EBL are needed. The recipe is given in appendix F. An overview of a device with the trench-gate design is shown in Fig. 5.8.

It is fabricated the following way: First alignment marks for positioning all the layers accurately are made. Trenches are etched and filed with Pt, and most of the substrate surface is then covered with SiO$_X$, see Fig. 5.8(a)-(c). Islands of catalyst are positioned a few microns from the gates and nanotubes are grown, see Fig. 5.8(c)-(d). The nanotubes grow in random directions and has typical lengths of up to ~10 µm and will therefore self-align across the gates, provided that the surface is smooth. An electrodes are placed on each side of the gates to contact the nanotube that hopefully has grown across, see fig. 5.8d. Finally, optical lithography is used to connect the EBL-defined electrodes and gates to contact pads, so the device can be bonded onto a chip carrier.

Fig. 5.9 shows some measurements on a trench-gate device. The nanotube in the device is semiconducting, as seen in (a) where the the center gate is kept at zero keeping the Fermi level of the center part of the nanotube in the band-gap and thus creating a tunnel barrier in the center part of the tube. The voltage applied to the two outer gates ($V_{G1}$ and $V_{G2}$) are swept in large ranges from hole filling at negative gate-voltage to electron filling at positive gate-voltage. The result is the pattern seen Fig. 5.9(a), where transport are possible in the four corners. A more detailed plot of a small region of the (h,h) region (i.e. the
Figure 5.8: Overview of the trench-gate design for making a double quantum dot in a nanotube. (a)-(c) are optical images and (d) is a scanning electron micrograph. Gates made of platinum are placed in etched trenches and covered in $\text{SiO}_x$. Then nanotubes are grown from islands of catalyst and contacted by titanium electrodes. Finally optical lithography is used to connect the leads to contact pads.
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Figure 5.9: Measurements on a trench-gate device at 350 mK. (a) Current at $V_{sd} = 6$ mV versus large-range voltages applied to G1 and G2, with CG and back-gate kept at 0 V. (b) Similar measurement at $V_{sd} = 5$ mV but with a much smaller range of gate voltages. Triangular shaped triple points with exited states are seen.

lower left corner of Fig. 5.9(a) where two hole dots are defined) is shown in Fig.5.9(b). Triangular shaped triple points with lines parallel to the sides of the triangle due to exited states are seen. The triple points are expected to increase in size to this triangular shape when the bias voltage is increased as described in Ref. [65].
Chapter 6

A single wall carbon nanotube double quantum dot

Abstract:\footnote{This chapter has been published in Applied Physics Letters 89, 232113 (2006)}
We report on two top-gate defined, coupled quantum dots in a semiconducting single wall carbon nanotube, constituting a tunable double quantum dot system. The single wall carbon nanotubes are contacted by titanium electrodes, and gated by three narrow top-gate electrodes as well as a back-gate. We show that a bias spectroscopy plot on just one of the two quantum dots can be used to extract the addition energy of both quantum dots. Furthermore, honeycomb charge stability diagrams are analyzed by an electrostatic capacitor model that includes cross capacitances, and we extract the coupling energy of the double quantum dot.
Electronic transport in single quantum dots (QDs) defined in single wall carbon nanotubes (SWCNTs) has been studied intensively over the last decade [8, 12, 14]. These devices are typically made by placing metal electrodes directly on top of a SWCNT resulting in tunnel barriers at each SWCNT-metal interface, and gated by using the substrate as one global gate. Recent studies have shown that it is possible to locally gate and locally deplete a small segment of a SWCNT [66, 67]. By placing several such local gates on top of a SWCNT, a double quantum dot (DQD) with tunable inter-dot coupling can be made [68, 69, 65, 70, 71]. A DQD is a desirable system since it can be used in the field of quantum computation as e.g. a single charge qubit or two interacting spin qubits [72]. The advantage of making DQDs in SWCNTs instead of other material systems such as GaAs/AlGaAs is that SWCNTs are thought to have a longer spin decoherence time. An important source of decoherence is the hyperfine coupling between the electron in the QD and the nuclei. $^{13}$C is the abundant isotope in natural carbon and has no net nuclear spin. The hyperfine coupling will therefore be highly reduced in SWCNTs.

In this chapter we present a fabrication scheme to contact and place three narrow local gates on top of a SWCNT. We show that a device fabricated by this method can be used to define two coupled QDs in series. The addition energies of both QDs are estimated from a low temperature bias spectroscopy plot on just one of the QDs. These addition energies are then used together with a honeycomb charge stability diagram to estimate the electrostatic coupling energy of the DQD.

### 6.1 Top-gate defined double quantum dots

The devices are made on a highly doped silicon substrate capped by a 0.5 $\mu$m thermally oxidized SiO$_2$ layer, and we use the substrate as a back-gate to tune the global potential of the SWCNT. A set of alignment marks are made by electron beam lithography (EBL), which are used to accurately position the following three steps of EBL. First, islands of catalyst material consisting of a suspension of aluminum oxide nanoparticles in methanol with dissolved iron nitrate and molybdenum acetate are placed at specific positions, see Fig. 6.1(a). For easy lift-off and an even distribution of the catalyst we use a thick double layered resist (9% copolymer, and 4% PMMA) and spin on the liquid catalyst at 1000 rpm for 150 s. The SWCNTs are then grown by chemical vapor deposition from the catalyst islands in a ceramic tube furnace at $\sim 900^\circ$C with a controlled flow of gasses, Ar: 1.1 L/min, $H_2$: 0.1 L/min, and $CH_4$: 0.5 L/min [41, 2]. Typically only a few or one SWCNT will grow several $\mu$m away from the island, see Fig. 6.1(b). The alignment marks are secondly used to position source and drain electrodes consisting of 50 nm titanium with a separation of 1.8 $\mu$m. Since the SWCNTs tend to bundle together into ropes within a distance of about $\sim 1 \mu$m from the island, the electrode nearest to the island are positioned $\sim 2 \mu$m from island, thus favoring contact to long straight SWCNTs. In about 30% of our devices only one tube is contacted. Third, three narrow gate electrodes are
6.1 Top-gate defined double quantum dots

defined by EBL using a thin double layer resist (6% copolymer, and 2% PMMA) and positioned between the source and drain electrodes, by use of the alignment marks. The gates consist of five evaporations of aluminum each 2 nm thick and oxidized in air for about 1 min, and a top layer of titanium. We contact the EBL-structures with a final step of optical lithography to be able to bond the device onto a chip-carrier.

The fabrication scheme presented here has good possibilities to be scaled up to produce several devices in each batch. In Fig. 6.1(a) we show a pattern with four potential devices. Several of these patterns could easily be made in each batch, where we currently make just two. An atomic force microscope (AFM) micrograph of a finished device is shown in Fig. 6.1(b) where only one SWCNT is contacted. The three gates are named G1, CG (center gate), and G2 starting from the source electrode. We apply source-drain voltage ($V_{sd}$) to the source electrode and keep the drain electrode at ground. The nanotube in the device that we present measurements on in this chapter has a height (diameter) measured with an AFM of about $\sim 1$ nm. It shows an ambipolar characteristic at room temperature as seen in Fig. 6.1(c), which suggests that it is a small band gap semiconducting SWCNT. We can thus use the back-gate to tune the global potential of the device from electron to hole transport. In the rest of the this chapter the measurements are made through the valence band with a

Figure 6.1: (a) Optical image of 4 potential devices consisting of one common source electrode, three common top-gate electrodes, and four individual drain electrodes. On the left hand side of the source electrode an island of catalyst material is positioned from where the carbon nanotubes grow. (b) Atomic force microscope micrograph of the region indicated by the black rectangle in (a). To the left (close to the catalyst island) several tubes can be seen, but only one tube has grown several $\mu$m away from the island (indicated with white arrows). Source and drain electrodes consisting of 50 nm titanium, and three top-gate electrodes consisting of five 2 nm layers of air-oxidized aluminum and 30 nm titanium, are positioned directly on top of the tube. Some resist residue can be seen around some of the leads. Insert: Schematic side view of the device. (c) Current through the device as function of voltage applied to the back-gate at room temperature, and with 1 mV source-drain voltage.
Figure 6.2: (a) Bias spectroscopy plot of differential conductance ($dI/dV$) versus source-drain voltage and voltage applied to G1, with $V_{CG} = 0$ V, $V_{G2} = 1.1$ V, and $V_{BG} = -6$ V at 300 mK. The white dashed lines are guidelines to the eye, indicating charge degeneracy lines. The addition energies of each quantum dot are indicated with green arrows. (b) Schematic figures of the hole transport through the double quantum dot at positions indicated with letters in (a). Solid and dashed lines are filled and empty hole states, respectively. The coupling energy is here neglected since it is much smaller than the addition energies (see below).

back-gate voltage of $V_{BG} = -6$ V, to ensure that transport is governed by holes.

