QUANTUM DOTS AND SUPERCONDUCTIVITY IN Ge-Si NANOWIRES

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DISSERTATION

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ABSTRACT

A quantum computer requires a quantum-mechanical two-level system with coherent control over its eigenstates. We investigate two different routes leading to quantum computational devices using Ge-Si core-shell nanowires in which holes are confined in one dimension: Normal-state quantum dots for spin qubits and proximity-induced superconductivity for Majorana fermions.

The nanowires have an extremely low defect density resulting in high mobilities and the ability to form intentional quantum dots of several lengths up to half a micron. The predicted strong direct-Rashba spin-orbit coupling allows the hole spin state to be controlled using electric-dipole spin-resonance and is at the same time a requirement for obtaining Majorana zero modes.

In a single quantum dot the g -factor is highly anisotropic with respect to the nanowire and electric field axis, which enables the tuning of the electric-dipole spin-resonance frequency. For a double quantum dot in Pauli-spin blockade we observe highly anisotropic leakage currents and identify spin-flip cotunnelling and spin-orbit interaction as the main sources of spin relaxation. Spin-lifetimes can therefore be optimised by carefully choosing the magnitude and direction of the magnetic field.

Highly transparent superconducting aluminium contacts to the nanowires are obtained by using a straightforward annealing procedure. We observe a Josephson current with a near ideal $I_C R_N$ product and the device shows Shapiro steps when exposed to microwaves. We thus confirm our device is a Josephson junction. Near depletion, we see a strongly coupled few-hole quantum dot supporting a supercurrent through single-particle levels, an important step towards realising Majorana fermions.

We find an additional superconducting phase with a lower T_C than of aluminium, most likely consisting of an Al/Si_x/Ge_y alloy. Near depletion, we observe a very hard induced superconducting gap, proof of a highly homogeneous superconductor-nanowire interface and indicating few in-gap states, which are detrimental for Majorana zero modes. We believe the hard gap, as well as the high interface transparencies, are the result of the presence of the superconducting alloy.

*The only difference
between screwing around and science
is writing it down.*
- Adam Savage

PREFACE

Before we start with the more complicated matters, I would like to give a little bit of personal background and express my gratitude to a multitude of people.

First of all, how do you end up doing a PhD in something like experimental quantum physics? Well, I guess you must be at least interested in technology and physics, but I think, most importantly, you must have a curiosity about how things work, right up to the point you want to take them apart to see what's inside. To see what makes it do what it does. It can't be magic, there has to be a logical explanation and you become determined to retrieve and understand this information.

For 'old fashioned' macroscopic machinery, doing exactly this works pretty well to, for instance, see which gears are connected, or which lever controls what axis. When electronics are involved, things become a little more abstract. You'll be tracing wires, drawing schemes and even though the basic principles are not too complicated, the total machine may very well be. When these electronics are integrated on a chip, things become so small that you cannot trace the lines or see the components with the naked eye any more, which adds another layer of abstraction and the technology becomes more and more like a black box. Still, the individual components function according to the same principles as their larger counterparts, but can form incredibly complex networks, take for instance, the processor in your computer or phone.

In order to reach the regime where things become quantum, we need to go to dimensions that are comparable to the smallest single component of the state-of-the-art of this integrated circuit technology so that we end up at length scales from 100 down to a few nanometers (1 nanometer = 10^{-9} meter). Here, all the laws and principles that worked so well on all the larger scales break down and we require a new set of laws that describe this peculiar behaviour of 'nature at the bottom'. The problem is that we all live in a very classical non-quantum world and the quantum world is so strange and fundamentally different that our intuition pretty much fails us, even though we can describe it with equations that are completely consistent. For example, in a quantum world, particles are described with a wavefunction that only tells you the odds of finding a particle. You cannot know where it is exactly until

you look for it and then find it, or not. It gets weirder, before you measure whether the particle is actually there, the particle is everywhere at the same time inside this wavefunction and only when you look for it, you force it to be somewhere specific, i. e., you collapse the wavefunction. Where it ends up exactly is random but consistent with the odds as described by the wavefunction. This is only a glimpse of what makes quantum mechanics both fascinating and also pretty weird, and I find it therefore a fascinating subject to dive into and explore.

Ever since I was very little, I have had kind of an obsession with technology. Back then, I could spend hours watching the washing machine spin around and watch the turntable play records. Of course, the latter meant my parents had to deal with a toddler that turned on their HiFi system at unexpected moments with a very random volume. I was perfectly content just watching all this magic unfolding before my eyes up to a certain age when I started to question what it is that makes these things move all by themselves. From that moment onwards I was fiddling with little electric motors, batteries and light bulbs to make little projects with a very mixed success rate. Then came Lego Technics where, together with my buddies *Marijn* and *Evert-Jan*, we made elevators, a self driving car (not a smart one but one that just drove into things) and helicopters that did not even come close to flying.

With my mother having a job in IT, I witnessed the rise of personal computers from almost the beginning and I still feel nostalgic whenever I see a 5.25" (actual) floppy. Gaming, and fixing and tuning computers soon became a hobby of mine. Programming only started when we got the infamous Texas Instruments TI-83 graphical calculators in secondary school, allowing you to write basic code with which you could make simple games. This provided much unnecessary distraction in math and physics classes and with the help of a little connect cable, together with *Michiel*, we would play multiplayer on two calculators, scoring mainly popularity points. After secondary school I moved from 'The North' to Enschede and started a Bachelor Advanced Technology at the University of Twente. Lots of friends from our class also made the choice to come to Twente, which made it easy to stay in touch and some of us even became housemates over the years.

During my master Nanotechnology and after much deliberation, I chose to perform my final Masters assignment on planar silicon quantum dots in the NanoElectronics group where *Filipp* immediately dragged me into the cleanroom and did an excellent job teaching me all

about micro and nano fabrication, and at his point I was fully unaware of the consequences that this would ultimately have.

This became clear right after my graduation where *Floris* kindly asked me if I would be interested in writing a booklet, that was to become the one you are holding right now, so we now know it all worked out in the end. Already knowing all the great people I would be working with, I gladly accepted.

Floris, you were a great supervisor and mentor and you always kept the silicon quantum electronics team running like a well-oiled machine. You were always quickly available for questions and assistance, which I highly appreciate and which not everyone has the luxury to experience during their PhD. Also, never stop making good bad jokes.

To our dear godfather and goedheiligman of the NE-family *Wilfred*, you always make sure that everyone in the group feels welcome and that everyone gets along. Thank you for letting me be a part of it.

Michel, your talks were always very interesting and explained the difficult stuff in a very intuitive way. Your lectures on spintronics during my masters were one of the reasons I got interested in NanoElectronics.

Peter, thank you for helping me see the error of my ways during error analysis and for getting those pesky typos out of my concept.

Herr Dr. Dipl.-Phys. Brauns or simply *Matthias* for friends, my companion in misdemeanour and coffee consumption. I had a lot of fun working together on our nanowires, even though we called them names or cursed them dearly from time to time. We spent countless hours experimenting, failing, trying again, getting frustrated and drinking beer, mostly in that order. Next to that, we've enjoyed numerous adventures on our many convocations. You inspired my 6th proposition and without you, this thesis would not have been what it is today.

Chris, the man of which his name became a verb. The ludicrous spaghetti coder, the uninhibited techno-hobo and fanatical tinkermeister. Your creative impulses and, well, impulsiveness in general, brought a refreshing spirit to the office. That is, when you were locatable. If not, you were probably going really fast. I greatly enjoyed our trip to half/double-baked country and later our indulgence in the Nippon culture where we never got lost and I never let us travel to the wrong hostel. Thank you for all the awesome.

Sergey, you were my loyal office mate since the beginning to pretty much the end. From you, I learned drinking smoked Russian Earl Grey, where to find the ugliest couch cover and a new definition of over-tightening. Next to that, your fabrication expertise and perseverance helped everyone make very pretty devices. They also worked as inten-

ded part of the time, which really is high praise in experimental science.

Ksenia, thank you for organising the great evenings in 'de Veste' and other get-togethers and for bringing *Kurt* along for the 'what if?' debates. *Elmer*, all the friday afternoon beers we drank and all the lively discussions in which you participated will be warmly remembered, although I cannot recall what they were all about.

Robert, my other long time office mate, your enticing enthusiasm really tied the room together, which was convenient since we had no rug. *Alejandro*, the best of luck with the heavy burden of continuing the silicon legacy. So long, and thanks for the all the jamon. *Dilu*, good luck with your DNA (no disrespect, I mean it's your research project). *Celestine*, good luck with the last straws. *Sander*, your enthusiasm in explaining your research makes the time fly. I now know something about complex oxides. At least I think I do. *Janine*, thank you for your fun and relaxing birthday parties around the fire. *Derya*, apart from the staff, we are the last ancient NE-veterans still standing and will now pass the torch to the new generation. *Tamer*, good luck with pushing the e-beam lithography tool to its limits. *Tao*, *Oğuzhan*, *Michal*, *Yigitcan*, *Pim*, *Frank* thanks for being excellent colleagues.

A lab will quickly degrade into chaos and anarchy without some fierce technicians. *Thijs* and *Johnny* thank you for keeping our dear Triton running and giving it the old rub and scrub now and then, and for keeping up the 3 o'clock teabreaks. *Thijs*, I am still waiting for the day we will stride to be the fastest in our virtual cars. *Martin*, you know a hell of a lot of electronics and join a for beer often, I like that.

Also, it is a blessing to have a secretary who knows all and fixes everything, within the bounds of the law obviously. *Karen* thank you for bringing your laughter to the office, although I think there is another who is winning the volume battle.

I have had the pleasure to work with many students who explored the scientific crossroads that I had no time for myself. *Yizhen*, *William*, *Gertjan* (twice), *Joren*, *Agung*, *Ton*, thank you for all your hard work and I hope I have been a satisfactory supervisor to you all.

Having students around is always a good thing, not only for the hard labour but also to keep the group young and fresh. *Bram*, *Thijs*, *Ren*, *Tom*, *Daan* and *Bas*, thanks and good luck with everything!

Last but not least, the *Cleanroom Staff* should be praised for running a tight ship, keeping all equipment running and doing their best to coerce all users to act like they are in their mother-in-laws kitchen, i. e., to be tidy and attentive. Also, kudo's for the practical joke to install the new e-beam lithography tool just when I'm leaving.

Even though a PhD can be time demanding, I did manage to maintain some social life outside the office.

Maarten, Ewoud, Henk, thanks for all the trips, games, concerts, beers, barbecues, movies and whisky over all the years. *Maarten* and *Marieke*, you will remain one of the last strongholds in Enschede. Make sure to hold the fortress for all future endeavours.

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Mom, Dad, without you I would not be here today, for multiple reasons, some more obvious than the other. You have always given me the opportunity to do, explore, and undertake everything, without ever pressuring me or having other expectations than that I am doing something I love doing. All while continuously supporting me and motivating me when needed. I could not wish for better parents. *Virna*, lets never stop having laughs, heated discussions and annoying each other.

---

¹ In a good way, don't punch me.

Also, listen carefully to the end of the speech during the ceremony. *Al-lard*, awesome job keeping it all together with my little sister.

Elena, thank you for all your love and support, for always being there for me, picking up the slack when I fail to, to give me a motivational ‘punt to the bottocks’ when required, and much more. I am very much looking forward to everything that is ahead and that we will experience together.

For now, lets get serious and begin the real story...

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ACRONYMS

ABS	Andreev bound state
AC	alternating current
AFM	atomic-force microscopy
ALD	atomic layer deposition
BCS	Bardeen-Cooper-Schrieffer
BHF	buffered hydrogen fluoride
CNT	carbon nanotube
DC	direct current
DOS	density of states
DRSOI	direct Rashba spin-orbit interaction
EBL	electron-beam lithography
EDSR	electric-dipole spin resonance
EDX	energy-dispersive X-ray spectroscopy
FET	field-effect transistor
FWHM	full-width at half-maximum
IPA	isopropyl alcohol
MAR	multiple Andreev reflection
MIBK	methyl isobutyl ketone
MOSFET	metal-oxide-semiconductor field-effect transistor
NDC	negative differential conductance
PMMA	poly(methyl methacrylate)
PSB	Pauli spin blockade
SEM	scanning electron microscopy

SOI	spin-orbit interaction
SQUID	superconducting quantum interference device
TEM	transmission electron microscopy
TMAH	tetramethylammoniumhydroxide
VLS	vapour-liquid-solid

INTRODUCTION

Technology and science are an inseparable symbiosis and it is baffling how fast our entire society has changed under their influence, especially in the last two centuries. After the invention of the steam engine unleashed the industrial revolution, physical labour for both man and animal slowly became obsolete which had a major impact of the daily lives of many people. The next invention that changed our lives in such a drastic manner is that of the digital computer where one could say it is an extension of our minds, allowing us to process and store astronomical amounts of information. Due to the subsequent rise of the internet, nearly everyone in the world can now instantly communicate and share information with each other over this global network, and these technologies are now so intertwined within our social and financial constructs that we can even speak of a strong dependency: civilisation all over the world can no longer function without computers.

