Single Charge and Spin Transport in Nanostructures

Jan Johansson

Section of Nanostructure Physics
Department of Physics
Royal Institute of Technology
Stockholm, Sweden
Akademisk avhandling som med tillstånd av Kungliga Tekniska Högskolan i Stockholm framlägges till offentlig granskning för avläggande av teknisk doktorsexamen fredagen den 23 Januari 2004 kl 10:00 i Seminarierum FA32 Roslagstullbacken 33, AlbaNova University Center, Kungliga Tekniska Högskolan, Stockholm.

Opponent: Prof. Dr. Ir. B.J. van Wees, Physics of Nanodevices University of Groningen, The Netherlands
Abstract

This thesis presents experimental and theoretical studies of single charge transport in mesoscopic systems. The first part of the thesis discusses, both theoretically and experimentally, the interplay between spin dependent tunneling and superconductivity in single electron transistors (SET). A model of transport of charge and spin in ferromagnet/superconductor/ferromagnet (F/S/F) structures is developed. Anti parallel alignment of the ferromagnetic electrodes produces a non equilibrium spin distribution on the superconducting island, which completely suppresses superconductivity at a critical spin accumulation. Large fluctuations of the spin accumulation, and thereby the superconducting gap are predicted, which reduce the effective gap both in the parallel and anti parallel states. It is shown that when the island volume is made small the critical spin accumulation is only a few spins. This makes the device a nearly ultimate spin sensor. Experimentally, F/S/F SET’s are fabricated and the transport properties are measured as a function of an external magnetic field and temperature. The large magnetoresistance measured is compared to the one predicted theoretically.

The second part of the thesis is about charging effects in one dimensional tunnel junction arrays. An experimental investigation of an alternative fabrication technique for single electron devices is conducted. We investigate the electrical transport properties of narrow titanium (Ti) wires which are anodically oxidized through a resist mask. The anodization process is described and transport measurements are presented. The current voltage characteristics of the resulting Ti/TiOx nano-wire exhibit a zero-current state, characteristic of the Coulomb blockade. The blockade region can be modulated by a capacitively coupled gate electrode. The observed experimental features of these samples can be explained in terms of a single electron tunneling model of a multi island array. The theoretical part of this work describes static background charges in one dimensional arrays. We have numerically studied the behavior of one dimensional tunnel junction arrays when random background charges are included using the “orthodox” theory of single electron tunneling. Random background charge distributions are varied in both amplitude and density. The use of a uniform array as a transistor is discussed both with and without random background charges. An analytic expression for the gain near zero gate voltage in a uniform array with no background charges is derived. The gate modulation with background charges present is simulated.
Contents

1 Introduction ............................................. 5
   1.1 Tunnel junctions and the charging energy .............. 5
   1.2 Spin dependent transport ............................... 7

2 Background topics ........................................ 9
   2.1 The single electron transistor ............................ 9
       2.1.1 The orthodox theory ............................... 11
       2.1.2 Calculating the transport properties ............... 14
   2.2 Ferromagnetic single electron transistors ............. 16
       2.2.1 Spin dependent tunneling ........................... 18
       2.2.2 Spin diffusion ....................................... 21
       2.2.3 The F/F/F and F/N/F SET .......................... 22
       2.2.4 The F/S/F structure .................................. 25

3 Experimental techniques .................................. 27
   3.1 Sample fabrication ....................................... 27
   3.2 Measurement setup ........................................ 28

4 Results ...................................................... 31
   4.1 Spin diffusion experiment ................................ 31
   4.2 The F/S/F SET ........................................... 33
       4.2.1 Theory ............................................... 35
       4.2.2 Experimental Results ................................ 40
       4.2.3 Interpretation and comparison to the model ....... 41
   4.3 One dimensional arrays ................................... 45
       4.3.1 Theory of the 1D array .............................. 45
       4.3.2 Gate effects in 1D arrays ............................ 48
       4.3.3 Background charges in 1D arrays .................... 52
CONTENTS

4.4 Anodization .................................................. 57
  4.4.1 Anodization results .................................. 60
  4.4.2 Results .................................................. 62
  4.4.3 Interpretation ......................................... 68

Acknowledgements .................................................. 73

Bibliography ......................................................... 75

Papers ................................................................ 87
Chapter 1

Introduction

This work is done in the Nanostructure group at KTH and hence it is about nanostructures. Nano in nanostructure refers to the length scale nanometer, nano is $10^{-9}$ and a nanometer (nm) is then $10^{-9}$ meter or one billionth meter. One nanometer is a very short distance, in metals the distance between atoms is roughly 0.5 nm and ha human hair is around 100000 nm in diameter. The nanostructures we fabricate have a smallest size of 30–100 nm so they are not small on the atomic scale but small on the human scale. The physics of nanostructures are also referred to as mesoscopic physics, meso means middle or in between, the physics of structures with size between microscopic structures like atoms and macroscopic objects like cars.

1.1 Tunnel junctions and the charging energy

When two metals are separated by a thin insulating barrier and a bias voltage is applied across the barrier, quantum mechanics allows electrons to tunnel through the barrier. Such a structure is commonly referred to as a tunnel junction or just junction. If we now consider two metal leads separated by two junctions with a small metal grain in between and a voltage applied across the double junction structure (the leads are commonly referred to as source and drain and the grain as the island). Electrons can tunnel through the junctions and must stay for some time on the middle grain. This fact that the electrons, on their journey from source to drain, makes the charge of the grain vary by the electron charge $e$ is the key ingredient in single charge transport. The electron charge is a very small quantity, but if the
central grain is very small the electrostatic energy associated with one extra
electron present on the island can be large. The electrostatic energy, or
charging energy, when a metallic grain is charged with $e$ is

$$E_c = \frac{e^2}{2C},$$

where $C$ is the capacitance of the grain. Thus the energy supplied by the
voltage source must overcome the charging energy in order for an electron to
enter on to the island, otherwise the energy of the circuit would be increased
which is forbidden. The fact that no electrons can travel through the dou-
ble junction structure for low voltages is known as Coulomb blockade. The
charging energy is a key ingredient in all Coulomb blockade phenomena, and
sets the relevant energy scale.

Using modern nano fabrication techniques, devices like the one described
here are possible to fabricated with known geometry and tunnel junctions.
The tunnel junctions often consists of a metal oxide of a few Å thickness (1 Å
is 0.1 nm or $10^{-10}$ m). The metal is usually Aluminum (Al) since Al forms a
well controlled barrier when exposed to pure Oxygen. The tunnel junctions
is formed where two metal electrodes are overlapping, hence the capacit-
ance of the island in the double junction device is approximately the sum of the
capacitances of the two junctions. In order to make the charging energy large
the capacitance must be small and therefore the junction must be small since
the capacitance scales as the junction area. A junction of $0.1 \times 0.1 \mu m^2$ ($1 \mu m$
is one millionth meter) gives a capacitance of around 0.5 fF and a charging
energy of 80 $\mu$eV, which is, both fabrication and measurement wise, well in
experimental reach.

There are two condition that must be fulfilled in order for the above to
be true. Firstly, there is one other energy scale that have to be considered:
the thermal energy. Thermal fluctuations must be smaller than the charging
energy:

$$k_B T < E_c,$$

where $T$ is the temperature and $k_B = 1.38 \cdot 10^{-23} \text{ J/K}$ is the Boltzmann
constant. The charging energy mentioned above of 80 $\mu$eV corresponds to a
temperature of 0.093 K, a temperature which is easy to go below experiment-
tally with dilution refrigerators who can go down to $\approx 0.01$ K.

Secondly, the island must only be connected to the leads via tunnel junc-
tions with a tunnel resistance greater than the quantum resistance, i.e.,

$$R \gg R_K = \hbar/e^2 \approx 25.8128 \text{ k}\Omega.$$  

(1.3)
1.2 Spin dependent transport

Conventional electronics uses only one property of the electron, namely the electrons charge. But with the progress in nano- and microfabrication and thin film technology the spin degree of freedom can play an important role in the transport process. Apart from mass and charge the electron has spin, which is a quantum mechanical property and is quantized to have two values along an external magnetic field, usually called up and down. A magnetic moment is associated with the spin. The field of spin electronics or spintronics studies spin dependent processes in various systems such as magnetic multilayers, semiconductors, magnetic oxides, superconductors and tunnel junctions.

In nonmagnetic metals there are equal numbers of spin up and spin down giving zero total magnetic moment, but in ferromagnetic metals there is an asymmetry between the two spin directions resulting in a nonzero macroscopic magnetization. Magnets come, as we know, in all sizes so there is not necessarily anything “nano” about them. The magnetization in a large ferromagnet is not constant throughout the magnet, it is broken up in so called domains and different domains have different directions of magnetization. But if the ferromagnet is small it will only contain a single domain, small here means \( \lesssim 100 \) nm. Thus, if one wants to have one well defined magnetization and fully exploit the asymmetry between the two spin directions one needs to fabricate ferromagnets with nanometer dimensions.

The asymmetry between the two spin directions has implications for the transport properties. The transport properties depend on the density of states of the electrons that participate in the transport and in ferromagnets the density of states for the two spin directions are shifted, giving larger density of states for majority spins (electrons with magnetic moment parallel to the magnetization) than minority spin (electrons with magnetic moment antiparallel to the magnetization). Hence the current in a ferromagnet is, to a large degree, carried by the majority spins.

The spin dependent transport is often studied in structures where two ferromagnets are separated by something nonmagnetic, such as a normal
CHAPTER 1. INTRODUCTION

metal or a tunnel barrier, in order to be able to set the magnetization of two ferromagnets differently (if the magnets are in contact the spins are exchange coupled across the contact and the magnetizations is not independent). When the ferromagnetic electrodes are separated by a tunnel junction the resistance depends on the relative orientation of the magnetization of the electrodes. If the magnetization of the electrodes are antiparallel, the majority electrons in one electrode tunnel into the like spin state in the other electrode, which are now the minority electron states, giving higher tunnel resistance than the parallel case where majority electrons tunnel into the majority states.

In case the ferromagnets are separated by a normal metal the current into the normal metal from the ferromagnet will introduce an asymmetry between the two spin directions in the normal metal. This surplus of one spin direction in a normal metal is known as “spin accumulation”. The spin accumulation is increased by the current but spin flip scattering decreases the spin accumulation. Thus, there will be an equilibrium situation between the influx of spins and the spin flip scattering. This in turn defines a characteristic length scale which describes how far into the normal metal the spin accumulation survives. It turns out that the spin accumulation decays exponentially into the normal metal on a length scale known as the “spin diffusion length”. The spin diffusion length for normal metals is between 10 nm and 1000 nm, depending on the metal and its purity. So again we find that it is necessary to fabricate nanostructures if the effect of spin accumulation is to be exploited.

One device making use of spin dependent transport, already used in commercially sold hard drives, is the Giant magnetoresistive (GMR) spin-valve. GMR, which was discovered in 1988 by Baibich et. al [1] and separately by Binasc et. al [2], is the “giant” dependence of resistance on an external magnetic field in magnetic/nonmagnetic multilayers. The external field switches the configuration of the ferromagnetic layers between antiparallel and parallel giving the change in resistance. Also spin dependent tunneling is making its way into industrial applications in magnetic random access memories (MRAM) [3, 4], which are being developed by several memory manufactures.
Chapter 2

Background topics

In this chapter the basic physical concepts and theories on which this thesis is built is introduced. First the concept of Coulomb blockade and charging energy is introduced with the help of the single electron transistor. The orthodox theory of single electron tunneling and the methods to calculate transport properties are described. The second part deals with spin dependent transport and gives a brief description of ferromagnetism, spin dependent tunneling and spin diffusion. In the last part the combination of spin dependent tunneling and charging effects and spin dependent tunneling and superconductivity is discussed.