6.2 Bias spectroscopy

Figure 6.2(a) shows a bias spectroscopy plot at 300 mK of the differential conductance versus $V_{sd}$ and voltage applied to G1 ($V_{G1}$), with CG and G2 kept constant at $V_{CG} = 0$ V, and $V_{G2} = 1.1$ V, respectively. That is, in Fig. 6.2(a) QD1 is probed by the source electrode from the left hand side, and a discreet energy level of QD2 from the right hand side. Around zero bias the device does not conduct, and the onset of conductance is asymmetric around zero bias. The onsets of conductance at point a and b in Fig. 6.2(a) are positioned at $V_{sd} \sim -1.5$ mV, and $V_{sd} \sim 2.1$ mV, respectively. The conductance gap is constant in the bias spectrum in Fig. 6.2(a), and also constant in the whole gate range that we measured ($V_{G1} = \pm 100$ mV). This gap in conductance is due to the DQD nature of the device, where the first QD (QD1) is tuned by G1, and the second QD (QD2) is tuned by G2. Both QDs have Coulomb blockade (CB) oscillations and since QD2 is in CB for the chosen gate voltage on G2,
transport is blocked whenever the bias is smaller than the addition energy of QD2. Since QD1 is probed from the right hand side by energy levels from QD2 and because the chemical potential of the drain lead is aligned asymmetrically between two successive energy levels of QD2, the conductance gap is asymmetric around zero bias (see Fig. 6.2(b)). At point a the energy levels of the two QDs are aligned with the chemical potential of the source lead, and we have hole transport from drain to source. From point a to point b the energy level of QD1 and the chemical potential of the source lead are kept aligned and shifted together, while QD2 is kept constant in CB, i.e., no sequential tunneling is possible. At point b the chemical potential of the source lead and the energy level of QD1 become aligned with the next energy level of QD2, which gives hole transport from source to drain. The conductance gap is therefore a measure of the addition energy of QD2 ($E_{\text{add}2}$).

Above and below the conductance gap structures similar to CB diamonds for a single QD are observed. These structures are due to CB in QD1 and illustrated from point b to point d through point c in Fig. 6.2(a). Along the line from point b to point d the ground level in each QD is kept aligned, while the chemical potential of the source electrode is shifted downwards to align with the next energy level of QD1. Because of the capacitive coupling between source and QD1, a negative compensating gate voltage on G1 is needed to keep the ground levels in QD1 and QD2 aligned. The distance from point b to point d in source-drain voltage is therefore a measure of the addition energy of QD1 ($E_{\text{add}1}$). Since no odd/even or four-period structures originating from the level spacings in the two dots are observed in either bias spectroscopy plots or honeycomb charge stability diagrams (see below), we estimate the level spacings to be much smaller than the charging energies. The two methods to read-off the addition energy of QD1 and QD2 give on average $E_{\text{add}1} \sim 2.2\text{ meV}$, and $E_{\text{add}2} \sim 3.6\text{ meV}$. At higher bias (above the level of point d) more structures are observed. A full explanation of these structures are outside the scope of this chapter, but an interesting subject for further study.

6.3 Honeycomb charge stability diagram

Figure 6.3 shows a charge stability diagram of current through the DQD as function $V_{G1}$, and $V_{G2}$. Honeycomb structures can be identified throughout the plot which is a clear sign of a DQD with inter-dot coupling. Within each honeycomb structure the number of holes in each QD is constant, as indicated with relative hole numbers (N,M) in Fig. 6.3(a) and (b). At the corners of these honeycombs so-called triple points are located, where three charge states are degenerate, e.g., (N,M), (N+1,M), and (N,M+1). At these triple points an increase in current is observed consistent with sequential tunneling becoming possible via the three degenerate charge states. Furthermore, the overall slope of the honeycombs as illustrated with white dashed lines in the right side of Fig. 6.3(a) can be used to estimate cross capacitances. When G2 is decreased by $\Delta V_{G21}$ (indicated in Fig. 6.3(a)) one hole is added to QD1, i.e., a cross
Figure 6.3: Charge stability diagrams at 300 mK of the measured current as function of $V_{G1}$, and $V_{G2}$, with $V_{CG} = 0$ V, $V_{BG} = -6$ V, and $V_{sd} = 2$ mV. (a) Honeycomb pattern with relative number of holes in each QD indicated with green numbers. The white dashed lines are guidelines to the eye. (b) Close-up of one set of triple points at the position indicated with the relative hole numbers.
capacitance from G2 to QD1 exists. Since the vertical distance ($\Delta V_{G12}$) between the two almost vertical lines to the right in Fig. 6.3(a) tends to infinity, there is almost zero cross capacitance from G1 to QD2.

### 6.4 Electrostatic coupling

The observed splitting of adjacent triple points, as shown in Fig. 6.3(b) is due to coupling between the QDs. The electrostatic coupling energy ($U_{Cm}$) is defined as the change in potential energy of QD1 when a hole is added to QD2, or vice versa. We have extended the electrostatic capacitor model in Ref. [14] to include cross capacitances. We find that the electrostatic coupling energy can be given in terms of quantities directly observable in a bias spectroscopy plot and in a honeycomb charge stability diagram:

$$U_{Cm} = E_{add1(2)} \cdot \frac{\Delta V_{m_{G1}(2)}}{\Delta V_{G1(2)}} \cdot \frac{\Delta V_{G12(21)}}{\Delta V_{G12(21)} - \Delta V_{G1(2)}}$$  \hspace{1cm} (6.1)

where $\Delta V_{G1(2)}$, and $\Delta V_{m_{G1}(2)}$, are the size of the honeycombs and the splitting of the triple points, as illustrated in Fig. 6.3(a) and (b), respectively. The last term in Eq. (6.1) accounts for the cross capacitances and goes to unity when there are no cross capacitances, i.e., $\Delta V_{G12(21)}$ goes to infinity. An average estimate of $\Delta V_{G1(2)}$ from all the honeycombs seen in Fig. 6.3(a) gives $\Delta V_{G1(2)} \sim 6(10)\, mV$. The estimated values of $\Delta V_{m_{G1}(2)}$ and $\Delta V_{G12(21)}$ are; $\Delta V_{m_{G1}(2)} \sim 1.25(1.10)\, mV$, and $\Delta V_{G12(21)} \sim 20\, mV$ ($\Delta V_{G12}$ tends to infinity). Since the gate voltages used in the bias spectrum in Fig. 6.2 and the gate voltages used in the charge stability diagram in Fig. 6.3 are chosen to be roughly the same, we can use the addition energies found above in the estimation of $U_{Cm}$. From these experimental values two consistent estimates of the electrostatic coupling energy are obtained, $U_{Cm} \sim 0.46(0.42)\, meV$.

### 6.5 Conclusion

In conclusion we have presented a fabrication scheme that in each batch can produce several devices for electronic transport in a SWCNT with three narrow top-gates. We show that a device fabricated by this method can be used to define two coupled QDs in series. From a bias spectroscopy plot of just one of the QDs the addition energies of both QDs are extracted. Furthermore, an electrostatic capacitor model that includes an observed cross capacitance is used on a honeycomb charge stability diagram to extract two consistent estimates of the coupling energy.