What will be the next influential leap in technology is uncertain, some believe it will be robotics and artificial intelligence, others believe humans and technology will come even closer in the form of transhumanism. Another, maybe less obvious contender, is a machine that applies the quantum mechanical world to perform a fundamentally different form of computation and has the power to solve problems that cannot be worked out within a reasonable time, even with all our current classical computers combined: the quantum computer.

1.1 QUANTUM COMPUTATION

The reason a quantum computer can solve certain problems on such different timescales is revealed when looking at the smallest building blocks that make up the system. Where bits in classical computers are always in either the 1 or 0 state (usually the ON and OFF state of a transistor), a quantum computer employs quantum bits, or so-called *qubits*. A qubit requires a quantum mechanical two-level system and

resides in a superposition of its two eigenstates $|0\rangle$ and $|1\rangle$. As a consequence, a quantum computer with n qubits can simultaneously be in a superposition of 2^n states, while a classical computer with n bits can only be in one of the 2^n resulting states. It is this *quantum parallelism* that only exists in the quantum world that allows certain algorithms to be evaluated much faster on a quantum computer than on a classical computer.

The full potential of the quantum computer is still a subject of research and for classical algorithms a quantum computer would not be intrinsically faster. However, two main highly relevant algorithms have been identified: (1) Shor's algorithm for factorising large numbers (Shor 1997) which would, if run on a universal quantum computer, render many commonly used cryptography methods based on prime number factorisation obsolete and (2) Grover's algorithm for searching large unsorted databases (Grover 1997) which would be of great practical use for anyone using large datasets. Other algorithms exist but are currently mainly extensions or evolutions of the two underlying principles on which these two algorithms are based. See Montanaro (2016) for an extensive review on the topic.

It therefore appears, as of now, that the first quantum computers would be deployed for specialised tasks and would function as an extension for classical computers which one could see as a quantum co-processor. Although a quantum computer is capable to perform the same tasks as a classical computer (it is a more general form of a Turing machine than a classical computer (Deutsch 1985)), it would likely be much slower since classical algorithms cannot benefit from quantum parallelism. Since quantum computing is still in its infancy, it is likely that many more algorithms will be discovered in the future which would give the quantum computer a more general role. For now, we are anxiously waiting for the first quantum processor that demonstrates *quantum supremacy*, meaning it shows a superpolynomial speedup over the competing classical algorithm.

Another application of a quantum computer would be the simulation of other quantum mechanical systems (Feynman 1982; Lloyd 1996). Where a classical computer would have to emulate quantum mechanics with software routines, a quantum computer would be able to use its 'quantum hardware' to natively perform these simulations. Not only would this be useful in a research environment, also more societal and commercial applications could be foreseen such as pharmaceuticals, catalytic processes, or design considerations in the semicon-

ductor industry. Current research efforts have yielded a trapped ion system with a few hundred entangled spins (Bohnet *et al.* 2016; Britton *et al.* 2012) on which basic simulations can be performed.

1.2 PHYSICAL IMPLEMENTATION

The quantum computer was first conceptualised in the 80's by Feynman *et al.* (1986) and Deutsch (1985). In the years that followed, various proposals and experiments resulted in the realisation of qubits in a wide variety of systems: cold trapped ions (Cirac *et al.* 1995), quantum electrodynamics on single atom birefringence (Turchette *et al.* 1995), nuclear spins in the liquid phase (Chuang *et al.* 1998) and atoms in an optical lattice (Brennen *et al.* 1999). The proposal of DiVincenzo (1995) marked the beginning of a more scalable approach and considered how a quantum computer and its individual qubits must be operated in the solid state. Based on the proposal of DiVincenzo (1995), two important proposals followed for electron spins in quantum dots (Loss *et al.* 1998) and donor spins in silicon (Kane 1998) which sparked the highly active field of spin-qubits.

1.2.1 *Top-down implementations for spin-qubits and superconducting qubits*

The first electron-spin qubits were realised in group III-V epitaxially grown heterostructures (Koppens 2007; Petta 2005), but the spin coherence in these materials are unavoidably limited by their finite hyperfine interaction due to their net nuclear spins. Effort now also moved towards group IV materials such as Si or Ge which offer a solution to this problem, since they predominantly consist of isotopes with zero net nuclear spin. Additionally, these device can be fabricated in a way that is compatible with CMOS technology. Being able to use the existing fabrication infrastructure is highly advantageous for a quick future transition from the laboratory to large scale fabrication.

The first single-atom spin qubit in silicon was realised by Pla *et al.* (2012), followed by the first qubit in a planar metal-oxide-semiconductor field-effect transistor (MOSFET) design (Veldhorst *et al.* 2014; Veldhorst *et al.* 2015a). The first fully CMOS compatible qubit only saw the light a few years ago (Maurand *et al.* 2016). By isotopically purifying Si and Ge by removing all isotopes with net spin (Itoh *et al.* 2003; Itoh *et al.* 1993), hyperfine interaction can be completely removed which results in spin coherence times of multiple

seconds (Muhonen *et al.* 2014; Veldhorst *et al.* 2015a; Veldhorst *et al.* 2015b), which is many orders of magnitude higher than the operation frequency of the qubits determined by their Rabi frequency. However, in these planar structures, the qubit wavefunction resides on the interface between the crystalline semiconductor and an amorphous oxide which is never fully homogeneous and prone to defects which are detrimental for device reproducibility. This issue can be solved by using Si/SiGe heterostructures (Takeda *et al.* 2016; Ward *et al.* 2016). These structures combine high quality epitaxially grown layers with low hyperfine interaction and can also be fabricated in a CMOS compatible way. In terms of the high potential spin coherence and possibilities for scalability, this is a highly promising system.

Another major branch of research focusses on superconducting qubits which use Josephson junctions and which exist in different flavours: flux qubits (Friedman *et al.* 2000; Mooij *et al.* 1999; van der Wal *et al.* 2000), charge qubits (Nakamura *et al.* 1999) and phase qubit (Martinis *et al.* 2002). For an extensive review of superconducting qubits see (Mooij 2004). These qubits are of a macroscopic size which puts less stringent conditions on the fabrication, but this is at the same time their great weakness in terms of scalability. Nevertheless, it is in these systems that preliminary testbed quantum computers have been realised which consist of a few tens of qubits (Song *et al.* 2017).

1.2.2 Bottom-up implementations for spin-qubits and topological qubits

Until now, we have treated top-down qubit systems where, in the case of spin qubits, confinement is usually realised by electrostatic potentials from nearby gates. A different approach is to use bottom-up grown structures, such as nanowires, in which the charges are radially confined by the dimensions of the structure itself, while charges are only free to move along the nanowire axis. Since we now effectively have a 1-dimensional channel, we only need confinement in the transport direction which can be achieved by gates oriented perpendicular to the nanowire axis.

The first experimental results on group III-V nanowires showing single quantum dots were reported by De Franceschi *et al.* (2003), followed by double quantum dots (Fasth *et al.* 2007; Fasth *et al.* 2005; Fuhrer *et al.* 2007) and, later on, by the first electron spin-orbit qubit (Nadj-Perge *et al.* 2010a). Next, single-hole spin control was demonstrated (Pribyl *et al.* 2013) where a hole can be seen as the

absence of an electron in the valence band of the semiconductor. For group IV materials, carbon nanotubes (CNTs) offered a successful platform for quantum dots and qubits (Jarillo-Herrero *et al.* 2004; Laird *et al.* 2013) as well as holes in Si nanowires (Zhong *et al.* 2005; Zwanenburg *et al.* 2009b). In Si-Ge core-shell nanowires, the system used in this thesis, double quantum dots, (Hu *et al.* 2007), spin filling (Roddaro *et al.* 2008), spin relaxation (Hu *et al.* 2012) and hole spin coherence (Higginbotham *et al.* 2014a) have been observed.

Another, very different, proposal by Kitaev (2000) described a way to create Majorana fermions in topological quantum wires. Although this is of high interest from a physics point of view, only more recently their implication for quantum computing applications highly robust against decoherence became apparent, which was immediately followed by proposals for the physical realisation (Alicia *et al.* 2010; Fu *et al.* 2008; Sau *et al.* 2010). The main required ingredients to create the required topological phase in a semiconducting nanowire are a sufficiently high g -factor and spin-orbit interaction and a hard induced superconducting gap. The first experimental signatures (Mourik *et al.* 2012) came in quick succession and research now focusses on obtaining harder induced superconducting gaps to obtain clear Majorana signals in different systems (Chang *et al.* 2015; Drachmann *et al.* 2017; Günel *et al.* 2014; Kjaergaard *et al.* 2016), and on realizing braiding operations (Alicia *et al.* 2011) to encode quantum information in a Majorana network.

Before the quest for Majorana fermions boosted the field of nanowires with proximity-induced supercurrent, much groundwork was already performed, mainly motivated by curiosity. The use of nanowires enables access to a new class of gate tunable Josephson junctions and the confinement imposed by their 1-dimensional character opened a realm in physics in which confined charge and/or quantum dots interact with the superconducting wavefunction. Experiments in InAs/InP nanowires (Doh *et al.* 2008; Lee *et al.* 2014), as well as in carbon nanotubes (Cleuziou *et al.* 2006; Eichler *et al.* 2007; Jarillo-Herrero *et al.* 2006; Jørgensen *et al.* 2009) display the rich physics in this system. For Si-Ge nanowires, only two reports in combination with superconducting contacts exist (Su *et al.* 2016; Xiang *et al.* 2006a), which leaves this system relatively unexplored.

1.3 GE-SI NANOWIRES

In this thesis we use Ge-Si core-shell nanowires as the workhorse and explore their suitability for two different quantum computation implementations where we expand on the existing results in literature: spin qubits and topological quantum computation using Majorana fermions.

This unique one-dimensional semiconducting system has many appealing properties: (1) Due to the staggered band gap between the Ge core and the Si shell, free holes accumulate in the core without the need for doping or externally applied electric fields (Lu *et al.* 2005; Zhang *et al.* 2010). (2) Ge and Si are group IV materials which have very low hyperfine interaction resulting in long spin coherence times. (3) A predicted enhanced direct Rashba spin-orbit interaction (DRSOI) for holes (Kloeffel *et al.* 2011) enables spin control via AC electric fields using electric-dipole spin resonance (EDSR) (Kloeffel *et al.* 2013b; Osika *et al.* 2013), a major technological advantage compared to AC magnetic fields. A strong spin-orbit interaction is also a requirement for Majorana zero modes (Stanescu *et al.* 2013). (4) A strongly anisotropic g -factor, which can be tuned using electric fields, makes it possible to bring individual qubits in and out of resonance with a constant AC electric field. Like spin-orbit interaction, a sufficiently high g -factor is also a requirement for Majorana fermions (Stanescu *et al.* 2013). (5) On the lengthscales of interest for qubits or Majorana fermions, these wires can be grown with virtually zero defect densities (Conesa-Boj *et al.* 2017) which greatly improves reproducibility and allows for better control over the electrostatic environment of the charge carriers. (6) From a physics point of view, the heavy-hole and light-hole mixing in the valence band edge is a rich and unexplored system (Csontos *et al.* 2009; Kloeffel *et al.* 2011)

1.4 OUTLINE

In this thesis I will present a wide variety of transport experiments with the purpose to confirm, explore or utilise the above properties where the final goal is to bring quantum computation a small step closer. We will now present an outline of this work.

In Chapter 2 we will discuss the theoretical concepts to give a basic intuitive description of the subjects. We start the discussion with quantum dots after which we proceed to the interaction between su-

perconductors and normal materials. Finally, we discuss a quantum dot with superconducting contacts.

Chapter 3 starts with a summary of nanowire growth and shows the different types of devices designed and fabricated during this research. The challenge in the normal quantum dot devices is to obtain small feature sizes with complex gate geometries and clean gate oxides. In that respect the superconducting devices are less complicated but the difficulty is the realisation of transparent contacts between the nanowires and a superconducting metal.

In Chapter 4 we present an elaborate method to extract the hole mobility from transport data and correlate the resulting mobilities to different crystal directions of the nanowires as determined by a transmission electron microscopy study. We find that the wires with the lowest diameter have the highest hole mobility and grow predominantly in the $[110]$ crystal direction which shows the least amount of defects.

