MAGNETIC FLUX MODULATION OF THE ENERGY GAP
IN NANOTUBE QUANTUM DOTS

BY

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We have studied the energy spectrum of multiwall carbon nanotubes using differential conductance measurements. The carbon nanotubes act as the Coulomb island of a single electron tunneling transistor. Temperature dependence of the conductivity curves shows that the electron wave functions extend between electrodes, and the energy levels are well-defined so they can be traced. Differential conductivity maps of the tubes are obtained as functions of the coaxial magnetic field, as well as of the perpendicular field. The maps for coaxial magnetic field show that a conductivity gap is induced as a function of the magnetic field, and the gap oscillates with a period of $h/e$, similar to that of Ajiki-Ando gap modulations. Ajiki and Ando predicted that a magnetic field can cause inter-conversion between metallic and semiconducting behavior of carbon nanotubes. Similarly, the measured density of states of the tubes also has a period of $h/e$. Tight binding calculations of carbon nanotube band structure are compared to the data.
To my wife Esra and son Talha.
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# Differential Conductance Oscillations of Multiwall Carbon Nanotubes with Magnetic Flux: Comparison to the Density of States Oscillations

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Chapter 1

Introduction

After their discovery 15 years ago\textsuperscript{1}, carbon nanotubes became the focus points of many researchers. Their properties including chemical, mechanical and electrical properties are studied extensively.

Carbon nanotubes look like rolled and seamed graphite sheets with two semi-spherical caps at the ends. Similar to a graphite sheet, carbon nanotubes have a hexagonal lattice structure. The way the graphite sheet is rolled and seamed defines the chirality vector of the carbon nanotube. The electronic properties of carbon nanotubes\textsuperscript{1,2,3} depend sensitively on the chirality vector. For example, depending on their chirality, the resulting single-electron energy spectra may or may not have gap at the Fermi energy, so the nanotubes can be either semiconducting or metallic\textsuperscript{4,5,6,7,8,9,10}. Currently, at the production level, there is no reliable way to produce a carbon nanotube with a chosen chirality. Although the chirality cannot be altered at will, the electronic properties — and, in particular, the energy gap — have been predicted to be tuneable by applying a coaxial magnetic field through the tube\textsuperscript{11}. The key point in understanding the coaxial magnetic field effect is the energy dispersion relation of 2D electrons in a graphite sheet, calculated by Wallace in 1947 (ref. 4). The Wallace energy band structure has six points in momentum space at which the energy gap goes to zero, i.e., the valence band touches the conduction band. The allowed energy states are distributed in reciprocal space as a result of periodic boundary conditions in the circumferential direction and the length of the tube. Application of a magnetic field alters the allowed momentum, and hence energy states. There is no other material known to date that can be periodically converted between
semiconducting, having a gap at Fermi Level, and metallic state, having no gap at Fermi level, by the magnetic field. This unexpected tunability of nanotubes was predicted by Ajiki and Ando (AA) in 1993 (ref. 11). Their predictions are further discussed in refs.12,13. The coaxial magnetic flux results in interconversion between semiconducting and metallic types of behavior, with the scale for the AA-gap and chirality-induced band gaps being comparable. The phenomenon is related to the Aharonov-Bohm (AB) coupling of the flux to the momentum of the electrons\(^{14}\). Application of the magnetic field along the tube, changes the momentum of the electrons, as predicted by the AB effect. At certain discrete values of the magnetic field, one of the momentum quantization lines cross one of the six points where the band gap is zero. Thus, for this field strength, a tube that is semiconducting in zero field could become metallic. Since momentum quantization lines are periodically spaced, one expects a periodic conversion of the tube between metallic and semiconducting states. This is the essence of the AA effect.

Multiwall carbon nanotubes (MWNT’s), unlike most other single molecules, have ideal size and structure for studying the effect of the AB-flux on the energy spectrum and conduction properties. So far, multiwall carbon nanotubes (MWNTs) remain as a single molecule, which can be treated with one full flux quantum by the accessible magnetic fields. Being concentric cylinders with outer radius \(R \approx 15\) nm, they allow penetration by an entire flux quantum \(\Phi_0 = h/e\) at accessible magnetic fields of about \(\Phi_0/\pi R^2 \approx 6\) T. In contrast, a typical single-wall nanotube\(^2\) with a radius of \(1.5\) nm would demand a \(\sim 600\) T magnetic field, which is currently not technically feasible. Note that previous experiments\(^{15,16}\) indicate that only the outermost shells contribute to the conduction. Thus \(R\) is in fact the relevant radial length scale, and one can expect to see AB modulations in, e.g., transport and spectral properties, such as the gap variation, over a field range of order \(10\) T.
Although the electronic properties of the tubes were studied previously as a function of magnetic fields, these works failed to provide convincing evidence for interconversion of the tubes and AA gap modulation. Some previous works focused on linear transport properties of long samples, typically several microns in length\textsuperscript{17,18}. Linear transport measurements showed oscillations with a period of $\hbar/2e$ due to weak localization effects\textsuperscript{19}. Other groups observed an oscillation with a period of $\hbar/3e$ and explained it by inner-shell contributions to the conductivity. Weak localization is characterized with negative magneto-resistance. Recently, positive magneto-resistance is also observed in transport measurements, which has caused disagreement among researchers\textsuperscript{20,21,22}. Theoretical papers show that in the ballistic regime\textsuperscript{23} where the mean free path is longer than the circumference of the tube. Positive magneto-resistance is also possible. Positive magneto-resistance has been observed depending on the electrostatic potential of the tube\textsuperscript{24}.

We performed experiments on quantum dot single-electron-tunnelling (SET) transistors\textsuperscript{25}, made out of a short multiwall nanotube (MWNT) and threaded by AB flux. By measuring the differential conductance, we observed a magnetically induced gap modulation at the Fermi level, thus providing evidence for the aforementioned interconversion with a period of $\Phi_0$.

In this dissertation, sample preparation, experimental setup, theoretical background, and experimental results will be presented.

Sample preparation is the most challenging part of this research. In order to test the AA effect, the tubes are supposed to be short and fat. However, it is very unlikely to have a tube with a length less than 500 nm sit across a 100 nm trench, which separates the two electrodes, and has large diameter at the same time. Chapter 2 will discuss how I prepare the samples,
choose the tubes, and how I connect electrodes and measure the samples. The parts of the experimental setup used during the measurements will be explained in details.

Depending on the electronic transparency, the conductivity of the contacts between carbon nanotubes and electrodes, the tube might act as a SET transistor. Theoretical background to understand the SET transistor is given in Chapter 3. The theoretical formulas are fitted with experimental results and prove that our tubes act as a SET transistor with well-defined energy levels.

Chapter 4 studies the conductance of the Coulomb island as a function of temperatures. This chapter focuses on the temperatures where the thermal fluctuations are larger than the charging energy. This regime is called weak Coulomb blockade. The conductance of the Coulomb island is explained using the theories developed for multiple tunnel junctions in this regime.

Chapter 5 provides theoretical calculations related to the band structure of carbon nanotubes. This chapter uses the tight binding method to calculate the band structure of a graphite sheet, which is the basic constituent of the nanotube. Later, by imposing additional constraints on the results, the band structure of the carbon nanotube is obtained. Theory predicted interconversion between metallic and semiconducting types of the nanotube with a period of $\Phi_0$. When the Zeeman splitting is included, the gap induced by the magnetic field slowly disappears.

Chapter 6 is a general review of Aharonov-Bohm effect on different systems. This chapter explains the previously done experiments on metal rings or cylinders as well as the carbon nanotube.
Chapter 7 shows the most important result of this thesis. The conductivity map of our SET transistors based on MWNT as a function of magnetic field and bias potential shows the energy levels of the tube oscillates with a period of $\Phi_0$. Furthermore, the conductivity maps show that the Coulomb blockade is effective only for the bias potentials smaller than $e/C$, where $C$ is the total capacitance. Further experiments confirmed that the magnetic field perpendicular to the axis of the tube did not produce anything like the magnetic field parallel to the tube.

Chapter 8 shows the energy spectrum of the carbon nanotubes studied using differential conductance measurements for different off-set bias potentials. A coaxial magnetic field was applied parallel to the tube. We find some indications that the energy spectrum of the tube oscillates with a period of magnetic flux quanta $\Phi_0$. The differential conductance shows different phases of oscillations, i.e. positive magneto-resistance, negative magneto-resistance for different offset bias potentials.

1.1. References


Chapter 2

Experimental Procedure

2.1. Introduction

The central theme of my thesis research project was a study of the effect of the magnetic field on the electronic properties of carbon nanotubes. This chapter provides an explanation to our choice of the nanotubes and the sample fabrication.

As predicted theoretically by Ajiki and Ando\(^1\), orbital magnetic effects in nanotubes become especially strong when the magnetic flux through the carbon nanotube reaches the value of one-half of the superconducting flux quantum, \(\hbar/2e\). In order to be able to inject a full flux quantum into the nanotube, we chose to work with multiwall carbon nanotubes. Because of their large diameter (~30 nm in many cases), the field required to achieve \(\hbar/2e\) magnetic flux is in the range of 5-10 T, which is technically possible to achieve using a regular superconducting solenoid. In the case of a small-diameter, single-wall carbon nanotube one needs to use a magnetic field of the order of 1000 T, which is not available in current laboratory conditions. Thus we do all measurements exclusively on the multiwall nanotubes.

In order to study the electronic spectrum, the carbon nanotubes have been placed on two independent electrodes, which act as the source and the drain of the single-electron-tunneling (SET) transistor, while the nanotube represents the Coulomb islands of the SET transistors. In most of the previous studies, only the single-wall carbon nanotubes were used in SET transistors\(^2,3,4\). Fabrication of a SET transistor based on a multiwall carbon nanotubes (MWNT) is complicated by the following factors: (i) Typically, a MWNT has a larger diameter, which
corresponds to a low charging energy, thus requiring a lower temperature for the observation of the SET effects. (ii) The large nanotube diameter leads to a low contact resistance, frequently lower than the quantum resistance, in which case the Coulomb blockade is suppressed and the SET transistor does not work properly. (iii) In most cases multiwall carbon nanotubes have a high level of defects, which applies additional restrictions on the dimensions of the SET device. In most of the experiments we used short nanotubes, typically shorter than 1 micron in length, in order to increase the Coulomb charging energy and make it larger than the thermal energy. The quality of nanotubes varies by the supplies, i.e., the average length, diameter, rigidity etc. We found that arc-produced tubes by Alfa-Aesar$^5$ are of good quality and many tubes appear to be shorter than 1 micron.

Samples are prepared on 4.8x4.8 mm silicon chips, each having a 100 nm wide trench. The trench allows the formation of two electrodes separated by a gap of ~100 nm. In short, to form the electrodes on which the nanotube is going to be positioned, we sputter metal over the trench, which cuts the film into two parts, separated by a gap of ~100 nm. Then photolithography is done to shape the electrodes and the contact pads. These two electrodes and a MWNT positioned over them form an SET transistor. The chips are mounted on plastic-based six-pin chip carriers$^6$. The measurements are done at temperatures down to 300 mK using He-3-SSV cryostat by Janis Research Co.$^7$ A detailed description of the sample fabrication and of the measurement setup are provided below.

### 2.2. Silicon chips

The wafer used to make chips with nanotube-based single electron transistors has a multiple-layer structure. This Si wafer (orientation 100) is 500 microns thick and is covered
with two insulating films: 500 nm of SiO$_2$ and a 60 nm low stress SiN film on the top. The SiO$_2$ oxide film itself consists of two layers, to insure good insulating characteristics: 400 nm thick wet SiO$_2$ and 100 nm dry SiO$_2$ oxide below it, right on top of the Si wafer. The surface of the Si wafer is heavily doped with phosphorus. The doped surface layer is about 10 microns thick and the doping concentration is $\sim 4 \times 10^{20}$ atoms/cc of P. The doping is done in order to ensure a high conductivity of Si even at cryogenic temperatures. This layer is used as the gate of the SET transistor. The nominal conductivity of this layer is $\sim 5 \times 10^4$ S/m. In order to produce trenches in the Si wafer, which serve as the electrode separation, we perform a step of standard electron beam lithography (the process is done by Gabriel Coeval in the Micro and Nanotechnology Laboratory of the Department of Electrical Engineering at UIUC). During this process the Si wafer is covered with an e-beam sensitive resist (PMMA). Later, a 100 nm wide line and some markers (5 micron by 5 micron size numbers positioned along the trench) are written on the resist. After developing the resist, the reactive ion etching (RIE) technique is used to etch trenches through the SiN film, by SF$_6$ plasma. The wafer was exposed to SF$_6$ for 3 minutes with a power of 150 W and 50 ccm gas flow. The RIE step engraves the trench and the markers in SiN film. After the RIE, the wafer is covered with a protective layer of photoresist (AZ5214) and diced into 4.8 mm by 4.8 mm square chips. Figure 2.1 shows a schematic picture of the chip.

After dicing, the chips are cleaned through the following steps: (i) The chips are sonicated in acetone for 5 minutes (we use an Aquasonic model 250T ultrasound bath for sonication), (ii) rinsed in deionized (DI) water, (iii) sonicated in 69.8% Nitric acid for 5 minutes, (iv) rinsed in DI water again. After the cleaning, we deepen the trench and create an undercut along the trench (into the SiO$_2$ and below the film of SiN), by immersing the chip.
Figure 2.1. The figure shows the chip (4.8x4.8 mm) with a protective resist on the top, which was used to prevent any dust contamination during the dicing process. Each color in the schematic shows a different layer. The chip contains the following layers (from the top to the bottom): 60 nm thick SiN, 500 nm thick SiO$_2$, and 10 micron thick doped Si$^{++}$ (P-doped) and the Si wafer itself, which is 0.5 mm thick. The trench and the markers are covered by the resist and not visible in the drawing.

into a 49% HF acid solution, for ~7-9 s. The HF etches the silicon oxide much faster (2300 nm/min) [9] than it etches the SiN (5 nm/min). Thus, by using HF we are able to remove some amount of SiO$_2$ inside the trench without changing the SiN film considerably. After this step, the edges of the SiN film near the trench become suspended and freestanding. After the HF, the samples are rinsed in DI water and then placed in nitric acid for another 5 minutes, without sonication, in order to remove HF and possible organic deposits. The nitric acid is rinsed with
Figure 2.2. The figure shows schematic pictures of the Si chip at different stages of the sample preparation process. (a) The trench and markers are put down by e-beam lithography in the following way: e-beam lithography is used to pattern the e-beam resist (the resist is not shown). The SiN film is etched by the SF$_6$ plasma in the regions which are not protected by the e-beam resist after e-beam lithography. The chip is sonicated in acetone, and nitric acid to remove the residual resist and any other organic contamination. Between the steps de-ionized water is used to rinse the chips. (b) The SiO$_2$ that is just below the open trench is etched by hydrofluoric acid (HF). This process creates an undercut around the trench, which is shown in the picture. Similar undercuts are expected through the markers but they are not shown in the drawing. (c) A metallic film is deposited over the chip. Usually a 3-5 nm Cr layer is deposited first and followed by 25 nm thick Au film. The picture shows the film covering the entire chip and illustrates how the trench produces a gap between the two electrodes, which are being formed on the banks of the trench. The chips covered with the Cr/Au bi-layer are ready for the deposition of nanotubes.
DI water. Finally, the chip is rinsed with 2-Propanol, and blown-dried in dry nitrogen gas. After cleaning and creating trench steps, the chips are ready for metal deposition. The entire chip is covered with a 3 nm Chromium sticking layer and a 30 nm Au film on the top. In most cases we use a DC Argon sputtering method (AJA Co-Sputter System) in order to deposit metallic films. Based on our experimental results, no significant difference on the contact resistance is observed between this type of electrodes and the electrodes made by evaporation. Figure 2.2 shows the schematic chips pictures after the removal of the protective resists, HF etching, and metal deposition. The chips shown are ready for the deposition of carbon nanotubes.

2.3. Tube deposition

The multiwall carbon nanotubes (MWNT) are bought from different suppliers. All the tubes are inspected under the Scanning Electron Microscope (SEM). Tubes from different suppliers may have different average lengths and diameters. They have different carbon contamination levels and even different rigidities. The ones supplied by Alfa Aesar are the most suitable for our type of experiments, because these tubes are short, relatively thick and straight. They have diameters of 20-40 nm and lengths of 500-5000 nm. The tubes were bought as soot clusters. After receiving the soot, a very small amount of the material is dispersed in a solvent. We typically use 1,2 Dichloroethane (DCE) or 2-Proponal to disperse nanotubes. In order to have a homogeneous suspension, the suspension with the tube soot is sonicated for about 5 minutes using an Aquasonic model 250T ultrasound bath. The shock waves created by the Aquasonic serve to separate the nanotubes from clusters and suspend the tubes in the liquid. At the end of sonication, we have a homogenous suspension. The darker the suspension, the larger the tube concentration in the suspension. A dark gray color liquid is the most suitable, as
**Figure 2.3.** Carbon Nanotubes are first sonicated in DCE before depositing on the chips with electrodes. (a) The carbon nanotube soot is shown, as received from the producer. (b) Tubes are dispersed in either 1,2 Dichloroethane or 2-Propanol. Such suspension is used in order to deposit tubes on the chip. The suspension initially is sonicated in our ultra-sound bath for about 5 minutes. Later, a drop of the suspension is placed on the Si chip and covers the entire top surface of the chip. After about 1 minute, the chip is blow-dried. Some tubes will stick to the surface.

It leaves a few tubes in every 100 µm² on the chip surface after the deposition process. Figure 2.3 shows the dust-like clusters of the carbon nanotubes as received, and the tube suspension.

After the tubes are suspended in the solvent and the suspension looks homogenous, the suspension can be used to deposit tubes on the surface of previously cleaned chips which have gold electrodes. Using a clean pipette, one or two drops of the suspension are placed on the surface of the chip in such way that the top surface of the chip is completely covered, while no liquid goes to the sides of the chip or to the bottom surface of the chip. This is done to prevent any contamination, as the bottom surface of the chip might not be as clean as the top surface.
Because of Van der Waals forces, some of the tubes will adhere to the surface of the chip. After waiting for about 1 minute, the suspension liquid is blown off the surface by dry nitrogen gas. Figure 2.4 shows the schematic picture of each step during the tube deposition.

2.4. Locating the tubes and forming the electrodes

A scanning electron microscope (SEM, Hitachi S-4700) is used to locate nanotubes. Hitachi S-4700 is a high-resolution field-emission microscope with a resolution of ~2 nm at 1kV accelerating voltage or 1.5 nm at 15kV acceleration voltage. During our work, most of the time 5 kV or 10 kV is used. The entire length of the trench on the chip is searched, 20 µm at a
Figure 2.5. The pictures show SEM (Hitachi S-4700) images of multiwall carbon nanotubes deposited on gold films on the top of silicon chips. The tubes are resting on the top of electrodes separated by the trench. The dark black region in the images is the trench separating the two electrodes. The tubes selected for measurements are those that are short, clean, and have a large diameter. All scale bars are 100 nm.

time, for suitable tubes. Positions of all the usable tubes are noted with respect to the markers. For experimental purposes, the shortest tubes, with the largest possible diameters, are chosen.
The reason behind choosing tubes as short as possible is that the shorter the tube, the shorter segment will be in contact with the electrodes, thus giving the highest possible contact resistance. High contact resistances are necessary for the Coulomb blockade phenomena. In addition, shorter tubes have a larger energy level spacing, which enables resolving single energy levels at ~300 mK. The reason behind choosing tubes with the largest available diameter is that it enables us to reach a higher value of the magnetic flux through the tube. Figure 2.5 shows some SEM images of tubes lying on the trench, which separates the two electrodes, which are the source and the drain of the SET transistor.