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A single wall carbon nanotube double quantum dot
Abstract:
We report on strongly coupled carbon nanotube double quantum dots with clear shell structures. Both 4-electron shells and 8-electron shells are observed. We show that nonlocal singlet and triplet states can be controllably created in an arbitrary shell. We furthermore show, in good agreement with theory, that it is possible to excite electrons between the singlet and triplet state via inelastic cotunneling.
7.1 Introduction

An artificial atom also called a quantum dot (QD) can today reproducibly be fabricated in a carbon nanotube (CNT) by placing two metal electrodes on top of the CNT. CNT QDs are ideal systems for exploring few electron physics because they have a very robust shell structure owing to the one-dimensional nature of the CNT [8, 14, 10, 73]. Recent experiments from several different groups have shown that it is possible to define two coupled QDs in one single CNT molecule [69, 68, 65, 70, 71, 3]. Both weakly coupled DQDs showing clear triangular triple points with excited states, as well as strongly coupled DQDs having molecular states have been shown. It has even been demonstrated that the inter-dot coupling can be tuned with a gate. DQDs are interesting structures to explore because they can be used as quantum bits (qubits). CNT DQD are very promising systems for realizing the spin-qubit because CNTs are expected to have very long spin coherence times (no hyperfine coupling due to zero nuclear spin in $^{12}$C, low spin-orbit coupling due to low atomic number). To realize the spin-qubit in a CNT DQD several steps are still needed, one of which is control of the number of interacting electrons. We show in this chapter that such control can be obtained by exploiting the shell structure which is intrinsic to CNT QDs. Each shell is typically separated with a level-spacing which is larger than the line width of each shell. Therefore, electrons filled into the one shell of a CNT DQD will not couple to electrons in other shells. In this chapter we show that nonlocal spin entangled 2-electron states (nonlocal singlet and triplet states) can be created in an arbitrary shell of a CNT DQD. We also show that excitation between the singlet and triplet state is possible via inelastic cotunneling, which corresponds well with theory.

7.2 Fabrication and Characterization

The devices are made on a highly doped silicon substrate with a top layer of silicon dioxide. The CNTs are grown by chemical vapor deposition from islands of catalyst material and subsequently contacted by 50 nm Titanium source and drain electrodes. Next, three narrow top-gate electrodes are fabricated between the source and drain electrodes, consisting of aluminum oxide and titanium. A more detailed description of the fabrication and CNT growth procedure is given in chapter 5. A schematic figure of the device together with the measurement setup is shown in Fig. 7.1(a). Source-drain voltage ($V_{sd}$) is applied to the source electrode and the drain electrode is grounded through a current-to-voltage amplifier. The three top-gate electrodes are named G1, CG (center gate), and G2 starting from the source electrode, and the voltages applied to them are $V_{G1}$, $V_{CG}$, and $V_{G2}$. All experimental data presented in this chapter are measured in a sorption pumped $^3$He cryostat at a temperature of 350 mK.

The device we analyze in this chapter has two coupled QDs in series as conformed by the observation of the so-called honeycomb pattern in current through the device ($I_{sd}$) versus $V_{G1}$ and $V_{G2}$ (see Fig. 7.1(b) and 7.2). The
Figure 7.1: Shell filling of a carbon nanotube double quantum dot. (a) Schematic figure of our devices consisting of a carbon nanotube (CNT) with titanium source and drain electrodes and three top-gate electrodes, named G1, CG (center gate), and G2, made of titanium and aluminum oxide. (b) Surface plot of current ($I_{sd}$) at constant bias ($V_{sd} = 0.2 \text{ mV}$) as function of voltage applied to G1 ($V_{G1}$) and G2 ($V_{G2}$). The numbers (N,M) are shell occupation numbers for one 4-electron shell. (c) Black lines: Schematic honeycomb diagram for a 4-electron shell with strong tunnel coupling, and a small cross capacitance. Gray lines: Same honeycomb diagram with negligible tunnel coupling. Dashed lines indicate where the line-traces in Fig. 7.3 are measured.
center gate is kept constant at $V_{CG} = 0 \text{V}$ in all the measurements presented here. The two QDs are strongly coupled indicated by the facts that we observe the entire edge the hexagons that make up the honeycomb diagram, and that the corners of the hexagons are rounded. We characterize dot 1 (dot 2) by measuring the differential conductance ($dI/dV_{sd}$) as function of $V_{sd}$ and $V_{G1}$ ($V_{G2}$). Such bias spectroscopy plots (not shown) show Coulomb blockade diamonds with charging energies $U_{C1} \sim 3 \text{meV}$ for dot 1, and $U_{C2} \sim 3.5 \text{meV}$ for dot 2. The level spacing of each dot is extracted from the honeycomb pattern (see below) yielding $\Delta E_{1} \sim 1.2 \text{meV}$ for dot 1, and $\Delta E_{2} \sim 1.5 \text{meV}$ for dot 2, hence the two dots are roughly equal in size. We also see that G2 has a much stronger gate-coupling than G1 probably because of a broken gate-geometry in G1 weakening its gate-coupling.

### 7.3 Shell filling

The number of electrons in each dot can be controlled by tuning $V_{G1}$ and $V_{G2}$. In the middle of a hexagon (in Fig. 7.1(b) or 7.2) a fixed number of electrons are localized in each dot, and electron transport is suppressed by Coulomb blockade.

---

**Figure 7.2:** *Eight-electron shell structure.* Surface plot of current ($I_{sd}$) at constant bias ($V_{sd} = 0.5 \text{mV}$) as function of voltage applied to G1 ($V_{G1}$) and G2 ($V_{G2}$). The numbers (N,M) are shell occupation numbers for one 8-electron shell. Two further eight electron shells were observed in connection the this shell, one below, and one to the right.
At the edge of a hexagon, transport is allowed through molecular states formed in the DQD. The height (width) of a hexagon corresponds to the energy required to add an extra electron in dot 1 (dot 2), called the addition energy. In Fig. 7.1(b) we see that the width and height of the hexagons alternates in size in a regular pattern between large and small due to two-fold spin degenerate energy levels in each dot. A 4-electron shell structure (two electrons in each dot) can therefore be identified in this plot. Shell occupation numbers (N,M), where N (M) is the shell occupation number in dot 1 (dot 2), for one of these shells are written onto the honeycomb diagram in Fig. 7.1(b). The height (width) of the hexagon with charge state (1,1) in Fig. 7.1(b) corresponds to \( U_{C1} \) (\( U_{C2} \)). The height (width) of the hexagon with charge state (0,0) corresponds to \( U_{C1} + \Delta E1 \) (\( U_{C1} + \Delta E1 \)), see schematic honeycomb diagram in Fig. 7.1(c). We have found the gate-couplings of G1 (G2) to dot 1 (dot 2) from bias spectroscopy plots and then used this method to extract charging energies and levels spacings for each dot with average values as stated above. A small cross capacitance is also present, indicated by the slight tilt of the honeycomb diagrams in Fig. 7.1(b) and 7.2.

A nanotube DQD can also have an 8-electron shell structure, as observed in Fig. 7.2. In Fig. 7.2 the four numbered hexagons are distinctively larger than the others. Between the larger hexagons three smaller hexagons are seen in both \( V_{G1} \) and \( V_{G2} \) direction, indicating that each dot has four-fold degenerate levels yielding an 8-electron shell structure of the DQD. Electron occupation in one of the shells is indicated with shell filling numbers (N,M). The four-fold degeneracy of each dot is due to spin and orbital degeneracy. We have observed both 4-electron and 8-electron shell structures in two different devices and at several different gate voltage ranges.

### 7.4 Magnetic field spectroscopy

We will in the rest of this chapter focus on the 4-electron shell structure, as seen in Fig. 7.1(b). We analyze the 4-electron shell structure by magnetic field spectroscopy and observe that the sizes of the nine hexagons that make up the honeycomb diagram for one shell (see Fig. 7.1(c)) depends very differently on magnetic field. The four large hexagons ((0,0), (2,0), (0,2), (2,2)) decreases in size as the magnetic field is increased, whereas the central small hexagon (1,1) first decreases in size up to \( \sim 2 - 3 \) T and then increases in size. The different magnetic field dependence will be used to identify the different spin-states present in the DQD.