2.1 The single electron transistor

The device described in section 1.1 is one of the simplest devices one can build with more than one junction, and also one of the most studied. The current through the junctions in this device is carried by single electrons and the gate electrode located close to the island can influence the transport by electrostatically coupling to the island. Hence this device is named the single electron transistor, or simply the SET. Fig. 2.1 shows a schematic picture together with a circuit diagram of a SET. The important energy scale of the SET is the charging energy, and is given by

\[ E_c = \frac{e^2}{C_\Sigma} (n + n_g)^2, \]  

(2.1)

where \( n \) is the number of excess electrons on the island and \( n_g \) is the induced charge on the island from the voltage sources \( n_g = (C_L V_L + C_R V_R + C_g V_g). \)
Figure 2.1: a) Artistic interpretation of a SET. b) The circuit diagram of a SET.

Figure 2.2: Simulated $I-V$ characteristics of a SET for $V_gC_g/e = 0, 0.25, 0.5$, co-tunneling neglected.
2.1. THE SINGLE ELECTRON TRANSISTOR

Figure 2.2 shows an example of simulated current-voltage ($I-V$) characteristics of a SET (using the orthodox theory and master equation method, see sections 2.1.1 and 2.1.2) for different values of the gate voltage. For small transport voltages there is no current flowing through the SET since any tunneling on or off the island would increase energy. This zero current state for small transport voltages is known as the *Coulomb blockade*. At a certain voltage this blockade is lifted and a finite current can flow through the SET. This voltage is the *threshold voltage*, $V_t$, which depends strongly on the gate voltage. The ability to modulate the threshold voltage and the current with a gate is the most important feature and makes the SET a useful device for applications such as electrometers and transistors. Another important property of the SET is that the modulation is a periodic function of the gate voltage. The period is given by $\Delta U_g = e/C_g$ because changing the gate by this amount induces a charge of one electron on the island and this induced charge may be exactly compensated for by one excess electron on the island.

In Fig. 2.3 the $I-V$ curve for an asymmetric SET is plotted. In an asymmetric SET, where the resistances of the two junctions are different, the Coulomb staircase becomes more pronounced. The Coulomb staircase is the steps occurring at $V = (2n - 1)e/C_\Sigma$. Also shown in the figure is a schematic way of drawing the energies of the leads and island, on the island the energy states are separated by the charging energy and in the leads the energy is set by the voltages. The origin of the steps is that more states on the island becomes available which increases the tunnel current, one state at the threshold voltage $V = e/C_\Sigma$, three states at $V = 3e/C_\Sigma$ and so on.

2.1.1 The orthodox theory

If the two inequalities 1.2 and 1.3 are fulfilled, then the effect of single electrons is observed in circuits of junctions. In order to calculate the transport through tunnel junction devices, the tunneling rates are needed. The approach used to calculate these rates for sequential tunneling uses the Fermi’s golden rule, and is called the “orthodox theory of single electron tunneling” [5, 6, 7]. The orthodox theory is based on the inequality (1.3), which assures that fluctuations of the charge across the junction is small, and hence, the charge makes a good quantum number to describe the circuit. Further, it is assumed that the tunneling process is instantaneous, the time spent by the electron in the barrier is much smaller than any other time scale, and that the energy relaxation time is much smaller than the average time between
Figure 2.3: Simulated $I-V$ characteristics of a asymmetric SET, co-tunneling neglected, and a schematic way of drawing the energies of the leads and island.
2.1. THE SINGLE Electron TRANSISTOR

successive tunnel events. The last assumption makes it possible to describe the electrons by the Fermi distribution function.

The tunneling rate from one of the states \( k \) in the left electrode into an available state \( q \) on the right electrode increasing the number of excess electrons on the right electrode from \( n \) to \( n + 1 \), is given by

\[
\Gamma^{L\rightarrow R} = \frac{1}{e^2 R_t} \int_{-\infty}^{\infty} d\epsilon_k \int_{-\infty}^{\infty} d\epsilon_q N_L(\epsilon_k) N_R(\epsilon_q) \times f_L(\epsilon_k) \left[ 1 - f_R(\epsilon_q) \right] \delta(\delta E_{ch} - (\epsilon_k - \epsilon_q)),
\]

(2.2)

where \( N_{L,R}(\epsilon) \) is the reduced (normalized to the densities of states at the Fermi energy, \( N_0 \), for the normal metal) densities of states of the island and leads, \( f_{L,R}(\epsilon) \) is the Fermi distribution function, \( R_t \) is the tunnel resistance of the junction separating the left and right electrode and the change in charging energy is given by \( \delta E_{ch} = E_{ch}(n + 1) - E_{ch}(n) - eV_i \). The energy of the tunneling process is conserved by the delta function, the change in electron energy \( \epsilon_k - \epsilon_q \) is equal to the change in charging energy \( \delta E_{ch} \). The rate equation, as written here, is valid for both superconducting and normal metal electrodes. In case the electrode is superconducting one should use the BCS density of state \( N_t(\epsilon)/N_0 = \Theta(|\epsilon| - \Delta)|\epsilon|/\sqrt{\epsilon^2 - \Delta^2} \) [8]. The tunnel resistance is given by

\[
\frac{1}{R_t} = 2 \frac{2\pi e^2}{\hbar} N_{0,L} \Omega_L N_{0,R} \Omega_R |T|^2,
\]

(2.3)

and depends on the tunnel matrix elements, \( T \), which are taken to be independent of energy, the density of states (DOS) per volume and spin at the Fermi energy, \( N_{0,L,R} \), and the volumes, \( \Omega_{L,R} \), of the leads. The factor of two in front is due to spin degeneracy, this will change when some or all parts of the device is made of ferromagnetic metals (see section 2.2). For a given temperature, the tunnel rates are only determined by the change in charging energy, which in turn depends on the bias voltage and gate voltage. At low temperatures \( k_B T \ll \delta E_{ch} \) only tunneling events that decrease the charging energy are allowed.

The tunneling of single electrons through single junctions is commonly referred to as sequential tunneling. In addition to sequential tunneling, there is co-tunneling, which is tunneling of several electrons through different junctions at the same time as a coherent quantum mechanical process. The rate of the co-tunneling process is crudely \( (R_K/R)^{N-1}\Gamma \) [7], where \( N \) is the number of junctions the electron tunneled through. Thus when the condition
$R \gg R_K$ is satisfied this rate is small, nevertheless it is clearly visible in experiments when $R \gtrsim R_K$ [9]. Also the case when the tunnel resistance is smaller than the quantum resistance is possible. The orthodox theory does not apply here and the transport must be calculated in a nonperturbative manner, which is done with a path integral formalism [10]. This limit is referred to as strong tunneling.

The orthodox theory as described here assumes that the device is in a very high impedance environment, i.e. the impedance of the environment is much larger than $R_K$. If the impedance seen by the device is finite the environment must be taken into account when calculating the rates, this is done by including a phenomenological quantum description of the environment, in terms of harmonic oscillators, into the Hamiltonian and then get the rates from the Fermi golden rule [11]. For a SET the island is protected from the environment by the junctions, which we already assumed are high impedance, so the rate equation (2.2) gives accurate results. For single junctions, on the other hand, the environment is crucial and charging energy effects are only present if a high impedance environment is provided [12].

### 2.1.2 Calculating the transport properties

The tunneling of electrons is a stochastic process, which means that it is not possible to write down an equation of motion to get the transport properties, i.e. the current as a function of the applied voltage. Instead a statistical approach is needed. Two different statistical methods are commonly used to calculate the current-voltage characteristics in tunnel junction structures, and they give the same result.

**The Master equation**

A master equation can be set up, which connects the tunneling rates with the probability $p(n, t)$ to find the system with $n$ electrons on the island at time $t$ (here a one island device is assumed, $p$ will generally depend on the number of islands of the device, $p(n, m, t)$ for two islands and so on).

$$\frac{dp(n, t)}{dt} = - [\Gamma_{L1}(n) + \Gamma_{IL}(n) + \Gamma_{RI}(n) + \Gamma_{IR}(n)]p(n, t) + [\Gamma_{L1}(n - 1) + \Gamma_{RI}(n - 1)]p(n - 1, t) + [\Gamma_{IL}(n + 1) + \Gamma_{IR}(n + 1)]p(n + 1, t).$$

(2.4)
2.1. THE SINGLE ELECTRON TRANSISTOR

The master equation together with the normalization condition, \( \sum_n p(n,t) = 1 \), gives the probabilities for all \( n \) from the tunneling rates. The first line in Eq. 2.4 describes the decrease of the probability to have \( n \) excess electrons on the island by tunneling from \( n \) to \( n \pm 1 \), and the two last row describes the increase in probability by tunneling from \( n \pm 1 \) to \( n \). In most cases one is interested in the current when the device is biased with a DC voltage. In that case, only the stationary solution (\( dp/dt = 0 \)) of the master equation is required. The rates and probabilities determine the current:

\[
I = e \sum_n [\Gamma_{LI}(n) - \Gamma_{IL}(n)]p(n) = e \sum_n [\Gamma_{RI}(n) - \Gamma_{IR}(n)]p(n),
\]

where the last equality holds for the stationary situation.

The master equation is fast and very well suited for simulation of the transport properties in structures with not too many island or states involved. The number of probabilities that have to be calculated are approximately \( S^N \), where \( N \) is the number of islands and \( S \) is the number of states with finite probability for each island. For a SET, \( N = 1 \) and \( S \) is the order of 10, but for a one dimensional array \( N \) can be over 100, so the master equation rapidly becomes unmanageable for large devices.

Monte Carlo simulations

One other approach for calculating the transport properties is the Monte Carlo method. The Monte Carlo method applied to one dimensional arrays is described by Bakhvalov et. al. [13]. It differs from the master equation approach, where the probabilities for all states involved in the transport are computed, in that only one state and the possible ways to change that state is considered at a time. In the SET, for example, one would be in some state \( \hat{n} \) and then only consider the rates to change that state to \( \hat{n} \pm 1 \) and then randomly choose one of these events, and so on. Or more precisely, the possible tunneling rates out of the present state are computed, then the interval zero to one is divided into segments with widths proportional to the tunneling rates and a random number \( \alpha \) in the interval \([0,1]\) is generated to choose one of the tunnel events, (Fig. 2.4). The probability of preserving the state of the system decays with time as

\[
P(t) = \exp[-\Gamma_\Sigma(t - t_0)], \quad \Gamma_\Sigma = \sum_n \Gamma_n,
\]
where \( t_0 \) is the time of the previous tunneling event leading to the present state and the sum runs over all tunnel rates changing the present state. A random number in the interval \([0,1]\) gives the time between successive tunnel events by inverting the equation for the probability. This procedure is then repeated until a stationary mean value of the current sets in.

For devices containing many islands and/or states the Monte Carlo method is better suited than the master equation approach. For example, a 1D array with \( N \) junctions only \( 2N \) rates changing the current state have to be considered at a time.

### 2.2 Ferromagnetic single electron transistors

Ferromagnetism results from the ability for the electrons in the material to lower their energy by lining up with parallel spins. The Pauli-principle prevents electrons with parallel spins to be close, thus the potential energy (Coulomb energy) is lowered if spins align parallel. The electrons with parallel spins can not have the same wave-vector, again according to the Pauli principle. Thus the reduction of the potential energy costs an increase in kinetic energy by going to larger wave-vector. In the 3d-ferromagnets, Cobalt, Iron and Nickel, the wide d-band is only partially filled and adding electrons in this band does not increase the kinetic energy very much, so there will be a net gain in energy by moving electrons from one spin-band to the other introducing a asymmetry between the spin up and spin down band. In the
Stoner model the asymmetry is a constant shift, \( \Delta = \epsilon_{k,\uparrow} - \epsilon_{k,\downarrow} \), between the spin-bands and the shape of the bands is preserved [14]. In figure 2.5 a schematic picture of a Stoner ferromagnet with the narrow s-band and the wide d-band band is drawn, in reality the band structure is much more complicated.

Figure 2.5: Schematics of the density of state of a ferromagnet showing the narrow s-band and the wide shifted d-band.