After locating all the tubes that can be used in experiments, according to the previously given criteria, the best tube is chosen. The electrode patterns are formed on the photoresist by the photolithography method. Photolithography is a method used to transfer images of a mask to the wafers, a Si chip in our case. Wet etchants, gold and chrome etchants, are used to remove the uncovered, exposed metals. In this process, the chip is baked at 100 Celsius for 5 minutes to remove any water molecules attached to the surface. It was later cooled down in continuously flowing dry nitrogen gas. The nitrogen gas flows through a reagent bottle with HMDS (Hexamethyldisilazane) at the bottom. HMDS is used (in some cases) to ensure that the photoresist does not lift off during the rest of the steps. After 5 minutes in dry nitrogen flow, the chip is placed on a spinner chuck. A positive photoresist (AZ 5214)\(^{10}\) is dripped on the chip till it covers the complete the top of the chip. Later, the chip is spun at 4000 rpm for 30 seconds, resulting in a 1.4 \(\mu\)m thick film. Spinning provides uniform photoresist films and removes the excess photo resist by centrifugal forces. The chip is placed on hotplate for 40 seconds at 100 Celsius to bake the photoresist. Afterward a photo mask which has different size electrode patterns on it is placed on the surface of chip. A suitable pattern is aligned with
Figure 2.6. Photolithography steps. (a) Initially, the chip is spin-coated with photoresist. The chip is spun at 4000 rpm for 30 seconds to form a 1.4 \( \mu \)m film of photoresist. (b) The photomask is placed on the top of the chip. The pattern of electrodes on the mask is aligned using the markers, under the microscope, in such a way that the selected tube stays under the pattern. (c) The chip is exposed to the 365 nm wavelength UV light for 6 seconds. The electrodes connected to the nanotube (not shown) are formed on photoresist after developing the exposed and chemically changed part of the photoresist.
respect to the markers of the chip, so that the electrodes can be formed where the nanotube is located. Later, the chip is exposed to 365 nm wavelength UV light for 6 seconds. UV light chemically changes the exposed part of the photoresist and makes it soluble in the appropriate developer. Immediately, a chemical developer (AZ developer 351) is used to etch the exposed regions of the photoresist for 40 seconds. The pure developer is diluted with DI water with 5:1 water:developer ratio before using it. Finally, the unexposed part of the photoresist will stay on the chip right on the top of the nanotube. The remaining photoresist forms a pattern of electrodes. Figure 2.6 summarizes the main photolithography steps. During this process, one of the two different photo masks is used to form the electrodes. This mask lets us choose one tube per chip and form only one set of electrodes on the chosen tube. A second mask is developed to allow more than one set of electrodes on different tubes on the same chip. The second mask consists of many different shapes. By using these shapes, one can expose all the desired regions with UV source, and form many electrodes, side by side.

After patterning the electrodes on the photoresist, chemical wet etchants are used to pattern the underlying metal by etching the unwanted parts of the metallic film. First, the chip is immersed in gold etchant\textsuperscript{11} (GE 8110), and kept there for 8 seconds, which is long enough to remove the gold film completely. After rinsing the chip in DI water, the chip is immersed in a chromium etchant\textsuperscript{12} (Cr-7) for 5 seconds. Ge 8110 etches 3 nm of gold per second, and Cr-7 etches 2 nm of Cr per second. Immediately after, the chip is rinsed with DI water and then blow-dried. After removing exposed metal, the conductivity of the chip surface is measured with a multimeter in the regions where the metal is supposed to have been removed. It is important to have no conductivity to ensure proper operation of the device. After the metal etching step is finished, the chips are washed with acetone to remove the remaining photoresist.
After photolithography, the chips are immersed into gold etchant (GE 8110), and then rinsed with water, and later immersed in chromium etchant (Cr-7). (b) The photoresist is removed by keeping the chip in acetone after etching the metals. The successful etching process is confirmed by measuring the resistance on the bare surface of the chip. Finally, the photoresist is removed with the acetone.

for 1 minute and then rinsed in DI water and later in 2-propanol just before drying. Figure 2.7 schematically explains these steps. At the end of this process the chip has the source and drain electrodes formed on it. The tube is ready to connect to the circuit and be measured using the electrodes.

2.5. Mounting silicon chips on chip carriers

Orienting the tube parallel to the applied magnetic field is very important. To ensure that the tube axis is parallel to the magnetic field, extra attention is paid. The SEM picture of the tube shows the orientation of the tube with respect to the trench. In addition, we know that the trench is parallel to the edge of the chip. So, the orientation of the tube with respect to the
edge of the chip is known. Using this information, we can orient the chip carrier, so that the tube is parallel to the magnetic field during the measurements.

The samples are placed on a chip carrier in order to mount them in our experimental setup. A couple of glass chips are glued to the chip carrier using stycast epoxy glue\(^{13}\) in order to elevate the sample. By doing that, we can raise the chip above the electrical pins available in the chip carrier, and thus obtain enough room in order to rotate the chip by any desired angle. Carbon tape is placed on the top of the glass pieces in order to hold the sample on the glass slides mounted on the chip (Figure 2.8). Soon after, gold wires are soldered to the electrical pins on the chip carrier. Gold wires\(^{14}\) are very thin, with a diameter of 50 µm. The gold can melt in contact with molten solder, if the temperature rises above 350°C, so the soldering needs to be done quickly, so that the solder solidifies exactly at the time when the gold wire is inserted into it. Figure 2.8 shows a chip carrier, which has gold wires attached to the pins. Next, the Si chip with electrodes and the nanotube is placed on the prepared chip carriers. The angle of the chip with respect to the chip carrier is chosen using the SEM pictures, as explained above, in order to make the tube parallel to the field during the measurement.

After preparing the chip carrier, the chip is placed on it. The SEM image of the tube is used to align the tube with respect to the holder. The total uncertainty of the alignment of the tube with respect to the magnetic field is estimated to be ±5°. In order to connect the gold wire to the gold leads, we use indium dots. The indium dots\(^{15}\) are spherical indium particles with an average diameter of 0.25 mm. Indium dots are placed on the surface of electrodes and pressed enough to make them stick to the gold surface. Before the connection, the length of each wire is measured and an extra length of the gold wire is cut to make a good fit. The wires need to have a low tension in order to prevent disconnection upon cooling. For this we allow some excess
Figure 2.8. A chip carrier is shown. The central part of the holder is raised using square pieces of a glass slide. The gold wires soldered to the pins of the chip carrier are used to connect the pins of the holder to the electrodes on the chip (not shown). A carbon tape is used to mount samples onto the glass chips.

length to the gold wires. The gold wire is placed on top of the squeezed indium dot and another indium dot is placed over the wire. The top dots are pressed on to trap the wire between the dots. This way the wire is connected to the electrodes, and the connection is robust and can survive many cooling cycles. Connecting the gold wire to the gate electrode is done using a
Figure 2.9. Pictures show a chip mounted on a chip carrier (top view). (a) Gold wires are used to connect the electrodes on the chip to the pins of the chip carrier. Initially, gold wires are soldered to the pins and then connected to the leads using the indium dots. Contacts between the gate electrode and the gold wire are made by silver paste on the side of the chip. (b) A closer look of electrodes is given here. The indium dots, wires and the electrodes can be seen. In the inset, the markers and the trench are also visible.

different method. The doped silicon layer is used as the back gate. A connection is made to the doped layer from the sides of the chip, using a silver paste\textsuperscript{16}. A small amount of the paste is applied to the side of the chip. Before the paste dries, the wire is pressed onto the paste. This way, the wire is glued to the side of the chip. The paste dries within minutes. It provides the connection between the doped Si layer and the gold wires. After connecting all the gold wires to the electrodes, the sample is ready for transport measurements. Figure 2.9 shows a chip sitting on the chip carrier and close-up shots of the electrodes. Markers and trenches are also visible in these optical pictures.
2.6. Electrical measurements

To measure the electrical properties, a low-distortion function generator (DS-360)\textsuperscript{17} is used, which supplies a small (0.05-0.5 mV peak-to-peak AC) potential difference between the two electrodes: the source and the drain. The resulting current is amplified by a low-noise current amplifier (SR-570)\textsuperscript{17}. At the same time, the potential between source and drain is amplified by a low-noise voltage amplifier (SR-560)\textsuperscript{17}. The amplified signals are recorded by computer using a Data Acquisition (DAQ) card (PCI-MIO-16XE-10)\textsuperscript{18} through a shielded connector block (BNC-2110)\textsuperscript{18}. A computer program is coded in LabView and is used in order to do various measurements by controlling and changing different variables through a high-performance GPIB Interface card (PCI-GPIB)\textsuperscript{18}. The program can also acquire large arrays of data and perform some preliminary analysis in addition to controlling the function generator, the superconducting magnet etc. A high-voltage source (Keithley-2400)\textsuperscript{19} is used to change the potentials of the gate electrode and controlled by the same LabView program. Another parameter, which is controlled in my experiments is the magnetic field applied to the tube (parallel to the tube in most cases). A superconducting magnet is used to generate magnetic fields up to 9T. Through the LabView program, a current source (CS-4)\textsuperscript{20} is controlled which provides current to the magnet. Based on the applied current, the applied magnetic field can be calculated. Up to 9 T can be applied to the sample, which corresponds to 40.33 A of current in the superconducting solenoid (at 4.2 K). In order to change the current through the superconducting magnet, a constant current of 50 mA is applied to the solenoid’s persistent switch. As the persistent switch is driven out of the superconducting state, current can be injected into the magnet. Figure 2.10 shows a schematic of the experiment.
Figure 2.10. A diagram of the electrical circuit of the experimental setup. The function generator, DS360, is used to generate the bias voltage. The voltage bias consists of a DC and AC components. The AC component has an amplitude of 100-500 μV. All the signals applied to or collected from the sample are filtered by a set of filters including π-filter, silver paste filter and RC filter. The current flowing through the sample is measured by a current amplifier (SR-570). A voltage amplifier (SR-560) measures the potential drop across the sample. The sample sits in a superconducting solenoid. The output of the current amplifier and the voltage amplifier are acquired by the DAQ card. The same program also controls the signal source, the current source for the magnet, and the gate voltage source.

The bias potential usually has independent AC and DC components. The AC component is a sinusoidal signal, having a peak-to-peak amplitude between 50μV to 500μV. The DC component provides the off-set bias voltage. In order to determine the differential
conductance, an I-V curve is measured at a fixed value of the DC bias and a very small AC sweep (50\(\mu\)V to 500\(\mu\)V). The slope of the I-V curve, which is, by definition, the differential conductance of the sample at the specified DC bias potential, is calculated by numerically fitting the measured I-V curve to a straight line. Differential conductance and the average current are recorded as a function of different variables, such as the temperature, gate potential, off-set bias potential, and magnetic field.

A thermometer is placed close to the sample, in a socket that is exactly analogous to the sample socket. Cooling of the sample and of the thermometer is achieved through the signal wires.

**2.7. Low Temperatures Measurements**

To do low temperature measurements two different setups are used. The first one is a home-made \(^4\)He system that can be cooled down to \(~1.5\)K. The second one is a commercial \(^3\)He system, purchased from Janis Research Co., which provides temperatures down to 280 mK.

I designed the \(^4\)He probe (Fig. 2.11) in order to do quick test measurements. It includes a 5.5 foot long stainless steel inner tube. The signal wires placed in the inner tube carry electrical signals to the sample positioned at the bottom of this tube. The signal wires are made out of a Teflon-coated Stablohm twisted pair\(^{21}\), which is made of a highly resistive alloy that has a very low temperature conductance coefficient. To protect the wires from dangling and breaking, they reside in a Teflon tube, which is inset into the inner tube. The Teflon tube is surrounded by stainless steel wool to reduce the electromagnetic noise. A copper tail piece (Fig. 2.11, arrow#1) is welded to the end of the inner tube. This tail piece holds a socket (2), which is
used to hold chip carriers and a Cernox thermometer\textsuperscript{13} (3). Copper has a high thermal conductivity at low temperatures. The signal wires are wound around the copper tail piece in order to provide thermalization of the signal wires. Some silver paste is applied to the wound part of the wires to filter the electromagnetic noise. Note that during normal operation the probe is filled with helium gas that serves as a thermal exchange gas and ensures thermalization of all the parts. Four of the wires are later soldered to the thermometer and the other 8 wires are soldered to the socket. The socket holds the chip carrier during the experiments. After the sample is placed on the chip carrier and connected to the socket, a screening copper box (cylindrical) engulfs the copper tail piece with the thermometer and the socket. The purpose of this box is to screen the external electromagnetic noise and to have uniform temperature around the sample and the thermometer.

The other end of the inner tube is welded into the KF-25 “T” (4) which enables us to connect the system to the outside world through a vacuum valve (5), a vacuum gauge (6), and a hermetically sealed MIL-C 26482 electrical connector\textsuperscript{22}. Another “T” (7) is used to increase the number of access entries. The inner tube has radiation-screening rings (8) soldered to its outer surface.

These serve to prevent thermal radiation from penetrating to the cold part of the probe. In order to change the temperature we use two different approaches: (i) slow cooling by pumping, and (ii) using a heater placed on the copper screening box. The heater for this purpose is made of a Stablohm wire having a 200 $\Omega$ resistance and wound around the copper box. These wires are used to heat the box and thus to increase the temperature of the sample. When a 90 mA current is applied to the heater, the temperature increases to \( \sim 20\text{K} \). Another 5.5 foot long stainless steel tube, which we call the “outer tube” (Fig. 2.11b), is used as an outer
shell for the inner tube with the copper tail piece, the socket, and the thermometer. This outer
tube, which serves as a Faraday cage for the sample and the signal wires and provides low
pressure environment for these parts, has the inner diameter of 0.93 inches and the outer
diameter of 1 inch. A copper disk (9) is welded to the bottom end of the outer tube and provides
a hermetically sealed bottom. The top end is welded to a KF-25 connector (10). The KF-25
connector enables us to connect it to the KF-25 ‘T’ (4) welded to the other end of the inner
tube. The outer tube has a KF-40 adaptor (11), sliding along its outer surface, which allows us
to place the tube in a Dewar and then seal the Dewar hermetically.

An aluminum box with 16 BNC connectors (12) and a MIL-C type MS3116F14-19S
electrical connector\textsuperscript{6} lets us connect measurement devices to the sample. The MS3116F14-19S
is compatible with the DT-1-H-14-19-P connecter. It is welded to the KF-25 flange using a
silver hard solder. This flange is connected to the ‘T’ and sealed with an O-ring. The Al box
has pi filters, placed to reduce the EM noise. Some switches (13) are placed on the box to
enable the grounding of all leads, as desired while mounting the sample or connecting electrical
cables.

Using the \(^4\)He probe is very easy and fast. First, the chip carrier is connected to the
socket with all the leads being grounded, and the copper screening box is placed over the
sample and the thermometer. Later, the inset is inserted into the stainless steel tube. Air is
pumped out of the probe using a rotary pump. Later, some He gas is introduced into the system
to have \(~5\) psi pressure inside. This amount of gas is sufficient enough to provide thermal
equilibrium with the outer shell and the sample, and to cool down the whole system. Later the
system is pre-cooled with liquid nitrogen and then inserted into a transport Dewar with liquid
helium. The sample temperature drops to 4.2K quickly after the bottom of the stainless steel
Figure 2.11. Major components of the He-4 Probe are shown in the picture. (a) The inner tube, which carries signal wires to the sample. The tube has rings (8) on it to reduce thermal radiation that might heat up the sample. The top insert shows the top part of the probe: The vacuum gauge (6), the vacuum valve (5), KF-25 “T”s (4, 7) and the connector box are shown. The connector box is made out of aluminum and has electrical filters in it to filter the EM noise. The BNC connectors (12) are used to connect low-noise voltmeters and other electrical devices. The box has an electrical connector that is compatible with the hermetically sealed connector attached to the KF-25 “T”. The second inset magnifies the bottom of the probe. It shows the Cu tail piece (1) with a socket (2) and the thermometer (3) connected to it. A copper box (not shown) covers the bottom of the probe. Some heater wires are wound around
the Cu box and used as a heater. The heater connectors can be seen in the picture. The wires are wound around the tail piece before they are soldered to the thermometer and the socket. The wires are covered with silver paste to filter thermal and electrical noise. (b) The outer shell is shown. It has a KF-25 adaptor (10) at the end, used to connect it to the KF-25 “T” welded on to the inner tube. The bottom is closed with copper disk (9). A sliding KF-40 adaptor (11) is also shown and it is used to hermetically connect the probe to the transport He Dewar.

comes into contact with helium. The temperature can be dropped further by pumping on the transport Dewar. By using this method, the temperature can be decreased down to ~1.5K. The temperature of the system is controlled either by changing the depth of insertion of the probe into the Dewar or by applying an electrical current to the heater.

The second setup is a $^3$He system, which has three major parts: (i) Dewar, (ii) superconducting solenoid, and (iii) $^3$He cryostat. The Dewar (Precision Cryogenic Systems, Inc.\cite{23}) is a LHe Superinsulated Dewar. The magnet is provided by Cryomagnetics, Inc.\cite{20}. The He-3 (Fig. 2.12) cryostat is supplied by Janis Research Co.\cite{7}. The cryostat has the following major parts, a sorption pump, a 1K-pot, and a $^3$He pot. This probe has two identical sockets: one for the sample and one for a Ruthenium Oxide Resistor thermometer\cite{13}. The sockets are connected to the $^3$He pot through signal wires, which are wound around a tail piece connected to the $^3$He pot. Thus, the thermometer and the sample are both cooled through signal wires and are both positioned equally with respect to the $^3$He pot and other cold parts. Under these conditions, the thermometer and the sample are expected to have the same temperature. We also note that the thermometer can be placed into any of the available sockets. In all cases, the lowest temperature measured was 280 mK. The two available sockets have different directions
Figure 2.12. Pictures of the $^3$He cryostat. (a) The top part of the insert. Some control valves and connectors can be seen. (b) The sockets with chip carriers inserted into them are visible. One of them is used to do electrical measurements on a sample, the other is used to measure the temperature using a Ruthenium Oxide resistor thermometer. Depending upon which socket is used, the magnetic field can be parallel or perpendicular to the surface of the sample. (c) Sorption pump, 1K pot and $^3$He pot. The sorption pump releases gas as it is heated up. The pump is filled with charcoal, which absorbs helium when it is cold and releases helium when it is heated to about 45K. The released gas is condensed into $^3$He pot. (d) The shells of the probe are put on the probe after mounting chip carriers and thermometers into the sockets.

of orientation so the chip surface can be parallel or perpendicular to the applied magnetic field, which is vertically directed.
Using this setup, the sample temperature can be dropped to 280 mK. During the operation the $^3\text{He}$ is condensed and then $^3\text{He}$ vapor is pumped out from the $^3\text{He}$ pot to decrease the temperature of the liquid $^3\text{He}$. During the $^3\text{He}$ condensing step the 1K pot is kept below 1.8K by pumping $^4\text{He}$ out of it while restricting the $^4\text{He}$ flow into it. At the same time, heating the sorption pump to 45 K releases $^3\text{He}$ gas absorbed in the charcoal inside the sorption pump. Applying nearly 5V through the sorption heater with 25 Ohm heater resistor is enough to raise the temperature of the sorption pump to 40 K. The 1K pot condenses the $^3\text{He}$ gas released by the sorption pump, and the condensed liquid $^3\text{He}$ is collected in the $^3\text{He}$ Pot. After condensing liquid $^3\text{He}$ for about 1 hour, the temperature of the sorption pump is reduced to 4.2 K. The $^3\text{He}$ vapor is pumped off by the sorption pump. This reduces the vapor pressure of the $^3\text{He}$ vapor and, to restore the equilibrium pressure, liquid $^3\text{He}$ vaporizes. As the highest energy atoms vaporize more easily, the remaining liquid is cooler.

2.8. References


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31
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Chapter 3

Coulomb Blockade Physics

3.1. Introduction

Coulomb islands are small metallic particles separated from the environment with tunneling junctions. Since they are small, the charging energy ($E_C = e^2/2C$, where $C$ is total capacitance of the island) can be much larger than the thermal fluctuation energy $k_B T$; where $T$ is temperature, $k_B$ is Boltzmann’s constant. Coulomb repulsion between electrons makes the charging of the island difficult. On the other hand, in order to pass a current through the island, it is necessary to increase the charge of the island at least by one electron. To understand how Coulomb islands work, let us consider two tunnel junctions to a small island, and for simplicity let us ignore the discreteness of the energy levels. Figure 3.1 shows the schematic of such a device. This kind of system is called a single electron tunneling (SET) transistor. At zero temperature, putting an electron to the island is blocked because of high charging energy\(^1\). Therefore the current does not flow. Blockage of the current is called Coulomb blockade. However, by increasing the potential difference between these two junctions, the Coulomb blockade of the current can be lifted. Another way to suppress the blockade is to change the potential of the island by applying a voltage to the gate electrode. To obtain conductivity certain conditions should be satisfied otherwise the conductivity is suppressed.