In Chapter 5 we show how our device design results in a high degree of control over the electrostatic environment of the charge carriers in the nanowire. We will electrostatically define quantum dots of various lengths up to 500 nm. Next, we define a highly stable double dot and extract the relevant energies and capacitances.

We now return to a single quantum dot in Chapter 6 and measure the g -factor by excited-state magnetospectroscopy. We perform 360° field rotations around all three main axes with respect to the nanowire and find, as predicted, a strong anisotropy with respect to the electric field axis and with respect to the nanowire orientation.

In Chapter 7 we investigate a double quantum dot in the weak coupling regime where we observe shell filling and find Pauli spin blockade for the charge degeneracies where one dot has a partially filled orbital. We furthermore identify various mechanisms responsible for leakage currents in the blocked state by performing a series of magnetospectroscopies. For low magnetic fields, spin-flip co-tunneling dominates the leakage current, while at higher magnetic fields we observe signatures of spin-orbit induced leakage.

We now come to the experiments in which aluminium superconducting leads induce a supercurrent in the nanowires. In Chapter 8 we show a finite Josephson current and multiple Andreev reflections, indicating highly transparent contacts from the superconductor to the nanowire.

When irradiating the device with microwaves we observe a large number of Shapiro steps, a direct manifestation of the AC Josephson effect. We conclude that our device is a true Josephson junction.

In Chapter 9 we investigate our nanowire Josephson junction as a function of applied electric field and identify two regimes: a highly transparent regime where multiple subbands participate in transport accompanied by a continuous supercurrent, and a single-hole tunneling regime where we observe supercurrent through single-particle levels of a few-hole quantum dot. Furthermore the quantum dot shows an even/odd filling up to depletion.

In Chapter 10 we investigate, next to aluminium, the presence of two additional superconducting phases in two different devices. These phases are the result of an alloying process during annealing and therefore they most likely consist of an alloy of Al/Ge and/or Al/Si. We discover the properties to vary per device: For a nanowire Josephson junction we find a superconducting material with lower critical temperature and a much higher critical field compared to the aluminium leads. For a device of which the nanowire channel was metallised during annealing, we find a critical temperature 3 times higher and a critical field that is 15 times higher than that of pure Al.

As a second topic we investigate the hardness of the induced superconducting gap near depletion, quantified by the ratio of the in-gap conductance to the out-of-gap conductance, which is a measure for the homogeneity of the superconductor-nanowire interface. We obtain current suppression values comparable to fabrication techniques which employ elaborate cleaning and etching procedures, or compared to devices that employ epitaxially grown aluminium contacts.

THEORETICAL CONCEPTS

This chapter will introduce theoretical concepts to provide a basic understanding of the research performed in this thesis. Where possible, references to more exhaustive literature will be given. We will first discuss single and double quantum dots after which we will focus on superconductivity and Josephson junctions. Finally, we treat the combination of quantum dots with superconducting contacts.

2.1 QUANTUM DOTS IN THE SOLID STATE

Quantum dots are small isolated islands of matter in which charges occupy well defined quantised energy levels. Quantisation of energy levels is realised by confinement in all three spatial directions (Kouwenhoven *et al.* 1997). Generally speaking, populating or depopulating such an island occurs in discrete steps of single charges. Since Ge-Si nanowires are hole conductors we will use the terms ‘charge’ and ‘hole’ interchangeably in this section. A more elaborate discussion on the subject of single and double quantum dots can be found in the review articles by Kouwenhoven *et al.* (1997), Kouwenhoven *et al.* (2001a), van der Wiel *et al.* (2002) and Hanson *et al.* (2007). Figure 2.1 to Figure 2.5 in this section are adapted with permission from Brauns (2016).

We need to consider two different energies when adding a charge to a quantum dot. First, there is the classical charging energy $E_C = e^2/C$ which is the energy needed to overcome the Coulomb repulsion between charges residing on the island. This energy is inversely proportional to the capacitance C of the dot: decreasing the dot size reduces C which increases E_C .

The second relevant energy scale has a completely quantummechanical origin and is the result of the spatial confinement of charges which leads to the quantisation of energy levels. These levels bear close

resemblance to the orbital wavefunctions in atoms and exhibit many of their properties, which has lead to the name ‘artificial atoms’. For instance, due to the Pauli exclusion principle, a single energy level may only be occupied by two charges of opposite spin, which leads to observable shell filling (Kouwenhoven *et al.* 2001a). Interestingly, quantum dots mostly have one or two spatial directions in which confinement is dominant. This results in an artificial atom with 1- or 2-dimensional orbital wavefunctions, a realm physically impossible to investigate in real atoms (Kouwenhoven *et al.* 2012). For example, the Ge-Si nanowires used in this thesis have a pronounced 1-dimensional character due to strong confinement in the radial direction.

The total energy for adding a charge to the dot thus becomes $E_{\text{add}} = E_C + E_{\text{orb}}$. Note that E_{orb} only contributes if a charge is added to an empty orbital. For a quantum dot, two requirements have to be fulfilled:

1. The charging energy and the orbital level spacing must be greater than the thermal energy: $E_C, E_{\text{orb}} \gg k_B T$ with k_B the Boltzmann constant and T the temperature. Since both E_{orb} and E_C increase with smaller dimension, we require a small island and low temperature.
2. The dot must be sufficiently isolated, i. e., the tunnel coupling Γ must be much lower than the charging energy and orbital level spacing: $E_C, E_{\text{orb}} \gg h\Gamma$ with h being Planck’s constant.

By measuring at typical temperatures of ~ 20 mK, we satisfy the first requirement. Small dimensions are realised by using Ge-Si nanowires with diameters less than 30 nm, which provide intrinsic confinement in the radial direction. In the third direction, (along the wire) we use gates to create tunable tunnel barriers to satisfy the second requirement.

Figure 2.1a shows a circuit diagram of a quantum dot connected to a source and drain and with a capacitive coupling to a gate. The gate voltage V_g changes the electrochemical potential of the consecutive charge levels μ_N , while applying a source drain bias V_{SD} shifts the electrochemical potential of source and drain (μ_S and μ_D). Measuring the current for a very small source-drain bias $\mu_S \approx \mu_D$ while sweeping V_g , results in peaks in current I whenever μ_N is resonant with μ_S and μ_D . For dots with constant E_{add} (i. e., with $E_C \gg E_{\text{orb}}$), this results in a series of evenly spaced peaks in current (Coulomb peaks), as shown in Figure 2.1b where ΔV_g is the gate voltage required to change the occupancy of the dot by one hole. When opening a larger bias window, i. e.,

Figure 2.1: (a) Circuit diagram of a single quantum dot connected to source, drain and gate. (b) Coulomb peaks in a plot of current I versus gate voltage V_g . (c) I is measured versus applied bias V_{SD} and V_g resulting in a bias spectroscopy showing diamond shaped regions where no current is measured. The red line indicates transport through an excited state, i. e., the next orbital level.

$\mu_S > \mu_D$, these sharp peaks evolve into wider regions of current until for $\mu_S - \mu_D > E_{add}$ at least always one μ_N is inside the bias window.

Measuring I versus both V_{SD} and V_g and performing the numerical derivative $\partial I / \partial V_{SD}$, we obtain a 2-dimensional conductance map (a so-called bias spectroscopy) as schematically drawn in Figure 2.1c for both positive and negative V_{SD} . The grey diamonds represent regions where transport is blocked ($I = 0$), since no dot charge level falls within the bias window and the charge occupation N is constant. When increasing V_g , we reach the $N - 1$ diamond where one charge is removed from the dot. The height of the diamond equals E_{add} and varies depending on the contribution of E_{orb} : in the $N - 1$ diamond we have assumed we are adding a charge to a partially filled orbital, hence the lower diamond height by E_{orb} . By inspecting the diamond heights we can thus determine both E_C and E_{orb} , assuming E_C remains constant. Additionally, we

Figure 2.2: (a) Circuit diagram of two tunnel coupled quantum dots with individual capacitively coupled gates. (b) Electrochemical potential diagram showing charge transport through a double quantum dot with finite bias.

can extract the gate lever arm $\alpha_g = E_{\text{add}}/\Delta V_g$, a measure for the capacitive coupling of the gate. When applying a bias $eV_{\text{SD}} > E_{\text{orb}}$, not only the lowest orbital level (ground state), but also a higher level orbital (excited state) may fall within the bias window. When on-resonance with μ_S or μ_D , this results in an enhanced tunnel probability which results in a conductance peak (red line in the bias spectroscopy in Figure 2.1c).

2.1.1.1 Double quantum dots

We now extend the circuit diagram of a single dot of Figure 2.1a to accommodate a second quantum dot in series, as shown in Figure 2.2a with dot occupancies M, N and electrochemical potentials μ_1, μ_2 for the left and right dot respectively. The dots are both tunnel coupled (modelled by $R_M \propto \Gamma$) and capacitively coupled (modelled by C_M). Additionally, both dots are capacitively coupled to a gate which changes μ_1 and μ_2 independently (we ignore gate cross-coupling in this case). Figure 2.2b shows the resulting electrochemical potential diagram where charge transport is only possible for $\mu_S \geq \mu_1 \geq \mu_2 > \mu_D$, i.e., for each tunnelling event we move down on the electrochemical potential ladder.

We first consider the situation where $C_M = 0$, which means that a change in μ_1 does not lead to a change in μ_2 . Quantum mechanically speaking, there is no wavefunction overlap between the charges

Figure 2.3: Stability diagrams of a double quantum dot with increasing mutual capacitance C_M . Diagrams are for the low bias condition $\mu_S \gtrsim \mu_D$. (a) Stability diagram for $C_M = 0$. Dots are uncoupled and transport is only possible at the black dots (a). (b) Stability diagram for intermediately coupled dots, the square grid evolves to a honeycomb pattern. (c) Fully coupled dots which effectively turn into a larger single dot. Transport is possible along diagonal black lines.

on both dots. The charge stability diagram for $C_M = 0$ is shown in Figure 2.3a, where we schematically show the hole occupation of both dots as function of V_1 and V_2 . On the vertical lines, the left dot is resonant with the source, while on the horizontal line, the right dot is resonant with the drain. Transport is only possible at their crossing where both dots are resonant with source and drain and four charge states are degenerate.

The other extreme is a strong mutual capacitance in the order of the gate capacitances $C_M \approx C_1, C_2$, as shown in Figure 2.3c. Since the charge is now highly delocalised and is spread out on both dots, we essentially obtain a single dot to which both gates couple equally. Transport is possible on the diagonal lines.

The most interesting case is for intermediate coupling, shown in Figure 2.3b, in which charge is mostly localised on individual dots, but there exists a finite wavefunction overlap. In this situation the charge degeneracy points in Figure 2.3a split to so-called triple points (three degenerate charge states) and the square lattice evolves into a honeycomb pattern. The splitting between the triple points, in terms of gate voltages, can be expressed as $\Delta V_{M,1(2)} = \Delta V_{1(2)} C_M / C_{2(1)}$ with $\Delta V_{1(2)}$ the difference between charge transitions for the left (right) dot, as denoted in Figure 2.3b (van der Wiel *et al.* 2002).

Figure 2.4: A zoom of a set of triple points at finite V_{SD} reveals a pair of bias triangles extending from the original triple points. Sketches (green) show the electrochemical potentials of source, drain and both dots in all three corners of the triangles. The red line (parallel to the base of the triangle) represent an excited state.

Figure 2.5: Electrochemical potential diagrams illustrating the mechanism of Pauli spin blockade at zero detuning ε . (a) shows the unblocked current direction while (b) shows the blocked transport direction.

For a finite source drain bias, i. e., $\mu_S > \mu_D$, the triple points turn into larger triangle shaped regions as seen in Figure 2.4. On the original triple points, the electrochemical potential of both dots is resonant with the drain. When moving along the basis of the triangle, i. e., from $(M+1, N+1)$ to (M, N) , the edge of the diamond marks the point where both dot levels are resonant with the source. Moving perpendicular to this axis, i. e., from $(M+1, N)$ to $(M, N+1)$, the dot electrochemical levels shift with respect to each other and we call this the detuning axis ε . As in a single dot, excited states can be present in the bias window which shows as an enhanced current in the triangles for $eV_{SD} > E_{orb}$, indicated by the red line in Figure 2.4.