The surplus of one spin direction results in a net magnetic moment in the ferromagnets. The direction and magnitude of the magnetization is determined by both material and geometry specific parameters [15]. The anisotropy constant defines a preferred direction for the magnetization and depends on the sample geometry (shape anisotropy) and crystal orientation (crystal anisotropy). The exchange stiffness parameter which is a measure of how strongly spins are coupled and hence gives the energy cost for any gradi-
ent in magnetization. The saturation magnetization is the magnetic moment in a fully aligned sample. Magnetostatic interactions favor a configuration with magnetic domains that have different direction of the magnetization. The magnetic configuration of a sample is governed by the total magnetic energy, which is the sum of the anisotropy energy, exchange energy, magnetostatic energy and the Zeeman energy describing the interaction with an applied field. The net magnetization and specific domain configuration is generally a complicated hysteretic function of the applied field. Fig. 2.6 and 2.7 show the magnetization as a function of an external field, calculated with the micro-magnetic code oommf [16] for two rectangular magnets with different widths. In this figure the effect of shape anisotropy can clearly be seen, the narrow magnet has an almost square hysteresis loop, and a uniform magnetization at zero external field. On the other hand the wide magnet, has a much more complicated switching behavior with large rotation of the magnetization around zero field and additional small hysteresis loops around ±80 mT. The field needed to switch the magnetization depends on the width; 30 mT for the wide magnet and 110 mT for the narrow one. This property is used when it is desired to control the magnetization of two magnetic electrodes independently, one electrode is made wider than the other and thus it will switch at a lower field. Hence there exists a window in external field where the two electrodes are antiparallel. For spin depended transport experiments it is also highly desirable to have a square hysteresis loop as in Fig. 2.7 to ensure a well defined magnetic state of the electrodes.

2.2.1 Spin dependent tunneling

The shift between the bands in a ferromagnet introduces spin dependent conductances, $G_\sigma$, given by the Einstein relation:

$$G_\sigma = e^2 N_0^\sigma D_\sigma,$$

where $N_0^\sigma$ is the spin dependent density of states at the Fermi level, $D_\sigma = 1/3v_F^\sigma l_\sigma$ is the spin dependent diffusion constant, $v_F^\sigma$ is the spin dependent Fermi velocity, $l_\sigma$ is the spin dependent mean free path and $\sigma = \uparrow, \downarrow$ is the spin index. Thus, the current carried by spin up electrons is not equal to the current carried by spin down electrons in a ferromagnet. The current in a bulk ferromagnet is spin polarized with the polarization defined by

$$P = \frac{j_\uparrow - j_\downarrow}{j_\uparrow + j_\downarrow}.$$
Figure 2.6: The hysteresis curve for a $500 \times 300 \times 10 \text{ nm}^3$ magnet, and the magnetization at three points.

Figure 2.7: The hysteresis curve for a $500 \times 70 \times 10 \text{ nm}^3$ magnet, and the magnetization at two points.
The difference of the density of states at the Fermi energy in a ferromagnet also affects the transport from one ferromagnet to another. In a tunnel junction structure with ferromagnetic leads, the tunneling conductance is dependent on the relative orientation of the magnetization of the two ferromagnetic leads. Here it is often assumed that the leads are single domain in the vicinity of the tunnel junction and also that the magnetization of the two leads are collinear, i.e. the magnetization of the left electrode can be either parallel (P) or antiparallel (AP) to the magnetization of the right electrode. Generally the tunnel conductance is a function of the angle between the two electrodes.

The first model of the magnetoresistance for a tunnel junction is due to Julliere [17] and based on arguments by Tedrow and Meservey [18]. The model is based on the assumption that the conductance, for each spin channel, is proportional to the product of the DOS at the Fermi energy, \( N_0 \), at each side of the barrier (see Eq. 2.3). The two spin channels are independent and the total tunnel conductance is the sum of the conductance of each spin channel. Furthermore, Julliere assumed that the density of states at the Fermi energy for each spin band is proportional to the number of conduction electrons in each spin band. Let \( \alpha_L \) be the fraction of electrons in the left electrode whose spin is in the direction of the magnetization and \( \alpha_R \) the corresponding quantity in the right electrode. Then, the spin dependent part of the conductance becomes, in the parallel and antiparallel alignment,

\[
G^P \propto \left[ \alpha_L \alpha_R + (1 - \alpha_L)(1 - \alpha_R) \right] N_{0,L} N_{0,R} \\
= \frac{1}{2} (1 + P_L P_R) N_{0,L} N_{0,R}
\]

\[
G^{AP} \propto \left[ \alpha_L (1 - \alpha_R) + (1 - \alpha_L) \alpha_R \right] N_{0,L} N_{0,R} \\
= \frac{1}{2} (1 - P_L P_R) N_{0,L} N_{0,R}
\]

(2.7)

where the polarization \( P_L = 2\alpha_L - 1 \) and \( P_R = 2\alpha_R - 1 \) is introduced. The polarization is the relative difference between the occupation of the two spin bands. The magnetoresistance (MR) is defined to be

\[
MR = \frac{\Delta R}{R_{AP}} = \frac{R_{AP} - R_P}{R_{AP}} = \frac{2 P_L P_R}{1 + P_L P_R}.
\]

(2.8)

This MR is sometimes called tunnel magnetoresistance (TMR) or junction magnetoresistance (JMR) and can also be defined with \( R_P \) in the denominator [19]. The model described here was the first model to explain spin
dependent tunneling. There exists more realistic theories of spin dependent tunneling which include effects ignored by the Julliere model. Some effects ignored in the Julliere model are: the DOS at the Fermi energy need not be proportional to the total polarization, the wave function can be different for the two spin directions which makes the tunnel matrix elements spin dependent, the DOS close to the barrier can be very different from the bulk density of state. The first attempt to overcome these problems was done by Slonczewski [20] who considered a plane wave description of the polarized electrons tunneling through a barrier. Other, more complicated models exist which take the full band structure into account [21], and it turn out that the band structure and the thickness and height of the barrier does affect the magnetoresistance.

The polarization of a F/N tunnel junction can be rewritten in terms of the tunnel conductance

\[ P = \frac{G^\uparrow - G^\downarrow}{G^\uparrow + G^\downarrow} \]  

(2.9)

This is the usual expression for the polarization of tunnel junctions. This expression correspond to the definition given by Meservey and Tedrow [22] for the polarization of the tunneling electrons in ferromagnet/superconductor (F/S) tunnel junctions. In experiments on F/S tunnel junctions a large in-plane magnetic field is applied, creating a Zeeman splitting between the spin up and spin down DOS in the superconductor. This splitting can be directly seen in the differential conductance of the F/S tunnel junction and the polarization can be extracted. Meservey and Tedrow measured a polarization of 40 % for Iron, 35 % for Cobalt and 23 % for Nickel and recently Monsma and Parkin [23] obtained P = 45 %, 42 % and 31 % for Iron, Cobalt and Nickel respectively using this method.

2.2.2 Spin diffusion

In a ferromagnet/normal metal (F/N) junction the tunnel electrons are spin polarized, which induces a non equilibrium spin density in the normal metal if a current is driven through the junction. The non equilibrium spin density is usually called spin accumulation. The equation governing the motion of non equilibrium spin density, \( n^\sigma \), in a diffusive metal is the so-called spin diffusion equation, and is usually given in terms of the chemical potential
\[ \mu^\uparrow - \mu^\downarrow = (n^\uparrow - n^\downarrow)/N_0. \]

In one dimension the diffusion equation is

\[
\frac{\partial^2 (\mu^\uparrow - \mu^\downarrow)}{\partial x^2} = \frac{(\mu^\uparrow - \mu^\downarrow)}{\lambda_{sf}^2}
\]

(2.10)

where \( \lambda_{sf} = \sqrt{D \tau_{sf}} \) is the spin relaxation length and \( D \) is the spin averaged diffusion constant given by \( 1/N_0 D = 1/N_0^\uparrow D^\uparrow + 1/N_0^\downarrow D^\downarrow \).

The spin diffusion equation can be derived in two ways; Valet and Fert [24] used the Boltzmann equation to show that the Eq. 2.10 is valid if the mean free path is much shorter than the spin relaxation length. They applied the model to describe the CPP-GMR (current perpendicular to the plane-giant magnetoresistance) effect including spin accumulation and spin relaxation. An alternative derivation based on thermodynamic arguments was given by Johnson and Silsbee [25] where equations for the charge, magnetization and heat currents in bulk and through interfaces are derived from the entropy production rate. In analogy to the electrical potential, a magnetic potential is introduced which drives the magnetization current. The key in deriving the spin diffusion equation is that the two spin populations are only weakly interacting, which allows for the two current (spin up and spin down) description of transport [26] and a separate chemical potential for each spin.

### 2.2.3 The F/F/F and F/N/F SET

Spin dependent tunneling described above can also be present in tunnel junction structures where charging effects are present. In these devices there will be an interplay between spin dependent tunneling and charging effects giving rise to new phenomena.

Extensive theoretical studies of SET’s with some or all parts made of ferromagnets have been conducted over the last few years. Various limits have been examined, concerning both the material composition of the SET and different tunnel mechanisms. In the sequential tunneling regime, a SET with all three electrodes made of ferromagnets (F/F/F) has been analyzed, and an enhancement of the MR as well as an MR oscillating with bias voltage was predicted [27, 28]. The SET is treated within the orthodox theory but the tunnel rates become spin dependent through the spin dependent tunnel resistance, in the simplest model the tunnel conductance acquires a spin dependence through the Julliere form (Eq. 2.7). The origin of the oscillating MR is the Coulomb staircase (see Fig. 2.3), the increase in tunnel resistance
from parallel and antiparallel makes the Coulomb staircase more pronounced giving peaks in the MR at \( V = (2n - 1) e/C_\Sigma \).

SET’s with two ferromagnetic outer electrodes and the central island made of a non-magnetic metal (F/N/F) have also been analyzed in the sequential tunneling regime [29, 30]. The antiparallel alignment of the ferromagnetic electrodes introduce a non equilibrium spin distribution in the normal island, which in turn shifts the chemical potential, \( \Delta \mu \), on the island. The shift in chemical potential effectively shifts the charging energy term entering the rate equation, \( \delta E_{ch} \rightarrow \delta E_{ch} \pm \Delta \mu \), where \( + ( - ) \) corresponds to spin up (down). The tunnel resistance in the two F/N junctions are spin dependent: \( R^t_i = 2R_i/(1 + P_i) \), \( R^d_i = 2R_i/(1 - P_i) \), according to the Julienne model Eq. 2.9. The spin accumulation is driven by the spin current, \( I^t_i, \), and relaxed by spin flip scattering, in the steady state a balance is govern by

\[
\Delta E_F N_0 V/\tau_{sf} = [I^t_L - I^t_R] / e,
\]

where \( N_0 \) is the density of states at the Fermi level, \( V \) is the volume and \( \tau_{sf} \) is the spin relaxation time. The spin dependent tunneling rates are plugged into the master equation which is solved self consistently together with the balance equation to obtain the transport properties. In figure 2.8 a) the shift in the chemical potential as a function of voltage is plotted. It is the spin accumulation that mediates the information of the magnetization of one electrode to the other one with a MR as a result (see Fig. 2.8 b). The spin accumulation, and thereby the MR, depends on the island volume and the spin relaxation time through the combination \( \alpha = \tau_{sf} / e^2 N_0 V (R_L + R_R) \).

If the volume is increased and/or the spin relaxation time is very short, \( \alpha \) approaches zero and the spin accumulation and MR vanish. The features in the spin accumulation curve around \( V = 3e/C_\Sigma, 5e/C_\Sigma, \ldots \) are a consequence of the Coulomb stair case (Fig. 2.3). The shift in chemical potential shifts the levels on the island by \( \pm \Delta \mu \), thus the location of the steps are different for the two spin directions.

Beyond the sequential tunneling limit, the MR is found to increase due to co-tunneling and higher order tunneling [31, 32] in F/F/F systems. The MR remained invariant for F/N/F SET’s [33] when co-tunneling was included.