Since the island is isolated from the surrounding, the number of the electrons on the island is an integer number. On the other hand, the electric potential of the island can be reduced or increased continuously via changing the gate potential. As a result, the equilibrium charge changes continuously\(^2,3,4,5\). However the island charge is discrete since it may have only
an integer number of electrons. Changing the gate potential makes the existing charging state unstable, and as the gate potential is changed further, one electron is forced to move out of or into the island depending on the gate potential change. When another charging state has a lower energy, the tunneling occurs and the second configuration is obtained. A current through the island appears when two charging states are degenerate, i.e. have the same energy, \( E_n = E_{n+1} \). In this case an electron can jump in and out without changing the energy. If the gate potential is kept at this specific value, when \( E_n = E_{n+1} \), the electrons will continue to jump on and off the island repeatedly, and a very small potential difference applied between the source and drain (called bias voltage) produces a net current flow. If the gate potential is increased further, the degeneracy of the two charging states will be lifted and the current will stop. However, by changing the gate potential further, the current will flow at a new degeneracy point where the following condition is true \( E_{n+1} = E_{n+2} \). Again, at this value of the gate potential a current peak will emerge. Thus, ramping the gate potential will result in a set of conductance peaks. Figure 3.2 illustrates how the gate potential produces conductance peaks. Figure 3.2a shows conductivity peaks for 0 bias potential. Figure 3.2b shows IV curves for different gate potentials.

Plotting the conductance as a function bias potential and gate potential reveals the diamond shape regions where the electrons do not have enough energy to charge the island in a cyclic process. These regions are called Coulomb blockade diamonds (Figure 3.3). The size of the diamonds depends on the energy required to charge the island. More detailed discussions of these processes will be given below.

The energy required to charge the island has two components. One of them is the charging energy, which is the energy change of the system when the charge of the island
Figure 3.1. The schematic circuit of an SET transistor. The coulomb island shown as a dashed box is isolated from the surrounding by tunneling junctions. A box at a junction represents tunneling is allowed at the specific location. At regular capacitors, at gate capacitor in our case, no tunneling is allowed. $U_b$ is the bias potential between source and drain electrodes, and $U_g$ is gate potential applied to the gate electrode. $V$, $C$, and $Q$ represent the potential drop, capacitance and charge of the corresponding junction. Loop 1 is used to show one of Kirchhoff’s loops. Path 1 and Path 2 are two paths used to bring the island to the equilibrium after charging events. More details on this issue will be in the following section 3.2.
changes by one electronic charge and the new electron is placed into the same orbital state as
the previously placed electron. Basically, the charging energy of the island is the energy that is
required to overcome the repulsion of the other electrons when a new electron is introduced. It
is equal to $E_c = e^2/2C$ where $C$ is the total geometric capacitance of the island. The other
contribution to the total energy change is the kinetic energy of the orbital state on which the
new electron is placed. Note that in a high magnetic field the Zeeman energy is also a
contributing factor.

A detailed circuit diagram for our SET transistor is given in Fig. 3.1. The Coulomb
island is separated by two tunneling junctions $C_1$, and $C_2$ from Source and drain electrodes. The
island is coupled to the gate electrode with a capacitance of $C_g$. Box-like representation on the
capacitors shows tunneling is allowed through $C_1$ and $C_2$ unlike $C_g$. $U_g$ is the gate potential used
to change the potential energy of the Coulomb island. $U_b$ is the bias potential used to induce
potential difference between the source and the drain electrodes. $V_1$, $V_2$, $V_g$ are the potential
drop at drain, source, and gate electrodes respectively. $Q_1$, $Q_2$, $Q_g$ are the accumulated charges
at the capacitors between island and drain, island and source, and island and gate electrodes
respectively.

In our measurements we use a single nanotube as Coulomb islands of a single-electron-
tunneling (SET) transistor. The operation of the SET transistor can be understood from the so-
called Orthodox theory of single-electron tunneling, by Likharev and Averin\textsuperscript{6}. In this section, I
will study the Coulomb Blockade event. Coulomb blockade diamonds will be determined using
Orthodox Theory. Orthodox theory requires the change of free energy when an electron tunnels
to calculate the current at a junction. The change in free energy will be calculated using two
different methods. These two methods are equivalent; they are simply two different ways to
Figure 3.2 IV curves and conductivity oscillations with gate potential of an SET transistor. (a) IV curve of an SET transistor for different gate potentials. The graphs are shifted vertically in order to provide clarity. The conductivity is blocked when the charging state is not degenerate. As gate potential is ramped, slowly the ground state will become degenerate with another state. Thus the conductivity is restored. If the gate potential changed further, the conductivity will be blocked again. These events are illustrated from bottom to top. The stepwise increase in the currents is due to two different contact resistances at the junctions. This will be explained later in section Coulomb Staircase. (b) When the island has a well defined number of electrons, the conductivity is blocked. The gate potential can lift the Coulomb Blockade by causing degeneracy in the ground state so the number of electrons on the island is not well defined. As the gate potential is ramped, the ground state will become
degenerate with another state and the conductivity will be restored. Later, degeneracy will be lifted, however degeneracy will be revealed again as the gate potential is increased. (from ref. 5)

solve the same problem. In addition, other results related to the peak height, peak width and peak minimum will be given and will be used to analyze our experimental data.

3.2. Orthodox theory

The tunneling rate through a junction can be calculated using Orthodox Theory. Pekola et al. derived the tunneling rate \( \Gamma = \frac{e^2 R_T}{(1 - e^{-\Delta F/k_B T})} \) where \( T \) is temperature, \( k_B \) is Boltzmann’s constant, and \( R_T \) is tunnel resistance. \( \Delta F \) is the free energy change before and after the tunneling event.

The current through the junction will depend on probability of finding \( n \) extra electrons on the junction. Thus the net electrical current can be formulated as \( I_n = e \sum_{n=-\infty}^{\infty} \sigma(n) \left[ \Gamma_1^+(n) - \Gamma_1^-(n) \right] \), which involves not only the forward tunneling event but also the backward tunneling events through a junction. In this formula, \( \sigma(n) \) represents the probability of finding \( n \) extra electrons on the island. The function \( \Gamma_1^+(n) \) shows the forward tunneling rate through the 1st junction, for a given number of electrons, \( n \), whereas \( \Gamma_1^-(n) \) shows the backward tunneling rate through the 1st junction when the island has \( n \) electrons. For the dynamic equilibrium case, the current through the 1st and the 2nd junctions are equal. Otherwise, a net charge will accumulate in the
island. A similar equation can be written down for the second junction also. To find the probability of finding \( n \) electrons on the island, the “Master” equation can be used:

\[
\left[ \Gamma_1^+ (n-1) + \Gamma_2^- (n-1) \right] \sigma(n-1) - \left[ \Gamma_1^+ (n) + \Gamma_1^- (n) + \Gamma_2^+ (n) + \Gamma_2^- (n) \right] \sigma(n) \\
+ \left[ \Gamma_1^- (n+1) + \Gamma_2^+ (n+1) \right] \sigma(n+1) = 0
\]

3.3. Free energy change

According to the Orthodox theory, the tunneling rate is determined by the free energy change, \( \Delta F \), when the tunneling occurs. The following two sections are dedicated to showing two ways to find the \( \Delta F \): (i) the potentials of the electrodes and (ii) the energy stored in capacitors. The first method uses the potential energy change of the electron as it tunnels from one electrode to the other one to calculate the \( \Delta F \). The second method uses the energy stored in each capacitor and the work done during the tunneling process to calculate the energy stored in each capacitances to calculate the \( \Delta F \).

3.3.1 Potentials of electrodes

This subsection uses the potential energies of each electrode to calculate the free energy change as an electron tunnels. \( \Delta F \) can be formulated in the following way using the average potential of the electrode where the electron resides before tunneling and the average potential of the electrode where the electrode resides after tunneling:

\[
\Delta F = e \left[ \frac{(\Phi_2 + \Phi_2')}{2} - \frac{(\Phi_1 + \Phi_1')}{2} \right],
\]

where \( \Phi_1, \Phi_1' \) are the initial and final potentials of the electrodes that the electron leaves, and \( \Phi_2, \Phi_2' \) are the initial and final potentials of the electrode to which the electron tunnels, respectively.
To find the $\Delta F$ we need to determine the initial and final potentials of the electrodes where the electrodes reside. The problem is solved for figure 3.1. From this figure, it is clear that the source electrode has a potential of $U_b$, a bias potential, all the time. The drain electrode is defined as ground, so its potential is $0V$. The potential of the island can be calculated by using Kirchhoff’s laws.

Loop 1 gives: $U_b - U_g = V_g + V_1$, and the loop around the circuit gives $U_b = V_1 + V_2$, where $U_g$ is applied gate potential, and $V_1$, $V_2$, and $V_g$ are the potential drop at first junction, second junction and gate electrode, respectively. The charge on the island is $ne = -C_1V_1 + C_2V_2 + C_gV_g$, where the $C_1$, $C_2$ and $C_g$ are the capacitance of first electrode, second electrode and gate electrode, respectively. Solving these three equations will lead to

$$V_1 = \frac{-ne + (C_2 + C_g)U_b - C_gU_g}{C_1 + C_2 + C_g}, \quad V_2 = \frac{ne + C_1U_b + C_gU_g}{C_1 + C_2 + C_g}, \quad \text{and} \quad V_g = \frac{ne + C_1U_b - (C_1 + C_2)U_g}{C_1 + C_2 + C_g}.$$

Thus the potential of the island equals $\Phi_{\text{island}}(n) = U_b - V_1 = V_2$. Remember that the potentials of source and drain are equal to $\Phi_{\text{source}} = U_b$ and $\Phi_{\text{drain}} = 0$ respectively. Now we can calculate the change in the free energy.

The change in the free energy when the electron tunnel from the source electrode to the island, which initially has $n$ electrons, $\Delta F^+_1(n)$, is:

$$\Delta F^+_1(n) = e \left[ \frac{\Phi_{\text{island}}(n + 1) + \Phi_{\text{island}}(n)}{2} - \frac{\Phi_{\text{source}} + \Phi_{\text{source}}}{2} \right] = e \left[ \frac{(2n + 1)e/2 + C_1U_b + C_gU_g}{C_1 + C_2 + C_g} - U_b \right].$$

Similarly, when an electron tunnels from the island to the source electrode, the free energy change $\Delta F^-_1(n)$ is
\[ \Delta F^{-}_1(n) = e^{\left[ \frac{\Phi_{\text{source}} + \Phi_{\text{source}}}{2} - \frac{\Phi_{\text{island}}(n) + \Phi_{\text{island}}(n-1)}{2} \right]} = e^{\left[ \frac{U_b}{2} - \frac{(2n-1)e / 2 + C_1 U_b + C_g U_g}{C_1 + C_2 + C_g} \right]}. \]

In a similar fashion, the change in the free energy, \( \Delta F^{+}_2(n) (\Delta F^{-}_2(n)) \), corresponding to events when an electron tunnels from (to) the island to (from) the drain electrode is

\[ \Delta F^{+}_2(n) = e^{\left[ \frac{\Phi_{\text{drain}} + \Phi_{\text{drain}}}{2} - \frac{\Phi_{\text{island}}(n) + \Phi_{\text{island}}(n-1)}{2} \right]} = e^{\left[ \frac{0 - (2n-1)e / 2 + C_1 U_b + C_g U_g}{C_1 + C_2 + C_g} \right]}, \]

\[ (\Delta F^{-}_2(n) = e^{\left[ \frac{\Phi_{\text{island}}(n+1) + \Phi_{\text{island}}(n)}{2} - \frac{\Phi_{\text{drain}} + \Phi_{\text{drain}}}{2} \right]} = e^{\left[ \frac{(2n+1)e / 2 + C_1 U_b + C_g U_g}{C_1 + C_2 + C_g} - 0 \right]}. \]

### 3.3.2 Energy stored in capacitors

This subsection uses the energy stored on each capacitor in addition to the work done during the tunneling event. This method calculates the energy difference in each of the three capacitors, and then adds the work done for reconfiguring the system.

Figure 3.1 is used to solve the electrostatic equation used to calculate the energies and work. Two of the three capacitors are the ones between the island and the source electrode, the island and drain electrode, with capacitances of \( C_1 \) and \( C_2 \) respectively, the electrons tunnel through these capacitors. The third one is between the gate electrode and the island.

The energy stored in each capacitance is \( E_i = \frac{C_i V_i^2}{2} \) where \( i \) is 1, 2 or g, referring to the first capacitor, second capacitor or gate capacitor, respectively. The capacitances are constant but the potential drops at the capacitors are variable.

The potential drops at junctions can be calculated using Kirchhoff’s laws, as calculated before. Here, I will copy the results for the potential drops; the details can be found in section 3.3.1:
The total energy stored in the capacitors is

\[ E_n = \frac{C_1 V_1^2 + C_2 V_2^2 + C_g V_g^2}{2} \]

Note that this expression gives the total energy of the system after ignoring any work done on the system. As the energy change between two configurations will be studied, it is not important to know the previously done work but only the work required to redistribute the charges after the tunneling event. Note that \( V_g \) and \( U_g \) are different and we need \( V_g \). The gate voltage \( U_g \) is equal to the voltage on the voltage source connected to the gate electrode. The voltage \( V_g = \frac{Q_g}{C_g} \) represents the voltage drop present on the gate capacitor. As our \( U_g \) is very large (up to 50 V) compared to the bias voltage and the charging energy, the voltage \( V_g \) is approximately equal to \( U_g \). On the other hand, as \( C_g \) is small, the charge induced on the island due to the effect of the gate electrode is small (a few electronic charges).

To understand the necessity of redistribution of charges in the system, let us consider a process in which one additional electron is added to the island through the first tunnel junction. Using the calculated \( V_1 \), \( V_2 \), and \( V_g \), we can find that the change in the amount of the charge on each capacitor by using the relationship between potential and charge for a given capacitor:

\[ \Delta Q_i = C_i \Delta V_i = e \frac{C_i}{C_1 + C_2 + C_g}, \text{ where } i \text{ is } 1, 2 \text{ or } g. \]

However, immediately after the tunneling process, the system is not yet in equilibrium because the first capacitor is charged by one unit.
change, and the rest is not affected. As can be seen from the previous equations, the charges of all the capacitances should change partially for an equilibrium case. So, some amount of charge has to be redistributed between the capacitors in order to reach the equilibrium charge distribution. During this equilibration process, some work \( W \) should be done by or against the potential sources. This work also contributes to the total change in energy of the system. The free energy change of the system is \( \Delta F(n) = E_{n+1} - E_n + W \).

In our case, as the tunneling takes place at the 1st junction, the charge on the second capacitor and the gate capacitor will change by the redistribution process. On the island, the charge can be redistributed to satisfy the equilibrium conditions without any work done, as no potential source exists on the island. However, in order to bring the charges to the negative terminal of the second electrode and gate electrode, some work should be done. The charge increases by \( \Delta Q_g = \frac{eC_g}{C_1 + C_2 + C_g} \) for the gate capacitor and by \( \Delta Q_2 = \frac{eC_2}{C_1 + C_2 + C_g} \) on the second junction. The negative sides of the capacitors require \(-\Delta Q_g\) and \(-\Delta Q_2\). Both \( U_b \) and \( U_g \) do work on \(-\Delta Q_g\), as \(-\Delta Q_g\) passes through the both potential sources as the charge is carried from the positive terminal of the 1st junction to the negative terminal of the gate capacitor (path 1 in Fig. 3.1). On the other hand, only \( U_b \) does work on \(-\Delta Q_2\) since \(-\Delta Q_2\) passes only through the bias potential source as the charge is carried from the positive terminal of the 1st junction to the negative terminal of the 2nd junction (path 2 in Fig. 3.1). Finally, the work done by the potential sources is \( W_{n1} = -\Delta Q_2 U_b - \Delta Q_g (U_b - U_g ) \) (“+1” refers to charging through the 1st junction.). So the total work done is calculated as: \( W_{n1} = -\frac{eC_2}{C_1 + C_2 + C_g} U_b - \frac{eC_g}{C_1 + C_2 + C_g} (U_b - U_g ) \).
Finally, the free energy change is found to be \( \Delta F_i^+ (n) = E_{n+1} - E_n + W_i \), which is equal to

\[
\Delta F_i^+ (n) = E_{n+1} - E_n + W_i = \frac{e^2 (2n + 1)}{2(C_1 + C_2 + C_g)} - \frac{e C_2}{C_1 + C_2 + C_g} U_b - \frac{e C_g}{C_1 + C_2 + C_g} (U_b - U_g)
\]

to

\[
= \frac{e (2ne + e - 2(C_2 + C_g) U_b + 2 C_g U_g)}{2(C_1 + C_2 + C_g)}
\]

(The index \(n\) stands for the number of electrons initially on the island and +1 represents an electron tunneling onto the island through the 1\textsuperscript{st} junction). Repeating same steps for different events, such as discharging through the 1\textsuperscript{st} junction or charging or discharging through the 2\textsuperscript{nd} junction, gives the result calculated in section 3.3.1.

### 3.4. Coulomb diamonds

Tunneling rate \( \Gamma \) is given as \( \Gamma = (e^2 R_T)^{-1} \frac{-\Delta F}{1 - e^{-\Delta F / k_B T}} \) in the previous section. At 0K, in order to have any tunneling take place, the free energy change, \( \Delta F \), should be negative. Otherwise the tunneling is forbidden and the current could be blocked. Following this understanding, 4 different constraints can be obtained for a Coulomb island using the calculated free energies to be stable with \( n \) electrons; i.e. the current is blocked:

\[
\Delta F_i^+ (n) > 0 \Rightarrow (C_2 + C_g) U_b - C_g U_g > (2n + 1)e / 2
\]

\[
\Delta F_i^- (n) > 0 \Rightarrow (2n - 1)e / 2 > (C_2 + C_g) U_b - C_g U_g
\]

\[
\Delta F_2^+ (n) > 0 \Rightarrow (2n - 1)e / 2 > -C_g U_b - C_g U_g
\]

\[
\Delta F_2^- (n) > 0 \Rightarrow -C_g U_b - C_g U_g > (2n + 1)e / 2
\]

These 4 constraints produce a diamond-shaped region defining the stable, non-conducting range of potentials for \( n \) electron on the island. Similar regions can be obtained for
**Figure 3.3.** Coulomb diamonds. The conductivity is blocked in the diamond shape regions because of Coulomb interaction. In these regions the number of the electrons is well defined as shown (i.e. $n=0$, $n=1$). Slopes of the lines defining the borders will be calculated in section Coulomb Diamonds (from ref. 5).

Different numbers of electrons located on the island. Figure 3.3 shows Coulomb diamonds formed by these 4 constrains. If the island’s quantum levels are not quantized, as assumed during the present calculation, the diamond-shaped regions are identical and monotonic repetitions of the same structure. When the quantization of the island is considered, instead of only potential energies of the electrodes being used to calculate the free energy change, kinetic energy changes should also be considered. As a result of the change in the kinetic energy, a quasi-periodic diamond structure reveals itself, depending on the energy level spacing and degeneracy of the quantized energies.
3.5. Coulomb staircase

To simplify the problem, let us ignore the effect of the gate electrode (its potential and capacitance). To simplify the case further, we ignore the backward leaking of the electrons, and only consider the electron flows in one direction. Modifying the previously derived free energy changes $\Delta F_s$, the equations give us the following conditions under which tunneling events can happen. Note that the neutrality of the island is also assumed:

$$\Delta F^+_1(n) < 0 \Rightarrow C_2 U_b < e/2,$$
$$\Delta F^+_2(n) < 0 \Rightarrow C_1 U_b < e/2.$$

If the capacitances of the junctions are taken to be equal then one junction starts charging the island at the bias voltage that the other junction starts discharging. Even though they have the same capacitances ($C_1=C_2=C$), they can have different junction resistances ($R_1<<R_2$), so they might charge the island at different frequencies (the tunneling rate $\Gamma$ depends on $R$ and $\Delta F$ thus $C$). As a result, one side of the junction can charge/discharge the island faster than the other one discharges/charges. However, at dynamic equilibrium, which is always assumed, the current through the first junction is equal to the current through the second junction. Otherwise, an accumulation of the charge on the island would have to occur. Thus, the slower junction acts as bottleneck and controls the frequency of the charging events. Initially, there will not be any current through the island. As the voltage becomes bigger than $e/2C$, current starts to flow. If the bottleneck is the drain junction then the island stays charged longer. The electrons cannot charge the island until an electron tunnels off the island through the drain junction. The island can be charged again after the extra charge leaves the island through the drain junction. The drain electrode controls the current. As the potential is further increased, a second electron will have enough energy to charge the island before the first
electron leaks through the drain. The conductivity will be increased because now two electrons contribute to the conductivity. This shows up as a stepwise increase in the IV curve. The Coulomb Staircase was theoretically calculated for $R_1=10R_2$ and $C_1=C_2=C$; the resultant IV curves are shown in Fig. 3.2a.