2.1.2 Pauli spin blockade

Pauli spin blockade (PSB) is a current rectifying mechanism that occurs in double quantum dots when one of the dots has a partially filled orbital below the Fermi level (Hanson *et al.* 2007) and can be utilised for spin readout using spin-to-charge conversion (Petta 2005). Figure 2.5a illustrates the non-blocked case for an orbital with one spin down hole in the left dot below the Fermi level. During transport, a spin up hole can tunnel from the source onto the left dot forming the $S(2,0)$ singlet state, after which it tunnels onto the right dot forming the $S(1,1)$ singlet and consequently tunnels to the drain, completing the transport cycle. For sufficiently high bias, a spin-down hole can tunnel from the source to the left dot forming the $T(2,0)$ triplet. This involves accessing the next orbital and the energy splitting between $S(2,0)$ and $T(2,0)$ $\Delta_{S-T} \approx E_{\text{orb}}$. Tunnelling to the right dot now results in the $T(1,1)$ state which is, apart from a negligible exchange energy, equal in energy to the $S(1,1)$ state (Hanson *et al.* 2007). The spin-down hole now tunnels into the drain, completing the transport cycle.

We now reverse the bias direction, as is shown in Figure 2.5b, while we keep the dots electrochemical potentials the same. The hole tunnelling from the drain into the right dot now has approximately equal probability to be spin-up or spin-down. A spin-up hole will form the $S(1,1)$ state, then the $S(2,0)$ state, and is consequently transported to the source. However, a spin-down hole will result in the $T(1,1)$ and with the $S(2,0)$ state unavailable due to the Pauli exclusion principle, tunnelling to the left dot is only possible via the $T(2,0)$ state. However, this state is Δ_{S-T} higher in energy and thus transport is blocked. Blockade can be lifted by applying a high enough bias $eV_{\text{SD}} > \Delta_{S-T} \approx E_{\text{orb}}$ or by spin flips induced by, e. g., microwaves (Hanson *et al.* 2007; van der Wiel *et al.* 2002)

In terms of the bias triangles in Figure 2.4, Pauli spin blockade would result in observing smaller triangles with missing bases, i. e., the trapezoid above the red line on the ε -axis would not show a current. In practice, multiple mechanisms induce spin flips such as spin-orbit interaction, hyperfine interaction and spin flips induced by the leads, which result in a finite leakage current in the trapezoid. In Section 7 these mechanisms are explained in more detail.

2.2 SUPERCONDUCTING PROXIMITY EFFECTS

Superconductivity is a phenomenon in which all electrons can be described by a single macroscopic wavefunction $\psi = \psi_0 e^{i\theta}$ with ψ_0 the amplitude and θ the phase. In a superconductor, long-range order is established by the formation of Cooper pairs: two coupled electrons with opposite spin and a bosonic character. The interaction strength of the Cooper pairs results in a finite range around E_F with a zero density of states (DOS) for electrons. The energy of the induced gap in the DOS is defined by the order parameter Δ . This material property is a direct measure for the critical temperature T_C , i. e., the maximum temperature for which the material remains superconducting. In this section we will highlight several subjects that describe the interaction between a mesoscopic normal material and a Type-I superconductor. The goal is to obtain a basic intuitive understanding while we refer to other literature for more detailed information.

2.2.1 Andreev reflection: the N-S interface

In Figure 2.6a, a schematic representation of a normal-superconductor (N-S) interface is shown. In the normal material the DOS is continuous up to E_F , while in the superconductor a gap with magnitude 2Δ is present. Inside the gap, only Cooper pairs are allowed at E_F , no states for electrons or holes are present and normal charge transport is therefore not possible. However, a more complicated scheme involving 4 charged particles enables transport through an N-S interface: Andreev reflection. As illustrated in Figure 2.6a, an electron with energy ε above E_F impinges on the interface and, at the same time, a hole is retro-reflected with energy ε below E_F , which phase-coherently retraces the original path of the electron. Since the reflected hole has opposite charge and momentum, a second electron is created which forms a Cooper pair with the original impinging electron, resulting in a total charge transfer of $2e$. The excess momentum due to the difference in energy between the original electron and the retro-reflected hole, is taken up by the Cooper pair $k_{\text{Cooper}} = k_e - k_h$ (Harmans 2003; Imry 2002).

The retro-reflected hole accumulates a phase shift φ_{e-h} due to the difference in momentum: $k_e - k_h = k_{e-h} = 2\varepsilon/\hbar v_F$ with v_F the Fermi velocity of the normal material. The electron and the hole remain phase

coherent for $k_{e-h} < \pi$ and one can show this leads to the definition for the phase coherence length

$$\xi_{\text{ball}} = \frac{\hbar v_F}{\pi \Delta}, \quad (2.1)$$

assuming a clean (ballistic) normal material. In the dirty (diffusive) limit this changes to

$$\xi_{\text{diff}} = \sqrt{\frac{\hbar v_F l_e}{\pi \Delta}} = \sqrt{\xi_{\text{ball}} l_e}, \quad (2.2)$$

with l_e the elastic scattering length of charge carriers in the normal material (holes in Ge-Si nanowires). Inspecting *Equation* (2.1) and *Equation* (2.2) we see that the coherence length mainly depends on the ratio v_F/Δ . This implies that a normal material with a high Fermi velocity combined with a low Δ (and therefore low T_C) results in longer coherence lengths (Tinkham 1996).

The conductance of an N-S interface for a single mode is given by

$$G_{\text{N-S}} = \frac{4e^2}{h} \frac{T^2}{(2-T)^2}, \quad (2.3)$$

where T is the transmission probability. Compared to the Landauer expression for normal interfaces $G_{\text{N-N}} = 2e^2 T/h$ and taking $T = 1$ we observe that $G_{\text{N-S}} = 2G_{\text{N-N}}$, i. e., there is a doubling of the conductance on a N-S interface consistent with the charge transfer of $2e$ at the interface. For $T \rightarrow 0$ however, $G_{\text{N-S}} \propto T^2$ which is a direct consequence of the fact that two charges need to traverse the interface simultaneously. This means a N-S interface is much more sensitive to reduced interface transparencies than a normal interface.

2.2.2 The S-N-S junction

We now add a second superconductor which results in a superconductor-normal-superconductor junction, as seen in Figure 2.6b. Transport through this junction is now facilitated by Andreev reflection processes on both interfaces: a Cooper pair dissociates on the S1-N interface and results in the simultaneous transport of an electron from S1 to S2, and a hole from S2 to S1. A charge of $2e$ is thus transferred to S2 which results in the formation of another Cooper pair in S2. This transport is phase-coherent, elastic and dissipationless, i. e., a supercurrent.

Figure 2.6: (a) Energy E versus DOS diagram of a normal-superconductor (N-S) interface showing the process of Andreev reflection. An impinging electron (solid) with energy ε and wavevector k_e results in a retro-reflected hole (hollow) with opposite spin, wavevector k_h and energy $-\varepsilon$, and an additional electron which forms a Cooper pair in the superconductor with the original electron. (b) S-N-S junction where Andreev reflection on both interfaces results in a phase-coherent Josephson current.

Quantum mechanically, this multiple Andreev process is quantised and will form bound states when we satisfy the condition that a particle acquires a phase of *modulo* 2π , after scattering and returning to its original position, i. e., when a particle completes the cycle $e \rightarrow \text{N-S}_2 \rightarrow h \rightarrow \text{N-S}_1 \rightarrow e$ it should acquire a total phase of $2\pi n$ with n an integer. There are two contributions to the phase of a particle undergoing this cycle: (1) the previously discussed difference in momentum multiplied by the junction length L contributes $\varphi_{e-h}^\pm = k_{e-h}^\pm L = 2\varepsilon^\pm L / \hbar v_F$, with $\pm$ indicating a left and right moving particle respectively, and (2) the phase difference between the superconducting condensate in S1 with phase φ_1 and in S2 with phase φ_2 , which results in $\varphi_{\text{N-S}} = \varphi_{1-2} - \cos^{-1}(\varepsilon^\pm / \Delta)$ with $\varphi_{1-2} = \varphi_1 - \varphi_2$. The $\cos^{-1}$ term is a consequence of the continuity conditions of the wavefunction (Harmans 2003; Imry 2002). This leads to the following condition for a phase coherent (Andreev) bound state:

$$\begin{aligned} \varphi_{e-h}^\pm + 2\varphi_{\text{N-S}} &= \frac{2\varepsilon_n^\pm L}{\hbar v_F} \pm \varphi_{1-2} + 2\cos^{-1}(\varepsilon_n^\pm / \Delta) = 2\pi n, \\ \varepsilon_n^\pm &= \frac{\hbar v_F}{2L} \left[2\cos^{-1}(\varepsilon_n^\pm / \Delta) \pm \varphi_{1-2} + 2\pi n \right], \end{aligned} \quad (2.4)$$

where $\varepsilon_n^\pm$ is the energy of the eigenstate. For $\varepsilon \ll \Delta$, Equation (2.4) simplifies to

$$\varepsilon_n^\pm = \frac{\hbar v_F}{2L} [(2\pi n + 1) \pm \varphi], \quad (2.5)$$

Figure 2.7: Energy vs DOS diagram showing the process of multiple Andreev reflection (MAR). Each scattering event results in the particle receiving $|\mu_S - \mu_D|$ of energy until it can tunnel into the DOS above the gap.

which reveals that for $\varphi = 0$, the left and right moving states are degenerate. This means there is no net transport, since the population probability for both left and right moving states are the same. For $\varphi \neq 0$, however, the left and right moving states differ in energy and a different current will be carried by the $+$ and the $-$ branch. This brings us to the conclusion that a S-N-S junction carries a supercurrent with a magnitude that is determined by the phase difference between the two superconductors. This is evident from the first Josephson equation $I_C = I_0 \sin(\varphi)$ with I_0 the intrinsic maximum Josephson current of the junction (Gross *et al.* 2005; Tinkham 1996).

For $\varphi > \pi$, the left ($-$) and right ($+$) moving states reverse direction: for $\pi < \varphi < 2\pi$, $\epsilon_0^\pm$ crosses the Fermi level and since there are no electron states below (or hole states above) the Fermi level, the system responds by changing the sign of the transport direction of the left and right moving states, thus reversing the sign of the supercurrent. The absolute current therefore has a maximum for $\varphi = \pi$ (or $\varphi = -\pi$). The current carried by a single bound state can be estimated as $I_C^\pm = ev_F/L$ (where v_F/L equals the number of transport events per second through the junction) in which we can substitute L with Equation (2.1) in the ballistic limit, resulting in a maximum current of $I_{C,\text{MAX}}^\pm \approx e\Delta\pi/\hbar$. A more rigorous calculation by Beenakker *et al.* (1991) gives the commonly accepted value $I_{C,\text{MAX}}^\pm \approx e\Delta/\hbar$.

2.2.3 Multiple Andreev reflections (finite bias)

In the previous discussion, μ_S was equal to μ_D , i. e., no voltage bias was applied. Figure 2.7 shows a schematic image of a S-N-S junction with a finite positive bias. A finite voltage drop implies there is no superconducting Josephson current. In fact, the difference in electrochemical potential of both superconductors prevents coherent Andreev processes involving Cooper pairs. Ordinary dissipative transport is only possible for $|eV_{SD}| = |\mu_S - \mu_D| > 2\Delta$ when the filled DOS peak of S₁ (S₂) and the empty DOS peak of S₂ (S₁) align for forward (reverse) bias and quasiparticles can directly tunnel from S₁ to S₂ (S₂ to S₁).

Another type of dissipative transport is possible for $|eV_{SD}| < 2\Delta$ which is illustrated in Figure 2.7: a quasiparticle from the full DOS peak in S₁ Andreev reflects at the N-S₂ interface and gains an energy eV_{SD} , traverses back and Andreev reflects again at the N-S₁ interface, while gaining another eV_{SD} of energy. These multiple Andreev reflection (MAR) repeat until the total gained energy $neV_{SD} > 2\Delta$ with n the number of undergone reflections, at which point the quasiparticle can escape to the finite DOS in the leads. When the bias satisfies $|eV_{SD}| = 2\Delta/n$, quasiparticles are transported between the sharp peaks in DOS in both superconductors which results in peaks of enhanced conductance. The number of n visible in transport can be suppressed by inelastic processes and therefore increases for higher homogeneity of the superconductor-normal interfaces and higher elastic scattering lengths of the normal material. Furthermore, MAR is only possible when the transmission T between superconductor and normal material is finite, i. e., there must be a finite probability of reflection (Flensberg *et al.* 1988).