Experimentally, there has been several successful observations of spin dependent transport together with charging effects. Extensive studies of SET’s fabricated of different magnetic materials showing the so called Magneto Coulomb Oscillations have bin conducted [34, 35, 36]. The magneto
Figure 2.8: a) The shift in chemical potential on the island as a function of voltage in a F/N/F SET. The inset shows a schematic of the SET with ferromagnetic leads and normal island. b) The $I-V$ characteristics in the parallel (dashed line) and antiparallel (solid line) state together with the MR (thin solid line). The parameters used are $C_L = C_R$, $R_L = 5R_R$, $P_L = P_R = 0.4$, $\alpha = \tau_{sf}/e^2N_0V(R_L + R_R) = 0.2$ and $k_B T = 0.01e^2/C_{\Sigma}$.
Coulomb oscillation is the oscillation of conductance in a SET with one or more electrodes made of ferromagnet, induced by a Zeeman shift of the Fermi levels for the spin up and spin down electrons when a external magnetic field is applied. Coulomb charging effects together with spin dependent tunneling have also been observed in composite systems. In discontinuous CoFe/HfO$_2$ multilayers [37] showing Coulomb blockade features, an enhanced MR was found. In small Cobalt clusters embedded in tunnel barriers [38, 39, 40] the MR was enhanced and showed oscillations with bias voltage. Arrays of Permalloy/AlO$_x$/Cobalt tunnel junctions [41] also have shown enhanced MR, which was attributed to Coulomb blockade.

### 2.2.4 The F/S/F structure

In superconductors two electrons with opposite spin form a Cooper pair with zero spin [42]. In ferromagnetic materials there is an asymmetry between the two spin orientations. Thus one would expect a competition between superconductivity and the induced non equilibrium spin density in a ferromagnet/superconductor/ferromagnet (F/S/F) structure. A F/S/F sandwich structure was theoretically studied by Takahashi et. al. [43]. The model used a phenomenological Hamiltonian and golden rule arguments to calculate the transport properties in the limit of no spin relaxation, $\tau_{sf} \rightarrow \infty$, and energy relaxation time $\tau_E$ shorter than the time between successive tunnel events $\tau_t$ in the superconductor. (In later articles [44, 45] finite spin relaxation time was included in the model.) The last assumption makes it possible to describe the quasi particles in the superconductor with the Fermi distribution function. It was shown that the antiparallel state produced a nonzero spin accumulation in the superconducting layer, and that the spin accumulation suppressed the gap. The fact that spin accumulation can completely suppress the superconducting gap can give rise to a very large magnetoresistance. In Fig. 2.9 the result of the transport equations from [43] is shown, in 2.9a) the $I$-$V$ characteristics for parallel and antiparallel, together with the magnetoresistance is plotted, and in 2.9b) the gap as a function of the applied voltage in the AP state and in 2.9c) the gap as a function of the non equilibrium chemical potential. As can be seen in the figure, the gap is a multivalued function of the chemical potential (2.9 c) but not multivalued in voltage (2.9 b). This is the case for higher temperatures ($T/T_c > 0.1$), for low temperatures ($T/T_c < 0.1$) the $\Delta(V)$ curve is also multivalued, which makes the $I$-$V$ characteristics multivalued.
Figure 2.9: The transport properties for a F/S/F sandwich structure at $T/T_c = 0.15$. a) The $I-V$ curves and the magnetoresistance. b) The gap as a function of the voltage. c) The gap as a function of the chemical potential.

Experiments by several groups have shown that a strong suppression of the gap follows when a spin polarized current is injected [46, 47, 48, 49, 50] into high $T_c$ superconductors. The model of Takahashi et al. fitted the measured curves of Ref. [47] very well [51]. In high $T_c$ superconductors the energy relaxation time is very short so the condition $\tau_E \ll \tau_l$ is fulfilled.

A single electron transistor with ferromagnetic leads and superconducting island have been investigated in theory by Imamura et al. [52] in the limit of charging energy larger than the superconducting gap, zero temperature and low voltage. They showed that in these limits the gap energy forbids any spin accumulation, but an MR still exists that is mediated by the single quasi particles which carry the current. The MR is, however, reduced from the Jullier value by spin relaxation on the island.
Chapter 3

Experimental techniques

In this chapter the sample fabrication procedure is described and the measurement setup is outlined. The ferromagnet/superconductor/ferromagnet single electron transistors was fabricated at the Nanofabrication Lab KTH, Stockholm. The samples used in the anodization experiments was manufactured in the Swedish Nanometer Laboratory (SnL) at Chalmers University of Technology in Göteborg. All measurements was conducted at KTH, Stockholm.

3.1 Sample fabrication

The electron beam lithography and shadow evaporation technique [53] used to fabricate nanometer sized tunnel junctions and other mesoscopic devices has been used for more than two decades and is well established. Here follows a short description of the major steps in the fabrication process used in this work.

1. Resist spinning: A silicon wafer covered with 1 μm SiO₂ is spin coted with a double layer of positive e-beam resist. The resist used in this work is PMGI SF7 for the bottom layer and ZEP 520 diluted 1:2 in Anisol for the top layer. The bottom layer is spun at 3000 rpm to give a thickness of 400 nm and 6000 rpm gives 63 nm for the top layer.

2. Electron beam lithography The chip is exposed in an electron beam lithography system (Raith Turnkey 150) to write the pattern. The fine structures together with leads and bonding pads is written in one
step. The larger structures, leads and bonding pads (structures with dimensions > 1 \( \mu \text{m} \)), is written with the larges aperture (120 \( \mu \text{m} \)) and the fine structures are written with a 7.5 \( \mu \text{m} \) aperture. The dose used is dependent on the structure but for the larges structures, the ones written with the 120 \( \mu \text{m} \) aperture, a dose of 50 \( \mu \text{C/cm}^2 \) was used. For the fine structures the dose used was between 100 \( \mu \text{C/cm}^2 \) and 2000 \( \mu \text{C/cm}^2 \). To remove the resist in the exposed areas the sample is selectively developed, first the top layer in p-xylene (70 s) and then the bottom layer in MF322:H\(_2\)O 3:2 (\( \approx 225 \) s). The development time for the bottom layer can vary, depending on the structure and the amount of undercut that is required. In Fig. 3.1 the resist spinning, lithography and development steps are schematically drawn.

3. **Shadow evaporation and lift off**: The sample circuit is formed by evaporation of metals using the standard shadow evaporation technique, see Fig. 3.2. Shadow evaporation is the technique where the metals are deposited from two angles forming an overlap structure. The desired metals are e-gun deposited in a UHV system with a base pressure of \( 10^{-9} \) mbar. In this work Aluminum, Cobalt and Titanium was used.

### 3.2 Measurement setup

The transport measurements were conducted in a Oxford Instrument Heliox\(^\text{VL}\) sorbtion pumped \(^3\)He refrigerator with a base temperature of 260 mK. The sample chips were mounted on a commercially available dual in line chip-carrier. Electrical connections were made with an ultrasonic bonder by bonding 25 \( \mu \text{m} \) thick aluminum wire from the chip-carrier to the pads on the chip. The signals were passed to the top of the cryostat by thermally anchored Constantan wires without any filtering. At room temperature homebuilt electronics was employed to amplify and measure the current and voltage in symmetric and asymmetric voltage bias configuration. The measurement and biasing setup electronics are described in references [54] and [55]. The output voltages where digitized by Keithley 2000 DMM mulimeters and sent to a computer via a GPIB connection.
Figure 3.1: A schematic of the resist spinning (a), (b) and (c), e-beam lithography (d) and development (e) and (f) for the two layer resist system.

Figure 3.2: A schematic of the shadow evaporation technique and the two layer resist system (only the top resist layer is shown).
Chapter 4

Results

In the first part of this chapter, both theoretical and experimental investigation of the ferromagnet/superconducting/ferromagnet single electron transistor is presented, followed by an interpretation of the measurements and comparison with the theory. In the second part the theoretical investigation of one dimensional arrays, the problem of random background charges, and the use of a 1D array as a transistor is presented. In the last part an alternative fabrication process of mesoscopic tunnel junction structures by means of anodization of Titanium is presented.

4.1 Spin diffusion experiment

By injecting spin polarized currents from a ferromagnet into a normal metal, a non-equilibrium spin accumulation is induced in the normal metal. The spin accumulation can be detected with a ferromagnetic probe located within the spin diffusion length of the injection point. With this kind of experiment, the spin diffusion length and polarization of the tunneling electrons is determined. The sample is shown in Fig. 4.1, the Cobalt electrodes which are 90, 130 and 190 nm wide and 50 nm thick, overlap a 10 nm thick and 100 nm wide Aluminum strip. The center to center separation of the Co electrodes is 280, 450 and 730 nm. The tunnel junctions have a resistance of 20 kΩ, 14 kΩ and 10 kΩ, the Al strip has a conductivity of $\sigma_{Al} = 1.5 \cdot 10^7 \ \Omega^{-1} m^{-1}$ at room temperature and $\sigma_{Al} = 2.4 \cdot 10^7 \ \Omega^{-1} m^{-1}$ at 4 K. The spin polarized current injected into the Aluminum from the Cobalt electrode induces a difference in the chemical potential between the spin up and spin down elec-
Figure 4.1: The sample for the spin diffusion experiments.

Figure 4.2: The signal measured in the non-local configuration. The numbers corresponds to the numbers in Fig. 4.1.
4.2. THE F/S/F SET

In this section the combined effects of superconductivity, charging and spin dependent tunneling in a ferromagnet/superconductor/ferromagnet single electron transistor (F/S/F SET) are investigated. A theory is developed and employed to model the transport of charge and spin in the device. Experiments on SET’s fabricated with Cobalt leads and an Aluminum island are presented and compared with the theory.
Figure 4.3: a) The single electron transistor consisting of two ferromagnet (F) electrodes and a superconducting (S) island. b) Schematic density of state for ferromagnet (left and right) and superconductor (middle) in the anti-parallel alignment of the electrodes when $V_L = V/2$ and $V_R = -V/2$. 
4.2. THE F/S/F SET

4.2.1 Theory

The model geometry shown in Fig. 4.3(a) is a standard SET geometry with two ferromagnetic electrodes connected to a superconducting island through tunnel junctions. The tunnel resistance is assumed to be much larger than the Quantum resistance. We assume collinear and singled domain magnetization of the two electrodes. The magnetization of the left electrode can be either parallel or anti-parallel to the magnetization of the right electrode. We investigate the regime where the energy relaxation time, $\tau_E$, is shorter than the average time between tunnel events, $\tau_t$. The spin accumulation is assumed to be spatially uniform, as is the case for sub-$\mu$m sized metal islands [63]. If $\tau_E < \tau_t$, the quasi particles thermalize on the island before tunnel out and are described by the Fermi distribution function. The validity of this assumption is discussed in Sec. 4.2.3. The resistance of the tunnel junctions is spin dependent through: $R^\uparrow_i = 2R_i/(1 + P_i)$, $R^\downarrow_i = 2R_i/(1 - P_i)$ (where $i = L$ (left) or $R$ (right) denotes the junction and $P_i$ is the polarization of the tunneling electrodes). The polarization is of the same sign ($P_L P_R > 0$) for the parallel alignment. By letting $P_L \rightarrow -P_L$ (or $P_R \rightarrow -P_R$) for reversed magnetization of the left (or right) electrode, we obtain the resistances in the anti-parallel alignment. Note that the total resistance for each F/S junction is spin independent ($(R^\uparrow_i)^{-1} + (R^\downarrow_i)^{-1} = R_i^{-1}$) as expected for an F/S junction. If the spin relaxation time is longer than the average time between tunnel events, $\tau_{sf} > \tau_t$, we expect a non equilibrium spin accumulation in the superconducting island, which will lead to a shift in the chemical potential, as was the case in the F/N/F SET (Sec. 2.2.3). We also expect an effect on the superconducting gap as for the F/S/F structure treated in section 2.2.4. Thus we need to modify the expression for the tunnel rates to include the non equilibrium shift in the chemical potential and gap, which will both depend on the spin accumulation. The state of the SET will be described by the number of non equilibrium spin up ($n^\uparrow$) and spin down ($n^\downarrow$) electrons on the island. The charge on the island is given by $ne = e(n^\uparrow + n^\downarrow)$ and the spin accumulation by $s = n^\uparrow - n^\downarrow$.