A similar case is valid if the capacitances are different. For the case $C_1<<C_2$ and $R_1=R_2=R$, the tunneling rates for the junctions will be different. For this specific case, the source charges faster than the drain discharges. Again, the drain becomes the bottleneck for charging events. When the bias voltage is smaller than $e/2C_2$, there will not be any current. As the bias voltage gets larger than $e/2C_2$, the one electron from the source will tunnel into the island. Eventually, the extra electron will leak into the drain. As soon as the extra electron tunnels to the drain, another electron from the source will tunnel onto the island. As a result, current starts to flow. As the bias voltage is increased further and reaches $3e/2C_2$, the island can become charged with two electrons at a time. As a result, a step will be observed in the IV curve. Increasing the bias potential further, more and more steps are observed.

Consequently, considering junctions with different parameters, the tunneling rates for them will be different. The tunneling rate of a junction depends on the capacitance of the junction and the resistance across it. It is nearly impossible to obtain two junctions having the same parameters. As a result, one of the junctions will charge/discharge the island faster than the other one discharges/charges it. As a result, there will be steps in the IV curve.

3.6. Amplitude and shape of conductance peaks as a function of gate potential

So far, all the derived equations are enough to understand the frequency of the conductivity peaks as a function of gate potential. Understanding the shape of the peaks
requires more rigorous work, which is beyond the scope of my research topic. In this section, I will summarize some of the previously published results.

Thermal activation energy, \( k_B T \), is a dominant factor defining the shape of the peaks. As the thermal activation energy becomes comparable to or less than the charging energy, \( e^2/C \), the Coulomb blockade phenomenon becomes observable. This regime is called the weak Coulomb Blockade regime. The maxima (\( G_{\text{max}} \)) and the minima (\( G_{\text{min}} \)) of the coulomb blockade oscillation have very similar values. The minima are calculated by Beenakker\(^1\) as:

\[
G_{\text{min}} \propto e^{\frac{\Delta E + e^2/C}{2kT}}
\]

If the thermal activation energy becomes less than the charging energy but larger than the average energy level spacing, \( \Delta E < kT < e^2/C \), this condition is called the classical Coulomb-blockade regime. The average number of levels contributing to the conductance, \( kT/\Delta E \), decreases as the temperature decreases. On the other hand, the tunneling rate through a single level is proportional to \( 1/T \), which means the conductivity of a single level increases as temperature decreases. These two factors cancel the effects of each other, and finally, in this regime, the maximum height of the conductance peak becomes temperature independent. The conductivity maxima are given by:

\[
G_{\text{max}} = \frac{e^2}{2\Delta E} \frac{\Gamma'^{\prime} \Gamma^{\prime}}{\Gamma'^{\prime} + \Gamma^{\prime}}
\]

where \( \Gamma \) is the tunneling rate of the left and right junction. The overall shape of the conductance vs. gate voltage curve is given by:

\[
G = \frac{e^2}{2\Delta E} \frac{\Gamma'^{\prime} \Gamma^{\prime}}{\Gamma'^{\prime} + \Gamma^{\prime}} \frac{\alpha \Delta V_g / kT}{\sinh(\alpha \Delta V_g / kT)} = \frac{e^2}{2\Delta E} \frac{\Gamma'^{\prime} \Gamma^{\prime}}{\Gamma'^{\prime} + \Gamma^{\prime}} \cosh^{-2} \left( \frac{\alpha \Delta V_g / 2.5kT}{2} \right).
\]

In the resonant tunneling regime, the thermal activation energy is smaller than the energy level spacing however larger than quantum fluctuation, \( h\Gamma << kT << \Delta E \). Similar to the previous case, the conductance through a single level has \( 1/T \) temperature dependence\(^5\). On the
Figure 3.4. Conductivity peak shapes and heights. (a) Conductivity peaks are shown as a function of gate potential in two different regimes. The dashed curve shows a conductivity peak in the classical Coulomb blockade. The solid curve shows the conductivity peak in the resonant tunneling regime. Graphs are normalized with peak height. (b) Peak maxima (solid) and peak minima (dashed) as a function of temperature is shown for $\Delta E=0.01E_C$. The conductivity initially drops (the weak Coulomb blockade), and later stabilizes (the classical Coulomb blockade), and starts to increase (resonant tunneling) (from ref. 5).
contrary, the number of levels is fixed at a non-zero integer, depending on the degeneracy of the level as \( k_B T / \Delta E \to 0 \). Finally, the overall shape \( (G) \) and peak height \( (G_{\text{max}}) \) are calculated as:

\[
G = \frac{e^2}{2kT} \frac{\Gamma' \Gamma''}{\Gamma' + \Gamma''} \cosh^2 \left( \frac{\alpha \Delta V_g}{2kT} \right) \quad \text{and} \quad G_{\text{max}} = \frac{e^2}{2kT} \frac{\Gamma' \Gamma''}{\Gamma' + \Gamma''}
\]

Figure 3.4a shows the conductivity peaks as a function of gate potential for resonant tunneling and classical Coulomb blockade regimes, and Figure 3.4b shows how the peak height and minima change with temperatures. As can be seen from figure 3.4a, the thermally broadened conductivity peaks have different shapes in the different regimes. Figure 3.4b shows that the conductivity peak drops as thermal fluctuations become comparable to the charging energy, \( E_C \), of the island. It becomes almost flat as the temperature drops further. As the temperature drops below the quantum energy level spacing, \( \Delta E \), of the island, the conductivity starts to increase. This argument holds when the distance where the electrons localize, the localization length, is comparable to or larger than the Coulomb island.

When thermal fluctuations are below quantum fluctuations, the conductance peaks are reduced by a factor of \( \Gamma / (\Gamma + \Gamma_{\text{in}}) \) and the width is increased by a factor \( (\Gamma + \Gamma_{\text{in}}) / \Gamma \), where \( \Gamma_{\text{in}} \) is the inelastic scattering rate.

3.7. Experiments

As a carbon nanotube is placed on the electrodes, naturally-formed tunneling barriers (possibly due to carbon contamination on the surface of the nanotube) isolate the tube from the electrodes. In this way a “Coulomb island” is formed. A schematic drawing of the system is shown in figure 3.1. Typically, we find many samples having the contact resistance, \( R_c \) (i.e. tube-electrode resistance) in the range comparable to or larger than the quantum resistance.
Figure 3.5. Conductivity of an SET transistor as a function of gate potential. At low temperatures, the conductivity is blocked because of Coulomb interactions. The gate potential can restore the conductivity of the island by changing the potential of the island. The gate potential results in aperiodic conductivity peaks, when the energy levels of the island are not ignorable. The peaks are separated by nearly 1.6V. The measurements are done at 0.3K.

\[ R_0 \equiv h/e^2 \approx 26 \text{k}\Omega \] . We find that \( R_c \) stays in the range between 10 kOhm and 300 kOhm for the majority of samples. Whenever the net contact resistance is larger than \( R_0 \), one expects the single-electron wave-functions to be localized on the tube, with a small overlap with the leads (i.e. electrodes). Thus, the electric charge on the tube is quantized. The conductivity is provided by electrons that pass through the tube on a one-by-one basis at 4K. Most of our samples show a Coulomb blockade effect at or below 4K.
Figure 3.6. IV and differential conductivity graphs of different SET transistors. (a) Two IV curves are shown for two different gate potentials. The red line shows the IV of a conductivity peak ($V_g = -5$ V). The blue line shows the IV curve at a valley ($V_g = -13$ V) where the conductivity is blocked. (b) The graph shows a stepwise increase in the current. The arrows indicate the steps. The steps for the negative potentials are separated more than the ones for the positive potentials. (c) The differential conductivity for a valley is shown for another sample. For negative potentials, the steps are more pronounced and separated than the positive ones.

A closely-positioned gate electrode (500 nm away from our nanotubes) can be used to change the potential of the tube. Although the Coulomb interactions block any increase or decrease in the number of electrons, which is required in order to have current flowing through the carbon nanotube, by changing the potential of the island, the carbon nanotube can be charged or discharged. As the gate potential is ramped up, the conductance is restored when degeneracy is induced between two different charging states, thus a conductance peak appears. Figure 3.5 shows the conductance as a function of the gate potential. For this sample, the conductance peaks are separated ~1.6V. The average spacing between two adjoining
Figure 3.7. The conductivity map as a function of bias potential and gate potential. The diamond-shaped regions show the regions where the conductance is blocked at 0 temperature. Using the border slopes and the sizes of the diamonds, many quantities of the SET transistor can be calculated. The size of the diamonds are almost constant, indicating that the quantum energy-level spacing is much smaller than the charging energy.

Conductance peaks varies between 0.5 V and 20 V for our samples. The peaks are separated from the adjoining ones with different potentials due to the quantum energy levels of the nanotube and spin degeneracy. On average, the gate capacitance, \( C_g \) is calculated for this nanotube as \( 1 \times 10^{-19} \) F.

Carbon nanotubes look like cylinders, so the geometric capacitance per unit length, \( C' = 2\pi\varepsilon / \ln(4D/d) \), between an infinitely long tube and a plane can be used to predict the gate capacitance of our tube. The quantity \( \varepsilon \) is the permittivity of environment (in our case the vacuum), \( D \) is the distance between the cylinder and the plane, and \( d \) is the diameter \( (D \gg d) \) of the tube. The capacitance of the tube is calculated as \( C_g = 1.5 \times 10^{-18} \) F for a length of 100 nm, and thus the Coulomb peaks are expected to have a period of 0.12 V. For most of our experiments, the potential difference between adjacent Coulomb peaks varies between 0.5 V
and 20 V, which is always greater than the theoretical prediction. This discrepancy might be due to the screening effect of the electrodes, since the electrodes are between the gate electrode and the tube, in addition to the applicability of the theory, which assumes an infinitely long tube and plane are infinitely large. Another way to lift the Coulomb blockade is to apply a large enough bias potential (up to $e/2C$). The required bias potential to lift the blockade depends on the gate potential. Figure 3.6a shows two IV curves for two gate potentials. The gate electrode can restore the conductivity by changing the potential of the island, thus causing degeneracy between two charging states. The red curve in the figure shows such a condition, however the blue curve shows that the conductance is blocked because there is no degeneracy. By increasing the bias potential more and more, the differential conductance shows a stepwise increase, as discussed in section 3.4 (Figs. 3.6b, c). Unlike the previously observed results, the steps are equally separated. The steps on one side (for negative bias potential) are separated further from each other than the ones on the other side (for the positive bias potential). This can be explained via different tunneling rates at the different junctions. Let us say that one of the junctions is much larger, and so is more strongly coupled, than the other one. The weakly-coupled junction will thus control the conductivity. If the tube is charged through the weakly coupled side, the island will stay discharged for longer, thus adding a quantum energy level to the conduction regime would result in a step in the current because the new level increases the probability of charging. If the tube is discharged through the weakly coupled side, the island will stay charged longer, thus adding a quantum energy level to the conduction regime, which will not result in a significant current change. When the island can host two extra electrons at a time, due to increase in potential difference of two electrodes, a step in the current will appear again. Thus, for one sign of the bias potential, the conductance will have steps with quantum
energy level spacing, for the other sign of the bias potential, the conductivity will have steps with the charging energy level spacing\(^8\). The charging energy is calculated as 2.8 mV and the quantum energy levels are calculated as 0.4 meV for our sample shown in Fig. 3.6b.

The conductance map as a function of gate potential and bias potential show diamond-shaped regions where the conductivity is low. Figure 3.7 shows such a map. Using this kind of map, the charging energy of the system is calculated to be 3 mV, which is in very good agreement with the prediction we obtain from the steps of the IV curve. The drain electrode is coupled to the tube with a capacitance of \(1.9 \times 10^{-17} \text{ F}\), and the source electrode is coupled with a capacitance of \(3.6 \times 10^{-17} \text{ F}\). Finally, the tube is coupled to the gate electrode with a capacitance of \(9.0 \times 10^{-20} \text{ F}\), which is very close to the result obtained using the differential conductivity as a function of gate potential shown in Fig. 3.5. The variance comes from assuming that the tube has a continuous spectrum, i.e. the spacing of the quantum energy levels can be neglected. The scaling factor \(\alpha\), which defines how strongly the gate capacitor couples to the tube, is found to be \(1.8 \times 10^{-3}\). The temperature dependence of our sample resembles figure 3.4b. A representation of our results for a different sample is shown in figure 3.8. Figure 3.8 shows how the conductivity peak-height changes as a function of temperature. Some, but not all of the conductivity peaks show this behavior. The rest of the conductivity peaks will decrease as the temperature decreases. As can be seen from figure 3.8, the conductivity peaks initially start to decrease with decreasing temperature. A further decrease in temperature results in a sudden rise of the conductivity, unlike the theoretical curve, in which the peak height stays almost constant over a range of temperatures, before rising again. This difference might arise from the fact that the theoretical curve is calculated for a charging energy that is 100 times the energy level splitting. The conductivity decreases until the thermal fluctuations are the same order of
Figure 3.8. The conductivity peak height as a function of temperature. The conductivity peak initially drops as the temperature drops, until a temperature of 0.5K, and then starts to increase as the temperature drops further. The fitting curve is a linear combination of fitting curves valid in different regimes including the weak Coulomb blockade, the classical Coulomb blockade or the resonant tunneling regimes. Each function is dominant in its own regimes.

magnitude as one tenth of the charging energy. The conductivity starts to increase when thermal fluctuations become comparable to the energy-level spacing. In our case, the charging energy is nearly 10 times the energy level-spitting, and thus the conductivity decreases with temperatures, down to a point where it should start to increase without any gap between these two temperatures. For our sample, all three regimes (i.e. weak Coulomb blockade, classical Coulomb blockade and resonant tunneling regimes) overlap each other and we observe all three phenomena within a very narrow range of temperatures. Thus, I fit the data with a linear combination of the three functions, as they are valid in different regimes. Considering that each fitting function dominates the others in their own regions, no problems should result. The
Figure 3.9. The conductivity peaks in different regimes. The conductivity peaks have different forms in the classical Coulomb blockade regime from that in the resonant tunneling regime. (a) The conductivity peak is fit by the function associated with the classical Coulomb blockade regime. The curve is measured at a temperature of 4.2K. (b) The conductivity peak is fit by the function valid for the resonant tunneling regime. The curve is measured at a temperature of 0.3K.

The Coulomb peaks have different shapes in the different regimes. Figure 3.9 shows the peaks of the same sample in two different regimes. Figure 3.9a shows the sample in the classical regime, and Fig. 3.9b shows it in the resonant tunneling regime. The fitting functions are chosen accordingly. The scaling factor, $\alpha = C_g/C$ (where $C$ is the total capacitance of the

charging energy is calculated as 0.15 meV by the fitting functions, which is calculated as 0.5 meV from the Coulomb diamonds and the IV curves. The energy level-spacing is calculated as a minimum of 0.05 meV.
(tube), is calculated as $2.4 \times 10^{-3}$ for the classical regime. Recall that the energy level-spacing is measured as 0.4 meV from IV curves, which is nearly equal to the thermal fluctuations at 4.2K. If the sample is assumed to be in the resonant tunneling regime, $\alpha$ could become $1.9 \times 10^{-3}$, which is much closer to the one calculated from the Coulomb diamonds. Thus, it is possible that Fig. 3.9a is another example of the resonant tunneling regime. The fitting parameter for $\alpha$ from Fig. 3.9b is calculated as $1.5 \times 10^{-3}$ for the fits of the resonant tunneling regime. From the Coulomb diamond size and shape, $\alpha$ is calculated as $1.8 \times 10^{-3}$ previously. Considering that the regimes are not well defined for our samples these results are in good agreement with their expected values.

3.8. Conclusion

We have made an SET transistor out of a single multiwall carbon nanotube. Our samples act as a Coulomb island at liquid helium temperatures. Electronic transport measurements show that the localization length is larger than the distance between electrodes, and that conduction occurs through single energy levels. The quantum energy level spacing is large enough to be observed during the experiments.

3.9. References


Chapter 4

Weak Coulomb Blockade (WCB) of Charge Transport in Multiwall Carbon Nanotubes used in Single-Electron-Tunneling Transistors (SET)

4.1. Abstract

The temperature and voltage dependence of the conductance of SET devices made out of multiwall carbon nanotubes is studied. To make an SET device we place a multiwall carbon nanotube over two gold electrodes separated by a ~100 nm wide gap. In many cases the tube forms a tunnel junction with each of the electrodes. The gate is provided by the substrate itself, which is doped. Thus an SET transistor is formed. At temperatures higher than the charging energy transport properties are determined by the phenomenon of WCB. Using expressions derived by Pekola et al. we are able to fit the experimental curves using the following four fitting parameters: conductance of each junction corresponding to the high temperature limits and capacitance of each junction. The fitting parameters extracted from fits of the temperature and voltage dependence of the conductance agree with each other. They also agree with the parameters extracted from the Coulomb diamonds measured at low temperatures. Thus we concluded that the operation of devices composed of a nanotubes connected to two electrodes is well explained by the theory of WCB.

4.2. Introduction

The WCB occurs in SET devices\(^1\), single tunnel junctions\(^2\), arrays of tunneling junctions\(^1\) and diffusive wires\(^3\) at temperatures higher than the charging energy. Thus WCB
occurs at temperatures high enough that such Coulomb oscillations are negligible. Thus one does not need to include the gate potential in the calculations. Yet, even at temperatures, $k_B T$, larger than the charging energy, $E_C$ the conductance of the devices listed above is suppressed due to the remaining WCB. The theory of WCB was developed in order to explain the operation of a primary thermometer, based on tunnel junctions. The full width at half minimum of the coulomb valley (Fig. 4.4) is found to scale with temperature and have a universal slope for all arrays of junctions with the same number of tunneling junctions. The theory also predicts that the normalized conductivity ($\gamma = G_t / (G_t - G)$) depends linearly on the temperature.

In my work, I studied the conductivity of a multiwall carbon nanotube forming a SET transistor with two gold electrodes as a function of temperature. The results are compared to theoretical calculations based on the Orthodox SET theory of Averin and Likharev. Calculation in the limit $E_C << k_B T$ gives a linear temperature dependence for the quantity $\gamma(T) \equiv G_T / (G_T - G(T))$ [ref. 5] where $G_T$ is the conductivity of the system at the high temperature limit at which the Coulomb blockade is suppressed. The slope is determined by the capacitance of a junction. Calculations of Pekola et al. are carried out in the first order approximation and give a linear dependence for the function $\gamma(T)$, which passes through the origin, without any offset. My experiments confirm the linear dependence but show that a nonzero offset is present for some of the samples. An offset is predicted by the theory developed by Golubev and Zaikin (GZ) which has the same order of magnitude as the experimentally measured offset. Unlike Pekola et al. theory, GZ is done up to the second order of corrections The fitting parameters are found to be consistent with the ones we obtain from Coulomb diamonds which were measured at much lower
temperatures. Our data shows that the Coulomb blockade starts to effect the conductivity below 100K for our samples.

4.3. Experiment

The samples are prepared using a multilayer Si chip (Fig. 4.1a). The top surface of the chip is a 60 nm SiN film with a trench in the middle of the surface. The trench is used to create an interruption in the Au film deposited over the surface of the chip. The doped layer present in the Si wafer is used as a gate electrode of the single electron tunneling transistor. SiN sits on a 500 nm thick SiO$_2$ layer, which helps to reduce any electrical leakage from the Au electrodes and gate electrode. The tubes are suspended in 1,2-Dichloroethane and sonicated for 5 minutes. The solution is dripped onto the surface of the chip with Au electrodes. After 1 minute, the solution is blow-dried. We locate the tubes connecting two sides of the trench using a scanning electron microscope (Hitachi S-4700). A sample micrograph is shown in Fig. 4.1b. The electrodes are defined in such a way that only one tube connects the resulting source and drain electrodes.

For transport measurements, two different cryostats, $^4$He and $^3$He cryostats, are used. The $^4$He cryostat provides temperatures down to 1.5K whereas the $^3$He cryostat provides temperatures down to 300 mK. A SRS-DS360 ultra-low distortion function generator is used to apply a small bias voltage (as low as 10 µV). The resulting current (I) through the tube is measured with a low noise current amplifier (SRS 570) while a low noise potential amplifier (SRS 560) measures the potential across the sample at the same time. The set-up allows us to measure current-voltage (I-V) and differential conductivity $G=dI/dV$ versus V curves at temperatures down to T=0.3K. Another separated high voltage source (K-2400) is
connected to the doped Si layer serving as gate electrode under the source and drain. The gate voltage determines the electrostatic potential of the nanotubes.