2.2.4 Quantum dot with superconducting leads

We will now discuss the combination of a quantum dot coupled to superconducting contacts as illustrated in Figure 2.8. Here, the tunnel broadening Γ is determined by the transparency of the superconductor-normal interface while the electrochemical potential of the individual dot levels can be adjusted by a gate voltage V_g . We classify and summarize three regimes of coupling as described in De Franceschi *et al.* (2010):

1. Weak coupling: $\Gamma \ll \Delta, E_{\text{add}}$
2. Strong coupling: $\Gamma \gg \Delta, E_{\text{add}}$ or $E_{\text{add}} > \Gamma \gg \Delta$
3. Intermediate coupling: $\Gamma \sim \Delta \sim E_{\text{add}}$

Figure 2.8: Energy vs DOS diagram showing a quantum dot with two tunnel barriers (blue) contacted with two superconductors. Coupling of the quantum dot is determined by the relative energy scales of the FWHM of the tunnel broadening Γ , the addition energy E_{add} and the order parameter Δ of the superconductor. The overlap between the single particle levels (yellow) on the quantum dot and the Cooper pair wave function in the superconductor is determined by the electrochemical potential of the individual dot levels ϵ_N .

(1) In the weak coupling regime, only quasiparticle transport is possible for $|eV_{SD}| > 2\Delta$, regardless of the relative position of ϵ_N , and MAR is mostly suppressed due to the very low interface transparencies. The strong DOS quasiparticle peaks of the superconductor result in a conductance peak at $|eV_{SD}| = 2\Delta$ which may be visible even in Coulomb blockade where no transport would be measured for normal contacts.

(2) The strong coupling regime allows superconducting transport through the junction. For the first case where $\Gamma \gg \Delta, E_{add}$, the magnitude of the supercurrent is

$$I_{C,MAX}(n) = \frac{ne\Delta}{\hbar} \quad (2.6)$$

as derived in Section 2.2.2 with n the number of parallel modes participating in transport. However, when single particle levels are well defined, i. e., $E_{add} > \Gamma \gg \Delta$ the supercurrent strongly depends on ϵ_N relative to the Fermi level. The following expression was derived by Galaktionov *et al.* (2002):

$$I_C = I_{C,MAX}(1) \left(1 - \sqrt{1 - T_{BW}}\right), \quad (2.7)$$

$$T_{BW} = \frac{\Gamma}{(\epsilon_N/h)^2 + \frac{1}{4}\Gamma^2},$$

where T_{BW} is the Breit-Wigner transmission probability. Although Equation (2.7) results in good qualitative fits of I_C as a function of

ε_N , the measured I_C is usually much lower than predicted from Equation (2.7). By taking the electromagnetic environment into account, which includes various noise sources such as the on-chip electronics, one can obtain a closer quantitative fit (De Franceschi *et al.* 2010; Jarillo-Herrero *et al.* 2006).

(3) In the intermediate coupling regime all relevant energy scales are comparable and competition between charges trapped on the quantum dot and superconductivity are most apparent. A complete description of this regime is beyond the scope of this work and can be found in De Franceschi *et al.* (2010)

3

DEVICE FABRICATION AND DESIGN CONSIDERATIONS

This chapter focuses on the fabrication of the devices used in this thesis. Using Ge-Si core-shell nanowires as the workhorse, experiments have been carried out on two types of devices: normal state devices (Section 3.2.2-Section 3.2.6) and devices with superconducting leads (Section 3.2.7). Special attention is given to electron-beam lithography (EBL), a tool quintessential in fabricating these nano-scale devices in a research environment. We focus especially on the application of cold development as a method to decrease feature size, while increasing reproducibility and robustness of the process. As a side note, all device metallisation steps are performed using electron-beam evaporation and is not further treated in this chapter. We start off by explaining the growth of Ge-Si nanowires.

3.1 NANOWIRE GROWTH

The Ge-Si core-shell nanowires used in this thesis have been grown by our collaborators¹ in the Photonics and Semiconductor Nanophysics group at the Eindhoven Technical University using the vapour-liquid-solid (VLS) method (Wagner *et al.* 1964). In this process, Au droplets of ~ 10 nm are dispersed on a pristine Ge [111] substrate and serve as a catalyst and precipitation site for a single nanowire. Germane (GeH_4), used as a precursor for the Ge core, dissociates at elevated temperatures on the Au droplet surface into Ge and H_2 . Ge is adsorbed by the Au until it becomes supersaturated, at which point Ge is deposited on the substrate, pushing the droplet upwards. This is an epitaxial process which, ideally, results in a mono-crystalline dislocation-free wire of several μm in length, with a diameter determined by the size of the initial droplet.

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¹ Ang Li, Erik Bakkers (PI)

Figure 3.1: TEM side view of a dislocation free Ge-Si core-shell nanowire segment with a core diameter of ~ 22 nm and a shell thickness of 3 nm.

Since the shell growth cannot be promoted by a catalyst droplet, a higher temperature is required to enable this radial growth. To prevent Au diffusion of the catalyst droplet from the top of the Ge core to the side walls at this elevated temperature, a Si segment is grown longitudinally on top of the core to physically separate the Au droplet. In this case, disilane (Si_2H_6) is used as precursor. Now, growth conditions are tuned to promote growth in the radial direction resulting in a Si shell. Figure 3.1 shows a TEM side-view of a defect-free nanowire segment where the core and shell are clearly discernible.

Apart from the preferential growth direction $[111]$, nanowires also tend to grow in the $[110]$ and $[211]$ orientation. Even though Si and Ge have a lattice mismatch of $\sim 4\%$, very low defect densities are realised in the $[110]$ direction, resulting in a heavily strained system (Conesa-Boj *et al.* 2017). In Chapter 4 we investigate the correlation between defect density and the resulting hole mobility of these different orientations. A more in-depth analysis on the strain is performed in a joint publication with our collaborators (Conesa-Boj *et al.* 2017).

3.2 DEVICE FABRICATION

This section highlights the relevant and novel elements used in device fabrication. We start with the least complex field-effect transistor (FET) nanowire devices and continue to devices used to define highly tunable quantum dots. During this research, several design and process improvements allowed us to decrease the gate distance, and therefore the quantum dot size, while increasing reproducibility. We will show

Figure 3.2: (a) FET device showing the nanowire (NW), the Ti/Pd source and drain and the SiO₂ covering the substrate. (b) Schematic device cross section.

three device generations that showcase the most important iterations. We now start by briefly describing the substrate used for all devices.

3.2.1 Wafer preparation

We use in-house thermally oxidised p^{++} Si [100] wafers with a 50, 100 or 200 nm thick layer of SiO₂. Using optical lithography, a total of 144 devices (dimensions 5 by 5 mm), each with 52 contact pads are defined on a single wafer. These contact pads are oriented around an area of 1.4 by 1.4 mm in the centre of each device, which serves as the active area.

3.2.2 Nanowire field-effect devices

Nanowire FET devices are used in Chapter 4 to correlate the hole mobility to the defect density and crystal orientation of the nanowires. Figure 3.2a shows an atomic-force microscopy (AFM) image while Figure 3.2b shows a schematic cross-section. In Figure 3.2a, the nanowire with a 23 nm diameter is covered on both sides by a source and drain, for which we use a stack of Ti (0.5 nm) and Pd (~50 nm) to obtain Ohmic contacts. The dark part surrounding source/drain is the SiO₂ covering the p^{++} Si substrate, which is used as a backgate. In these devices, nanowires are deposited on arbitrary locations on the substrate after which they are located and contacted.

Figure 3.3: (a) Schematic cross section of a highly tunable quantum dot device showing six gates between source and drain. (b) Bottom panel: AFM image of the first generation of quantum dot devices with a 100 nm gate pitch. Top panel: The height profile taken at the green line shows the gate protruding ~ 8 nm from the SiO_2 . (c) AFM image of the second device generation with a 60 nm gate pitch and only ~ 3 nm gate protrusion. In both (b) and (c), we see the NW, the Ti/Pd source drain, gates 1 to 6, the Al_2O_3 covering the gates, and the SiO_2 covering the substrate.

3.2.3 Highly tunable quantum dot devices

To be able to electrostatically define quantum dots, we need local electrostatic potential variations along the nanowire. To this goal, we use a set of six bottom gates oriented perpendicular to the nanowire, which are embedded in the SiO_2 that covers the substrate, as shown in Figure 3.3a. Embedding the gates increases electrostatic coupling to the wire and improves mechanical stability during wire deposition, where wire adhesion is determined by van der Waals interaction. The gates are defined using EBL and are embedded by an accurately timed buffered hydrogen fluoride (BHF) etch after developing the poly(methyl methacrylate) (PMMA)². We use atomic layer deposition (ALD) Al_2O_3 as a dielectric between the bottom gates and the nanowire. The nanowires are placed deterministically on top of a single set of gates using a micro-manipulator and an optical microscope.

When comparing the height profiles of a first generation device in Figure 3.3b and a second generation device in Figure 3.3c, the most important difference is the decrease in gate pitch from 100 to 60 nm and gate width from ~ 25 to ~ 15 nm. This was realised using cold development of PMMA (Cord *et al.* 2007) which we will discuss further in Section 3.2.6. From the height profiles we also notice a reduction in gate protrusion out of the substrate due to an optimised embedding process. Additionally, we (1) reduced the length of the bottom gates (a decision reversed in the next generation), (2) extended the Al_2O_3 to cover a larger portion of the gate contacts and (3) switched from BHF to tetramethylammoniumhydroxide (TMAH) as a more gentle and selective etching agent for Al_2O_3 , preventing over-etching.

3.2.4 Large gate array devices

The third device iteration is shown in Figure 3.4, where the gate lengths have increased from 4 μm to 18 μm , while the number of parallel gates has increased from 6 to 50 as can be seen in Figure 3.4b. The main advantage is that much less precision is needed when positioning nanowires, as long as they are oriented more or less perpendicularly to the bottom gate array. Since nanowire deposition is the most labour-intensive fabrication step, this greatly increases device throughput. After deposition, the gates with the most favourable positioning can be picked and contacted in a subsequent step. As default we increased the number of gates per wire from 6 to 8, while connecting

² We use PMMA 950k which is the most commonly used positive EBL resist

Figure 3.4: (a) Third generation device, gate length is increased from 4 μm to 18 μm and the number of parallel gates is increased from 6 to 50. Indicated are the Ti/Pd source and drain, gates 1 to 8, the Al₂O₃ covering the gates, and the SiO₂ covering the substrate. (b) Zoom in the blue rectangle in (a) where we see a 16 nm diameter nanowire. 8 gates run perpendicular to the wire below the Al₂O₃. (d) Zoom of the green rectangle in (a) taken before Al₂O₃ deposition showing an array of 50 parallel gates with a 60 nm pitch and a ~12 nm width. (c) Height profile over the gates with a pitch of 60 nm in (d) taken at the yellow line.

more gates is possible if the length of the nanowire is sufficient. Even though the length of the gates increased and the width of the gates is only ~ 12 nm, the yields are high. All gates have two contact pads and can be connected from both sides of the device and $\sim 90\%$ of the gates show conductivity in this configuration³. Since the gate that runs below the nanowire under investigation can usually connect to at least one gate contact, in most devices all gates could be used. This shows the high level of control on this part of the device fabrication.

3.2.5 Preliminary measurements: A comparison

The results presented in Chapter 5, Chapter 6 and Chapter 7 of this thesis have all been performed on the first generation of devices. However, for all three device generations we successfully defined quantum dots in a nanowire. Figure 3.5 shows gate versus gate plots for devices of all three generations. In Figure 3.5a current to drain I is plotted versus V_{g4} and V_{g6} while the gate in-between $V_{g5} = 0$ V. The exact gate numbers are arbitrary and a similar configuration is used for the second (Figure 3.5b) and third (Figure 3.5c) generation.

In all plots we observe diagonal lines characteristic for a single quantum dot defined between the two gates (see Chapter 2). The diagonal lines are spaced closer together for the first generation device compared to the others two, which at equal electrostatic coupling to the gates is consistent with a larger gate pitch, i. e., a larger quantum dot. A more quantitative comparison proves difficult because of different thickness of Al_2O_3 across devices, and asymmetric gate coupling to the quantum dot evident from the lines of increased current being not perfectly diagonal.

We notice other differences: the second and third generation require much higher operation voltages and, especially for the third generation device, more charge switching events are present. We cannot correlate these negative properties to changes in the device design and they must therefore be attributed to either the nanowires themselves or to the quality of the used Al_2O_3 . We note these measurements are performed at $T = 4$ K without any filtering at low temperature, and we have observed drastic changes in devices properties for first-generation devices (lower operation voltages and less charge switching events) when measuring these devices in a dilution refrigerator. Unfortunately, time con-

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³ Nanowire deposition may result in damaged gates. This statistic is only based on substrates that were undamaged.