The relation between the number of non equilibrium spins and the shift in chemical potential are related by

$$n^\sigma = N_0 V \int_{-\infty}^{\infty} dE N(E) [f(E - \Delta \mu^\sigma) - f(E)],$$  \hspace{1cm} (4.2)

where $N(E) = \Theta(E - \Delta) \frac{|E|}{\sqrt{E^2 - \Delta^2}}$ is the normalized BCS density of states, $N_0$ is the density of states per spin at the Fermi energy in the normal state,
Figure 4.4: The self-consistent solution of Eq. (4.2) and (4.3). The chemical potential (top panels) and gap (bottom panels) as a function of the spin imbalance (for the case $n^+ = -n^-$). The left panels show the temperature dependence ($N_0 V = 3 \cdot 10^6$ [eV]$^{-1}$) and the right panels show the dependence of volume ($T = 150$ mK).

Figure 4.5: Current voltage characteristics in the P and AP state and $MR = (I_{AP} - I_P)/I_{AP}$ of F/S/F SET. The device parameters are $R_L = R_R = 30$ kΩ, $C_L = C_R = 0.8$ fF, $P_L = P_R = 0.35$, $N_0 V = 2 \cdot 10^6$ [eV]$^{-1}$, $T = 50$ mK, $\tau_{sf} = 1$ ns, $V_g = 0$. 
4.2. THE F/S/F SET

\(f(E)\) is the Fermi distribution function, \(V\) is the island volume, and \(\sigma = \uparrow, \downarrow\) is the spin index. If \(n^\sigma = 1\) this equation reduces to the equation for the odd-even free energy difference \([64]\) used in studies of the parity effect in \(N/S/N\) SET's \([65, 66]\). Equation 4.2 gives the shift in chemical potential on the superconducting island for a given spin accumulation. The shift in the chemical potential influences the gap through the BCS gap equation \([42]\)

\[
\ln \left( \frac{\Delta_0}{\Delta} \right) = \int_0^{\hbar \omega_D} \frac{dE}{\xi} \left[ f(\xi + \Delta \mu^\uparrow) + f(\xi + \Delta \mu^\downarrow) \right],
\]

(4.3)

where \(\xi = \sqrt{E^2 + \Delta^2}\) and \(\omega_D\) is the Debye frequency. For arbitrary spin accumulation on the island, (arbitrary \(n^\uparrow\) and \(n^\downarrow\)), solving equations (4.2) and (4.3) self-consistently determines the shift in the chemical potential, \(\Delta \mu^\sigma\), and the gap, \(\Delta\), uniquely. In Fig. 4.4 the chemical potential and the gap as a function of the spin imbalance, \(s \equiv n^\uparrow - n^\downarrow\), are shown for the case when \(n^\uparrow = -n^\downarrow\). When the spin imbalance increases, there will be a critical value of \(s\) where the gap will discontinuously drop to zero and the chemical potential will drop to a lower value. After the complete suppression of the gap the chemical potential will increase linearly with the slope of \(1/N_0V\) as in the normal conducting case. Note that the gap and chemical potential do not depend on \(n^\uparrow\) and \(n^\downarrow\) only through the combination \(s = n^\uparrow - n^\downarrow\). Different \(n^\uparrow\) and \(n^\downarrow\) having the same \(s\) can produce different \(\Delta\) and \(\Delta \mu^\sigma\).

The transport properties are determined by the rates for tunneling on and off the island. The tunneling rates depend on the charge of the island through \(ne = e(n^\uparrow + n^\downarrow)\), as well as \(n^\uparrow\) and \(n^\downarrow\) separately, through the dependence of \(\Delta\) and \(\Delta \mu^\sigma\) on \(n^\uparrow\) and \(n^\downarrow\). The tunneling rates are also spin dependent through the spin dependent resistances:

\[
\Gamma^{\text{on}, \sigma}_i(n) = \frac{1}{e^2 R_i^2} \int_{-\infty}^{\infty} dE N(E) f(E - \delta E^{\text{on}}_i(n)) \left[ 1 - f(E + \Delta \mu^\sigma) \right]
\]

\[
\Gamma^{\text{off}, \sigma}_i(n) = \frac{1}{e^2 R_i^2} \int_{-\infty}^{\infty} dE N(E) f(E - \Delta \mu^\sigma) \left[ 1 - f(E - \delta E^{\text{off}}_i(n)) \right]
\]

(4.4)

where \(\delta E^{\text{on/loff}}_i(n)\) is the change in the charging energy for tunneling one electron on or off the island through the left or right junction. With the tunneling rates obtained as described above, the Monte Carlo method described in Sec. 2.1.2 is employed to calculate the transport of charge and spin through the F/S/F SET. The spin flip process is accounted for in a phenomenological way by letting \(s \rightarrow s \exp[-(t - t_0)/\tau_s]\), where \(\tau_s\) is the spin flip time. There
Figure 4.6: a) The time average of the spin accumulation, $\langle s \rangle$, and the standard deviation of the spin accumulation $\delta s = [\langle s^2 \rangle - \langle s \rangle^2]^{1/2}$, b) the time average of the gap, $\langle \Delta \rangle$, and the standard deviation of the gap, $\delta \Delta = [\langle \Delta^2 \rangle - \langle \Delta \rangle^2]^{1/2}$, when $V_g C_g/e = 1/2$.

Figure 4.7: Time trace of the gap (solid line) and the spin accumulation (dots) at a bias voltage of 0.49 mV. The parameters are the same as in Fig. 4.5. The dashed horizontal line is the critical spin accumulation for gap suppression.
are several reasons to use the Monte Carlo method instead of the Master equation approach:

- for our island size $n^\uparrow$ can be as large as hundreds and the Master equation would become of great numerical complexity with $N = 2$ and $S = 100$, see Sec. 2.1.2.

- the intertwined dependence of the gap and chemical potential on $n^\uparrow$ and $n^\downarrow$ can be included into the Monte Carlo method directly. Eq’s 4.2 and 4.3 are solved ones and tabulated, and thereby decoupled from the calculation of the tunneling rates.

- both the time dependence and time average can be obtained directly from the Monte Carlo method since the evolution of the system is followed in time.

The drawback of the Monte Carlo method is that it is slow. Good statistics (i.e. smooth curves) are obtained after $10^5$-$10^6$ tunnel events.

An example of the simulated transport characteristics are shown in Fig. 4.5, where the $I$-$V$ curves for the parallel and antiparallel states are plotted together with the magnetoresistance, $MR = (I_P - I_{AP})/I_{AP}$. The effect of gap suppression in the AP state can clearly be seen both in the IV and MR curve. The time average of the gap starts to decrease at approximately 0.5 mV, giving a substantially higher current in the AP state and a maximum MR of 70 %. In Fig. 4.6 a) the time average of the spin accumulation, $\langle s \rangle$, and the standard deviation of the spin accumulation $\delta s = [\langle s^2 \rangle - \langle s \rangle^2]^{1/2}$ are plotted. There is no spin accumulation or fluctuations of the spin accumulation below the threshold voltage, as expected. However, above the threshold voltage the current starts to flow through the SET and spins start to accumulate on the island in the AP state, but not in the P state. The fluctuations of $s$ are larger in the parallel state than in the antiparallel state. A similar effect was found in the F/N/F SET [67]. In Fig. 4.6 b) the time average of the gap, $\langle \Delta \rangle$, and the standard deviation of the gap, $\delta \Delta = [\langle \Delta^2 \rangle - \langle \Delta \rangle^2]^{1/2}$ are shown. In Fig. 4.6 the importance of fluctuations of the spin accumulation is clearly visible. In the parallel state $\langle s \rangle$ is zero but the fluctuations of $s$ are larger than the critical spin accumulation (the critical spin accumulation for this island zise and temperature is around 18) which leads to a partial gap suppression even in the parallel state. In Fig. 4.7 a time trace of the
spin accumulation and the gap are plotted. The spin accumulation fluctuates around the critical value, causing the gap to fluctuate between a finite value and zero, giving \( \langle \Delta \rangle \approx \Delta / 2 = 100 \ \mu \text{eV}. \)

4.2.2 Experimental Results

The F/S/F SET’s, consisting of an Al island separating two Co electrodes as shown in Fig. 4.8, fabricated using e-beam lithography and the two-angle shadow evaporation technique, Sec. 3. The island was 15 nm thick and oxidized in 100 mTorr of O\(_2\) for 2 minutes, to form the tunnel junctions, prior to deposition of 40 nm thick Co. The Co electrodes, 60 and 80 nm wide, where spaced by \( \sim 400 \ \text{nm} \). The difference in width resulted in different magnetostatic shape anisotropy, which determined the switching field of the electrodes. The orientation and length of the Co fingers was chosen so as to minimize the stray fields through the Al due to the open ends, and minimize the effect of magnetization curling in the junction area [68, 69]. The effect of stray magnetic field from the electrodes can strongly suppress the superconductivity if the ends of the ferromagnets are located on top of, or close to, the superconductor [70, 71, 72]. The external field was applied along the Co electrodes, perpendicular to the longer side of the Al island. In Fig. 4.9 the resistance as a function of the external field is shown for two sweep speeds, 4 and 15 Oe/s. Three different effects are visible in these curves: the direct effect of the external field on gap, the external field suppresses the superconducting gap which decreases the resistance and at high fields drives the island normal. A sweep rate dependent variation of the resistance and a sweep rate independent variation of the resistance. The minimum in the resistance at relatively low fields (± 150 Oe) are sweep rate dependent. The change in resistance at the minimum is a factor of three smaller when the field is swept at 4 Oe/s, compared to 15 Oe/s. If the field sweep is stopped at ± 150 Oe the resistance decays to the zero field resistance value in a few seconds. On the other hand, the minimum located around ±1 kOe is sweep rate independent. If the sweep is stopped in this field window and the external field is removed the device remains in this low resistance state. Thus, this latter resistance change is interpreted as to originate from an antiparallel alignment of the ferromagnetic electrodes. In Fig. 4.10 the \( I-V \) characteristics in the parallel and antiparallel state are shown. The curves are measured after preparing the device in one of the states and then removing the external field. From the \( I-V \) and current versus gate voltage data taken in the parallel state the
4.2. THE F/S/F SET

![SEM Image of the F/S/F SET](image)

Figure 4.8: SEM image of the F/S/F SET

Sample parameters are estimated to be: $\Delta = 200 \, \mu\text{eV}$, $R_L = R_R = 42.5 \, k\Omega$, $C_L = C_R = 0.57 \, \text{fF}$ and $C_g = 1 \, \text{aF}$. From the $I-V$ curves in the P and AP state the magnetoresistance was calculated and is shown in Fig. 4.11 for three temperatures.

4.2.3 Interpretation and comparison to the model

Our model for the F/S/F SET does not fit the details of the measured MR, as can be seen by comparing Fig. 4.10 and Fig. 4.5. There are several points where the model and the measured magnetoresistance differ:

- The measured $I-V$ curve in the antiparallel state has a substantially lower threshold voltage than the $I-V$ curve in the parallel state. In the model, this behavior is not possible since spin accumulation is what makes the P and AP state different, and the spin accumulation is proportional to the current, i.e. absent below $V_{th}$ (see Fig. 4.5 and
Figure 4.9: Magnetoresistance of the Al/Co/Al SET measured with two sweep speeds, 4 Oe/s and 15 Oe/s.

- The relatively high temperature, $T = 250$ mK, and large island, $N_0 V \approx 10^7 \text{[eV]}^{-1}$, gives a critical spin accumulation of 540. The current and thereby the voltage needed to produce this spin accumulation would have to be large and hence the maximum MR should be at a voltage much above the threshold voltage, rather than below $V_{th}$.

- The temperature dependence in Fig. 4.11 does not agree with the predicted one. The critical spin accumulation increases with increasing temperature, hence the maximum MR should occur at a higher voltage for a higher temperature, not at the same voltage.