We start by analyzing the width of the hexagon with charge state (0,1), i.e., the two Coulomb peaks from (0,0) to (0,1), and from (0,1) to (0,2) (see Fig. 7.3(a) and (c)). The chemical potential for the first Coulomb peak (\( \mu_{01\rightarrow00} = E_{01} - E_{00} \)) is given by the eigenenergy of the state in region (0,1) (\( E_{01} \)), which at finite magnetic field is a spin down state \( |\downarrow_2\rangle \), minus the eigenenergy of the state in region (0,0) (\( E_{00} \)). The chemical potential for the second Coulomb peak is given analogously by \( \mu_{S(02)\rightarrow01} = E_{S(02)} - E_{01} \), where \( E_{S(02)} \) is the eigenenergy of the local singlet state in the (0,2) region (S(02)). We have here
Figure 7.3: **Magnetic field spectroscopy of Singlet and Triplet states.**

(a) and (b) Theoretical magnetic field dependence of the chemical potentials used for electron transport in (c)-(f). The line traces in (c)-(f) are extracted from eight honeycomb diagrams measured at $B = 0, 1, 2, \ldots, 7\, T$ in a gate range where the DQD had the same four electron shell structure as in Fig. 7.1(b). (c) and (e) Horizontal and vertical line traces through the hexagons with charge state $(0,1)$ and $(1,0)$ as function of electrostatic potential in dot 2 ($E_2$) and dot 1 ($E_1$), respectively. (d) and (f) Horizontal and vertical line traces through the central hexagon with charge state $(1,1)$. Each line in (c)-(f) is offset 0.2 nA, 0.3 nA, 0.3 nA, and 0.5 nA respectively for clarity, and the left-most peak is centered at zero since we were only able to measure relative positions.
neglected the triplet state $T_{0}(02)$ because it has $\Delta S_{T}(02) \sim \Delta E_{2}$ higher energy than $S(02)$. The magnetic field dependence of the two chemical potentials is found from the magnetic field dependence of the corresponding eigenenergies: $\mu_{01-00} = E_{01} - E_{00} \propto -1/2g\mu_{B}B$ and $\mu_{S(02)\rightarrow 01} = E_{S(02)} - E_{01} \propto 1/2g\mu_{B}B$ (see Fig. 7.3(a)). Therefore, as long as $T(02)$ is not ground state (i.e., for magnetic fields $B \lesssim \Delta E_{2}/g\mu_{B} \sim 13T$) we expect the two chemical potentials, as sketched in Fig. 7.3(a), to separate with a total separation-energy of $g\mu_{B}B$. The same analysis can be used on the two Coulomb peaks going from (0,0) to (1,0), and from (1,0) to (2,0), i.e., the height of hexagon (1,0). Measurement of the relative separation of these Coulomb peaks are shown in Fig. 7.3(c) and (e) (absolute position cannot be measured since each line trace is extracted from different honeycomb diagrams). The measurement shows good quantitative agreement with theory. The measured full splitting at $7T$ is $0.95\text{meV}$ and $0.8\text{meV}$ in (c) and (e), respectively, where theory predicts $g\mu_{B}B = 0.81\text{meV}$ with $g = 2$ for nanotubes.

### 7.5 Nonlocal Singlet and Triplet states

Nonlocal spin-entanglement between two electrons can be explored in a DQD by filling an electron into each dot. The nonlocal singlet ($S(11)$) and triplets ($T_{-}(11), T_{0}(11)$, and $T_{+}(11)$) are very interesting states to explore because they constitute nonlocally spin-entangled two-electron systems (with one electron in each dot). Nonlocally spin-entangled systems, like the $S(11)$ and $T_{0}(11)$, are prime candidates for making qubit operations because they constitute two spatially separated entangled spin-qubits. We will in the following show that these states can be controllably formed in an arbitrary 4-electron shell of a CNT DQD by filling in two electrons (one in each dot) into an otherwise empty shell.

In the (1,1) region of the honeycomb diagram we have one electron in each dot and the nonlocal singlet and triplets are therefore expected to participate in transport in this region. The separation between $S(11)$ and $T_{0}(11)$ is an exchange energy ($J$), and the ground state for $g\mu_{B}B < J$ ($g\mu_{B}B > J$) is $S(11)$ ($T_{-}(11)$).

To identify $S(11)$ and $T_{-}(11)$ we make horizontal and vertical line traces through hexagon (1,1) at increasing magnetic fields (see Fig. 7.3(d) and (f)). When going from region (1,0) to region (1,1) we fill, at low (high) magnetic fields, an electron into the $S(11)$ ($T_{-}(11)$) state. The magnetic field dependence of the chemical potential for the first Coulomb peak in the horizontal line trace is therefore expected to be given by:

\[
\mu_{S(11)\rightarrow 10} = E_{S(11)} - E_{10} \propto +1/2g\mu_{B}B, \quad \text{for} \quad g\mu_{B}B < J
\]

\[
\mu_{T_{-}(11)\rightarrow 10} = E_{T_{-}(11)} - E_{10} \propto -1/2g\mu_{B}B, \quad \text{for} \quad g\mu_{B}B > J
\]

(7.1)

where $E_{S(11)}, E_{10},$ and $E_{T_{-}(11)}$ are eigenenergies. The first equation describes transport at low magnetic fields when $S(11)$ is ground state, and the second equation describes transport at high magnetic fields when $T_{-}(11)$ is ground state. When going from region (1,1) to region (1,2) the initial state is $S(11)$
or T_{-1}(11) (depending on magnetic field) and we fill in the third electron into state $|↓_1↑_2↓_2⟩$. The magnetic field dependence of the chemical potential for the second Coulomb peak is therefore expected to be given by:

$$\mu_{12-S(11)} = E_{12} - E_{S(11)} \propto -1/2g\mu_B B,$$

for $g\mu_B B < J$,

$$\mu_{12-T-(11)} = E_{12} - E_{T-(11)} \propto +1/2g\mu_B B,$$

for $g\mu_B B > J$. (7.2)

The same magnetic field dependence of the chemical potentials (Coulomb peaks) is expected for the vertical line trace through region (1,1). The theoretical prediction (Fig. 7.3(b)) is in good agreement with the measurement in Fig. 7.3(d) and (f). Best fit between experiment and theory is obtained with the bend occurring around $\sim 2 - 3$ T which correspond to an exchange energy of $J \sim 0.23 - 0.35$ meV.

The exchange energy can also be estimated from the tunnel coupling strength $(t)$ using $J = 4t^2/U_C$ [74], which is valid at large detuning ($\varepsilon = E2 - E1$, centered at the anti-crossing of (11) and (02) see Fig. 7.4(d)), i.e., valid for this measurement. We estimate $t$ and mutual charging energy $(U_{Cm})$ by fitting Eq. (5.18) and (5.32) to the separation between the upper and lower wings at the four anti-crossing points for the shell analyzed in Fig. 7.3 yielding an average of $t \sim 0.45$ meV and $U_{Cm} \sim 0.4$ meV. This gives a consistent estimate of the exchange energy $J = 4t^2/U_C \sim 0.27$ meV.

### 7.6 Singlet - Triplet Excitation

The singlet and triplet states are especially interesting to analyze at the anti-crossing point between (1,1) and (0,2) because their chemical potentials cross at elevated magnetic fields, which can be used to excite electrons between the singlet and the triplet state via inelastic cotunneling at finite bias voltage. At elevated magnetic field and finite bias voltage we identify two distinct inelastic cotunneling lines, where the first is due to excitation of electrons from triplet to singlet and the second is due to excitation from singlet to triplet.

At the anti-crossing between (1,1) and (0,2) the states $S(11)$ and $S(02)$ hybridizes into a bonding state, which we call the singlet bonding state $S_B = \frac{1}{\sqrt{2}}(S(11) + S(02))$. The same is occurs for the triplet states, called triplet bonding state; $T_B = \frac{1}{\sqrt{2}}(T_{-1}(11) + T_{-1}(02))$, where the triplet state with $S_Z = -1$ is chosen because it has the lowest energy at finite magnetic field. The chemical potential for adding an electron into $S_B$ or $T_B$ along the black dashed line in Fig. 7.4(a) (given one electron in state $|↓_2⟩$) is given by (see section 5.2.4):

$$\mu_{S_B-01}(\varepsilon, B) = E_{S_B} - E_{01} = -\frac{1}{2}\sqrt{(2t)^2 + \varepsilon^2} + \frac{1}{2}g\mu_B B$$

$$\mu_{T_B-01}(\varepsilon, B) = E_{T_B} - E_{01} = -\frac{1}{2}\sqrt{(2t)^2 + (\varepsilon - \Delta E2)^2 - \Delta E2} - \frac{1}{2}g\mu_B B$$

where $E_{S_B}$, $E_{T_B}$, and $E_{01}$ are eigenenergies of $S_B$, $T_B$, and $|↓_2⟩$. We plot Eq. (7.3) and (7.4) in Fig. 7.4(d) with $B = 0$ T solid green and blue lines, and
Figure 7.4: **Excitation between singlet and triplet.** (a) Small section of the honeycomb diagram analyzed in Fig. 7.3 with $V_{sd} = 50 \mu V$ at $B = 0$ T (left) and $B = 6.5$ T (right). The numbers $(N,M)$ indicate the electron occupation for one shell. (b) Surface plot of current ($I_{sd}$) at $V_{sd} = 0.2$ mV versus detuning ($\varepsilon$) and magnetic field ($B$) at the black dashed line in (a). White lines are calculated positions of three cotunneling processes. (c) Schematic transport diagrams for elastic cotunneling (A) and inelastic cotunneling (B and C). (d) Chemical potential for adding an electron to the singlet bonding and triplet bonding state with $B = 0$ T (solid green and blue lines) and with $B = 6$ T (dashed green and blue lines) calculated using Eq. (7.3) and (7.4). The parameters used are $t = 0.45$, $\Delta E2 = 1.5$ meV, and $g = 2$.