Figure 3.5: Current I vs V_{gi} and V_{gj} measured at $T = 4$ K where voltages are applied only the gates i, j while a bias $V_{SD} = 1$ mV is applied and (a) First generation device showing V_{g4} vs V_{g6} , (b) Second generation device showing V_{g6} vs V_{g4} , (c) Third generation device showing V_{g6} vs V_{g8} . Diagonal lines of peaks in current indicate an intentional quantum dot between the two gates.

Figure 3.6: SEM image of a non-embedded gate structure with a 40 nm pitch and ~ 12 nm gate width. The PMMA layer used was 55 nm. Gates are evaporated Ti/Pd (1/11 nm) and the substrate is 100 nm SiO₂ in Si.

straints have not allowed us to measure second or third-generation devices at temperatures lower than 4 K.

Although the third device generation still has to prove itself, we believe the design is a large improvement in terms of flexibility (any of the 50 gates can be connected as desired) and ease of fabrication (nanowires can be deposited anywhere on the gate array).

3.2.6 Decreased pitch and cold development

All devices are fabricated using an EBL system equipped with a 30 keV electron beam, a voltage where proximity effects cause background exposure of resist (in this case PMMA) up to microns away from the actual exposed region (Liming Ren *et al.* 2004). Patterning different structures within this proximity zone causes cross-exposure of the structures resulting in unwanted broadening of the exposed pattern. One way to correct for this is to use proximity correction, which can be performed

Figure 3.7: (a) Nanowire FET device with superconducting Al leads. The nanowire can be seen between source and drain with a channel length of 150 nm. (b) Schematic cross section of the device in (a).

in various ways (Li 2015). However, this does not solve the underlying problem of the background exposure itself and cannot completely eliminate broadening effects.

Alternatively, we use cold development of PMMA. By cooling down the developer (methyl isobutyl ketone (MIBK)-isopropyl alcohol (IPA) 1:3) to -15°C it becomes much more selective towards shorter PMMA chains, i. e., highly exposed regions. This has three effects: (1) the PMMA exposed by the proximity effect is no longer developed and broadening of exposed areas is eliminated (Cord *et al.* 2007), (2) the required exposure dose to clear PMMA increases while structures become considerably less sensitive to variations in exposure dose, and this thus boosts reproducibility and (3) the shape of the developed channels has almost no undercut which is beneficial for creating embedded gates with our isotropic etching process.

To show the limits of this technique for our 30 kV EBL system, we show a 40 nm pitch gate structure in Figure 3.6 with non-embedded gates⁴, a feature size that is normally only available using high kV EBL systems. These structures can be routinely fabricated, even for a variety of exposure doses, underlining the robustness of this development technique.

⁴ Since gates are embedded using an isotropic BHF etch, this reduces the effective pitch by approximately the depth of etching.

3.2.7 *Superconductor-semiconductor nanowire devices*

Figure 3.6 shows the nanowire Josephson FET device used in Chapter 8, Chapter 9 and Chapter 10. Compared to the normal state FET devices in Figure 3.2 these devices have a shorter channel length of ~ 150 nm in the order of the coherence length of the semiconducting nanowire, while the aluminium source/drain serve to induce a proximity supercurrent. Each source and drain contact has two independent connections to enable four-terminal measurements.

Transparent contacts from the Al source and drain to the Ge-Si nanowire have been realised by performing an annealing step: devices are placed on a hotplate in ambient conditions at 180°C for 10 min. Annealing of super-semi devices will be discussed further in Chapter 10.

HOLE MOBILITY IN NANOWIRE FIELD-EFFECT TRANSISTORS

In this chapter, we determine the field-effect hole mobility μ of Ge-Si core-shell nanowires and look for correlation with the wire diameter and the crystal orientation. μ is a measure for the amount of scattering sites in the wire (Sze 1995) and therefore serves as a method to compare wire quality. In previous studies, Ge-Si core-shell nanowires have shown mobilities between 800 and 1000 cm²/Vs at liquid helium temperature ($T = 4$ K) (Hao *et al.* 2010; Higginbotham *et al.* 2014a; Higginbotham *et al.* 2014b) while at room temperature ($T = 300$ K) mobilities between 100 and 730 cm²/Vs have been reported (Hao *et al.* 2010; Nguyen *et al.* 2014; Smith *et al.* 2010; Xiang *et al.* 2006a; Zhao *et al.* 2011). There is another report of mobilities up to 1800 cm²/Vs at 77 K for modulation doped wires (Dillen *et al.* 2014). Here, we find mobilities that are much higher with a record mobility of 4268 cm²/Vs at 4 K and 1535 cm²/Vs at 300 K. Additionally, we present a method to improve the accuracy of the mobility extracted from measurement data: for every device we average μ for a wide range of applied bias voltages where we only include data where a linear transconductance is observed as required for the used model (Lundstrom 2001). Together with extensive error calculations, this proves especially useful to obtain accurate values of μ at low temperatures.[†]

After electrical characterisation, nine test devices were inspected using TEM, for which we can relate μ to the crystal orientation of the wire. As explained in Section 3.1, three crystal orientations are known to exist for these wires: [111], [110] and [112] of which only the first two were found on devices that have been electrically characterised. We find that wires with a [110] orientation have the smallest diameter, the

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[†] Part of this work was published in ‘Boosting Hole Mobility in Coherently Strained [110]-Oriented Ge-Si Core-Shell Nanowires’, S. Conesa-Boj, A. Li, S. Koelling, M. Brauns, J. Ridderbos, T. T. Nguyen, M. A. Verheijen, P. M. Koenraad, F. A. Zwanenburg and E. Bakkers, Nano Letters 17.4 (2017)

lowest defect density and the highest mobility (Chu *et al.* 2012). Finally, we will estimate the charge carrier density in the nanowires.

In our measurements we use nanowire FETs consisting of a nanowire connected by ohmic source-drain contacts lying on a thermally oxidised silicon wafer, which functions as a backgate. A more detailed description of the devices is given in Section 3.2.2. Two-point resistances between source and drain typically vary from 20 to 200 k Ω , and we estimate the contact resistance to be ~ 4 k Ω per contact, by comparing to four-probe measurements.

We extract the mobility in the linear regime of operation where we apply a source-drain bias V_{SD} while measuring the drain current I as a function of gate voltage V_{BG} . Since we assume an (inverse) linear relation between I and V_{BG} , we can define the transconductance as $G_t = -\Delta I / \Delta V_{BG}$, which is directly proportional to μ (Galup-Montoro *et al.* 2007; Malik 1995):

$$\mu = \frac{G_t L^2}{V_{SD} C_G} \quad (4.1)$$

with L the channel length of the nanowire transistor and C_G the nanowire-to-backgate capacitance (Sze 1995; Wunnicke 2006) which we estimate by simulation using a finite-element method (see Section 4.2). In literature, often a linear fit to a single I - V_{BG} curve is used to estimate μ . At 300 K μ is independent of V_{SD} and this approach is valid but at 4 K, Coulomb effects and the limited number of subbands participating in transport can cause G_t to fluctuate as a function of applied gate voltage, especially at low V_{SD} . Therefore, our method extracts an averaged value of μ from many I - V_{BG} characteristics taken in a large range of biases $|V_{SD}| < 10$ mV for every device. Next to this, we remove the data where the linear field-effect mobility model no longer applies, for example when transport is dominated by Coulomb oscillations (see Section 4.1 and Section 4.2, grey curves in Figure 4.1). To clarify the method, Figure 4.1 shows I - V_{BG} curves for four different devices at 4 K (left) and at 300 K (right). For each curve, G_t is determined as the slope of a least-squares linear fit between the fitting limits.

This chapter consists of 3 sections:

- Section 4.1 starts with the limitations and restrictions of the used model and continues with all steps taken to determine μ and the

corresponding standard deviation σ . We start with calculating individual mobility values μ_i and a standard deviation σ_i for each V_{SD} in Section 4.1.1. This is followed by the selection of the bias regime in Section 4.1.2. Next, we calculate the averaged μ in Section 4.1.3 and end with the estimation of the total standard deviation σ in Section 4.1.4. Both Figure 4.1 and Figure 4.2 include the measurements of our highest mobility nanowire, Device #9 ($\mu = 4268 \text{ cm}^2/\text{Vs}$ at 4 K).

- Section 4.2 is dedicated to the simulation of the nanowire-to-backgate capacitance C_G which is tailored to the specific geometry of each device.
- The resulting mobilities are presented in Section 4.3 and related to their wire diameter and crystal orientation.
- Section 4.4 shows how the hole density in the nanowires is calculated based on μ

4.1 EXTRACTING μ FROM MEASUREMENT DATA

Before discussing the analysis of the measurement data, let us consider the constraints imposed by the linear model in Equation (4.1): As said, the device should operate in the linear regime, i. e., V_{SD} must be far away from saturation so that I scales linearly with V_{SD} (Galup-Montoro *et al.* 2007). This holds for the used range $|V_{SD}| < 10 \text{ mV}$, however at 4 K we need to consider Coulomb effects, as we will see in Section 4.1.2. Another requirement is that $V_{SD} \ll V_{BG}$, so that V_{SD} does not significantly contribute to the effect of the backgate. Since V_{BG} is operated over a range of several volts, this requirement is met. Next, the channel length should be much larger than its own width and the thickness of the used dielectric (Malik 1995). Our devices satisfy this condition: the channel lengths are between 700 and 1 000 nm, the oxide thickness is between 50 and 100 nm and the wire diameter is between 15 and 45 nm.

4.1.1 Calculating μ_i and σ_i

Here, we explain how $\mu_i(V_{SD})$, the individual mobility for each value of V_{SD} , and its standard deviation σ_i are extracted from the I - V_{BG} plots in Figure 4.1. We inspect 4 devices: Device #9 and #5 with a $[110]$ orientation and Device #1 and #2 with a $[111]$ orientation. All devices are measured at 4 K and 300 K. In these turn-off curves, the devices

Figure 4.1: (a), (c), (e) and (g) I versus V_{BG} at 4 K for Devices #9, #5, #1 and #2; (b), (d), (f) and (h) I versus V_{BG} at 300 K for Devices #9, #5, #1 and #2. V_{SD} is varied from 1 mV to 10 mV in steps of 1 mV. For each curve, a least-squares linear fit (dotted lines) is performed between the fitting limits denoted by the vertical lines and arrows. Only the blue curves and the corresponding fits are used in the final calculation of μ .

are switched from an ON to an OFF state by increasing V_{BG} . A least-squares linear fit of the data between the fitting limits (the two vertical black lines) yields an average value of G_t . Equation (4.1) then gives the corresponding mobility $\mu_i(V_{SD})$, as shown in Figure 4.2a at 4 K and in Figure 4.2b at 300 K for both Device #9 [110] and Device #1 [111]. The precise position of fitting limits is determined by visual inspection in order to select the largest part of the I - V_{BG} curve that is still considered linear. At 300 K this can be easily estimated, however, at 4 K defining a linear region is more ambiguous. The larger deviation from linearity also results in a larger standard deviation σ_i , which is estimated as

$$\sigma_i = \sqrt{\frac{\sum_{r=1}^{N_i} (I_{r,i} - \mathbf{I}_{r,i})^2}{(N_i - 1)}} \frac{L^2}{\Delta V_{G,i} V_{SD,i} C_G} \quad (4.2)$$

where $I_{r,i}$ is the measured current, $\mathbf{I}_{r,i}$ is the current predicted by the linear fit, N_i is the number of measurements of $I_{r,i}$ within the fitting limits, and $\Delta V_{G,i}$ is the range of V_{BG} within the fitting limits. The square-root term is the standard deviation over the residuals of the fit $(I_{r,i} - \mathbf{I}_{r,i})^2$, in units of I . This standard deviation is then converted to units of μ_i by the second term in Equation (4.2) and plotted together with μ in Figure 4.2a (4 K) and Figure 4.2b (300 K) for Device #9 and #1.

4.1.2 Bias range selection

We now come to determine which $\mu_i(V_{SD})$ will be averaged to obtain the final nanowire mobility μ . At 300 K nearly the complete dataset can be used because both μ_i and σ_i show no significant variation as a function of V_{SD} as can be seen for Device #9 and Device #1 in Figure 4.2b. Only when $|V_{SD}| < 0.3$ mV, measurement noise starts to obscure the signal, and the corresponding μ_i are excluded. Figure 4.2d and Figure 4.2f show the complete dataset for these devices at 300 K plotted as numerical differential conductance dI/dV_{SD} . The conductance is independent of V_{SD} , indicating we are operating in the linear regime of the device.