One possible explanation for these discrepancies is that the model only takes into account sequential tunneling, in the experiment the actual mechanism for transport below and at the threshold voltage is co-tunneling. Can
co-tunneling produce enough spin accumulation to suppress the gap? For an F/N/F SET co-tunneling was included [33] and it was shown that co-tunneling did not produce more spin accumulation than sequential tunneling. The co-tunneling current is small, of the order of 1 pA. Thus, it is unlikely that the small current carried by co-tunneling can produce enough spin accumulation to affect the gap. One other possibility is Andreev reflections. Below the threshold voltage, the Andreev tunneling mechanism can carry current. One electron tunnels, a hole is reflected, and a Cooper pair is formed on the island. The reversed process occurs in the other junction. In this process only Cooper pairs are involved in the transport on the island and thus no spin accumulation can occur.

A second assumption of the model is that the energy relaxation time is shorter than the average time between tunnel events. If the opposite was the case it would not explain the MR since it has been shown that no gap
Figure 4.11: $MR = (R_{AP} - R_P)/R_{AP}$ of the Al/Co/Al SET.
suppression from spin accumulation is expected in an F/S/F structure [73] in this limit. It is, in fact, probable that the energy relaxation time is comparable to or longer than the average time between two tunneling events when the island is made of Aluminum having $\tau_E(Al) \approx 10^{-8}$ s [74]. Other superconductors can have much shorter energy relaxation times, for example Lead where $\tau_E(Pb) \approx 5 \cdot 10^{-11}$ s [74].

Other reported experiments on F/S/F SET’s [75, 76, 77] are faced with even greater interpretational difficulties [78]. For example, no stable anti parallel state was observed, large fringing fields through the superconducting island were present, and the same MR was observed for the inverted S/F/S device [79]. More experiments are needed to resolve these controversies.

4.3 One dimensional arrays

In this section the theoretical work on one dimensional (1D) arrays is presented. First is a brief overview of the theory of 1D arrays, followed by the investigation of the performance of the 1D arrays as a transistor. Presented last in this section is a study of the ability to compensate for random background charges in the 1D array by the reorganization of electrons between the islands.

4.3.1 Theory of the 1D array

The one dimensional (1D) array consists of $N - 1$ small islands connected in series by tunnel junctions. Each junction is characterized in terms of its resistance $R_i$ and capacitance $C_i$. It is assumed that the condition $R \gg R_K$ holds for each junction so that the orthodox theory applies, and the rate equation (2.2) is valid. Here the charging energy used is the energy change in the whole circuit which is the “global” view of tunneling, as opposed to the “local” view where only the difference in charging energy of the single junction before and after tunneling is considered [11]. As for the single electron transistor, the gate voltages $V_{g,i}$ is coupled through the capacitances $C_{g,i}$ to each island. A transport voltage is applied across the array by the external voltages $V_L$ and $V_R$. An equivalent circuit is shown in Fig. 4.12. The charge and potential on island $i$ are denoted by $q_i$ and $\varphi_i$ ($\varphi_0 = V_L$ and $\varphi_N = V_R$). The charge configuration of the array is the vector $\vec{q} = (q_1, q_2, \ldots, q_{N-1})$. To be able to calculate the tunneling rates through the junctions of the array
it is necessary to derive an expression for the free energy of the 1D array. A simple expression for the free energy can be found in terms of the potentials on the islands:

\[ F = \frac{1}{2} \sum_{i=1}^{N} C_i (\varphi_i - \varphi_{i-1})^2 + \frac{1}{2} \sum_{i=1}^{N-1} C_{g,i} (\varphi_i - V_{g,i})^2 - V_L Q_L - V_R Q_R - \sum_{i=1}^{N-1} V_{g,i} Q_{g,i}. \]  

(4.5)

Here \( Q_L \) and \( Q_R \) are the induced charge on the first and last capacitor of the array, and \( Q_{g,i} \) is the induced charge on each gate capacitor, and \( m_R (m_L) \) is the number of electrons that have tunnelled into the array through the right-hand (left-hand) side. When calculating tunneling rates one requires the difference in free energy when one electron tunnels through one junction in the array. Such an expression is particularly simple in terms of the potential [13]:

\[ \Delta F = F' - F = \frac{e}{2} [(\varphi_{k+1} + \varphi'_{k+1}) - (\varphi_k + \varphi'_{k})], \]  

(4.6)

giving \( \Delta F \) when one electron tunnels from island \( k \) to island \( k \pm 1 \). To be able to use this expression when calculating the tunneling rates, the relation between the charges on the islands and the potentials is needed. This is a straightforward electrostatic problem, conservation of charge gives the relation between the charge on each island and the surrounding potentials. The
standard way is to only take the capacitance between neighboring islands and gate capacitance into account, the so called tridiagonal model, as shown in figure 4.12. Taking all capacitances into account only gives small corrections to the tridiagonal model [80]. The charge on island \( i \) is thus the sum of the charges on the three nearby capacitor plates:

\[-C_i \phi_{i-1} + (C_i + C_{i+1} + C_{g,i}) \phi_i - C_{i+1} \phi_{i+1} = q_i \quad (4.7)\]

This island charge \( q_i \) consists of the number of electrons or holes that tunneled onto the island \((n_e)e\), the background charge induced on the island \((q_{0,i})\) and the gate induced charge \((n_{g,i}e = C_{g,i}V_{g,i})\). The number of charges on an island \( n_i \) can only take integer values, but the background charge and charge induced by the gate is a continuous variables. We will return to the background charge in sec. 4.3.3. Until then we can imagine that the background charges are zero. The equations above are valid for general arrays with arbitrary junction capacitances and resistances. From now on we restrict ourselves to homogeneous arrays, where all junctions have equal capacitances and resistances. For homogeneous arrays the equation system 4.7 can be inverted analytically [81] and it is thus possible to derive an expression for the potential as a function of the charges, \( \bar{\phi} = C^{-1} \bar{q} \). The components of the symmetric matrix \( C^{-1} \) are

\[ C^{-1}_{ij} = \frac{e \cosh(N - |i - j|)\lambda - \cosh(N - i - j)\lambda}{2 \sinh \lambda \sinh N\lambda} \quad (4.8)\]

where \( \lambda = \text{arch}(1 + C_g/2C) \). From this equation we get the soliton like potential from one excess electron in an initially neutral array, as shown in Fig. 4.13. The parameter \( \lambda \) introduced in ref. [13] is related to the width of this soliton, \( \lambda^{-1} \) being the so called soliton length. The soliton length is the screening length, giving the distance over which the potential from one excess electron in an infinite array is screened to \( e^{-1} \) of its value by the freely moving charges in the ground plane. The soliton length is thus the number of islands over which charges in the array have a direct influence on the potential. When \( C_g \ll C \) the expression for the inverse soliton length reduces to \( \lambda = \sqrt{C_g/C} \). A weak gate coupling thus gives a long soliton and a strong gate coupling \((C_g \approx C)\) gives a short soliton, \( \lambda \approx 1 \).

In figure 4.14 the current voltage characteristics of a 50 junction array are plotted for two different lengths of the soliton, \( \lambda = 0.2 \) and \( \lambda = 0.01 \). These curves were simulated in the case \( T = 0 \) using the Monte Carlo method
described in section 2.1.2. The simulated $I$-$V$ curves of arrays resemble the ones for the single electron transistor, with a Coulomb blockade region and the sharp onset of current at the threshold voltage. As can be seen, the shape of the $I$-$V$ curves for small currents depends strongly on $\lambda$. Also plotted in this figure is the line $I=V/(NR)$ which is the slope of the $I$-$V$ curves for large voltages $V \gg V_t$.

### 4.3.2 Gate effects in 1D arrays

In the single electron transistor we saw that the $I$-$V$ characteristics were strongly effected by the capacitively coupled gate. In 1D arrays, gates can also be coupled to one or more islands to effect the operation. For example, in the turnstile configuration [82] one has a gate coupled only to the middle island of the array. By applying an oscillating signal to that gate one can
4.3. ONE DIMENSIONAL ARRAYS

Figure 4.14: Simulated $I-V$ characteristics of a 50 junction array of tunnel junctions for two different soliton lengths at $T = 0$. The straight line is $I=V/(NR)$.

make one electron go through the array per cycle [83]. A more complicated configuration is the single electron pump, where there is one gate per island and every gate is individually controlled. In this setup one applies a pulse to the gates one after the other, so the gate pulse travels through the array with one electron following the pulse. Thus, the electron is “pumped” through the array [82, 84]. The single electron pump can be very accurate and is being used to implement a new capacitance standard [85]. The 1D array can also be used as a trap for electrons if one of the tunnel junctions at the end is replaced by a gate capacitor. It is possible to trap electrons by applying a gate voltage to pull one electron into the array, and then hold the electron for long times in the array with no gate applied [86, 87]. One dimensional arrays can also be used as thermometers [88]. In the case considered here the gate voltage is uniformly applied ($V_{g,i} = V_g$ and $C_{g,i} = C_g$). As was the case for the SET, the current voltage characteristics are strongly affected
near the threshold. In Fig. 4.15 the threshold voltage is plotted as a function of the induced gate charge for different soliton lengths. One sees that the threshold is a complicated function of the gate, and that the amount of modulation depends on $\lambda$. The irregular pattern obtained is due to pinning of electrons or holes in the array by the applied gate. This trapping of integer charges is more frequent when the soliton is long. The gate modulation is periodic in the induced gate charge if the gate voltage is uniformly applied. The period is one electron induced on each island, as was the case with the SET. In figure 4.15 one sees that the threshold voltage for zero gate voltage gets larger with increasing soliton length. Further it can be seen that the threshold is more sensitive to the gate when the soliton length is large.

The initial linear decrease of $V_I$ with $n_g$ can be examined analytically because the gate voltage is not large enough to pin any charges inside the array. When no charges are localized in the array (electrons, holes, or background charges) it was shown in ref. [81] that the threshold voltage is determined by the tunneling rates through the two end junctions. With the expression for the inverse capacitance matrix (Eq. 4.8) it is possible to find the relevant potentials to calculate the tunneling rate through the end junction

$$\varphi_1 = \frac{e}{C} \left[ C^{-1}_{1,1} (1 + CV_L/e) + n_g \sum_{j=1}^{N-1} C^{-1}_{1,j} \right],$$

$$\varphi'_1 = \frac{e}{C} \left[ C^{-1}_{1,1} V_L C/e + n_g \sum_{j=1}^{N-1} C^{-1}_{1,j} \right],$$

$$\varphi_0 = \varphi'_0 = V_L.$$

Here it has been assumed that a hole will tunnel in from the left, which corresponds to $V_L > 0$, $V_R = 0$ and $V_g < 0$. The tunneling rates are obtained from the difference in free energy (Eq. 4.6). Finally, it is possible to obtain the threshold voltage as a function of $n_g$ by setting the tunneling rate to zero and solving for $V_L$:

$$V_I = \frac{e}{C} \left[ \frac{\sinh(N-1)\lambda}{\sinh N\lambda - \sinh(N-1)\lambda} - \frac{\sinh[N/2(N-1)\lambda] \sinh N\lambda/2}{\sinh^2 \lambda/2 \cosh(N - \frac{1}{2})\lambda - n_g} \right]. \quad (4.9)$$

The first term in the brackets gives the threshold voltage when the gate voltage is zero and the second term in the brackets gives the initial linear decrease in threshold voltage as a function of $n_g$. This prefactor of the $n_g$
term explains the very steep slope of the decrease in threshold voltage, for a large soliton length and/or number of junctions. From the first term one obtains a threshold voltage for zero gate voltage which grows with \( N \), but in the limit \( N \lambda \gg 1 \) is given by

\[
V_t(N \lambda \gg 1, n_g = 0) = \frac{e}{2C}(e^\lambda - 1)^{-1}.
\]  

(4.10)

![Graph](image)

Figure 4.15: The effect of the gate on the threshold voltage for an array with no background charges present and \( N = 100 \), for four values of \( \lambda \).