4.4. Results and discussion

As the temperature of a sample drops, initially the conductivity of the SET device starts to decrease. Figure 4.1c shows how the conductivity of one of the samples (sample A) studied drops as temperatures is decreased. If the temperature is decreased further, the conductance of the coulomb peaks start to increase. The phenomena of the conductivity increase with decreasing the temperature happens when the energy level spacing becomes comparable to thermal fluctuations\textsuperscript{6,7}. Figure 4.1d shows a result where the height of a coulomb peak (the red data points in Fig. 4.1d) increases as temperatures drops below 10K for a multiwall nanotube length of 250 nm (Sample D). Usually, the increase of the Coulomb peak heights occurs at 500 mK for some of the peaks. The increase in the coulomb peaks is interpreted as the localization length being larger than the electrode spacing of 100 nm. The conductivity at a coulomb valley (the blue data points in Fig. 4.1d) decreases as temperature decreases.

To understand the temperature dependence of our results we compare our data to the theoretical predictions for an array of multiple tunneling junctions\textsuperscript{1,2,3,5,8,9,10,11} (Fig. 4.2). The theory is developed for the WCB regime ($k_B T > E_C$). The differential conductivity for an array of $N$ tunneling junctions is given as a function of temperature as:\textsuperscript{1}

\[
\frac{dI}{dV} = G_t \left[ 1 - 2 \sum_{i=1}^{N} \frac{R_{t,i}}{R_e} \Delta_i \left( \frac{R_{t,i}}{R_e} \frac{eV}{k_B T} \right) \right] \tag{4.1}
\]

where $V$ is the bias potential across the tunneling junction array, the $R_{t,i}$ is the high
Figure 4.1. Schematic sample picture, a scanning electron micrograph, and two temperature dependence curves are shown. (a) The chips used to prepare have multi layer structures. The gap in SiN film creates an interruption between electrodes. (b) A scanning electron micrograph of sample D is shown. The dark region shows the trench in the Si chip. (c) The temperature dependence of sample A is shown for $V_{\text{gate}}=0$ V. (d) The conductivity of sample D is shown. Although the conductivity of the carbon nanotube is suppressed by the Coulomb interactions as the temperature drops, the conductivity for the Coulomb peaks (red dots, $V_{\text{gate}}=0$ V) start to increase when the thermal fluctuations become comparable to the energy level spacing. On the other hand, the conductivity of the valley keeps dropping (blue dots, $V_{\text{gate}}=-11$ V). For the temperatures larger than the charging energy, the Coulomb
oscillations disappear and the conductivity data for peaks and valleys merges into each other. The charging energies are 0.4 meV, 0.4 meV, 2.7 meV, and 3.5 meV for samples A, B, C, and D respectively.

temperature resistance limit of the tunneling resistance of $i^{th}$ junction, $R_x$, is the summation of the $R_{x,i}$'s, $G_T$ is asymptotic value of $G$ when $V \rightarrow \pm \infty$ which is equal to the inverse of the corresponding resistance, i.e. $G_{x,i} = R_{x,i}^{-1}$, and $e$ is electron charge.

$$g(x) = \left[x \sinh(x) - 4 \sinh^2(x/2)\right] \left[8 \sinh^4(x/2)\right]$$ is the Pekola function and $\Delta_i$, the Coulomb blockade threshold for the $i^{th}$ junction and originates from the inverse capacitance matrix of arrays\(^8\): $\Delta_i = ((C^-)_{i-1,i-1} + (C^-)_{i,i} + (C^-)_{i,i+1})e^2/2$.

Capacitance matrix $C$ is defined in ref. 7 as:

$$C_{i,j} = \begin{cases} 
C_i + C_{i+1} & \text{for } i = j \\
-C_{i+1} & \text{for } i = j - 1 \\
-C_i & \text{for } i = j + 1 \\
0 & \text{for else} 
\end{cases}$$

For our samples, we know we have two tunneling junctions: the junction of the tube with source and the junction of the tube with drain (our experiments show that the localization length is larger than the distance between source and drain electrode). So the capacitance matrix $C$ for our sample is $C = \begin{bmatrix} C_1 + C_2 & -C_2 \\ C_2 & C_2 \end{bmatrix}$. Inverse matrix $C^{-1}$ is

$$C^{-1} = \begin{bmatrix} 
\frac{1}{C_1} & \frac{1}{C_1} \\
\frac{1}{C_1} & \frac{C_1 + C_2}{C_1 C_2} 
\end{bmatrix}$$

Finally the conductivity for our two junction system is calculated as
Figure 4.2. Schematics of an N-junction array. Each junction has tunneling resistance of $R_T$ at high temperatures and has coupling capacitance of $C_i$. The islands between two neighboring junctions are coupled to the gate by capacitance $C_{0,i}$. For our system there are only two junctions, and $C_{0,i}$ is ignored since it is 3 orders of magnitude smaller than the coupling capacitances $C_i$ (from ref. 1).

\[
G = G_T \left[ 1 - \frac{R_{T,1}}{R_{\Sigma}} \frac{e^2}{C_i k_B T} g \left( \frac{R_{T,1} eV}{R_{\Sigma} k_B T} \right) - \frac{R_{T,2}}{R_{\Sigma}} \frac{e^2}{C_2 k_B T} g \left( \frac{R_{T,2} eV}{R_{\Sigma} k_B T} \right) \right] \tag{4.2}
\]

and the formula becomes, for zero bias potential ($\lim_{x \to 0} g(x) = 1/6$), with

\[
C_{\text{eff}} = (R_{T,1} + R_{T,2}) \left( \frac{R_{T,1}}{C_1} + \frac{R_{T,2}}{C_2} \right)^{-1}
\]

as:

\[
G = G_T \left[ 1 - \frac{e^2}{6C_{\text{eff}} k_B T} \right] \tag{4.3}
\]

The equation 4.3 can be rewritten in the following form:

\[
\frac{G_T}{G_T - G} = \frac{6C_{\text{eff}} k_B T}{e^2} \tag{4.4}
\]

In figure 4.3, $R(T)$ curves are shown for 4 different samples. The data is plotted according to Eq. 4.4. The slopes of the lines are determined with effective capacitance. The
Figure 4.3. Normalized conductance change as a function of temperature for 4 different samples (sample A, B, C, and D). The data are shown as circles while the theoretical fits are obtained using equation 4.4. The high temperature conductivity is found as 308 $\mu$S, 32 $\mu$S, 3.64 $\mu$S and 3.0 $\mu$S for samples A, B, C, and D respectively. The capacitances are 200 aF, 247 aF, 77 aF and 55 aF respectively.

The linearity of the data strongly depends on the choice of the limiting high-temperature conductivity $G_T$. The results are listed in table 1. For some of our samples, the high temperature limit is reached between 60 K and 100 K. Apparently beyond this limiting temperature, some other factor, or factors dominate the temperature dependence of the conductivity, since we observe the resistance raise above the high temperature limit $G_T$ of this temperature. It is possible that the intrinsic resistance of the tube becomes dominant at
Figure 4.4. The differential conductance as a function of bias potential is shown for sample B. The experimental data (squares for 1.4K and circles for 4.2K) are fit by Eq. 4.2. The capacitances calculated from the fits are $3.9 \times 10^{-16} \text{F}$ and $2.4 \times 10^{-16} \text{F}$. Conductivity of junctions is calculated as $72 \ \mu\text{S}$ and $56 \ \mu\text{S}$. The asymmetry in the data especially for 1.4K, which emerges from the Coulomb blockade theory is due to different junction capacitances.

For a given gate potential, this asymmetry can be observed in Fig. 4.5.

higher T. The linear fit for sample C and D intercepts the y-axis for T=0 K at almost 0 as the theory of arrays of tunneling junctions predicts however the fits for sample A and B have offsets that cannot be explained by the theory.

Figure 4.4 shows the normalized differential conductance for sample B for two different temperatures. The data are shown as squares (T=1.4 K) and circles (T=4.2 K). The
Table 4.1. The fitting parameters used to fit the data to the different theoretical curves and Coulomb diamonds are shown. The last column shows the charging energies in terms of temperatures obtained from Coulomb diamonds. All the capacitances are given in units of atto-Farad ($10^{-18}$ F). All of the conductivity data are given in units of micro-Siemens ($10^{-6}$ S). All data obtained with three different fits are consistent with each other.

Data fits are made using Eq. 4.2. At 4.2 K (solid red line), the theoretical curve agrees with the data perfectly well. However, at 1.4 K (solid blue line), deviates at bias potentials higher than 1 mV. The data is not symmetric for this temperature. This observation can be explained by the asymmetric junction capacitances (this will be explained in detailed below). The fitting parameters for these two temperature data sets (1.4 K and 4.2 K) are very close to each other. The theoretical fitting curve requires 4 fitting parameters, ($C_1$, $C_2$, $G_1$, and $G_2$), and the fitting parameters are given in table 4.1.

Figure 4.5 shows the differential conductivity vs. bias potential and gate potential at 300 mK for sample B. The conductivity is blocked in the blue regions. The borders of the rhombic blue islands are defined by the capacitance of each junction. More details on this subject can be found in chapter 3. Two such rhombic islands are shown in Fig. 4.5. The
Figure 4.5. The Coulomb diamonds measured using sample B at 300 mK. In the blue islands, the conductivity is suppressed by the Coulomb blockade. Using the slopes of the lines around the blue islands the capacitance of each island can be calculated. Evident from the two adjunct islands at -1V or at 6V the different electron waves couple to the electrodes differently. Average capacitance calculated for this sample are $3.5 \times 10^{-16}$ F and $2.0 \times 10^{-16}$ F. The data for the other samples are shown in table 4.1.

The capacitance of the junctions calculated out of the Coulomb diamonds are shown in table 4.1. Because the border slopes of each island are different, the capacitances calculated using the information extracted from two different islands is slightly different from each other for the same tube. This becomes more obvious if the border slopes of the two adjunct islands at $V_{gate}=-1$ V or the border slopes of another two adjunct islands at $V_{gate}=6$ V are compared. The capacitance of a junction depends on the geometrical parameters of the contact and also on how strongly the electrons waves couple to the electrodes. On the other hand, for the data
Table 4.2. Effective capacitance, and high temperature $R_T$ is given for different fitting methods. The second column shows the $C_{eff}$ calculated using the slopes of fitting lines in Fig.4.2. the third column shows the $C_{eff}$ calculated by the definition using the capacitance and conductivity of each junction obtained from $G(V)$ data. The fourth column shows the $C_{eff}$’s calculated using the capacitance obtained from the Coulomb diamonds. During this calculations, the conductance of the juctions are assumed to be constant, and total conductance is the one calculated from Fig. 4.3. The fifth column shows the high temperature limit resistance $R_T$ calculated from slopes of the best fit of Fig. 4.3. The sixth column shows the high temperature limit resistance $R_T$ calculated from fitting parameters of $G(V)$ curves. The last column shows the room temperature limits.

we obtained at high temperatures ($k_B T > E_C$), many electrons with different waves contribute to the conductivity so observed capacitance is the average of all capacitances due to all different electron waves contributing to the conductivity. Table 4.1 shows the average of capacitances calculated.

For a given gate potential, the conductivity of an island as a function of bias potential will be asymmetric for junctions with different capacitances. This can be observed from Fig.
4.5 in terms of the border slopes. This is why the differential conductivity vs. bias potential is not totally symmetric for the data shown for 1.4K. At higher temperatures \((k_B T > E_C)\), when the Coulomb oscillations and corresponding Coulomb diamonds get suppressed by thermal fluctuations, the asymmetry in the \(G(V)\) curve disappears although zero-bias resistance peak persists at higher temperatures.

Table 4.2 shows the \(C_{\text{eff}}\), and high temperature resistances \((R_T = 1/G_T)\) obtained using different experiments. The first column shows the data calculated slopes of the fits in Fig. 4.3. The second column shows the \(C_{\text{eff}}\) calculated using the capacitances obtained from the \(G(V)\) fits similar to Fig. 4.4. The third column shows the calculated \(C_{\text{eff}}\) by using the capacitances obtained from diamonds and assuming both junctions have equal conductivity (conductivity of each junction cannot be extracted from Coulomb diamonds using available data). All the calculated \(C_{\text{eff}}\) calculated from \(G_T(T)\) and \(G(V)\) are very consistent with each other however, the \(C_{\text{eff}}\) calculated from diamonds are slightly off maybe since we do not know the exact value of conductivity.

4.5. Conclusion

In summary, we studied the transport properties of multiwall carbon nanotubes positioned as Coulomb islands in single electron transistors. Our samples act as a single electron tunneling transistor below 4K. The high temperature transport properties of the samples are analyzed using the theories developed for arrays of tunneling junctions\(^{1,5}\). The temperature dependent and voltage dependent data agree well with the theory. The fitting parameters obtained from graphs are consistent with the ones calculated from coulomb diamond patterns. Thus, we conclude that the transport mechanism in our single electron
tunneling transistors can be explained by WCB developed for the array of junctions for the temperatures higher than charging energy.

4.6. References

Chapter 5

Theory of Nanotube Band Structure

5.1. Introduction

A carbon nanotube (CN)\textsuperscript{1} is a graphite sheet rolled into a tube and seamed. The way it is rolled defines the chirality of the tube. The chirality defines whether or not the tube is metallic or semiconducting. Since currently there is no reliable way to separate them, both kinds of nanotubes exist among our samples, and both kinds of nanotubes are experimentally studied. A graphite sheet has a two-dimensional crystal structure. To calculate the band structure of the nanotube, one can use the information about the band structure of graphite. The tight-binding method will be used to find the energy levels of the graphite and later the result will be modified to satisfy the extra constraints imposed by the boundary conditions associated with the nanotube.

Multiwall carbon nanotubes are much more complicated systems, compared to their cousin, the single-wall carbon nanotubes. They are composed of many concentric nanotubes, and these nanotubes may also have different chiralities. The inclusion of the interaction between different layers of the tubes makes determination of the eigenfunctions of the Hamiltonian of the carbon nanotubes quite complicated. In addition, as the electrons are supposed to tunnel into the inner layers, the conductivity through the inner layers will be low because of an extra tunneling barrier between the outer shell and the inner shells of the multiwall carbon nanotubes. In addition, previous experiments show that only the outermost shell contributes to the conductance\textsuperscript{2,3}. Thus, only the outermost shell is assumed to be
conducting, and the results of the tight binding model for the outermost shell will be representative of those of the multiwall carbon nanotube.

5.2. Bloch waves

For a system with translation symmetry, i.e. crystal lattices like graphite sheet, the energy eigenfunctions can be taken to satisfy Bloch’s Theorem. This indicates that for a system with a periodic potential \( V(\vec{x}) = V(\vec{x} + \vec{R}) \) where \( \vec{R} \) is any displacement which is a linear combination of unit vectors of the lattice the energy eigenfunction should have the following form: \( \Psi_k(\vec{r}) = e^{i\vec{k} \cdot \vec{r}} u_k(\vec{r}) \) where \( \vec{k} \) is the reciprocal lattice vector and \( u_k(\vec{r}) \) periodic functions i.e. \( u_k(\vec{r}) = u_k(\vec{r} + \vec{R}) \). Thus, \( \Psi_k(\vec{r}) \) satisfies the Bloch’s theorem:

\[
\Psi_k(\vec{r} + \vec{R}) = e^{i\vec{k} \cdot (\vec{r} + \vec{R})} u_k(\vec{r} + \vec{R}) = e^{i\vec{k} \cdot \vec{R}} e^{i\vec{k} \cdot \vec{r}} u_k(\vec{r}) = e^{i\vec{k} \cdot \vec{R}} \Psi_k(\vec{r})
\]  

(5.1).

5.3. Tight binding model

Knowing that the wave function of the electron in a periodic potential should satisfy Bloch’s theorem, does not help much in finding the eigenfunction of the electron in the lattice. In order to construct the eigenfunctions, the atomic wave functions can be used to form the Bloch wave functions. The simplest approach is to assume that the atomic wave functions are still valid and do not overlap strongly as they are combined to obtain the Bloch wave function of the electrons in periodic potential. The following wave function is used for describing the behavior of the electrons in the lattice:
\[ \Psi_{j,k} = \frac{1}{\sqrt{N}} \sum_{\vec{r}} e^{i\vec{k}\cdot\vec{R}_j} (\vec{r} - \vec{R}_j), \] where \( j \) represents the \( j^{th} \) solutions for the atomic orbital. The wave function is normalized by the factor \( \frac{1}{\sqrt{N}} \) since \( N \) lattice sites exist. This wave function satisfies Eq. 5.1: \( \Psi_{j,k}(\vec{k}, \vec{r} + \vec{a}) = e^{i\vec{a}\cdot\vec{R}} \Psi_{j,k}(\vec{k}, \vec{r}), \) where \( \vec{a} \) is any displacement that is a linear combination of unit vectors of the lattice.\(^4\)

\[
\Psi_{j,k}(k, \vec{r} + \vec{a}) = \frac{1}{\sqrt{N}} \sum_{\vec{r}} e^{i\vec{k}\cdot\vec{R}_j} (\vec{r} + \vec{a} - \vec{R}_j) = \frac{1}{\sqrt{N}} \sum_{\vec{R} = \vec{R} - \vec{a}} e^{i\vec{k}\cdot(\vec{R} + \vec{a})} \phi_j(\vec{r} - \vec{R}') = e^{i\vec{a}\cdot\vec{R}} \Psi_{j,k}(k, \vec{r})
\]

The energy of the system is: \( E_{j,k} = \frac{\langle \Psi_{j,k}(k, \vec{r}) | \hat{H} | \Psi_{j,k}(k, \vec{r}) \rangle}{\langle \Psi_{j,k}(k, \vec{r}) | \Psi_{j,k}(k, \vec{r}) \rangle}. \) The tight binding calculation simplifies the problem using the fact that the electronic states of each atom are localized. There will be very little spatial overlap between wave functions of different atoms such that only the interaction of the nearest neighboring atoms are important.

### 5.4. Band structure of graphite

The structure of each wall of a carbon nanotube looks very similar to the structure of a graphene sheet. Thus, the bandstructure of carbon nanotubes are expected to resemble that of the graphite.

Each graphite sheet is made out of hexagonal lattice of carbon atoms. Figure 5.1 shows the crystalline structure and reciprocal lattice of a graphite sheet. A carbon atom sits on each corner of the hexagon shown in Fig. 5.1a. Nearest neighboring carbon atoms have a spacing of \( a_{C-C} = 1.42 \ \text{Å} \) between themselves. This lattice can be defined by two lattice vectors...
Figure 5.1. The unit cell and Brillouin zone of graphite. (a) \( a_1 \) and \( a_2 \) are unit vectors of the graphite. The dashed rhombus shows a unit cell having two atoms in it. (b) \( b_1 \) and \( b_2 \) are reciprocal lattice vectors. The shaded area is the first Brillouin zone of graphite. The energy dispersion relation for the dotted line connecting \( K, M \) and \( \Gamma \) is given in Fig. 5.2. (from ref. 7).

\[
\vec{a}_1 = \left( \frac{1}{2} a \sqrt{3}, -\frac{1}{2} a \right) \quad \text{and} \quad \vec{a}_2 = \left( \frac{1}{2} a \sqrt{3}, \frac{1}{2} a \right) \quad \text{where} \quad a = \sqrt{3} a_c. \quad \text{A unit cell of the lattice is shown in Fig. 5.1a as dotted rhombus.}
\]

The reciprocal lattice vectors \( \vec{b}_1 \) and \( \vec{b}_2 \) can be calculated as

\[
\vec{b}_1 = \left( \frac{2\pi}{a \sqrt{3}}, \frac{2\pi}{a} \right) \quad \text{and} \quad \vec{b}_2 = \left( \frac{2\pi}{a \sqrt{3}}, \frac{2\pi}{a} \right) \quad \text{using the relation} \quad \vec{a}_i \cdot \vec{b}_j = 2\pi \delta_{ij} \quad \text{(ref. 5).} \quad \text{The first Brillouin zone is shown as the shaded region in Fig. 5.1b.}
\]

The energy dispersion relation for the graphite can be found by using the orbital wave function of \( p_z \) electrons\(^6,7\) as:

\[
E(k_x, k_y) = \pm t \sqrt{1 + 4 \cos^2 \left( \frac{1}{2} k_y a \right) + 4 \cos \left( \frac{1}{2} k_x a \right) \cos^2 \left( \frac{1}{2} \sqrt{3} k_y a \right) \quad \text{where} \quad t \quad (=3.033 \text{ eV}) \quad \text{is}
\]
Figure 5.2. The energy dispersion relation for a graphite sheet. The inset shows the energy dispersion relation along the dotted line in Fig. 5.1 (from ref. 7).