At 4 K, the values of $\mu_i(V_{SD})$ and $\sigma_i(V_{SD})$ start to fluctuate when $|V_{SD}| < 3.5$ mV for Device #9 and when $|V_{SD}| < 5$ mV for Device #1 (Figure 4.2a). When inspecting the corresponding conductance maps in Figure 4.2c and Figure 4.2e, Coulomb blockaded regimes can be identified in this specific range of V_{SD} . For all devices, we exclude the range of V_{SD} , which shows clear Coulomb blockade since the lin-

ear field-effect mobility model (Equation (4.1)) no longer applies here. Next, we combine the obtained values $\mu_i(V_{SD})$ within the selected bias range to obtain an averaged value μ .

4.1.3 The final value for the mobility μ

To extract the final mobility value μ , we take a weighted average over the selected $\mu_i(V_{SD})$ values (see Figure 4.2a and Figure 4.2b) as

$$\mu = \frac{\sum_{i=1}^n \mu_i w_i}{\sum_{i=1}^n w_i} \quad (4.3)$$

Here, n is the number of selected values of $\mu_i(V_{SD})$ and w_i are the corresponding weighting factors, defined as $w_i = 1/\sigma_i^2$, i. e., the inverse of the variance of μ_i . Values of μ_i with smaller standard deviations thus have a bigger contribution to μ . For Device #9 we obtain $\mu = 1535 \text{ cm}^2/\text{Vs}$ at 300 K and $\mu = 4268 \text{ cm}^2/\text{Vs}$ at 4 K, while for Device #1 we obtain $\mu = 369 \text{ cm}^2/\text{Vs}$ at 300 K and $\mu = 1209 \text{ cm}^2/\text{Vs}$ at 4 K

4.1.4 Calculation of the total standard deviation σ

To calculate the total standard deviation σ of the previously extracted μ , we need a sum with the same weighting factors as in Equation (4.3) over the residuals of all selected measurement data with respect to the reference function $D_{r,\mu}$ which must be a linear function that yields μ if plugged into Equation (4.1). This results in $D_{r,\mu} = G_{t,\mu} V_{BG} + \beta$, where $G_{t,\mu}$ is the normalised transconductance and β is the intercept at $V_{BG} = 0$, both normalised to V_{SD} . To compute the residuals, we also normalise the measurement data to V_{SD} so they have the same unit as $D_{r,\mu}$. This gives the following expression for the total standard deviation σ as

$$\sigma = \sqrt{\frac{\sum_{i=1}^n \sum_{r=1}^{N_i} \left(\frac{I_{r,i}}{V_{SD,i} \Delta V_{G,i}} - \frac{D_r}{\Delta V_{G,i}} \right)^2 w_i}{\sum_{i=1}^n w_i (N_i - 1)}} \frac{L^2}{C_G} \quad (4.4)$$

Comparing with Equation (4.4) with Equation (4.2) we see that the square-root term now contains the standard deviation of the residuals in units $I/V_{SD} V_{BG} = G_t/V_{SD}$ and that the weighting factor w_i is assigned for every set of residuals. Since the first term is now in units of G_t/V_{SD} , the second term converts to units of μ . For Device #9 we now obtain $\mu = 1535 \pm 31 \text{ cm}^2/\text{Vs}$ at 300 K and $\mu = 4268 \pm 219 \text{ cm}^2/\text{Vs}$ at 4 K, while for Device #1 we obtain $\mu = 1209 \pm 188 \text{ cm}^2/\text{Vs}$ at 300 K and $\mu = 369 \pm 2 \text{ cm}^2/\text{Vs}$ at 4 K.

Figure 4.2: (a) μ_i versus V_{SD} at 4 K and (b) at 300 K (blue dots) for Device #9 and Device #1. The blue shaded area is the corresponding standard deviation σ_i . The black vertical lines denote the selected bias region for determining μ and σ . (c) Numerical differential conductance $|dI/dV_{SD}|$ versus V_{SD} and V_{BG} at 300 K for Device #9. (d) The same plot as (c) now at 300 K. (e) $|dI/dV_{SD}|$ versus V_{SD} and V_{BG} at 300 K for Device #1. (f) The same plot as (e) now at 300 K.

Table 4.1: Polynomial fits of the simulated capacitance to the wire diameter for the three combinations of channel length and oxide thickness as shown in Figure 4.3b.

	L (nm)	SiO ₂ (nm)	2nd order polynomial fit:	R ² :
A	800	55	$C_G = -2.95 \cdot 10^{-3} d^2 + 0.685d + 19.8$	0.999
B	1000	105	$C_G = -2.75 \cdot 10^{-3} d^2 + 0.594d + 21.4$	0.999
C	700	105	$C_G = -1.85 \cdot 10^{-3} d^2 + 0.395d + 14.1$	0.998

4.2 MODELING C_G USING FINITE-ELEMENTS

In this section, we describe how we determine the nanowire-to-backgate capacitance using a finite element approach, which is shown to agree well with direct measurements of nanowire capacitances (Tu *et al.* 2007). Since all charge carriers are localised in the Ge, it is this volume that defines the capacitance. We model the wire as a Ge core with a Si shell thickness of 2.5 nm and a 1 nm thick native SiO₂ layer, both estimated from TEM data of multiple wires. We define a 3-dimensional model, in which material properties are assigned as shown in Figure 4.3, except for the Ge core and the Si substrate, because they are modelled as metallic terminals for the electrostatics

Figure 4.3: (a) Schematic top view (upper panel) of the nanowire device as used in the finite-element simulation. The black dotted line shows the cross-section in the lower panel. The Ge core and the Si substrate (denoted by *) are configured as metallic terminals as is required for the electrostatics calculations. This is a valid assumption based on the high charge carrier density (see Section 4.4). (b) Capacitance versus wire diameter for three configurations of channel length and oxide thickness as calculated by the finite-element method.

calculation. This approximation is valid because of the high charge carrier density in both materials, even at 4 K (see Section 4.4).

We calculate the capacitance for three types of devices with varying SiO_2 thickness (d_{ox}) and channel length (L) that correspond to our devices, and plot them as a function of total wire diameter (Ge core with a Si shell and native SiO_2) in Figure 4.3b. Next, a 2nd-order polynomial fit on the capacitances (see Table 4.1) is used to interpolate the capacitance for any nanowire diameter between 12 and 77 nm. The last column shows the correlation constant R^2 which is very close to a perfect fit ($R^2 = 1$).

To conclude, the finite element calculation on the 3-dimensional model resembles the real device as close as can be reasonably realised and together with the excellent quality of the fit, we obtain nanowire capacitances with a high accuracy for any wire diameter between 12 and 77 nm.

4.3 MOBILITY μ VERSUS WIRE DIAMETER AND CRYSTAL DIRECTION

Figure 4.4a and Figure 4.4b show mobilities and errors for all measured devices at 4 K and 300 K respectively. In both plots, the data points form two clusters with respect to the wire diameter. Wires with low diameter have a higher mobility for which we have only detected a $[110]$ crystal orientation. The larger diameter wires only appear in the $[111]$ direction and have a much lower mobility.

Figure 4.4c shows the defect density as determined from TEM, versus the wire diameter, for a large number of wires, which were not electrically characterised. The crystal orientation is determined from high-resolution TEM images such as Figure 4.4d, which shows a $[110]$ wire without dislocations between the Ge mono-crystalline core and Si shell. In Figure 4.4c we also find the $[112]$ crystal direction which, on average, shows a lower defect density and diameter than the $[111]$ orientation. The fact that we did not find this orientation in the electrically characterised devices in Figure 4.4a and Figure 4.4b is most likely due to the low sample size. Most strikingly, for all three wires with $[110]$ crystal direction, no dislocation faults within the inspected wire segment was found, indicating that these wires are of very high quality and must be highly strained to accommodate the $\sim 4\%$ lattice mismatch between the Ge core and the Si shell. This strain modifies the semiconducting band structure in the wire which is predicted to further increase the mobil-

Figure 4.4: (a) μ vs wire diameter at 4 K. The crystal orientation is denoted on the TEM inspected devices (red for [110], blue for [111]). For the grey datapoints no crystal orientation is available. (b) μ vs wire diameter at 300 K for the same wires as (a). (c) Defect density vs nanowire diameter for three crystal orientations. (d) High-resolution TEM image of a [110] nanowire segment.

ity (Dayeh *et al.* 2013; Li *et al.* 2014; Peköz *et al.* 2009). For an in-depth analysis on the strain in these wires see ref. (Conesa-Boj *et al.* 2017).

4.4 ESTIMATION OF THE HOLE DENSITY

We take Device #9 at 300 K in Figure 4.1b as an example to calculate the hole density $n(V_{\text{BG}})$, which can be approximated in two ways. First we calculate the total accumulated charge relative to depletion ($n = 0$ at $V_{\text{BG}} \approx 13$ V). Assuming the number of holes increases linearly with decreasing V_{BG} , the total charge on the wire is given by $\Delta Q = -C_G \cdot \Delta V_{\text{BG}}$ with ΔV_{BG} the voltage below depletion. We calculate n at $V_{\text{BG}} = 9$ V, i.e., at the left fitting limit of the linear fit in Figure 4.1b so $\Delta V_{\text{BG}} = -4$ V. For this device, $C_G \approx 20$ aF which gives

$\Delta Q \approx 7.9 \cdot 10^{-17}$ Coulomb. We divide by the volume of the nanowire to find the hole density $n_{300K} = \Delta Q / (\pi r^2 L e) \approx 1.4 \cdot 10^{19} \text{ cm}^{-3}$ with L the channel length (700 nm) and r the radius of the Ge core (4 nm)¹. Using the same procedure for the data at 4 K (Figure 4.1a), a value of $n_{4K} \approx 1.0 \cdot 10^{19} \text{ cm}^{-3}$ is obtained.

The second method uses the relation $\kappa = ne\mu$ or $n = \kappa / e\mu$ which relates the conductivity of a material κ to the mobility μ . $\kappa = GL / (\pi r^2)$ where G is the conductance extracted from measurements. In Figure 4.1a (300 K), at the left-most boundary of the linear fit ($V_{BG} = 9 \text{ V}$), $G = 4.4 \cdot 10^{-5} \text{ S}$, $\kappa = 6 \cdot 10^5 \text{ S/m}$ while $\mu = 1535 \text{ cm}^2/\text{Vs}$, resulting in $n_{300K} \approx 2.4 \cdot 10^{19} \text{ cm}^{-3}$. At 4 K (Figure 4.1b, $V_{BG} = 2 \text{ V}$) $\kappa = 9 \cdot 10^5 \text{ S/m}$ and $\mu = 4268 \text{ cm}^2/\text{Vs}$ resulting in $n_{4K} \approx 1.3 \cdot 10^{19} \text{ cm}^{-3}$, which are very high values for undoped semiconducting materials (Schroder 2005) and is an effect of the staggered band alignment between the Ge core and the Si shell in combination with strain (Peng *et al.* 2011). Since we can increase the number of holes in the wire for more negative V_{BG} (although not with a linear relation to V_{BG}), our maximum hole density is likely to be higher.

4.5 CONCLUSION

To summarize, we have demonstrated a technique to determine the mobility with a careful choice of fitting limits and a consistent method for selection of the physically relevant data from a large dataset. In addition, we use a weighted average based on the quality of the linear fits to obtain a reliable value for μ and use a detailed finite-element method to accurately determine the nanowire to backgate capacitance C_G . We correlate wire diameter with crystal orientation and observe that wires with diameters between 15 - 25 nm have the highest mobility and, as predicted (Chu *et al.* 2012), predominantly grow in the $[110]$ direction. To our knowledge, this is the most thorough study for determining mobilities at low temperatures in semiconductor nanowire devices where, in addition, TEM studies are correlated to the electrical measurements.

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¹ As explained in Section 4.3, for a total wire diameter of 15 nm, we subtract the native SiO_2 ($2 \times 1 \text{ nm}$) and the Si shell ($2 \times 2.5 \text{ nm}$), resulting in a core diameter (radius) of 8 (4) nm respectively.