with $B = 6$ T dashed green and blue lines. We see that $S_B$ is ground state at $B = 0$ T, and that the chemical potentials cross at elevated magnetic fields.

In Fig. 7.4(a) a broad peak in conductance versus detuning between the upper and lower wing in the gap between (1,1) and (0,2) is seen (white arrow marked A) due to elastic cotunneling via $S_B$. The elastic cotunneling process, schematically shown in Fig. 7.4(c) (mark A), takes place via two tunnel processes; $S_B \Rightarrow |01\rangle$ (an electron tunnels to drain), and $|01\rangle \Rightarrow S_B$ (an electron
tunnels into the DQD from source), where the charge state $|01\rangle$ becomes $|\downarrow 2\rangle$ at finite magnetic field. The elastic cotunneling peak is expected to be centered around $\varepsilon = 0$ due to equal weight of $S(11)$ and $S(02)$ at this point. At elevated magnetic field $(g\mu_B B \gtrsim t)$ $T_B$ becomes ground state for $\varepsilon < 0$ and the elastic cotunneling is therefore expected to become suppressed. Therefore, elastic cotunneling centered around $\varepsilon = 0$ is expected for $B \lesssim t/(g\mu_B)$ (white line marked A in Fig. 7.4(b)).

Fig. 7.4(b) show a surface plot of $I_{sd}$ versus $\varepsilon$ and $B$ along the black dashed line in Fig. 7.4(a), and theoretically calculated positions of elastic and inelastic cotunneling processes (white lines) in the same coordinate system. At elevated magnetic field the chemical potentials for adding an electron to $S_B$ and $T_B$ cross, which (with finite $V_{sd}$ applied) leads to inelastic cotunneling at both sides of the crossing (see Fig. 7.4(c) and (d)). The conditions for these two inelastic cotunneling processes are:

$$\mu_{S_B \rightarrow 01}(\varepsilon, B) - \mu_{T_B \rightarrow 01}(\varepsilon, B) = \pm V_{sd} \quad (7.5)$$

These two conditions give two $\varepsilon(B)$ expressions, which are plotted as white lines marked B and C in Fig. 7.4(b). In the first cotunneling process (B in Fig. 7.4) an electron is excited from triplet to singlet whereas is the second process (C in Fig. 7.4) an electron is excited from singlet to triplet (see Fig. 7.4(c)). Note that no fitting parameter is used in the calculation of these two lines (the parameters used, $t = 0.45\text{ meV}$, $\Delta E = 1.5\text{ meV}$ and $V_{sd} = 0.2\text{ mV}$, are all found from the analysis above or set during measurement). The calculated positions of the inelastic and elastic cotunneling processes seem to capture the measurement in Fig. 7.4(b) well.

### 7.7 Conclusion

In conclusion we have fabricated strongly coupled double quantum dots in carbon nanotubes exhibiting both 4-electron and 8-electron shell structures. We showed that nonlocal singlet and triplet states can be created in an arbitrary shell. We extracted an exchange energy, $J \sim 0.23 - 0.35\text{ meV}$, consistent with an estimated tunnel coupling strength, $t \sim 0.45\text{ meV}$. We furthermore showed, in good agreement with theory, that electrons can be excited between singlet and triplet states via inelastic cotunneling.

### Acknowledgements

We wish to acknowledge the support of the EU-STREP ULTRA-1D program.
Chapter 8

Summary and Outlook

Summary

The main achievement in the first part of this thesis is the observation of the Josephson effect (supercurrent) in all of the three well-established contact regimes; open regime (Fabry-Perot regime) in chapter 2, intermediate regime (Kondo regime) in chapter 3, and closed regime (Coulomb blockade regime) in chapter 4. The first to report supercurrent in SWCNTs was Kasumov et. al. in 1999 [17]. They reported a supercurrent much higher than theoretically predicted. The next report on supercurrent in SWCNTs was not until February 2006 by the Delft group [5] and our own in May 2006 [1] (chapter 2). Now a supercurrent much smaller than theoretically predicted was found! In chapter 1 we show that the circuit surrounding the superconductor-nanotube-superconductor junction is the cause of the heavily suppressed supercurrent, and in chapter 4 we shown that by carefully designing the surrounding circuit a supercurrent matching theoretical predictions can be found.

The main achievement in the second part of this thesis is the observation of shell structures in nanotube DQDs because it lead to our most interesting finding namely that it is possible to create nonlocally spin-entangled 2-electron states in an arbitrary shell (chapter 7). Developing a fabricating scheme for these devices proved very difficult, but after many many failed attempts we finally in December 2006 showed our first working device (Ref. [3] or chapter 6). It was made with a top-gate design, which we since have developed further into a promising trench-gate design (see chapter 5). In the initial phase of this project much time was spend on optimizing the nanotube growth procedure to obtain high quality nanotubes (see the three recipes in the appendix).
Outlook

An interesting device under recent investigation made from nanotube Josephson junctions is the nanotube Superconducting QUantum Interference Device (SQUID). SQUIDs can be used to make superconducting qubits [60, 61, 62], where the advantage of the nanotube SQUID (over other types of SQUIDs made in e.g. Al-AlO$_x$) is the high degree of tunability of the critical current in each Josephson junction, even reversal of the critical current is possible as shown in chapter 4.

By coupling a high frequency signal to a Josephson junction at its resonance frequency the supercurrent branch can be divided into one or more steps depending on the amplitude of the high frequency signal. This effect, called "Shapiro steps", has only very recently been investigated in nanotube Josephson junctions [75].

Spin blockade is an interesting effect in DQDs that has not yet been observed in nanotube systems. Spin blockade rectifies current at certain charge configurations because the local singlet-triplet splitting is much larger the the nonlocal singlet-triplet splitting. This difference in splitting is observed in chapter 7 in an arbitrary shell of a nanotube DQD, which indicates that spin blockade should be possible to observe in a nanotube DQD.

The singlet and triplet states can also constitute the two states in a spin-qubit as shown in Ref. [76]. A similar spin-qubit made in a nanotube DQD is also very interesting since the spin relaxation time is expected to be much longer in nanotubes.

Interesting devices can also be made by combining the josephson junction and the DQD. One idea from Refs. [77, 78] is to make three contacts to a nanotube with the middle electrode being a superconductor and the two outer electrodes normal metals, forming a QD on both sides of the superconductor. Cooper pairs injected from the superconductor will split into two electrons, one in each dot, thus creating a nonlocally spin-entangled 2-electron state.
Appendix A

Publication list

1. Critical current $0-\pi$ transition in designed Josephson quantum dot junctions
   H. Ingerslev Jørgensen, T. Novotný, K. Grove-Rasmussen, K. Flensberg, and P. E. Lindelof
   *NanoLetters* (Accepted for publication).

2. Electronic transport in single wall carbon nanotube weak links in the Fabry-Perot regime
   H. Ingerslev Jørgensen, K. Grove-Rasmussen, T. Novotný, K. Flensberg, and P. E. Lindelof

3. Single wall carbon nanotube double quantum dot
   H. Ingerslev Jørgensen, K. Grove-Rasmussen, J. R. Hauptmann, and P. E. Lindelof

4. Carbon nanotube Josephson junctions
   H. Ingerslev Jørgensen, K. Grove-Rasmussen, and P. E. Lindelof
   NSTI Nanotech 2006 Conference, Boston (May 7-11 2006), oral presentation.