**The array as a transistor**

The gate voltage dependence with a large threshold voltage modulation implies that, for appropriate values of \( N \) and \( \lambda \), an array would make a good transistor or switch. To use the array with uniform gate coupling as a switch would be good because of the initial steep dependence of \( V_t \) on \( n_g \), and the wide region with a weaker dependence. The sensitivity to gate voltage is greatest when \( N \) is large but \( N < \lambda^{-1} \). In this case the threshold voltage is large at zero gate voltage, and the gate drives the threshold almost to zero.

Eq. (4.9) can be used to calculate the voltage gain of the uniform array. If the array is current biased very close to zero, and it is assumed that the gate voltage drives the threshold voltage to zero, the voltage gain is given by

\[
\eta = \frac{\mathrm{d}V_{\text{out}}}{\mathrm{d}V_g} \approx \frac{C_g}{e} \frac{\mathrm{d}V_t}{\mathrm{d}n_g} = 4 \frac{\sinh \left[ \frac{1}{2} (N - 1) \lambda \right] \sinh N \lambda / 2}{\cosh (N - \frac{1}{2}) \lambda}.
\]  

(4.11)
The gain is plotted in Fig. 4.16 for different values of \( \lambda \). The gain approaches 2 when \( \lambda N \gg 5 \). This behavior resembles the voltage gain in the uniform single electron transistor where one needs \( C_g \gg C \) to get a maximum gain of 2.

![Figure 4.16: The voltage gain near zero gate voltage of an array plotted versus \( N \) for different values of \( \lambda \). The analytic expression is given in Eq. 4.11 valid for arbitrary \( N \) and \( \lambda \).](image)

4.3.3 Background charges in 1D arrays

We now return to the effect of background charges on the transport properties of the 1D array. The background charges are caused by charges being trapped in the substrate or in the tunnel barriers, thereby inducing an excess charge on the islands, \( q_i \rightarrow q_i + q_{0i} \). The background charge \( q_{0i} \), is between \(-e/2\) and \( e/2 \) because any value outside this interval would be compensated for by one electron or hole tunneling on to the island. The background charges may be static, causing a shift of the device operation point, or dynamic, causing noise in the device [89, 90, 91]. The effect of background charges has been observed in experiments with small arrays, the influence of the background
4.3. ONE DIMENSIONAL ARRAYS

charge shows up as an offset in the current versus gate voltage curves [92]. It has also been shown that the threshold voltage could be maximized by tuning gates to each island and thereby compensating for the background charge [93]. In experiments with SET’s, the current versus gate voltage curves showed jumps which was attributed to the movement of background charges [94], the interpretation was that the gate caused background charges to move. In single electron traps, the observed discrepancy between experiments and theory was likely to be due to random background charges [87]. Theoretical studies of the effect of random background charges in one dimensional arrays have examined how the threshold voltage [95, 96, 97, 98] and noise properties [99] are affected when background charges are included.

Background charge distributions

Starting from a random background charge configuration tunneling events can, under certain conditions, lower the free energy by adding or rearranging electrons or holes. This possibility to relax the random potential variations depends on the length of the array, the soliton length, and on the actual configuration of the background charges. In figure 4.17 the potential before and after relaxation is shown for two different values of λ. One can see by looking at figure 4.17 that the ability to reduce the potential variation by moving integer charges, depends strongly on the soliton length. For a short soliton length the potential is nearly unchanged by the relaxation process (Fig. 4.17a). However for a long soliton length (Fig. 4.17b) we can see that the relaxation reduces the variation of the potential. The potentials plotted in figure 4.17 for different λ are normalized by the threshold voltage, since the threshold voltage is a measure of the Coulomb blockade strength.

The random background charge configuration can be described by two parameters: the number of islands having an induced charge \( q_{0,i} \neq 0 \), the density of induced charges \( \rho \in \{0, N-1\} \), and the amplitude \( A \) of the induced charges \( q_{0,i} \in \{-Ae, +Ae\} \) with \( 0 \leq A \leq 0.5 \). A maximally disordered array would have \( \rho = N - 1 \) and \( A = 0.5 \). In Fig. 4.18 the standard deviation, std \( \varphi = \sqrt{\langle \varphi^2 \rangle - \langle \varphi \rangle^2} \), of the potential along the array after relaxation is plotted versus \( \rho \) and \( A \), \( \langle \ldots \rangle \) denotes averaging over 1000 realizations of the background charge configuration. Because we have uncorrelated background charges, \( \langle \text{std } \varphi \rangle \) is a good measure of the roughness of the potential. It is also a measure of the electrostatic energy in the array, because the electrostatic energy is proportional to the gradient of the potential squared. Thus,
Figure 4.17: The potential in a 50 junction array as a function of the island number for \( \lambda = 1 \) and \( \lambda = 0.01 \). The solid line is before the free energy has been minimized by single charge tunneling, and the dashed line is after minimizing.

lowering the energy means smoothing the potential. In figure 4.18 \( \langle \text{std} \ \varphi \rangle \) is plotted as a function of \( A \) and \( \rho \) for different \( \lambda \) and \( N = 100 \). Here it can be seen that the smoothing of the potential becomes more effective when the soliton length increases. When the soliton length is comparable to the array length the value of \( \langle \text{std} \ \varphi \rangle \) is weakly dependent on \( \lambda \).

**Gate modulation with background charges present**

In figure 4.19 the gate modulation for an array with random background charges is shown. Comparing with the gate modulation in the absence of background charges (Fig. 4.15) it can be seen that the presence of background charges have a drastic effect on the gate modulation. The large peak and high sensitivity around zero gate voltage disappears, and the largest modulation can occur near any value of \( n_g \). The maximum modulation \( (V_i|_{\text{max}} - V_i|_{\text{min}}) \) also becomes smaller than in the no-background-charge case. In Fig. 4.20 the maximum modulation (the brackets denote averaging over 1000 background charge configurations) is plotted as a function of number of junctions for different values of the soliton length. The error bars plotted for several points give the standard deviation of the modulation, or the spread in modulation depending on the particular configuration of background charges. As in the
4.3. ONE DIMENSIONAL ARRAYS

Figure 4.18: The mean value of the standard deviation of the potential in the array of 100 junctions for different values of $\lambda$.

In the no-background-charge case, the modulation becomes larger with increasing $N$ and $\lambda^{-1}$.

Unfortunately, the ability to smoothen the random potential due to random background charges by rearranging electrons, does not remove the problem of background charges. The long soliton length needed to reduce the random potential also makes the array more sensitive, and thus the two effects cancel each other. This means that the use of the array as a transistor and the gain behavior near zero gate voltage discussed in the previous section, is destroyed by random background charges. The simulations show that it is still possible to modulate the threshold voltage with gate voltage when background charges are present, however the modulation changes, becoming strongly dependent on the actual background charge distribution.
Figure 4.19: The effect of the gate on the threshold voltage for an array with background charges present and \( N = 100 \) for four values of \( \lambda \).

Figure 4.20: The average modulation of the threshold voltage plotted versus \( N \) for various \( \lambda \). The error bars give the standard deviation calculated from 1000 random background charge configurations. The solid lines is the modulation without background charges present.
4.4 Anodization

In this section the fabrication of nanostructures by anodization is described. Transport measurements of the fabricated structures are presented together with an interpretation of the results.

Nanofabrication by anodization

The anodization process refers to the growth of oxide when a metal is submerged in an electrolyte and is anodically biased with respect to a counter electrode. If, as in our case, Titanium (Ti) is anodized, then Titanium oxide will grow on the surface of the metal, thereby reducing the amount of metal. The type of oxide that will grow (and the crystal structure) depends on the type of electrolyte used, the temperature and whether the anodization is done by applying a current or voltage [100]. Anodization is most commonly used to obtain a protecting oxide film on metals. In our case the objective of anodization is the reduction of metal to oxide and fabrication of a tunnel barrier or weak conducting link.

The technique of fabricating nanostructures by means of anodization has been utilized by many groups. In aluminum SET’s the method of anodization was used to reduce the dimensions beyond the limits of EB lithography [101], which allowed room temperature operation [102]. Another anodization technique is the oxidation of thin Ti surfaces using a scanning tunneling microscope (STM) tip [103, 104, 105]. With this so called nano-oxidation technique the pattern and tunnel barriers are written directly with the STM tip by oxidizing through the metal. With this approach room temperature operation of SET’s has also been achieved [106, 107, 108]. Niobium Josephson junctions are commercially fabricated by anodizing a prefabricated trilayer sandwich through a photo lithography mask [109]. The methods described above are used to miniaturize devices with prefabricated tunnel junctions, or making the tunnel junctions by anodization. A third method is to anodize prefabricated nanostructures without any tunnel barriers. This has been done with Nb nanostructures [110] where charging effects could be seen. Silicon wires subjected to dry thermal oxidation also showed Coulomb blockade effects [111]. In this work the last method is used to fabricate nanoscale devices showing charging effects, by anodization of continuous Titanium strips.
**Fabrication of the samples**

The samples are fabricated using electron beam lithography and the shadow evaporation technique described in chapter 3. In this work two different sample geometries are examined. The first is a 100 nm wide and 30 nm thick wire and the second is a similar wire with two weak links. The two EB lithography masks are schematically drawn in Fig. 4.21 a) and b). The only difference between the two masks is that there is a suspended bridge in the top resist layer in b). By first evaporating Ti at an angle $\alpha$ from the normal incidence the Ti is deposited halfway under the top resist layer (the suspended bridge) and then evaporating at an angle $-\alpha$ a shift is caused. The resulting structure viewed from the side is shown in Fig. 4.21 c). The hanging bridge was 100 nm wide and the shift was such that the weak links were separated by approximately 100 nm. Hereafter the samples with these two weak links will be referred to as **SET-like samples**, because of the resemblance of the SET with one island separated from the leads by the two weak links. And samples without weak links will be referred to as **wire-like samples**.

**The anodization mask**

After complete lift off a new layer of 2 $\mu$m thick resist was spun on the chip and subsequently exposed in a second step of e-beam lithography. This step defined the anodization mask (Fig. 4.22) used for the anodization process. The mask covered the whole chip except for an opening over a part of the Ti and the counter electrode, this opening will be called the anodization window. The anodization window was approximately 3 $\mu$m wide. In this step it was important to use mark detection and pattern alignment to get the window in the right place over both the Ti nanostructure and the gold counter electrode. The resist used for the anodization mask was relatively thick to prevent dielectric breakdown of the resist during the subsequent anodization process.

**The anodization setup**

The setup for the local anodization of nanostructures was first used by Henning *et. al.* [110] to fabricate Niobium nanostructures. A sketch of the experimental setup for the anodization of the Ti nanostructures is shown in figure 4.22. The electrochemical cell is built up by the Ti structure, a counter electrode and a drop of electrolyte. The integrated on-chip cathode close to
Figure 4.21: Schematics of the EB pattern, a) shows the mask for the wire-like samples and b) the mask for the SET-like samples. c) schematic of the resulting structure of the SET-like sample, viewed from the side.
the structure to be anodized has the advantage that there is no need to place an external cathode close to the structure and that only a very small (Ø 1 mm) drop of electrolyte has to be used. Only the electrical contacts to the pads on the chip have to be made. The electrolyte used for all samples is acetic acid diluted 3:1 with deionized water.

With this setup the resistance was monitored while the anodization proceeded, enabling in situ control of the resistance. The anodization voltage was applied by a Yokogawa 7651 voltage source and the resistance was measured by a lock-in technique. We used a Stanford SR830 DSP lock-in amplifier in current measuring mode to measure the current through the sample. A sine shaped voltage excitation with an amplitude of 0.3 mV and a frequency of 1 kHz was applied to the sample. The anodization voltage and current through the sample was logged by a computer via a GPIB connections.