The transfer integral between the two nearest neighbors. During this calculation, the energy of an electron in $p_z$ state is assumed to be 0.

The energy dispersion for a graphite sheet is shown in figure 5.2 for the first Brillouin zone.

5.5. The Band structure of carbon nanotubes

Since carbon nanotubes can be thought of as a rolled up graphite sheet, therefore if one ignores the effects of the curvature of the shell, the energy dispersion relation of the graphite sheet should work as well for the carbon nanotubes.

The geometrical structure of carbon nanotubes can be defined by a chirality vector $(\vec{C}_h)$, and translation vector $(\vec{T})$. The chirality vector shows which two atoms in the graphite sheet will overlap along the circumferential direction as carbon nanotubes formed. Translation vector can be defined as one of the unit vectors of the carbon nanotubes along
Figure 5.3. A carbon nanotube is unrolled to show the similarity between the nanotube and the graphite sheet. \( \vec{a}_1 \) and \( \vec{a}_2 \) are the unit vectors of graphite. \( \overrightarrow{OA} \) or \( \overrightarrow{BB'} \) defines the chirality vector \( \vec{C}_h \) for (4,2) CN. \( \overrightarrow{AB'} \) or \( \overrightarrow{AB} \) defines the translational vector \( \vec{T} \). Unit cell and Brillouin zone of graphite (from ref. 7).

The axis of the carbon nanotube. Figure 5.3 shows an example of a chirality vector \( \vec{C}_h = 4\vec{a}_1 + 2\vec{a}_2 \) or (4,2) CN, and translation vector for (4,2) CN. Because of the periodic boundary conditions induced by the chirality vector, only special wave vectors \( \vec{k} \) satisfying the following conditions are allowed: \( \vec{C}_h \cdot \vec{k} = 2\pi n \) where \( n \) is an integer. The allowed wave vectors all depend on the chirality vector. The finite length of the tubes will induce another constraint on the wave vector.

The armchair carbon nanotubes, \((n,n)\) CNs, do not have any energy gap between the valence and conductance bands. One of the allowed wave vectors of the armchair nanotubes coincides with the corners of the first Brillouin zones, where the gap is zero.
Figure 5.4. The metallic and semiconducting tubes are shown with respect to their chirality vector \((n,m)\) (from ref. 7).

Some of the zigzag nanotubes, \((n,0)\) CNs, i.e. \((4,0)\) CN, have a gap between the valance and conductance bands. The allowed wave vectors of some of the zigzag nanotubes do not coincide with the corners of the first Brillouin zones, so they are semiconducting. The rest of the zigzag tubes are metallic.

Figure 5.4 shows which CNs are metallic and which are semiconducting.

A tight binding calculation predicts the energy gap for semiconducting carbon the nanotube as

\[
E_g = \frac{|t|A_{c-c}}{d_t}
\]

where \(t\) is the overlap integral, \(A_{c-c}\) is the distance between nearest neighbor carbon atoms, and \(d_t\) is the diameter\(^7,8\).

The system we study here is an ideal one. It does not involve any disorder. In order to have a complete understanding of the band structure, we need to know many other factors like the effects of the substrate and the effects of the inner tubes and also the effects of the coupling between the leads and tubes. Coulomb interactions, disorder, and dopant levels
should be also involved in order to understand the band structure and electrical properties of the nanotubes. Many of these factors still require further analysis.

5.6. Ajiki-Ando gap

Ajiki and Ando\(^9\) made an important prediction about the band structure of the nanotubes: A coaxial magnetic field can change the allowed wave vectors, thus drastically changing the band structure. Consequently the electrical properties of a given nanotube can be controlled by a coaxial magnetic field.

In the presence of a magnetic field, the wave vector of a given system is related to that without a field by
\[
\vec{k} \rightarrow \vec{k} + \frac{\Phi}{R\Phi_0} \vec{K}_1\text{,}
\]
where \(\vec{k}\) is the wave vector, \(R\) is the radius of the tube, \(\Phi\) is the flux through the tube, \(\Phi_0=h/e\) is the flux quanta, and \(\vec{K}_1\) is the reciprocal lattice vector along the circumferential direction. The chosen gauge is \(A = \mathbf{B} \times \mathbf{r} / 2\) for our calculation. As a result, the band structure changes with a period of one flux quantum \(\Phi_0=h/e\). Figure 5.5 shows how the energy spectrum of a nanotube changes with the magnetic field for some low-laying energy levels.

Figure 5.5a shows a result for a metallic tube. The undoped metallic tube does not have any gap between the valance and conductance bands. As the co-axial magnetic field is applied to the tube, a band gap is created. The gap reaches its a maximum at \(\Phi_0/2\) and then goes back to 0 for \(\Phi=\Phi_0\). This continues in a periodic manner with maximum gap opening for \(\Phi = \frac{2n+1}{2}\Phi_0\) and no opening for \(\Phi = n\Phi_0\) (n is an integer).

Figure 5.5b shows a result for the semiconducting tube. There is a gap on the order of \(E_{max}\) for the 0 magnetic field (and thus flux). As the flux increased, the gap starts to close,
Figure 5.5. Energy spectrum of (a) a metallic (200,200) CN and (b) a semiconducting (346,0) CN tube calculated with tight binding method. The graphs show how the band gaps change as a function of magnetic flux.

and becomes 0 for $\Phi=\Phi_0/3$. Later, a smaller gap opens with a maximum at $\Phi=\Phi_0/2$, which will close back to 0 at $\Phi=2\Phi_0/3$. Beyond $\Phi=2\Phi_0/3$, the gap restores itself to the size at $\Phi=0$. The whole process continues similarly beyond $\Phi=\Phi_0$.

5.7. Zeeman splitting

Ajiki-Ando predicted the interconversion of types of the nanotubes between metallic and semiconducting nanotubes with a period of $\Phi_0$. As a magnetic field applied to the nanotubes, the Zeeman splitting will also occur. The Zeeman splitting will mix up the energy levels of the nanotubes. With large magnetic fields, the gap of semiconducting tubes gets smaller and the interconversion become less and less prominent.
Figure 5.6. The energy spectrum of a metallic tube with (200,200) chirality vector with Zeeman splitting. The Zeeman splitting shown is stronger than it actually is in order to emphasize its importance on the periodicity of energy spectrum with magnetic flux.

Figure 5.6 shows the result of a tight-binding model for metallic tubes with Zeeman splitting. Zeeman splitting will destroy the periodicity of the Ajiki-Ando gap modulation.

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I acknowledge very fruitful discussions with Dr. T-C. Wei. Dr. Wei wrote a program to calculate the band structure of the nanotubes.

5.8. References


Chapter 6

Aharonov-Bohm Effect

6.1. Introduction

Consider a beam of electrons that is separated into two parts and the two parts travel through the two different paths, before they are combined together to form an interference pattern at the end. Even though the beams do not even travel in the magnetic field the interference pattern can be altered by changing the amount of the magnetic field enclosed by these paths. This phenomenon is known as the Aharonov-Bohm (AB) effect, which demonstrates how much the phase of the electrons is sensitive to the vector potential $\vec{A}$. The AB effect was reviewed by many authors$^{1,2,3}$. Here we follow the review of Aronov and Sharvin$^4$ closely.

Ehrenberg and Siday proposed the experiment shown in figure 6.1 to test the mentioned phenomenon in 1949 [5]. The schematic of the experiment is presented in the Fig. 6.1a. The beam emitted by the source S is separated into two parts, and the two parts travel through the paths I and II before they form an interference pattern on the screen AB. There is a magnetic field confined in region “f” surrounded by the paths. The electron beams do not touch the magnetic field, as they travel though magnetic field free paths I and II around region “f”. Finally, the interference pattern (Figure 6.1b) is proposed to shift by changing the magnetic field in the region “f” thus the magnetic flux. Aharonov and Bohm explained this quantum mechanical phenomenon in 1959 [6] unaware of the previous works.
Figure 6.1. (a) Schematic of the experiment proposed by Ehrenberg and Siday in 1949 [ref. 5]. The two electron-beams emitted by source S take two different paths before creating the interference pattern on screen AB. The two paths engulf the region “f”, which has a magnetic field. (b) Interference pattern is shown for the two electron-beams of different paths.

Historically, already in 1939, Franz\(^7\) mentioned that the phase difference between two paths was changed by the magnetic flux enclosed by the paths.

The phase difference between the two paths can be calculated by the following formula: \( \Delta \varphi = \frac{e}{\hbar} \int \vec{A} \, dl = 2\pi \frac{\Phi}{\Phi_0} \), where \( \vec{A} \) is vector potential, \( \hbar = 2\pi\hbar \) is Planks constant, \( e \) is unit charge and \( \Phi_0 = h/e \) is the flux quantum. The phase difference will depend on the magnetic flux, thus on the total magnetic field surrounded by the paths.

Let us consider a case of a ring with radius R and a freely moving electron on it to understand how the AB effect changes the phase and the energy of the systems. Figure 6.2a
Figure 6.2. The schematic of the one-dimensional ring, and the quantum energy levels of the ring as a function of magnetic field. (a) The shaded region confines the magnetic field. The electrons are free to move on the ring. (b) The lower lying energy levels of the electrons are shown as a function of magnetic flux. Energies of the levels change by changing the magnetic flux in the system. The splitting occurs due to a weak arbitrary potential (from ref. 4).

illustrates the geometry under consideration. The shaded area represents the region where the magnetic field exists. The wave function solutions of the Hamiltonian will be in the following form:

$$\psi_n = e^{iny/R},$$

where $n$ is any integer, and $y$ is the coordinate of the electron on the ring.

The wave function is quantized due to the periodic boundary condition: $y$ and $y+2\pi R$ are referring to the same position.
By solving the Schrödinger equation, one obtains the energy of the system as

\[ E_n = \frac{\hbar^2}{2mR^2} \left[ n - \frac{\Phi}{\Phi_0} \right]^2, \]

where \( n \) is an integer, and \( m \) is the mass of the electron.

Figure 6.2b shows the energy of the ground state as a function of the magnetic flux. Magnetic flux is given in units of the flux quantum. Considering a weak arbitrary potential, there will be a splitting as shown in Fig. 6.2b when two energy levels cross each other. Overall, the ground state, energy spectrum and other properties of the system expressed by the energy spectrum of the system will be a periodic function of the magnetic flux with a period of \( \Phi_0 / 8, 9 \).

Up to now many different magnetic field experiments have been done. These experiments show that the conductivity and the magnetic moment of the samples depend in an oscillatory manner on the magnetic flux\(^{10,11,12,13,14}\). These oscillations are called Dingle oscillations\(^{13}\) and are understood in terms of geometric size effects in the momentum space. The oscillations are periodic with \( \Phi_0 / S_i \) where \( S_i \) is the area of “regular polygons with an arbitrary number of sides inscribed into the circular cross section of a cylinder, the polygons representing close projections of the electron trajectories on the cylinder cross section”.

Following works emphasize the localization of the electrons on the surface of the structure\(^{15,16,17}\), here the structure is a cylinder, thus the relatively significant contribution of oscillations with the period of \( B = \Phi_0 / \pi R^2 \) are expected. The first experiments investigating the oscillations with \( B = \Phi_0 / \pi R^2 \) were done by Brant et al\(^{18,19}\). Their analyses of a very complex spectrum of oscillations revealed an oscillation with a period close to \( B = \Phi_0 / \pi R^2 \).

Meanwhile, another class of oscillations of transport measurements for multiply connected samples are predicted for a diffusive regime first by Altshuler et al\(^{20}\). These oscillations
have a period of $B = \Phi_0/2\pi R^2$. Such oscillations are firstly observed by Sharvin and Sharvin in the same year\textsuperscript{21}. The works done on quazi-one dimensional rings led to another class of oscillations for tiny conductors with a period of $B = \Phi_0/\pi R^2$ [22,23,24,25,26,27,28].

6.2. Resistivity oscillations in multiply connected disordered conductors

The resistivity of the disordered conductors oscillates as a function of the magnetic field with a period of $B = \Phi_0/2\pi R^2$ due to the Aharonov-Bohm effect in the form of a phenomenon called weak localization\textsuperscript{21,29,30,31,32}.

To understand the weak localization phenomena, let us calculate the probability of transferring a particle from one point to another one, and see how it changes as a function of the magnetic field. The schematic of the problem is given in figure 6.3. The probability of transferring a particle between two points, “P” and “Q”, (Figure 6.3) depends on the sum of the probabilities of all possible trajectories between the two points. Some of the trajectories will be self-crossing. One such self-crossing trajectory is shown in the figure. The electrons which are travelling along the second trajectory have two options at point “O”. For an electron, the probability of coming back to point “O” through such trajectories can be calculated as:

$$|A_1 + A_2|^2 = |A_1|^2 + |A_2|^2 + 2 \text{Re} A_1^* A_2 = 4|A_1|^2,$$

where $A_i$ are the phase differences for each path. Note that the phase of the electrons travelling through the paths will be the same when there is no magnetic field. The calculated probability is twice the sum of the squared amplitude moduli. High probability of returning to the position “O” means less probability of the electron transfer from point “P” to “Q”.
Figure 6.3. Three trajectories between “P” and “Q” are shown. The 2nd trajectory has a self crossing point “O”.

When the magnetic field is applied to the system, the electrons will have a different phase at the end of each path. For the loop shown in figure 6.3 on the second trajectory, the phase for the electrons travelling $i^{th}$ path will be as follows:

$$A_1 \rightarrow A_1 e^{ir_i/h A_{dil}} = A_1 e^{i2\pi \phi_i/\Phi_0}$$

$$A_2 \rightarrow A_2 e^{-ir_i/h A_{dil}} = A_2 e^{-i2\pi \phi_i/\Phi_0}$$

So there will be an additional phase difference of $\Delta \phi = 2\pi \Phi / \Phi_0$ while for the case of zero magnetic field there is no phase difference between the two paths. The phase induced by the magnetic field can destroy the interference pattern thus the localization of the electrons.
Figure 6.4. The resistance change of a cylindrical lithium film is shown as a function of magnetic field. The sample is measured at 1.1 K. The diameter of the cylindrical structure is 1.3 µm, and the film thickness is 127nm. The theoretical calculation is shown with dashed lines (from ref. 32).

The value of \( |A_1 + A_2|^2 \) might even go to zero, showing the total destruction of the localization. Therefore the conductivity will increase as a result of these kinds of destructive interferences on the self crossing trajectories. Thus, the resistance will decrease, and the negative magneto-resistance will be observed\(^{33,34,35}\). Since the interference will become constructive for large enough magnetic fluxes, which is destructive initially, the resistivity.
Figure 6.5. The resistance oscillations of the planer structures. The measurements are done at 0.13 K on two different structures, necklace and mesh with different sizes. The schematics of the structures referred as necklace and mesh are shown in the inset. The upper one is necklace. The resistance oscillations are shown as a function of magnetic field. The first curve from at the top is a control sample with a large side length “S”. The next top three show the data for necklace pattern with different side lengths as noted next to the curves in the graph. The lowest lying two curves show the data for mesh patterns with different side lengths. “S” is given in units of µm. (from ref. 36)

will increase. As magnetic flux is increased further, the interference will become destructive again. The Resistivity will oscillate as a function of magnetic flux with a period of $\Phi_0/2$

Some of the first experiments$^{21,31,32}$ are done on cylindrical films deposited on quartz filaments. Figure 6.4 shows an experimental result of 10-100 nm thick lithium films deposited on 1µm diameter quartz filaments$^{3,32}$. The conductivity is measured at the liquid
helium temperatures with a DC potentiometer and a coaxial magnetic field. The resistivity of the film oscillates in an oscillatory manner with the magnetic field, thus the magnetic flux. The period of the oscillations is $\Phi_0/2$ in terms of the magnetic flux. The maximum of the resistance did not appear for zero magnetic field unlike the expectations. This observation is explained by the magnetic field trapped in the film. The detailed study of the weak localization theory predicts that the amplitude of the oscillations will drop and the negative magneto-resistance will be observed in a large scale. All these features can be observed in Fig. 6.4.

Figure 6.5 shows multiply connected planer structures. The experiment is done by Dolan et al. The structure consists of arrays of squares connected to the next one through the corners. The resistance oscillations have a period of $\Phi_0/2$ where $\Phi_0$ is the magnetic flux of a single square.

6.3. Experiments on mesoscopic samples

Theoretically, the oscillations with a period of $\Phi_0$ are also studied. It is found that if the system is one dimensional, the conductance of the system has a period of $\Phi_0$. In addition, for these systems, other oscillations with a period of $\Phi_0/2$ also exists due to interference of the time reversal of the waves. However, in the one dimensional case, the oscillations with a period of $\Phi_0/2$ have smaller amplitude: If the transmission coefficient for one direction is $t$, then the amplitude of the oscillation with the period of $\Phi_0$ will be the order of $t$. On the other hand, the amplitude of the oscillations with the period of $\Phi_0/2$ will be $t^2$ [39] which makes the oscillations with the period of $\Phi_0$ stand out.
Figure 6.6. The conductivity of a gold ring as a function of magnetic field. The diameter of the ring is 784 nm where the thickness is 41 nm. (a) the resistance of the ring as a function of magnetic field. (b) the Fourier transform of the resistance data. The peak at $131 T^{-1}$ corresponds to the resistance oscillations with a period of $\Phi_0$ (from ref. 28).

Figure 6.6 shows the conductivity of a gold ring as a function of magnetic flux$^{28}$. The conductance has many fluctuations with the magnetic field. The Fourier power spectrum of the fluctuations shows a peak at magnetic flux $\Phi_0$. The experiment is performed on a ring with a diameter of 784 nm and a width of 41 nm. Chandrasekhar et al$^{40}$ successfully reproduce oscillations with period of $\Phi_0$ on silver rings in addition to the ones with $\Phi_0/2$. 
Figure 6.7. The magneto-resistance of a multiwall nanotube. As temperatures get high, because of the temperature dependent phase-coherence length, the oscillations disappear. The conductance of the tube oscillates with a period of $\Phi_0/2$ magnetic flux. The radius calculated as 8.6 nm from the data agrees well with the radius of 8.0 nm measured by atomic force micrograph (from ref. 41).
6.4. Aharonov-Bohm oscillations in carbon nanotubes

Carbon nanotubes have cylinder like structure with a diameter in the range of 1 nm to 50 nm. Their sizes make them ideal to observe the Aharonov-Bohm oscillations in carbon nanotubes. One of the first experiments on the conductivity of the carbon nanotubes is done by Bachtold et al.\textsuperscript{41}. They observe the conductance oscillations with a period of $\Phi_0/2$ magnetic flux. Figure 6.7 shows the conductance oscillations observed on a nanotube with a radius of 8 nm at various temperatures. The oscillations are well fitted by the theoretical predictions for the Aharonov-Bohm effect in hollow cylinders. The oscillations disappear at high enough temperatures due to temperature dependent phase-coherence length. Using the conductance data of 1.8 K as a function of magnetic field, the phase-coherence length is determined as 54 nm which has the same order of magnitude with the circumference of the tube and with the length of the part that carries current between electrodes. The comparison of the phase-coherence length and the dimensions of the tube suggests that the conductivity is diffusive in the carbon nanotubes. The period of the oscillations suggest that only the outer most shell or shells are contributing to the conductivity.

Some samples show superimposed short periodic oscillation on oscillations mentioned above. Similar fast oscillations are also observed by other researchers\textsuperscript{42,43}. The source of the oscillations are not clear and the radius calculated from the period of the oscillations turns out to be much larger than the radius of the nanotube.
6.5. Conclusion

- Multiply-connected planer structures show conductivity oscillations with magnetic flux with a period of \( \frac{\Phi_0}{2} \), which is a manifestation of the AB effect combined with weak localization phenomenon.

- Very thin metallic wire loops show conductivity oscillations with magnetic flux with a period of \( \Phi_0 \), which was the original most simple prediction of the AB physics.

- Conductance oscillations with a period of \( \Phi_0/2 \) are observed in multiwall carbon nanotubes, proving that transport properties of disordered nanotubes are described in terms of the same physics as disordered metallic cylinders in parallel magnetic field.