5. Kondo resonance enhanced supercurrent in single wall carbon nanotube Josephson junctions
   K. Grove-Rasmussen, H. Ingerslev Jørgensen and P. E. Lindelof

6. Fabry-Perot interference, Kondo effect and Coulomb blockade in carbon nanotubes
   K. Grove-Rasmussen, H. Ingerslev Jørgensen and P. E. Lindelof
   *Physica E* (Accepted for publication).
7. Single Wall carbon nanotube weak links
   K. Grove-Rasmussen, H. I. Jørgensen and P. E. Lindelof

8. Kondo-Enhanced Andreev Tunneling in InAs Nanowire Quantum Dots
   Submitted to Physical Review Letters.
Appendix B

List of contributions at conferences, symposia, etc:

1. European Workshop on Electron Interactions in Ultra 1D nanostructures (ULTRA-1D Workshop), France, Nice (15 - 20 May 2007)
   Oral presentation:
   Shell filling in carbon nanotube double quantum dots

   Poster presentation:
   Single Wall Carbon Nanotube weak links

   Oral presentation:
   Carbon nanotube Josephson junctions & double quantum dots

4. NSTI Nanotech 2006, USA, Boston (7 - 11 May 2006)
   Oral presentation:
   Carbon nanotube Josephson junctions

   Poster presentation:
   Single Wall Carbon Nanotube weak links proceeding paper

   Oral presentation:
   Electron beam lithography and nanotubes with superconductors
7. 4th Øresund workshop on quantum transport
   Denmark, Niels Bohr Institute, (19 Dec. 2005)
   Oral presentation:
   Single wall carbon nanotube Josephson junctions

8. Solid state quantum transport and quantum information processing,
   Denmark, Niels Bohr Institute (17 Nov. 2005)
   Oral presentation:
   Single wall carbon nanotube weak links in the Fabry-Perot regime

9. Quantum coherence and decoherence at the nanoscale, (ULTRA-1D Workshop),
   Greece, Corfu (28 Aug. - 2 sep. 2005)
   Oral presentation:
   Single wall carbon nanotubes between superconductors

10. NT05 - sixth international conference on the science and application of
    nanotubes,
    Sweeden, Chalmers university of technology, Gothenburg (26 June - 1 July 2005)
    Poster presentation:
    Single-Walled Carbon nanotubes contacted to superconducting leads

11. NANO2005 - 11th Workshop on Electron Beam Lithography for Applications in Nanotechnology,
    Germany Dortmund, Raith (28 Feb - 1. March 2005)
    invited talk:
    Contacts and gates to carbon nanotubes made by E-beam lithography

12. Nanoscale Dynamics and Quantum Coherence,
    Germany, Hamburg University (19 - 23 Sep. 2004)
    Poster presentation
    Contacting single wall carbon nanotubes
Appendix C

Catalyst ”LFeMo5” for nanotube growth

This is the recipe for the catalyst ”LFeMo5” used to grow nanotube:

- 76 mg Al₂O₃ (Degussa) in 35 ml Methanol sonicated for 10 min.
- 23.9 mg MoO₂(CH₃COO)₂ in 10 ml Methanol sonicated for 10 min.
- 102.9 mg Fe(NO₃)₃ in 10 ml Methanol sonicated for 10 min.

The three solutions are mixed in a 100 ml flask and sonicated for ~ 3 min. For transfer 20 ml Methanol was used i.e. a total of 75 ml of Methanol is in the solution. The solution was left with mechanical steering for a weekend. Approximately one drop is spun onto the sample at 1000 rpm with the spinnor set to a slow start-up acceleration.
Catalyst "LFeMo5" for nanotube growth
Appendix D

Recipe for the CNT Josephson QD junction

1. 19x10mm Si/SiO$_2$ chip, 6x3 areas, $\Delta x = \Delta y = 3$ mm.

2. Cleaning: acetone and sonication > 2 min., methanol, IPA. Check that the surface is clean in optical microscope.

3. UVL1: Alignment marks:
   - LOR3B 4000 rpm 40 s, 185°C 4 min.
     AZ1505 4000 rpm 40 s, 115°C 45 s.
   - Expose edge with mask HIJ01, Edge 45 s.
   - Development, AZ400K 90 s, rinse in water.
   - Expose with mask HIJ01, pattern A.marks 5 s
   - Development, AZ400K 25 s, rinse in water.
   - Ash 20 s.
   - Metal evaporate 5 nm Ti and 40 nm Pt (to see them clearly after CVD-growth)
   - Lift-off: Remover PG + sonication.

4. Cut/cleave chip in appropriate sizes. (e.g. 3x2 areas on each)

5. UVL2: Catalyst islands:
   - LOR3B 4000rpm 40s, 185°C 4min.
     AZ1505 4000rpm 40s, 115°C 45s.
   - Expose edge with mask: KGRHIJ01 or HIJ02, Edge 45 s. (then rotate sample $\pi$ and expose again!)
   - Development, AZ400K 90s, rinse in water.
Recipe for the CNT Josephson QD junction

- Expose with mask HIJ01, pattern Cat. Top gate 5s., Align very precisely!
- Development, AZ400K 25s, rinse in water.
- Do not ash!
- Spin on catalyst "LFeMo5":
  - i Sonicate LFeMo5 5min, magnet stirring 2min.
  - ii Clean suction in spin-coater.
  - iii Apply one drop of LFeMo5, start spinner at 1000rpm with acceleration = 1/2 for 150s.
  - iv Heat plate 185°C 3min.
- Lift-off: Remover PG ~ 15 min, flush hard, sonicate 30s, rinse.

6. Growth of Carbon nanotubes:

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<tbody>
<tr>
<td>Flush</td>
<td>2.8 (100%)</td>
<td>0</td>
<td>0</td>
<td>T ~ 20°C</td>
<td>3min</td>
</tr>
<tr>
<td>Heating</td>
<td>1.1 (40%)</td>
<td>0.1 (20%)</td>
<td>0</td>
<td>T_{set} = 880°C</td>
<td>25min</td>
</tr>
<tr>
<td>Growth</td>
<td>1.1 (40%)</td>
<td>0.1 (20%)</td>
<td>0.5 (15%)</td>
<td>T = 880°C</td>
<td>10min</td>
</tr>
<tr>
<td>Cooling</td>
<td>1.1 (40%)</td>
<td>0</td>
<td>0</td>
<td>T ~ 35°C</td>
<td>30min</td>
</tr>
</tbody>
</table>

7. Check for CNTs in test areas with SEM. There has to be a lot of tubes, a carpet of tubes on the islands. If not, grow again with another temperature ±25°C.

8. EBL1: Normal metal, resistive leads:

- 6% Copolymer, 4000rpm 45s, 185°C 90s.
- 2% PMMA, 4000rpm 45s, 185°C 90s.

- Make a scratch in the lower right corner of the chip (for focusing).
- Elphy parameters:
  - i Mask: UV-HIJ01, JJ...
  - ii Sensitivity=200 µC/cm², step-size=2pixels, Work area 200 µm.
  - iii Reset alignment in Elphy,
- SEM parameters:
  - i 30KV, A. nr=4, CL=4, current constant, Time after flash > 1h ⇒ I_{beam} ~ 25 pA.
  - ii Focus in scratch.
  - iii Align rotation with metal bar (from UVL1) by using joystick.
  - iv Align "scan rotate" also with metal bar (~ 10 deg.).
  - v 400x , slow scan (10), beam blanker, extern control.
  - vi Expose (-1,-1)(test area), (-2,-2), (-2,-5)..., ∆x = ∆y = 3 mm.
    * Focus in middle of each pattern (Remember to read-of the current)
    * Align to alignment marks
* Expose layer 5
  - Development: MIBK:IPA 1:3 45s, rinse in IPA
  - Metal evaporate 15nm Cr, 15nm Au.
  - Lift-off: acetone
    Important: do not use sonication (it will destroy your tubes!!)