4.4.1 Anodization results

Prior to anodization both SET- and wire-like samples displayed a resistance in the range 20–30 kΩ. The subsequent behavior when anodized was the same for the two sample types. The resistance started to slowly increase irreversibly when the anodization voltage exceeded a couple of volts and there was a strong correlation between the applied anodization voltage and the rate of change in the resistance. The measured resistance dynamics during anodic oxidation is a complicated function of the applied voltage and time for a given sample geometry and metal thickness. In figures 4.23 and 4.24 examples of the anodization of Ti nanostructures are shown. The resistance of the sample is plotted as a function of the anodization voltage and time. In both figures the strong correlation between the applied voltage and the rate of change of the resistance can be seen, and that this correlation extends over all resistances examined (20–400 kΩ). One also sees that the rate of change in the resistance goes almost to zero when the anodization voltage is set to zero (Fig. 4.23). In Fig. 4.24b the resistance was measured for forty hours after the voltage was turned off, during which the resistance continued to increase, albeit with a continuously decreasing rate. This behavior was seen in all samples: we stopped at some value of resistance and after a couple of days the resistance had increased, but did not change thereafter. This subsequent change in resistance depended on how high in resistance the sample was anodized, and on the sample type. In wire-like samples the change was not as dramatic as in the SET-like samples. For the wire-like
samples the post-anodization resistance change was at most 30% (300 kΩ → 405 kΩ). For SET-like samples the change was greater, for example 70 kΩ → 50 MΩ and 310 kΩ → infinite zero bias resistance. Sometimes the resistance suddenly started to increase exponentially during the anodization process. This behaviour was frequently observed when anodizing to higher resistances (≥ 400 kΩ). As soon as the sample went into this very rapid increase in resistance, the process could not be stopped and the sample went insulating. Before the exponential increase of resistance sets in (below 400 kΩ) the resistance could be changed in a controlled manner.

The behavior described above can be qualitatively understood in terms of
Figure 4.23: Example of anodic oxidation of a wire-like sample anodized to 300 kΩ: total resistance as a function of the applied voltage and time.

The simple picture of an oxide film being grown at the interface between the Titanium and the electrolyte [112]. The anodization voltage sets up an electric field in the present oxide causing oxygen ions to diffuse through the oxide and thereby increasing the oxide film thickness. The observed dependence of the resistance on voltage and time is then plausible. The resistance increase after the anodization voltage is set to zero can be explained by the oxygen ions sitting halfway in the oxide when the anodization voltage is turned off, since they will continue to diffuse through the oxide and further increase the oxide film. A possible approach to avoid this increase in resistance after anodization might be to reverse the polarity of the applied voltage instead of setting it to zero, thereby driving the oxygen ions out of the oxide.

4.4.2 Results

In this chapter the results of the transport measurements on the anodized Ti nanostructures are presented. The measurements on SET-like samples
and wire-like samples are presented. In the last section a discussion and interpretation of the results is given.

**SET-like samples**

All anodized SET-like samples showed a nonlinear $I-V$ characteristic at low temperatures and some even at room temperature. Figure 4.25 to 4.27 show examples of $I-V$ curves for three SET-like samples anodized to different resistances. The first one (Fig. 4.25) is the sample from Fig. 4.23 which was anodized to 30 kΩ and then monitored for 40 hours after anodization. This relatively low resistance sample had a perfectly ohmic behavior at room temperature, but as the temperature was decreased below 150 K, a nonlinearity in the $I-V$ characteristics appeared. For the second sample, shown in
Fig. 4.26, the anodization was stopped at 70 kΩ after which the resistance increased to \( \sim 50 \) MΩ. As can be seen in Fig. 4.26, this sample has a pronounced nonlinear \( I-V \) characteristics at room temperature and at 4.2 K. The third sample shown in Fig. 4.27 was anodized to 310 kΩ. This sample displayed a large (10 V) zero current state at room temperatures. Comprising these three samples we see that there is a strong correlation between the anodized zero bias resistance and the extent of the nonlinearity in the \( I-V \) characteristics.

![Figure 4.25: Temperature dependence of the \( I-V \) characteristics between 10 K and room temperature for a SET-like sample anodized to a resistance of 30 kΩ.](image)

All samples measured had a gate electrode close to the anodized region, but the applied gate voltage had no effect on the \( I-V \) characteristics for the SET-like samples. This lack of a gate effect might indicate that the observed nonlinear \( I-V \) curves in these SET-like samples is not only due to charging (Coulomb blockade) effects.
Wire-like samples

All the wire-like samples showed perfect Ohmic behavior at room temperatures. This might be due to the fact that none of these samples had a zero bias resistance higher than 410 kΩ at room temperature. However, as the temperature was decreased to below 5 K the $I$–$V$ characteristics began to display nonlinearities around zero bias, and for temperatures around and below 1 K a zero current state for small bias voltages was evident. A typical temperature dependence of the $I$–$V$ curve for wire-like samples is exemplified in fig 4.28, where one can clearly see the nonlinearity develop as the temperature is lowered. It can also be seen in this figure that the current voltage relation is almost linear above the threshold voltage.

A closer examination of the $I$–$V$ curves revealed that the current jumps at some values of the voltage and the current corresponding to the two different sweep directions can differ by a small amount. Biasing the wire at a fixed
Figure 4.27: $I$–$V$ characteristics of a SET-like sample with infinite room temperature resistance.

voltage showed that the current was randomly jumping between different values. Consequently the $I$–$V$ characteristics exhibited occasional discontinuities and hysteresis. This behavior is shown in Fig. 4.29 where three selected $I$–$V$ curves with large and frequent switching and hysteresis are plotted. It can be seen that the system jumps between a number of different branches with equal slope in the conducting regime. This is a strong indication that charges moving in the vicinity of the wire have a large effect on the conduction.

Background charges changing the conduction of the wire suggest that modulation of the $I$–$V$ characteristics with a gate electrode should be possible. In Fig. 4.30 $I$–$V$ curves for different gate voltages are shown. A clear modulation of the current can be seen and the threshold voltage changed by a factor of two as the gate potential was varied between -2 and +2 V. Comparing figures 4.29 and 4.30 shows that the magnitude of the switching and the gate controlled change of the $I$–$V$ characteristics is of the same size.
Figure 4.28: Temperature dependence of the $I-V$ characteristics between $T=4.5$ K and 0.265 K for a wire-like sample. The curves have been offset by 1 nA for each temperature.

Figure 4.29: Three $I-V$ curves showing the jumping and hysteretic behavior.
Figure 4.30: $I-V$ characteristics for different gate voltages at $T=265$ mK.

The described switching behavior coexisted with the gate modulation, preventing reproducible gate characteristics. One could however see that the current changed continuously (between jumps) when biasing the wire at a fixed voltage and sweeping the gate.

4.4.3 Interpretation

In this section a interpretation of the transport measurements is given. Since the behavior of the wire-like and SET-like samples was quite different we treat them separately.

SET-like samples

We have seen that the SET-like samples have a highly nonlinear $I-V$ characteristics with a zero current state for sufficiently low temperatures. However, the observed data does not provide sufficient experimental evidence to unam-
4.4. ANODIZATION

biguously attribute the features in the $I-V$ curves to single electron charging effects. Firstly, we were not able to modulate the current with a capacitively coupled gate, which would be a characteristic signature of single charging effects. Secondly, as can be seen from the $I-V$ curves the large voltage behavior is not linear but curved, at least in the range examined here. This is not what one would expect either theoretically [13] or experimentally [113, 114]. For an array of tunnel junctions the asymptotic current voltage relation should be linear (Fig. 4.14). The nonlinear $I-V$ curves might be due to barrier suppression effects like Fowler-Nordheim tunneling [115].

The wire-like samples

As already suggested, the transport behavior of the anodized wire-like samples can be interpreted in terms of a Coulomb blockade model. The $I-V$ characteristics of the anodized wires shown in Fig. 4.28 show features that are typical of transport behavior observed in one dimensional arrays of tunnel junctions [116]. The measured $I-V$ curves also compare favorably with the simulated $I-V$ curve (Fig.4.14) for a one dimensional array. A question arises about the nature of the tunnel junctions and islands. There are no designed barriers or junctions in the anodized wire-like samples. Thus the island formation must be due to some spontaneous mechanism during the anodization process. We assume that the anodization of the Ti wire creates a structure of capacitively and multiply coupled grains, which act as an array of tunnel junctions. Evaporated thin metal films often are granular. The grains could be the seeds for the island formation during anodization.

In other experiments on high ohmic wires without any obvious tunnel barriers there has been evidence of single electron charging effects. In $\text{In}_2\text{O}_3-x$ wires [117, 118] experiments have shown gate controlled Coulomb blockade. The presence of islands in these samples where thought to be due to localized electron states. In narrow silicon wires evidence of charging effects also has been reported [111, 119, 120]. In these wires, partially isolated island formation is believed to yield multiple tunnel junctions, which are responsible for the charging effects. Such islands have been observed with a transmission electron microscope [119]. In anodized Niobium wires with two weak links, similar to our SET-like samples, Coulomb blockade of single electrons as well as Cooper pairs has been reported [110]. It is likely that the islands formed during the anodization process are of varying size, thereby having different capacitances and tunnel resistances. It is also likely that charge traps and
Figure 4.31: Simulation of the $I-V$ characteristics for the schematically shown network of tunnel junctions at $T=0$. The junction parameters are: $C$ from 20 aF to 4 fF and $R$ from 100 kΩ to 1 GΩ.

Figure 4.32: Simulation of the $I-V$ characteristics for a inhomogeneous array of 20 tunnel junctions at $T=0$. The junction parameters are: $C$ from 0.7 aF to 3.5 aF, $C_g$ from 0.02 aF to 0.8 aF and $R$ from 40 kΩ to 300 kΩ.
incomplete conduction paths are formed during the oxidation process. These charge traps and incomplete conduction paths are believed to be responsible for the observed switching behavior (fig 4.29). Figure 4.31 shows a simulated $I$–$V$ curve of the model circuit shown in the inset. As can be seen, the jumping and hysteretic behavior of the measured $I$–$V$ characteristics (Fig. 4.29) can be imitated by a simple model of a short array capacitively coupled to incomplete conduction paths. The gate modulation of the threshold voltage and current further suggests that charging effects are responsible for the features in the measured $I$–$V$ curves. The modulation of the current state in the $I$–$V$ characteristics of the anodized Ti wires was not observed to be periodic. As was shown theoretically in chapter 3, the gate modulation of an array with uniform gate coupling is a periodic function of the applied gate voltage. In our Ti wires the island size is likely to vary and thereby the gate coupling to different islands will be different. Simulations of such arrays showed that the threshold voltage as a function of the gate voltage was aperiodic. In figure 4.32 the voltage, at a current close to zero, of a inhomogeneous 20 junction array is plotted as a function of the gate voltage. In this figure one can see that the Coulomb blockade is not completely lifted for any value of the gate voltage, as observed experimentally in the anodized Ti wires.
Acknowledgements

I have been part of the Nanostructure group at KTH since the middle of 1998. During this time I think I have learned more than during any other period of my life. I have learned a lot of new and exciting physics, but also a lot about people and working together with others. All of this would, of course, not have been possible without all the talented and helpful members of the group who I had the privilege to work in. First of all I would like to thank my supervisors, David Haviland for always being positive and encouraging, and Vlad Korenivski for always taking the time to discuss and insisting that there must be a physical reason for everything. I would also like to thank my old roommates Volker and Peter and new roommates Mattias and Silvia for making every day much more fun. Anders Liljeborg deserves a thanks, for explaining and showing me how my computer really works. In fact, I want to thank every one in the Nanostructure physics group.

On a personal level I would like to thank my family, Jessica, Karl and Elsa, thanks for always being there. I love you, you make everything worthwhile.
Bibliography


Papers

Appended papers


II Spin injection in Ferromagnet - superconductor / normal - Ferromagnet structures M. Urech, Jan Johansson, V. Korenivski and D. B. Haviland, accepted for publication, JMMM, (2003).


My contribution to the papers: In paper I, I developed the model, set up the simulations and wrote the paper. In paper II was part of the sample fabrication and measurement together with Mattias Urech. In III I wrote the first version of the comment and took part in writing the final version. In paper IV I made the samples and conducted the measurements. In V, I did all calculations and simulations and wrote the paper. In paper IV and IIIV, I worked together with Karin Andersson and Volker Schöllmann on sample fabrication. Together with Volker, I built up the anodization setup, conducted the anodization and made the transport measurements of the samples.

Other publications, not appended