- The \( \Phi_0 \)-period oscillations have not been observed on nanotubes prior to our work, which is presented in the next chapter. This was the main motivation for my research project.

6.6 References


Chapter 7

*h/e Magnetic Flux Modulation of the Energy Gap in Nanotube Quantum Dots*

7.1. Summary

The electronic properties of carbon nanotubes\(^1,2,3\) depend sensitively on the manner in which their constituent graphite sheets are rolled and seamed, i.e. their chirality. For example, depending on whether or not the resulting single-electron energy spectra have gaps at the Fermi energy, they can be either semiconducting or metallic\(^4,5,6,7,8,9,10\). Although the chirality can not be altered at will, the electronic properties — and, in particular, the energy gap — have been predicted to be tunable by applying a coaxial magnetic field through the tube. There is no other material that can be periodically converted between the semiconducting and the metallic state by the magnetic field. This unexpected tunability of nanotubes was predicted by Ajiki and Ando (AA) in 1993 (ref. 11). The predictions are further discussed in refs.12,13. The key prediction is that the magnetic flux applied along and inside the nanotube results in interconversion between semiconducting and metallic types of behaviour, with the scale for the AA- and chirality-induced band gaps being comparable. The phenomenon is related to the Aharonov-Bohm (AB) coupling of the flux to the momentum of the electrons\(^14\). The key point in understanding the AA effect is the energy dispersion relation of 2D electrons in graphite sheet, calculated by Wallace in 1947. This Wallace energy-band structure has six points in the momentum space at which the energy gap goes to zero and valance band touches the conduction band. Application of the
magnetic field along the tube, changes the momentum of the electrons, as predicted by the
AB effect. At some values of the magnetic field one of the momentum quantization lines
crosses one of the six points where the band gap is zero. Thus, for this field, the tube, which
is semiconducting in zero field, becomes metallic. As momentum quantization lines are
periodically spaced, one expects a periodic conversion of the tube between metallic and
semiconducting states. This is the essence of AA effect.

We performed experiments on quantum dot single-electron-tunneling transistors\textsuperscript{15},
made out of a short multiwall nanotubes (MWNT) and threaded by AB flux. By measuring
the differential conductance, we observe a magnetically induced gap modulation at the
Fermi level, thus providing evidence for the aforementioned interconversion. Whereas the
linear conductance of long MWNT’s is known to yield period $h/2e$ oscillations\textsuperscript{16}, for short
MWNT’s in the quantum-dot regime we find experimental evidence for $h/e$ variations in the
energy spectrum, in broad agreement with simple tight-binding calculations. This suggests
an observation of the AA physics in nanotube quantum dots.

\textbf{7.2. Introduction}

Multiwall carbon nanotubes (MWNT’s), unlike most other single molecules, have
the ideal size and structure for studying the effect of the AB-flux on the energy spectrum
and conduction properties. Being concentric cylinders with outer radius $R \approx 15$ nm, they
allow penetration by an entire flux quantum $\Phi_0=h/e$ at accessible magnetic fields of about
$\Phi_0/\pi R^2 \approx 6$ T. In contrast, a typical single-wall nanotube\textsuperscript{2} with radius of 1.5nm would
demand a $\sim 600$ T magnetic field, which is currently infeasible. Previous experiments\textsuperscript{17,18}
indicate that only the outermost shells contribute to conduction. Thus $R$ is in fact the
relevant radial length scale, and one can expect to see AB modulations in, e.g., transport and spectral properties, such as the gap variation, over a field range of order 10 T. Although AB physics has been studied in MWNT’s, to date, attention has been focused on linear transport properties of long samples, typically several microns in length$^{19,20}$. Here we exploit a second aspect of nanotubes, namely their ability, when they are short (for our purposes, about 1 µm or shorter), to exhibit quantum dot physics. Thus, short MWNT’s furnish us with the opportunity to explore AB physics in quantum dots.

7.3. Experiment

In our experiments we position individual MWNT’s$^{21}$ over two Au electrodes, thus creating single-electron tunneling transistors, because the tube-to-gold contact resistance can be sufficiently high (Fig. 7.1a). The source and drain electrodes, between which the bias voltage ($V_B$) is applied, are separated by a =100 nm gap. The substrate is a doped Si wafer, covered with a SiO$_2$/SiN double-layer on which the Au electrodes are fabricated lithographically. Being doped, the wafer can subsequently be used as a gate electrode$^{22}$. An important feature of the sample is the trench, which is cut through the SiN film before Au is deposited, and served to create a gap between the source and drain electrodes. Two samples, A and B, of respective lengths $L_A$=800 nm and $L_B$=500 nm and radii $R_A$=15 nm and $R_B$=13 nm, have been investigated. A scanning electron microscope micrograph for sample B is shown in Fig. 7.1b.
Figure 7.1. Fabrication and measurements on quantum dot devices made of nanotubes. (a) Schematic of a single electron transistor built from a single nanotube. The
sample is fabricated on a highly doped Si wafer (which acts as gate electrode) covered with 500 nm SiO$_2$ and a 60 nm SiN film. The source and drain electrodes are formed by evaporating a strip of gold film across a 100 nm wide trenched etched into SiN/ SiO$_2$ double-layer$^{22}$. Multiwall carbon nanotubes$^{21}$ are deposited onto the Au electrodes from a dichloroethane solution. The magnetic field is parallel to the tube axis. (b) A SEM image of a representative sample. (c) Quasi-periodic Coulomb conductance peaks for sample B. (d) The temperature dependence of one of the conductance peaks height (sample B). The increasing conductance suggests resonant tunnelling through a single energy level. (e) Coulomb blockade diamond measured on sample A.

7.4. Results and discussion

The samples are initially probed by applying a small amplitude ($\approx$0.05 mV, 7 Hz) AC voltage bias between the source and the drain, for a range of gate voltages, and measuring the resulting AC current, which is translated into the differential conductance ($dI/dV$), normalized by the conductance quantum $e^2/h$. These initial measurements are carried out at $T$=0.3 K. Representative results are shown in Fig. 7.1c and 7.1e. The former shows the conductance variation vs. gate voltage for sample B; the latter presents a color-coded differential conductance map plotted vs. the gate and bias voltages for sample A. Both samples exhibit characteristic Coulomb blockade peaks and diamonds. This confirms that the devices are operating as single-electron tunnelling transistors. As is well known, in the Coulomb blockade regime, conduction through the tube occurs at low bias voltage only for those specific values of the gate voltage for which it is comparably probable to have $N$ or $N+1$ electrons on the tube, resulting in the expected Coulomb blockade conductance.
peaks\textsuperscript{23,24}, as shown in Fig. 7.1c. The gate-voltage differences between Coulomb peaks are proportional to $E_C + \Delta E$ — the sum of the charging energy and the single-particle level spacing. The pattern of Coulomb blockade diamonds\textsuperscript{25} in Fig. 7.1e is a typical feature observed on quantum dot devices, which occurs due to single-electron transport. Since the distance between the corners of the Coulomb diamonds in the $V_B$ direction is roughly $2E_C$ (if the level splitting is neglected), we determine that $E_{CA} \approx 0.4$ meV and $E_{CB} \approx 0.6$ meV (for samples A and B). These values are comparable to an estimate of the charging energy of a nanotube, $E_C \approx 1.4eV/L$[nm] (ref. 26), which gives 1.75 meV and 2.8 meV for samples A and B. A smaller charging energy is observed, possibly due to a strong capacitive coupling of the nanotube to the electrodes. The temperature dependence of the Coulomb-peak height is illustrated in Fig. 7.1d, which shows an increase at low temperature. This feature is observed on some, but not all Coulomb peaks, and suggests resonant tunneling through individual energy states of the tube\textsuperscript{6,7}. The increase begins at $T \approx 0.5$K, indicating that the level spacing is about $\sim 0.05$meV or larger.

Our main results are displayed as color-coded maps of the differential conductance, plotted vs. bias voltage and magnetic flux, as shown in Figs. 7.2 (sample A) and Fig. 7.3 (sample B). The flux, $\Phi = \pi R^2 B$, is calculated from the applied magnetic field $B$ and the measured outer radius of each tube ($R_A = 15 \pm 1$ nm and $R_B = 13 \pm 1$ nm). The gate voltage is kept constant during each conductance measurement. The resulting conductance maps are always symmetric with respect to the magnetic field sign. First let us discuss sample A, for which two measurements are shown: one for $V_g = -4.1$ V (Fig. 7.2a), corresponding to the maximum of one of the Coulomb peaks at zero flux and zero bias; the other for $V_g = -35$ V (Fig. 7.2b), is slightly off another (zero-flux, zero-bias) maximum. These measurements
Figure 7.2. Color-coded differential conductance of a MWNT quantum dot plotted versus bias voltage and magnetic flux. The Differential conductance measured at (a) Coulomb peak (in zero field) with fixed $V_g=-4.1V$. (b) Sample A differential conductance measured slightly off a Coulomb peak, with fixed $V_g=-35V$. The inset shows the same data in black-and-white format in order to emphasize the Zeeman slitting of a single energy level. The gray lines are to guide to the eye, marking the level splitting.
show the same trend: The conductance gap (i.e. the blue region, where the conductance is very low) continues to widen with increasing magnitude of the AB flux, starting small, until a half flux quantum is reached. After that, the trend reverses until a full flux quantum is reached, at which flux the induced gap reaches a pronounced minimum.

To understand the origin of the field-induced gap structure and related features of the conductance maps, we have compared them with predictions of a simple tight-binding model for the pi-electrons of carbon nanotubes of finite length. These are summarized in Fig. 7.4, which shows the variation, with flux, of the low-lying single particle energy levels for a metallic armchair tube (Fig. 7.4a) and a semiconducting zigzag tube (Fig. 7.4b). The most prominent feature is the variation, and even creation, of a gap induced by the magnetic flux and periodic in it, with period $h/e$ (the gap is the diamond-shaped region from which all lines are expelled). For metallic tubes, the magnetic flux opens a gap and results in a sequence of identical gap diamonds; this gap closes at integral magnetic flux quanta. By contrast, for semiconducting tubes, the magnetic flux tends to shrink the gap, leading to gap closure at pairs of magnetic flux values straddling half-flux quanta, resulting in gap diamonds of two distinct sizes. A simple band structure estimate of the maximum field-induced (metallic case) gap gives $2E_M = \sqrt{3}ta/(2R) = 36$ meV, where $a \approx 2.49$ Å is the lattice constant and $t \approx 2.5$ eV is the inter-site hopping amplitude for graphene; $E_M$ is comparable in scale to that of a chirality-induced gap of the same radius.

To make the comparison between the experimental differential conductance maps and the theoretical predictions, we note that the former are essentially a measure of density of electronic states at a given energy, provided the gate voltage compensates for the charging energy. Figure 7.4 also characterizes this quantity, inasmuch as the density of lines
Figure 7.3. Color-coded differential conductance of a MWNT quantum dot plotted versus bias voltage and magnetic flux. (a) Differential conductance measured for a Coulomb peak (in zero field with zero bias potential) with fixed $V_g=-25.3V$. (b) Differential
conductance measured for a valley between two Coulomb peaks, with fixed \( V_g = -21.6 \text{V} \). (c) Differential conductance measured for a Coulomb peak (in zero field with zero bias potential) with fixed \( V_g = -25.3 \text{V} \). The images are symmetric with respect to the sign change of the magnetic field and almost symmetric with respect to the sign change of the bias voltage. For high bias potentials, the graphs look very similar to each other. The most striking difference appears in the blue region for zero bias case. For the peaks, a light blue island in the middle of the dark blue is seen corresponding to the peaks, however for the valley, the central part of the graph is dark blue showing no response to the coaxial magnetic field.

reflects the density of states. We see from Figs. 7.2a and 7.2b (sample A) that our measurements qualitatively resemble the broad predictions shown in Fig. 7.4a.

We now turn to sample B, for which three measurements are shown: one for \( V_g = -25.2 \text{V} \) (Fig. 7.3a), corresponding to the maximum of one of the Coulomb peaks at zero flux and zero bias; the other one for \( V_g = -21.6 \text{V} \) (Fig. 7.3b), corresponding to a valley between two (zero-flux, zero-bias) maximum, the last one for \( V_g = -2.8 \text{V} \) (Fig. 7.3c), corresponding to the maximum of another Coulomb peaks (zero-flux, zero-bias). One of the most salient feature of the maps is the similarity between the graphs. Another most salient feature is the high degree of periodicity in the flux dependence, with period \( \Phi_0 \), especially at bias voltages higher than roughly 0.5 mV. All the curves show the same trend. In contrast with sample A, the resulting tendency is for higher conductance near half-odd-integer quanta of magnetic flux. As in the present experiment, the differential conductance reflects the underlying density of states, this tendency suggests that the density of states is smaller at integer flux...
quanta, also in contrast with sample A. Another prominent feature is the near-perfect symmetry with respect to bias voltage, observed in sample B but certainly not in sample A. Two origins of this symmetry present themselves: symmetry between tunnel barriers and/or accidental half-filling (the latter being unlikely). Notice, too, the narrow but pronounced region of low conductance at low bias, $V_B<0.4$ mV (indicated in dark blue in Fig. 7.3). For all but very low magnetic fluxes, the width of this region is particularly insensitive to the magnetic field. We speculate that conductance in this region occurs via localized states, generated by surface impurities, which are not able to wrap around the tube and thus they can not sense the magnetic flux. Moreover, such states would have much smaller matrix elements with electrode electronic states and should give low conductance. By contrast, conduction at higher bias is dominated by more extended states, which are multiply connected and thus sensitive to magnetic flux. Thus we observe strong oscillations of the conductance with a period of one flux quantum at higher bias.

One plausible interpretation of the above observations is that sample B is a semiconducting tube, which has become doped by surface impurities due to prior exposure to air, which shifts the Fermi level away from half-filling. Let us explore this point by comparing the data (Fig. 7.3) with the results from simple tight-binding calculations for a semiconducting tube (Fig. 7.4b). Provided the Fermi-level shift is small, the data and the calculations essentially agree with regard to the phase of the flux dependence of the conductance. However, if the shift is very small, the calculated maximum energy gap would exceed the observed one (~1.5 meV) by an order of magnitude. Moreover, the secondary gap diamonds, expected for half-filling, are not observed. The secondary gap diamonds are the ones, which appears between $h/3e$ and $2h/3e$. Taken together, these imply
Figure 7.4. The energy levels for nanotubes plotted versus magnetic flux threaded through the nanotube. The simulations are for nanotubes of length approximately 730 nm, diameter 28 nm and two different chiralities: (a) (200,200) armchair and (b) (346,0) zigzag respectively. The energy scale is in unit of $E_M$. A dashed horizontal line in figure (a) indicates a position of the Fermi level, shifted by doping. In this case we observe a lower density of states (at the Fermi level) at integer flux values and higher values of the density of states at half-integer flux values (assuming equal participation of the states above and below the Fermi level). The dispersions are calculated within the standard single-particle tight-binding model with the hopping strength $t \approx 2.5 \text{ eV}$ (ref. 8).

that substantial doping has occurred. Another plausible interpretation is that the tube is a metallic tube. In this case also, if a strong doping occurs, the shift of the Fermi level away from half-filling could produce a diamond structure reflecting high-conductance near half-
odd-integer quanta of flux. An example of such a Fermi level is indicated in Fig. 7.4a by a horizontal dashed line. The available data do not allow us to draw a definite conclusion about the chirality of the tube. However, the results definitely demonstrate that the period of the flux response is $\hbar/e$ rather than $\hbar/2e$.

The Zeeman effect can also be observed with our SET devices. An example is shown in Fig. 7.2b (black-and-white inset). Here we see a single energy level, which splits in the magnetic field by an amount that agrees with the g-factor reported previously for single-wall nanotubes\textsuperscript{6}. Indeed, consider, e.g., the splitting at $\Phi=0.5\Phi_0$ (which corresponds to a field of 2.9 T), which is about 0.32 meV. This translates into a g-factor of 1.95, in agreement with previous reports\textsuperscript{6}. From this data it is possible to estimate that the Zeeman splitting at $B\sim10$ T roughly equals the maximum gap. Therefore the gap structure is not periodic with flux, but rather becomes smeared. This effect is clearly seen in Fig. 7.2a, where the periodicity with flux breaks down at high fields.

7.5. The role of the Coulomb blockade and perpendicular field measurements

In order to eliminate any confusion for the interpretation of the experimental results, and to show the important of the coaxial magnetic field, sample B is tested with perpendicular field. In the following subsection, the results of these experiments will be given.

Coulomb interactions dominate the conductivity for low bias potentials. It is not immediately clear what part of our the results are due to the magnetic field, and what part of the results are due to the Coulomb blockade. Experiments for different gate potentials will be shown in the following subsection.
**Figure 7.5.** Differential Conductance map with a magnetic field perpendicular to the tube axis. The map is measured for a conductivity peak at zero bias and zero field ($V_g = -22.85$ V). The data is rescaled since the contacts have become more transparent during the sample warm-up in order to rotate it. Previously observed $h/e$ oscillations are not observed for perpendicular field. There is a slight overall increase in the conductivity as a function of magnetic field.

### 7.5.1 Energy spectrum for perpendicular magnetic field

As further confirmation of the relevance of the AB flux we have tested the effect of a perpendicular field. The sample is warmed to the room temperature to have access to it. The sample is placed in another orientation so that the magnetic field is perpendicular to the tube rather than parallel as we had in experiments presented above. The sample was slightly changed during the warming up and cooling down process. The conductivity of the tube is increased by a factor of 5 for both conductivity at Coulomb peaks and Coulomb valleys. The results suggest that the contacts between our nanotube and the Au electrodes become more
transparent after second cooling down. The resultant conductivity map is normalized by a factor of 5 in order to compare it to the parallel field measurements. Figure 7.5 shows the conductivity map measured for perpendicular magnetic field. The gate potential is set for a Coulomb peak for zero-bias and zero-field case (Vg=-22.85V). The dominant $h/e$ magnetic flux periodic oscillations observed before are not observed with perpendicular field as predicted. The conductivity of the carbon nanotube stays mostly the same as it is for zero magnetic field, for non-zero transverse magnetic fields. There is a slight increase in the data that might be understood in terms of weak localization.

7.5.2 Energy spectrum for different gate potentials

Coulomb blockade physics controls the conductivity of our samples. In order to gain a deeper understanding of AA physics and the Coulomb blockade more measurements have been done with different gate potentials for sample B. The gate potential can lift the blockage as shown in figure 7.1c. The data shown in figure 7.3a and 7.3c are obtained for Coulomb peaks. On the other hand, the data shown in figure 7.3b are obtained for a coulomb valley where the conductivity is minimal.

Even thought the graphs looks very similar to each other for high bias potentials, the major difference shows up in the center of the map (at zero bias, zero field). For measurements done at Coulomb peaks (Fig. 7.3a and 7.3c), a high conducting island (light blue region) shows up which moves away as magnetic field is applied. For the maps measured by setting the gate potential in such a way that the Coulomb blockade dominates the conductivity for zero bias potential, there are no such high conductance island observed (Fig. 7.3c). Other than that, the graphs looks very similar to each other, even some very fine
features are reproduced for two measurements with the two different potentials. Consequently, we concluded that, it is not very important to set the gate potential so that we obtain the data for a Coulomb peak or a valley to study the AA energy spectrum modulation.

The gate voltage changes the potential energy of the island. As the potential of the island becomes favorable for another charging configuration, the electrons tunnel into or out of the island, thus the number of the electrons on the island changes. In some sense this shifts the Fermi level of the island, thus the effect is somewhat similar to that of a dopant. Previously published results showed that the Fermi level could be shifted dramatically by applying a gate potential\textsuperscript{27}. For our case, the gate potential difference of the Coulomb peaks on which Fig. 7.3a and 7.3b measured is nearly 23 V. A change of 23 V in the gate potential corresponds to a 3.6 mV ($\alpha=1.56\times10^{-4}$) potential change of the island due to weak gate-tube coupling. Considering only 4 electrons are added to the island as the gate potential is changed by 23 V, and charging energy is a minimum of 0.5 meV, then the maximum possible energy shift in the Fermi level is 1.5 meV. This shift is one order of magnitude smaller than the chirality induced gap, so we do not expect to see any significant change in the energy spectrum in our case due to the change at the gate potential. For our case, even though a very strong gate potential is applied to the tube (up to ±50V), because of weak coupling between the gate electrode and the tube and relatively large charging energy, the Fermi Level does not shift dramatically.
7.6. Experimental observation of the AA energy gap modulation by other research groups

There are two other groups which studied the AA gap modulations\textsuperscript{28,29} in parallel with ours, but using different methods. Minot \textit{et al.}\textsuperscript{28} of Cornell University and Zaric \textit{et al.}\textsuperscript{29} of Rice University confirmed the predictions made by Ajiki and Ando, however they could not apply a full $\Phi_0$, which is the full period of the AA gap modulations.