9. EBL2: Superconducting leads:
  - 6% Copolymer, 4000rpm 45s, 185°C 90s.
    2% PMMA, 4000rpm 45s, 185°C 90s.
  - Make a scratch in the lower right corner of the chip (for focusing).
  - Elphy parameters:
    i. Mask: UV-HIJ01, JJ...
    ii. Sensitivity=200 μC/cm², step-size=2 pixels, Work area 200 μm.
    iii. Reset alignment in Elphy,
  - SEM parameters:
    i. 30KV, A. nr=4, CL=4, current constant, Time after flash > 1h
      ⇒ \( I_{beam} \sim 25 \text{ pA} \).
    ii. Focus in scratch.
    iii. Align rotation with metal bar (from UVL1) by using joystick.
    iv. Align ”scan rotate” also with metal bar (~ 10 deg.).
    v. 400x, slow scan (10), beam blanker, extern control.
    vi. Expose (-1,-1)(test area), (-2,-2), (-2,-5).... \( \Delta x = \Delta y = 3 \text{ mm} \).
      * Focus in middle of each pattern (Remember to read-of the current)
      * Align to alignment marks
      * Expose layer 3
  - Development: MIBK:IPA 1:3 45s, rinse in IPA
  - Metal evaporate 5nm Ti, 60nm Al, 5nm Ti. Switch fast between metals to get a good alloy.
  - Lift-off: acetone (patience is a virtue)
    Important: do not use sonication (it will destroy your tubes!!)

10. UVL3: Contact pads:
  - AZ4511 4000rpm 40s, 115°C 45s.
  - Expose edge with mask KGRHIJ01 or HIJ02, Edge 45s. (then rotate sample π and expose again!)
  - Development, AZ400K 90s, rinse in water.
  - Expose with mask HIJ01, pattern 24pin 11-13s (test time!)
  - Development, AZ400K 60s, rinse in water.
Recipe for the CNT Josephson QD junction

- **Ash 25s**
- Metal evaporate 30nm Cr and 150nm Au.
- Lift off: acetone > 1 h.


12. Bond devices that work.
   Bond parameters: 80°C, First Bond: 3, 3, 4.5, Second bond: 5, 5, 5
   MANUAL BALL - Make sure the gold wire are properly grounded.
Appendix E

Recipe for the CNT double quantum dot with top-gates

1. 19x10mm Si/SiO$_2$ chip, 6x3 areas, $\Delta x = \Delta y = 3$ mm.

2. Cleaning: acetone and sonication > 2 min., methanol, IPA. Check that the surface is clean in optical microscope.

3. UVL1: Alignment marks:
   - LOR3B 4000 rpm 40 s, 185°C 4 min.
     AZ1505 4000 rpm 40 s, 115°C 45 s.
   - Expose edge with mask HIJ01, Edge 45 s.
   - Development, AZ400K 90 s, rinse in water.
   - Expose with mask HIJ01, pattern A.marks 5 s
   - Development, AZ400K 25 s, rinse in water.
   - Ash 20 s.
   - Metal evaporate 5 nm Ti and 40 nm Pt (to see them clearly after CVD-growth)
   - Lift-off: Remover PG + sonication.

4. Cut/cleave chip in appropriate sizes. (e.g. 3x2 areas on each)

5. UVL2: Catalyst islands:
   - LOR3B 4000 rpm 40 s, 185°C 4 min.
     AZ1505 4000 rpm 40 s, 115°C 45 s.
   - Expose edge with mask: KGRHIJ01 or HIJ02, Edge 45 s. (then rotate sample $\pi$ and expose again!)
   - Development, AZ400K 90 s, rinse in water.
Recipe for the CNT double quantum dot with top-gates

- Expose with mask HIJ01, pattern Cat. Top gate 5s., Align very precisely!
- Development, AZ400K 25s, rinse in water.
- Do not ash!
- Spin on catalyst "LFeMo5":
  i. Sonicate LFeMo5 5min, magnet stirring 2min.
  ii. Clean suction in spin-coater.
  iii. Apply one drop of LFeMo5, start spinner at 1000rpm with acceleration = 1/2 for 150s.
  iv. Heat plate 185°C 3min.
- Lift-off: Remover PG ~ 15 min, flush hard, sonicate 30s, rinse.

6. Growth of Carbon nanotubes:

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<td>3min</td>
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<tr>
<td>Heating</td>
<td>1.1 (40%)</td>
<td>0.1 (20%)</td>
<td>0</td>
<td>T_set = 880°C</td>
<td>25 min</td>
</tr>
<tr>
<td>Growth</td>
<td>1.1 (40%)</td>
<td>0.1 (20%)</td>
<td>0.5 (13%)</td>
<td>T = 880°C</td>
<td>10 min</td>
</tr>
<tr>
<td>Cooling</td>
<td>1.1 (40%)</td>
<td>0</td>
<td>0</td>
<td>T ~ 35°C</td>
<td>30 min</td>
</tr>
</tbody>
</table>

7. Check for CNTs in test areas with SEM. There has to be a lot of tubes, a carpet of tubes on the islands. If not, grow again with another temperature ±25°C.

8. EBL1: Electrodes and precision alignment marks:

- 6% Copolymer, 4000rpm 45s, 185°C 90s.
- 2% PMMA, 4000rpm 45s, 185°C 90s.
- Make a scratch in the lower right corner of the chip (for focusing).
- Elphy parameters:
  i. Mask: UV-HIJ01, TopGate_24pin_A
  ii. Sensitivity=200 µC/cm², step-size=2pixels, Work area 200 µm.
  iii. Reset alignment in Elphy,
- SEM parameters:
  i. 30KV, A. nr=4, CL=4, current constant, Time after flash > 1h ⇒ I_{beam} ~ 25 pA.
  ii. Focus in scratch.
  iii. Align rotation with metal bar (from UVL1) by using joystick.
  iv. Align "scan rotate" also with metal bar (~ 10 deg.).
  v. 400x , slow scan (10), beam blanker, extern control.
  vi. Expose (-1,-1)(test area), (-2,-2), (-2,-5)..., ∆x = ∆y = 3 mm.
  * Focus in middle of each pattern (Remember to read-of the current)
  * Align to alignment marks
* Expose layer 5
  - Development: MIBK:IPA 1:3 45s, rinse in IPA
  - Metal evaporate 10nm Ti, 30nm Au.
    Evaporate with low pressure $< 5 \cdot 10^{-7}$ Torr, and low rate 1-3Å/sec.
  - Lift-off: acetone
    Important: do not use sonication (it will destroy your tubes!!)

EBL2: Local gates / Top Gates:
  - 6% Copolymer, 4000rpm 45s, 185°C 90s.
  - 2% PMMA, 4000rpm 45s, 185°C 90s.
  - Make a scratch in the lower right corner of the chip (for focusing).
  - Elphy parameters:
    i. Mask: UV-HIJ01, TopGate, 24pin
    ii. Sensitivity=200 $\mu$C/cm$^2$, step-size=2pixels, Work area 200 $\mu$m.
    iii. Reset alignment in Elphy.
  - SEM parameters:
    i. 30KV, A. nr=4, CL=4, current constant, Time after flash $> 1$ h
      $\Rightarrow I_{beam} \sim 25$ pA.
    ii. Focus in scratch.
    iii. Align rotation with metal bar (from UVL1) by using joystick.
    iv. Align "scan rotate" also with metal bar ($\sim$ 10 deg.).
    v. 400x, slow scan (10), beam blanker, extern control.
    vi. Expose (-1,-1)(test area), (-2,-2), (-2,-5)..., $\Delta x = \Delta y = 3$ mm.
      * Focus in middle of each pattern (Remember to read-of the current)
      * Align to alignment marks
      * Expose layer 5
  - Development: MIBK:IPA 1:3 45s, rinse in IPA
  - Metal evaporate 50nm SiO (or 5x(2nm Al, vent in load-lock)) and 30nm Ti, align SiO(Al)-boat and Ti-E-gun with gate leads to prevent shadow evaporation.
    Degas SiO and evaporate $\sim 8$ Å/s, low pressure $< 5 \cdot 10^{-7}$ Torr. Ti: rate 1-3Å/sec
  - Lift-off: acetone
    Important: do not use sonication (it will destroy your tubes!!)

UVL3: Contact pads:
  - AZ4511 4000rpm 40s, 115°C 45s.
  - Expose edge with mask KGRHIJ01 or HIJ02, Edge 45s. (then rotate sample $\pi$ and expose again!)
Recipe for the CNT double quantum dot with top-gates

- Development, AZ400K 90s, rinse in water.
- Expose with mask HIJ01, pattern 24pin 11-13s (test time!)
- Development, AZ400K 60s, rinse in water.
- **Ash 25s**
- Metal evaporate 30nm Cr and 150nm Au.
- Liftoff: acetone > 1 h.


10. Bond devices that work.
    Bond parameters: 80°C, First Bond: 3, 3, 4.5, Second bond: 5, 5, 5
    **MANUAL BALL** - Make sure the gold wire are properly grounded.
Appendix F

Recipe for the CNT double quantum dot with trench-gates

1. 19x10mm Si/SiO₂ chip, 6x3 areas, $\Delta x = \Delta y = 3$ mm.

2. Cleaning: acetone and sonication $> 2$ min., methanol, IPA. Check that the surface is clean in optical microscope.

3. UVL1: Alignment marks:
   - LOR3B 4000 rpm 40 s, 185°C 4 min.
     AZ1505 4000 rpm 40 s, 115°C 45 s.
   - Expose edge with mask HIJ01, Edge 45 s.
   - Development, AZ400K 90 s, rinse in water.
   - Expose with mask HIJ01, pattern A.marks 5 s.
   - Development, AZ400K 25 s, rinse in water.
   - Ash 20 s.
   - Metal evaporate 5 nm Ti and 40 nm Pt (to see them clearly after CVD-growth).
   - Lift-off: Remover PG + sonication.

4. Cut/cleave chip in appropriate sizes. (e.g. 3x2 areas on each)

5. EBL2: Trench-gates and precision alignment marks:
   - 4% PMMA, 4000rpm 45 s, 185°C 90 s.
   - Make a scratch in the lower right corner of the chip (for focusing).
   - Elphy parameters:
Recipe for the CNT double quantum dot with trench-gates

1. Mask: UV-HIJ01, TopGate, 24pin, A layer 3
   ii Sensitivity=300 μC/cm², step-size=2 pixels, Work area 200 μm.
   iii Reset alignment in Elphy,

   - **SEM parameters:**
     i 30KV, A. nr=4, CL=4, current constant, Time after flash > 1h ⇒ \( I_{\text{beam}} \sim 25 \text{ pA} \).
     ii Focus in scratch.
     iii Align rotation with metal bar (from UVL1) by using joystick.
     iv Align "scan rotate" also with metal bar (≈ 10 deg.).
     v 400x, slow scan (10), beam blanker, extern control.
     vi Expose (-1,-1) (test area), (-2,-2), (-2,-5), ..., \( \Delta x = \Delta y = 3 \text{ mm} \).
       * Focus in middle of each pattern (Remember to read-off the current)
       * Align to alignment marks
       * Expose layer 5

   - Development: MIBK:IPA 1:3 45s, rinse in IPA
   - Ash 25 s.
   - Etch i buffed HF 7:1 20 s ⇒ 35 nm (40 sec gives about 70 nm).
   - Metal evaporate 5 nm Ti and 30 nm Pt, align Ti-boat and Pt-E-gun with gate leads to prevent shadow evaporation.
     Evaporate with low pressure < 5 \( \cdot 10^{-7} \) Torr. and low rate 1-3 Å/sec
   - Lift-off: acetone
     Important: do not use sonication (it will destroy your tubes!!)

2. UVL2: Gate oxide:
   - LOR3B 4000 rpm 40 s, 185°C 4 min.
     AZ1505 4000 rpm 40 s, 115°C 45 s.
   - Expose with mask HIJ02, TopGate3 (wrong name on UV-mask!) 5 s.
   - Development, AZ400K 25 s, rinse in water.
   - Ash 20 s.
   - Evaporate 60 nm-120 nm (which gives 50 nm-100 nm) SiO. Degas until \( P < 10^{-6} \) Torr. (might take more than 10 min!). Use high rate 1-3 Å/sec.
   - Lift-off: Remover PG.

3. UVL3: Catalyst islands:
   - LOR3B 4000 rpm 40 s, 185°C 4 min.
     AZ1505 4000 rpm 40 s, 115°C 45 s.
   - Expose edge with mask: KGRHIJ01 or HIJ02, Edge 45 s. (then rotate sample π and expose again!)
• Development, AZ400K 90s, rinse in water.
• Expose with mask HIJ01, pattern Cat. Top gate 5s., Align very precisely!
• Development, AZ400K 25s, rinse in water.
• Do not ash!
• Spin on catalyst “LFeMo5”:
  i. Sonicate LFeMo5 5min, magnet stirring 2min.
  ii. Clean suction in spin-coater.
  iii. Apply one drop of LFeMo5, start spinner at 1000rpm with acceleration = 1/2 for 150s.
  iv. Heat plate 185°C 3min.
• Lift-off: Remover PG ∼ 15 min, flush hard, sonicate 30s, rinse.

8. Growth of Carbon nanotubes:

<table>
<thead>
<tr>
<th></th>
<th>[L/min]</th>
<th>[L/min]</th>
<th>[L/min]</th>
<th>Temp.</th>
<th>Time</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flush</td>
<td>2.8 (100%)</td>
<td>0</td>
<td>0</td>
<td>T ~ 20°C</td>
<td>30min</td>
</tr>
<tr>
<td>Heating</td>
<td>1.1 (40%)</td>
<td>0.1 (20%)</td>
<td>0</td>
<td>T_set = 880°C</td>
<td>25min</td>
</tr>
<tr>
<td>Growth</td>
<td>1.1 (40%)</td>
<td>0.1 (20%)</td>
<td>0.5 (13%)</td>
<td>T = 880°C</td>
<td>10min</td>
</tr>
<tr>
<td>Cooling</td>
<td>1.1 (40%)</td>
<td>0</td>
<td>0</td>
<td>T ~ 35°C</td>
<td>30min</td>
</tr>
</tbody>
</table>

9. Check for CNTs in test areas with SEM. There has to be a lot of tubes, a carpet of tubes on the islands. If not, grow again with another temperature ±25°C.

10. EBL2: Electrodes:

• 6% Copolymer, 4000rpm 45s, 185°C 90s.
  2% PMMA, 4000rpm 45s, 185°C 90s.
• Make a scratch in the lower right corner of the chip (for focusing).
• Elphy parameters:
  i. Mask: UV-HIJ01, TopGate, 24pin, B layer 5
  ii. Sensitivity=200 μC/cm², step-size=2pixels, Work area 200 μm.
  iii. Reset alignment in Elphy,
• SEM parameters:
  i. 30KV, A. nr=4, CL=4, current constant, Time after flash > 1h ⇒ I_{beam} ~ 25 pA.
  ii. Focus in scratch.
  iii. Align rotation with metal bar (from UVL1) by using joystick.
  iv. Align ”scan rotate” also with metal bar (∼ 10 deg.).
  v. 400x, slow scan (10), beam blanker, extern control.
  vi. Expose (-1,-1)(test area), (-2,-2), (-2,-5),..., Δx = Δy = 3mm.
    * Focus in middle of each pattern (Remember to read-off the current)
100 Recipe for the CNT double quantum dot with trench-gates

* Align to alignment marks
* Expose layer 5

- Development: MIBK:IPA 1:3 45s, rinse in IPA
- Metal evaporate. Choose your metal! (50nm Ti is nice)
  Evaporate with low pressure < $5 \cdot 10^{-7}$ Torr, and low rate 1-3Å/sec.
- Lift-off: acetone
  Important: do not use sonication (it will destroy your tubes!!)

UVL4: Contact pads:

- AZ4511 4000rpm 40s, 115°C 45s.
- Expose edge with mask KGRHLJ01 or HIJ02, Edge 45s. (then rotate sample $\pi$ and expose again!)
- Development, AZ400K 90s, rinse in water.
- Expose with mask HIJ02, pattern 24pin BP 11-13s (test time!)
- Development, AZ400K 60s, rinse in water.
- Ash 25s
  - Metal evaporate 30nm Cr and 150nm Au.
  - Lift-off: acetone > 1h.


12. Bond devices that work.
   Bond parameters: 80°C, First Bond: 3, 3, 4.5, Second bond: 5, 5, 5
   MANUAL BALL - Make sure the gold wire are properly grounded.