Minot \textit{et al.} used small diameter (2-5 nm) nanotubes to study AA physics. They measured the transport properties of the individual tubes in the magnetic field at liquid helium temperatures. The conductance of their samples is blocked by the Coulomb interactions at liquid helium temperatures. They used a nearby gate electrode to change the electrostatic potential of the tube and to restore the conductance. At a zero bias potential, the conductance is restored for some specific gate potentials so the conductance peaks are observed as the gate potential is ramped. By varying the gate and bias potentials, they obtained the Coulomb diamonds, which revealed the chirality induced gap. The chirality induced gap is much larger than the gaps resulted from the Coulomb interactions. They applied a coaxial magnetic field to modify the chirality induced gap.

AA predicted that the chirality induced gap can be closed by applying magnetic field\textsuperscript{11}. Since their tubes had very small diameters, an inaccessibly large magnetic field is required to close the chirality induced gap completely. However, they proved that what they observed was the early steps of the gap modulation predicted by AA. To prove AA gap modulations, they plotted the conductance of the nanotubes as a function of magnetic field and gate potential for zero bias potential. Using the relationship between the coordinates of the conductance peaks on the gate potential and magnetic field axes, the orbital angular
momentum of the electron levels is measured. They found out that the orbital angular momentum they measured from the experimental data was very close to the one predicted by the theoretical calculations.

Zaric et al. chose to study optical properties of the single wall carbon nanotubes to observe the AA gap modulation. They measured the photoluminescence and absorption as a function of the magnetic field. They observed the splitting of the some of the first subband. The splitting in the absorption and photoluminescence data are explained by the splitting of a level and its time reversal as a function of magnetic field. This splitting is the essence of the AA gap modulation, and the size of the splitting depends on the magnetic field. Although they applied less then 1% of the field required to see full cycle of the AA gap modulation, their existing data presents an evidence for AA physics.

7.7. Conclusion

In summary, we have fabricated single electron tunneling transistors from multiwall carbon nanotubes, and probed the electronic states of the constituent nanotubes via conductance measurements. We have observed variations of the conductance gap with magnetic flux, and found them to be approximately periodic. In accordance with general quantum mechanical principles associated with the Aharonov-Bohm effect, the observed period is one magnetic flux quantum. The overall form of the conductance maps is consistent with the tight-binding results for the band structure. However, the absolute scale of gaps is smaller than would be expected for an ideal nanotube. Plausible origins for this discrepancy include the effects of disorder and doping.
7.8 References

A similar version of Chapter 7 appeared in Science 304, 1132 (2004).


21 Alfa Aesar, Multiwall nanotubes, Stock # 43197, Lot # L03J42.


Chapter 8


8.1. Abstract

The energy spectrum of carbon nanotubes is studied using differential conductance measurements for different offset bias potentials. A coaxial magnetic field was applied parallel to the tube. We find some indications that the energy spectrum of the tube oscillates with a period of magnetic flux quanta $\Phi_0$ ($\Phi_0=h/e$, where “$h$” is Planck’s constant and “$e$” is elementary charge). We probe the energy spectrum of the nanotube by fixing the offset bias potential to different values on our single electron tunneling transistor. The differential conductance shows different phases of oscillations, i.e. positive and negative magneto-resistance. The oscillations are qualitatively explained by density of states derived from tight binding model calculations.

8.2. Introduction

The electrical properties of carbon nanotubes depend on not only their chirality and the crystal structure but many other things such as doping level, electrode material, and the electrostatic potential of the tube. Depending on the chirality, the nanotubes show either metallic or semiconducting behavior. These behaviors originate from the distribution of the allowed energy states in reciprocal space. The distribution is determined by the crystal structure of the graphene forming the tube and the periodic boundary conditions in the
circumferential direction. The Fermi level can be pushed away from half filing case by doping the tube, or by changing the potential energy of the tube. Dopants can come from the electrodes, and/or surface surfactants. They introduce additional charge carriers into the tube. An electrostatic potential also controls the charge carrier concentration. Therefore, the electronic properties of the tube can be changed by changing the doping level or the electrostatic potential of the tube. In addition, coaxial magnetic field couples with the energy spectrum of the tube and strongly modifies it.

Previously, the linear electronic transport properties of carbon nanotubes have been studied as a function of magnetic fields. The AB effect has been observed in MWNTs, in the context of linear conductance measurements performed on tubes longer than the phase-breaking length. Conductance oscillations reflect the sensitivity of the electrical properties to the AB flux threading the tube in terms of weak localization, typically yielding a $\Phi_0/2$ period. Weak localization is characterized by negative magneto-resistance. Later, another interpretation is proposed suggesting the density of states as a dominant factor in magneto-transport measurements, especially in the ballistic regime rather than the weak localization in the diffusive regime. The last article caused fierce arguments among the researchers. Later theoretical works showed that positive magneto-resistance is also possible if the phase-breaking length is longer than the size of the tubes, in other words, if the dominant factor is the density of states. Furthermore, the calculations show that the conductivity of the same nanotube in the ballistic regime can increase or decrease as the magnetic field is ramped, depending on the Fermi-level location. Density of states calculations predict the variation with a period of magnetic flux quanta ($\Phi_0$), not half flux quanta ($\Phi_0/2$) as in the weak localization calculations. Although, the weak localization has a component with a
period of $\Phi_0$ which becomes significant as the phase-breaking length becomes comparable to the circumference of the tube, the weak localization always predicts negative magneto-resistance unlike the density of states, which says both positive and negative magneto-resistance is possible for a 0 magnetic field. When the electron transport is ballistic, the density of the states controls the magneto-transport measurements, thus the electrical potential and/or the dopant concentration of the tube controls the magneto-resistance by changing the Fermi Level. Lately, positive magneto-resistance has been observed, depending on the electrostatic potential of the tube. In our experiment we scan different energy levels by using an AC signal on the top of a DC bias potential, imitating the effect of doping.

So far, multiwall carbon nanotubes (MWNTs) remain as unique molecules, as they can be treated with one full flux quantum, by the magnetic fields accessible in today’s laboratories. A nanotube with an outer radius $R$ in the range of 10-20 nm will allow penetration by an entire magnetic flux quantum for experimentally accessible magnetic fields in the range of 3.3 T to 13.2 T. Note that previous experiments indicate that only the outermost shell contributes to conduction, so the theoretical predictions can be tested in the laboratory.

Here, we provide experimental results for the different phases of differential conductivity (and thus magneto-resistance) as a function of bias potential and coaxial magnetic field. This way, we trace the density of states for different energies. We placed a single MWNT between two gold electrodes. A bias potential was applied between these two electrodes. The bias potential has two separate components: DC and AC. The DC component of the signal induces an offset for the AC component, and is used to trace
different energy levels, thus imitating different doping concentrations and/or different electrostatic potentials. The AC component helps to obtain density of states for the given DC offset potential.

Depending on the transparency of contacts, the carbon tubes might act as a single-electron tunneling (SET) transistor or just a regular wire. In both cases, we studied the differential conductivity of the nanotube as a function of offset bias potential and magnetic field. A magnetic field up to 9T is applied, which is less than the field required to apply \( \Phi_0 \) magnetic flux for the first tube (with a radius of 8 nm), and more than \( 2\Phi_0 \) for the second tube (with a radius of 18 nm). Upon subjecting the nanotubes to a parallel field, we find the sign of the differential conductance can be positive or negative, depending on the offset bias potential, and is independent of the transparency of contacts. Qualitatively speaking, the magnetic field changes the kinetic energies of the electrons, so the energy levels might move away or towards the focused energy level regions. Since each energy level responds to the magnetic field differently through its angular momentum, the density of the states for some specific doping levels might increase or decrease. The differential conductance is proportional to the density of states, and is expected to vary as the magnetic field changes the density of states. In order to understand the observed magnetic-transport measurements, we calculated the density of states using a tight-binding calculation. The tight binding calculation also predicts positive and negative slopes for magneto-resistance with a period of \( \Phi_0 \), as a function of the Fermi level location. It also predicts that slopes will change back and forth from positive to negative. The tight-binding model overestimates the offset potential required to change the phase of magneto-resistance.
Figure 8.1. Chip layers and a scanning electron micrograph are shown. (a) The Si chip used during the experiment has multiple layers. (b) Micrograph shows an experimental grade tube sitting on top of two electrodes. The dark region shows the gap between the electrodes.

8.3. Experiment

The carbon nanotubes were placed between two electrodes separated with a trench etched in the underlying Si chip. The chip has multiplayer structure. The chip geometry, with electrodes and a nanotube, is shown in Fig. 8.1a. The Si wafer has a 60 nm SiN film on the top of an insulating 500 nm SiO$_2$ layer. The Si substrate layer under SiO$_2$, is strongly doped with phosphorus in order to ensure its electrical conductivity down to low temperatures. This allows us to use the Si substrate wafer as the gate electrode of the SET transistor.

The fabrication starts with an e-beam lithography on a Si chip. The e-beam lithography and following reactive ion etching steps creates a trench and markers along the trench in SiN film. Hydrofluoric (HF) acid is used to deepen the openings into SiO$_2$. HF etching leaves two hanging SiN edges$^{12}$. A strip of gold is then evaporated across this
trench. The trench causes a well-defined interruption (a "gap") of about 120 nm in width in the gold film. Thus we obtain two electrically disconnected gold electrodes separated by a distance of ~120 nm. Commercially available carbon nanotubes\(^\text{13}\) are ultrasonically dispersed in Dichloroethane or 2-propanol and then deposited over the surface of the Si chip with Au electrodes. A scanning electron microscope (SEM) (Hitachi S4700) is used to locate the nanotubes. The positions of the tubes noted with respect to the markers are to be used for positioning the mask while doing photolithography later. The tubes with large diameters are most suitable for magnetic field experiments so the thickest tubes are chosen to work on. We can thread the large tubes with higher magnetic flux than we can thread with much smaller tubes. After defining electrodes by photolithography, the wet etchants are used to remove extra metals from the surface of the Si chip. The samples presented in this section are thicker than 15 nm in diameter.

For transport measurements a SRS-DS360 ultra-low distortion function generator is used to apply a small bias voltage (V) between the source and the drain electrodes. The function generator is capable of supplying an AC signal (down to 10 µV) on top of a DC offset potential. The resulting current (I) through the tube is measured with a low-noise current amplifier (SRS 570). A low-noise potential amplifier (SRS 560) measures the potential across the sample simultaneously. The set-up allows us to measure current-voltage (I-V) and differential conductivity \(G=dI/dV\) vs. \(V\) curves at temperatures down to \(T=0.3\text{K}\). Another separate high-voltage source is connected to the doped Si wafer under the source and drain. The doped layer is used as a gate electrode if the sample acts as an SET transistor. The gate voltage determines the electrostatic potential of the nanotubes.
8.4. Results and Discussion

Our main results are shown in Fig. 8.2 (Sample A) and Fig. 8.3 (Sample B). The graphs show the differential conductivity of the samples, normalized by the quantum of conductance \( (e^2/h) \). The flux \( \Phi = \pi R^2 B \) is calculated from the applied magnetic field \( B \) and the measured outer radius of each tube \( (R_A = 8 \pm 1 \text{ nm} \text{ and } R_B = 18 \pm 1 \text{ nm}) \). The gate voltage is kept constant during each conductance measurement.

First let us discuss sample A, for which differential conductivity is shown as a function of the magnetic field (and thus magnetic flux) for three different offset bias potentials. The conductivity of Sample A is blocked at low temperatures. The data are taken at 300 mK. The charging energy is 2 meV. Ramping the gate potential results in conductance peaks that arise from the Coulomb blockade effect. Throughout our measurements, the gate potential is kept constant at 18.1 V (corresponding to a conductivity peak at 9T). The magnetic flux is calculated by using the radius of the sample, measured using a scanning electron micrograph of the sample. The radius is calculated as 8 nm. In order to have one flux quantum \( (\Phi_0) \) passing through the tube, 23 T should be applied parallel to the tube. Our limit is 9 T. Figure 8.2 shows the differential conductivity for different offset bias potentials, DC, with the same AC component (0.1 mV). The graphs are shifted vertically for clarity. As the magnetic field is varied, the differential conductivity is either increased or decreased monotonically. Note that, for this specific tube, half of the full flux quantum \( (\Phi_0/2) \) could not be reached. The lowest bias potential is chosen such that the Coulomb blockade is totally suppressed \( (V_b \text{ is } 2.5 \text{ mV}) \). For this potential, the differential conductivity decreased as the magnetic field is ramped. For a larger bias potential \( (V_{bias} = 4.6 \text{ mV}) \), the differential potential increased as the magnitude of the magnetic field
Figure 8.2. The differential conductivity as a function of coaxial magnetic field for sample A. The DC component of the signal is set to different values, however the AC component is set to 0.1 mV. The lowest DC offset is 2.5 mV which is higher than the charging energy, 2 meV. The graphs are shifted vertically to have clarity. The ticks are separated by 0.02 $e^2/h$ from each other.

increased. When the bias potential is increased further, the slope of the conductivity changed one more time and becomes negative again.

Sample B is a much thicker and longer nanotube than sample A is. The outer diameter of sample B is 36 nm, which should enclose one full quantum flux at ~4 T. Sample B has more transparent contacts with the electrodes than sample A had. The total resistance is roughly 10 kOhm at room temperature and did not change much as temperatures dropped to 300 mK. As a result of the high conductance through the contacts, the Coulomb blockade is not observed for this tube. The differential conductivity of sample B is shown in Figure 8.3 for 2 different bias potentials. As with sample A, sample B also
Figure 8.3. The differential conductivity as a function of coaxial magnetic field for sample B. The DC component of the signal is set to different values, however the AC component is fixed to 0.1 mV. Graphs are shifted vertically in order to have clarity. The ticks are separated by $0.2 \, e^2/h$ from each other.

shows two different signs for the slope of the differential conductivity as a function of the magnetic field. For zero bias potential, the data have a negative slope initially; however, for 1 mV the graph has a positive slope initially. For 0 DC offset bias potential, initially the differential conductivity drops and then starts to increase, as the magnetic flux becomes bigger than half flux quantum. The conductivity keeps increasing until one full magnetic flux quantum is applied. Beyond one full flux quantum, the conductivity starts to drop again. It continues through this cyclic process. The height of the oscillation for $\Phi=\Phi_0$ is strikingly large compared to the $\Phi=0$ and $\Phi=2\Phi_0$ height. The differential conductance increases as the magnetic field is ramped for $V_b=1$ mV, and starts to drop for magnetic fluxes larger than
\( \Phi_0/2 \). This trend changed later when one full flux quanta (\( \Phi_0 \)) was applied. The above-mentioned process continues up to 2 full flux quanta.

In order to understand these magnetic effects, we perform a simple tight-binding calculation under ideal conditions, corresponding to a single isolated tube that is free of disorder. Note that the previous studies showed that the current is transferred mainly by the outer-most shell of the MWNT\(^2,1^6\). Thus, we neglect the possible effects of inner shells.

Each shell of a carbon nanotube can be thought of as a graphite sheet rolled up and welded seamlessly. Thus the energy spectrum of a carbon nanotube is expected to be similar to that of graphite. The graphite sheet is made out of a hexagonal lattice of carbon atoms. Starting with a non-interacting Hamiltonian for \( p_z \) electrons hopping on the graphite lattice sites, considering interactions in terms of their eigenbases and tremendously simplifying Hamiltonian, the problem become relatively easy. The calculated results from this model show many qualitative effects observed in the experiment. Details of this method can be found in chapter 5.

The energy dispersion is given for a 2-d hexagonal lattice of graphene by the Wallace formula\(^ {14}\) (i.e., the graphite sheet):

\[
E_{g2D}(\vec{k}) = \pm t \sqrt{1 + 4 \cos \frac{k_xa}{2} + 4 \cos^2 \frac{k_ya}{2}}
\]

where \( k_x \) is the component of the momentum parallel to one side of the hexagon, \( k_y \) is the component orthogonal to \( k_x \), and \( a \) is the next-nearest neighbor spacing between carbon atoms. Rolling up the sheet seamlessly into a tube of radius \( R \) leads to the quantization of momentum. \( \vec{k} = k_xi + k_yj = v\vec{K}_r + \mu\vec{K}_l \)

where \( \vec{K}_r \) is the reciprocal lattice vector that points along the circumferential direction and \( \vec{K}_l \) is the one along the axial direction, and \( \mu \) and \( v \) are integers associated with quantization.
Figure 8.4 The energy levels of nanotubes versus magnetic flux threaded through the nanotube is plotted. The simulations are for tubes of lengths approximately 730 nm, diameter 28 nm and two different chiralities: (a), (200,200) armchair and (b), (346,0) zigzag respectively. The energy scale is in units of $E_M$. The dashed horizontal lines in figures indicates the positions of the Fermi level, for which the density of states are calculated.

along the circumference and the finite length of the tube. When the tube is threaded with the coaxial magnetic field, the energy dispersion is related to that without a field by

$$\vec{k} \rightarrow \vec{k} + \frac{\Phi}{R\Phi_0} \vec{K},$$

where $\vec{k}$ is the quantum state, $R$ is the radius of the tube, and $\Phi$ is the flux through the tube (the gauge we choose is $A = B \times \vec{r} / 2$).

Figures 8.4a and b show energy levels of two tubes with distinct chiralities [(200,200) and (346,0)] as a function of magnetic flux. The tubes have a radius of ~14 nm.
These two graphs are the only two distinct graphs one can get from hexagonal lattice up to a
different number of levels and energies. Figures 8.4a and 8.4b show how the energy levels
of a metallic and semiconducting tube respond to the magnetic field, respectively. Figures
8.5a and 8.5b show the density of states at proposed Fermi levels shown in Figs. 8.4a, and
Fig. 8.4b respectively. The density of states is calculated by counting the number of the
levels within a window around the proposed Fermi Level (a₁, a₂ etc.). The width of the
window is ±10% of $E_M$, based on the fact that the periodicity of experimental data is 2 mV
and the AC component of the bias potential is 0.2 mV. The resulting graphs are periodic as a
function of flux with $\Phi_0$, and the graphs may have positive or negative slopes at a zero
magnetic field, depending upon the location of the Fermi level. Our calculation predicts that
the slope of the density of the states, which changed with the magnetic field between
positive and negative values periodically, as the Fermi level is moved up or down. Even
though the tight-binding calculation predicts that the slopes will shift on the order of 40
meV, the experiment shows a periodicity with one order of magnitude smaller energies. One
might argue that the level splitting is large, compared to the AC signal, and we might align
Fermi levels with single energy levels, or a gap between two levels within the same sub-
band. However, in that case, we would then predict much faster oscillations, which we do
not see.

The difference between the experimental result and the theory might stem from the
simplicity of the theory. A more realistic result can be obtained by adding other factors, such
as curvature of lattice structure, effects of substrate and coupling of different shells, exact
coupling between the leads and the tubes, Zeeman coupling, Coulomb interactions, disorder,
and dopants, all in the presence of a magnetic field.
**Figure 8.5** The total number of levels within a 6 meV interval centered at energy levels that are indicated with dashed lines in Fig. 8.4a and 8.4b is shown. (a) The number of levels of metallic tubes for some of the possible Fermi levels. (b) The number of levels of semiconducting tubes for some of the possible Fermi levels. The ticks on the graph axes are separated by 20 for number of states axis.

### 8.5. Conclusion

Our main result shows that the slope of differential conductivity can be positive or negative, unlike the case for the diffusive regime, and thus weak localization always produces positive differential conductivity. The periodicity of the conductance oscillations is $\Phi_0$ (not $\Phi_0/2$) for weak localization. The tight binding method was used to analyze the data. The tight binding method reproduces the overall shape; however, it fails to predict the periodicity of the phase shifts.

### 8.6. Acknowledgments

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8.7 References


13 Alfa Aesar, Fullerene, buckytube/nanotube, multi-walled, ground core, Stock #43197, Lot #L03J42, 26 Parkridge Road, Ward Hill, MA 01835.

Author’s Biography

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Ulas started graduate school at the University of Illinois at Urbana-Champaign in 1999. In January of 2001, Ulas joined the research group of Alexey Bezryadin and began studying electrical transport properties of multiwall carbon nanotubes.

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