HIGHLY TUNEABLE HOLE QUANTUM DOTS IN GE-SI CORE-SHELL NANOWIRES

We define single quantum dots of lengths varying from 60 nanometers up to nearly half a micron in Ge-Si core-shell nanowires. The charging energies scale inversely with the quantum dot length between 18 and 4 meV. Subsequently we split up a long dot into a double quantum dot with separate control over the tunnel couplings and the electrochemical potential of each dot. Both single and double quantum dot configurations prove to be very stable and show excellent control over the electrostatic environment of the dots, making this system a highly versatile platform for spin-based quantum computing.[†]

[†] This work was published as ‘Highly tuneable hole quantum dots in Ge-Si core-shell nanowires’, M. Brauns, J. Ridderbos, W. G. van der Wiel, A. Li, E. P. A. M. Bakkers and F. A. Zwanenburg, *Applied Physics Letters* 109.14 (2015)

5.1 INTRODUCTION

For spin-based quantum computing (Loss *et al.* 1998), increasing research efforts have focused in recent years on C, Si, and Ge (Amato *et al.* 2014; Laird *et al.* 2015; Zwanenburg *et al.* 2013) because they can be isotopically enriched to only contain nuclei with zero spin (Itoh *et al.* 2003; Itoh *et al.* 1993) and thus exhibit exceptionally long spin lifetimes (Muhonen *et al.* 2014; Veldhorst *et al.* 2014). The one-dimensional character of Ge-Si core-shell nanowires leads to unique electronic properties in the valence band, where heavy and light hole states are mixed (Csontos *et al.* 2009; Csontos *et al.* 2007; Kloeffer *et al.* 2011). Early experiments in Ge-Si core-shell nanowires include experiments on double quantum dots (Hu *et al.* 2007) and spin relaxation times (Hu *et al.* 2012). The band mixing causes an enhanced DRSOI (Kloeffer *et al.* 2011), which can be exploited for efficient spin manipulation (Kloeffer *et al.* 2013b). Therefore, Ge-Si core-shell nanowires are an ideal platform for future quantum computation applications.

In this Letter, we define single quantum dots of several lengths in a Ge-Si core-shell nanowire. We controllably split longer quantum dots up into double quantum dots with tuneable interdot tunnel coupling. Both single and double quantum dots show an exceptional degree of measurement stability.

5.2 DEVICE DESIGN

We will discuss measurements in two different devices $D1$ and $D2$ (see Figure 5.1) on two different chips, which have been fabricated in the same way: A p^{++} -doped Si substrate is covered with 200 nm SiO_2 , on which six bottom gates $g1$ - $g6$ with 100 nm pitch are patterned with EBL. Before metallization of the bottom gates, a 13 s buffered hydrofluoric acid dip etches 20 nm deep trenches into the SiO_2 , so that the bottom gates (approximately 20 nm thick) are sunken into the SiO_2 for an improved planarity. The gates are covered with 10 nm Al_2O_3 grown with ALD at 100 °C. Two single nanowires with a Si shell thickness of 2.5 nm and a Ge core radius of 8 nm ($D1$) and 9 nm ($D2$) are deterministically placed on top of the gate structure with a micromanipulator. Based on TEM studies of similar wires, both the core and the shell are monocrystalline, and their axis is likely pointed along the $[110]$ crystal axis (Conesa-Boj *et al.* 2017). Subsequently we define ohmic contacts to the nanowires and gate contacts made of Ti/Pd (0.5/50 nm) with EBL. The nanowire parts above the bottom gates are at no point exposed to the electron beam, preventing carbon deposition and introduction of

Figure 5.1: False-colour AFM image of device (a) $D1$, and (b) $D2$. (c) Schematic cross-section displaying the p^{++} -doped Si substrate (grey) with 200 nm of SiO_2 (dark red), six bottom gates $g1$ - $g6$ (light red), each 35 nm wide and with a pitch of 100 nm. The bottom gates are buried under 10 nm of Al_2O_3 (yellow), on top of which the nanowire is deposited (green) and ohmic contacts (0.5/50 nm Ti/Pd, blue) are defined.

defects into the otherwise defect-free Ge core. All measurements are performed using direct current (DC) electronic equipment in a dilution refrigerator with a base temperature of 8 mK. A bias voltage V_{SD} is applied to source, the current I is measured at the drain contact. An effective hole temperature of $T_{\text{hole}} \approx 30$ mK has been determined in one of the devices by measuring the temperature dependence of the Coulomb peak width (Goldhaber-Gordon *et al.* 1998; Mueller *et al.* 2013).

5.3 SINGLE QUANTUM DOTS OF VARYING LENGTH

By using different gates to induce tunnel barriers we can form quantum dots in our nanowire with lengths varying from very long quantum dots (using $g1$ and $g6$) to very short dots (using adjacent gates). This flexible quantum dot length together with a tuneable tunnel coupling between the quantum dot and the reservoirs is a great improvement compared to using lateral heterostructures (Fuhrer *et al.* 2007; Roddaro *et al.* 2011), or Schottky barriers at the nanowire-metal interface with the contacts (Nilsson *et al.* 2011; Zwanenburg *et al.* 2009a).

We assume the length of our gate-defined quantum dots to be the the distance between the inner edges of the barrier gates. Using a gate

width of $\sim 40\text{nm}$ this results in quantum dot lengths of $\sim 60\text{nm}$ for adjacent barrier gates, $\sim 160\text{nm}$ for barrier gates with one plunger gate in between, $\sim 260\text{nm}$ for two plunger gates, $\sim 360\text{nm}$ for three plunger gates, and $\sim 460\text{nm}$ for four plunger gates, i. e., we are able to tune the dot length over almost an order of magnitude.

In Figure 5.2a-Figure 5.2e we plot $dI/dV \equiv dI/dV_{SD}$ versus V_{SD} and the voltage on the plunger gate V_p . The formation of quantum dots of five different lengths is reflected in the clear Coulomb diamonds. The shortest quantum dot is formed in device $D1$ [Figure 5.2a]. The quantum dots formed with one up to four plunger gates are formed in both devices $D1$ and $D2$, Figure 5.2b-Figure 5.2e display bias spectroscopies of quantum dots formed in $D2$.

We extract the respective charging energies E_C for both devices from the Coulomb diamond height and find a decreasing E_C from 18.3 meV to 4.2 meV , inversely proportional to the increasing dot length [see Figure 5.2f and Table 5.1]. Since E_C is linked to the total capacitance C of the quantum dot via $E_C = e^2/C$ (Kouwenhoven *et al.* 1997), C is directly proportional to the quantum dot length. E_C and C are highly consistent for the two devices.

For the quantum dot configurations with a dedicated plunger gate [Figure 5.2b-Figure 5.2e], E_C as well as the shape of the Coulomb diamonds stay constant over several charge transitions, reflecting the validity of the constant interaction model. In Figure 5.2a, E_C increases significantly from 16.7 meV to 20.0 meV and also the slopes of the Coulomb diamond edges change. We attribute this to using the right barrier gate as a plunger, leading to a decreasing dot size and changing capacitive couplings to this barrier gate and the adjacent reservoir. Therefore, the constant interaction model is not valid in this configuration, and the tuneability of the quantum dot is limited compared to the longer quantum dots with a dedicated plunger gate. We extract the values for E_C and C for the zero-plunger configuration from the middle Coulomb diamond, for which they are in line with those for the longer dots.

The constant charging energies over several Coulomb diamonds in Figure 5.2b-Figure 5.2e are accompanied by constant Coulomb peak spacings ΔV_g at $V_{SD} = 0$, indicating a constant gate capacitance C_g over several charge transitions, another indication for the validity of the constant interaction model. If we now compare the plunger gate capacitances between Figure 5.2b and Figure 5.2e, we find them to be all very similar, ($\sim 5.5\text{ aF}$), while the total capacitance increases linearly by $\sim 7.5\text{ aF}$ per additional plunger gate [see Figure 5.2f]. The discrepancy of $\sim 2\text{ aF}$ can be explained by the finite capacitance of the global back gate which increases with the dot length and the change in the self-capacitance of the quantum dot. The linearly increasing total capa-

Figure 5.2: Bias spectroscopy of gate-defined single quantum dots formed with (a) zero, (b) one, (c) two, (d) three, and (e) four gates between the barrier gates (indicated in blue). (a) is measured in $D1$, (b)-(e) in $D2$. (f) Charging energy E_C (left) and total capacitance C (right) of the dot plotted versus the dot length l . Red triangles measured in $D1$, black circles in $D2$.

Table 5.1: Parameters for electrostatically defined quantum dots of varying length as extracted from Figure 5.2a-Figure 5.2e.

l (nm)	E_C (meV)	C (aF)	ΔV_g (mV)	C_g (aF)
60	18.3(2)	8.8(2)	104(1)	1.54(2)
160	10.2(2)	15.7(3)	31.5(2)	5.09(3)
260	6.8(2)	23.5(5)	29.6(4)	5.41(7)
360	5.2(1)	30.8(6)	28.6(4)	5.63(8)
460	4.2(1)	38.6(9)	29.7(4)	5.39(7)

citance indicates equal coupling of all gates, consistent with the gate geometry (equal width and distance to the nanowire). In Figure 5.2f we also plot E_C and C for quantum dots formed in $D1$ with at least one plunger gate alongside the data for $D2$. The consistency between the data and therefore demonstrates a high degree of control over the electrostatic environment of the gate-defined quantum dot.

5.4 TUNEABLE DOUBLE QUANTUM DOTS

Tuneable double quantum dots are essential for spin readout via Pauli spin blockade (Ono *et al.* 2002). For a fully tuneable double quantum dot we need five gates: Three barrier gates to form tunnel barriers, and two plunger gates to tune the electrochemical potential of each dot separately. We use device $D1$ starting from a situation equivalent to Figure 5.2d, and increase the voltage on the middle gate V_{g4} . When approaching the pinch-off voltage, a tunnel barrier is formed and the single quantum dot splits up into two tunnel-coupled quantum dots.

The charge stability diagrams at four different V_{g4} are plotted in Figure 5.3a. We keep the outer barrier gates at constant voltages ($V_{g2} = 2490$ mV, $V_{g6} = 2940$ mV), and plot the current at a fixed $V_{SD} = 1$ mV. For $V_{g4} = 0$ mV we observe the typical stability diagram of a single quantum dot (van der Wiel *et al.* 2002). The spacing of the diagonal, parallel lines of finite current along the respective plunger gate axis is directly related to the capacitance between the quantum dot and this gate: $C_g = e/\Delta V_g$. We observe $\Delta V_g \approx 27$ mV for both $g3$ and $g5$, i.e. both gates have the same capacitance $C_g \approx 5.8$ aF to the quantum dot. This indicates that the quantum dot indeed stretches over the whole distance between the tunnel barriers above gates $g2$ and $g6$ and is also in agreement with the gate capacitances in Table 5.1.

Figure 5.3: (a) Charge stability diagrams with current I plotted versus V_{g3} and V_{g5} for varying voltages on g_4 at fixed $V_{g2} = 2490$ mV and $V_{g6} = 2940$ mV. (b) Charge stability diagram of a double quantum dot at $V_{g2} = 2500$ mV, $V_{g4} = 2100$ mV, and $V_{g6} = 3180$ mV. (k, l) denote the charge occupation numbers on the left (k) and right dot (l). All measurements performed on device $D1$.

At $V_{g4} = 2000\text{mV}$ the straight lines have evolved into a regular honeycomb pattern with two distinct slopes that form the long edges of each honeycomb, indicating the formation of a strongly coupled double quantum dot (van der Wiel *et al.* 2002). From the distance between adjacent parallel lines we extract the voltages needed to add a hole to the left (right) dot ΔV_{g3} , (ΔV_{g5}) and calculate the corresponding capacitances. For the left dot we find $\Delta V_{g3} = 25.1(5)\text{mV}$ and for the right dot $\Delta V_{g5} = 26.1(5)\text{mV}$, resulting in gate capacitances $C_{g3} = 6.3(2)\text{aF}$ and $C_{g5} = 6.1(2)\text{aF}$. The sets of honeycomb edges representing the addition of a hole to either the left or the right dot are both significantly slanted because of the mutual capacitive coupling C_M between the two dots that leads to a separation between the two triple points (van der Wiel *et al.* 2002). We express this shift in terms of gate voltages and find $\Delta V_{g3,M} = 9.2(5)\text{mV}$ and $\Delta V_{g5,M} = 10.9(5)\text{mV}$. Using the expression $C_{g,M} = C_g \Delta V_{g,M} / \Delta V_g$ (van der Wiel *et al.* 2002), we obtain $C_{g3,M} = 2.3(3)\text{aF}$ and $C_{g5,M} = 2.5(3)\text{aF}$. A second mechanism affecting the slopes of the honeycomb edges is the finite cross capacitance between $g3$ and the right dot $C_{g3,C}$, and $g5$ and the left dot $C_{g5,C}$. This cross capacitance leads to a shift of the triple points along the $g3$ gate axis while changing the charge occupation of the right dot, and along the $g5$ gate axis while changing the number of holes on the left dot. This effect is very weak, and we extract $C_{g3,C} \approx C_{g5,C} \approx 0.1\text{aF}$.

Increasing the voltage on the interdot barrier gate to $V_{g4} = 2700\text{mV}$ only slightly changes the gate capacitances to $C_{g3} = 5.9(2)\text{aF}$ and $C_{g5} = 5.7(2)\text{aF}$. For the mutual gate capacitances we find a much stronger relative change to $C_{g3,M} = 0.6(1)\text{aF}$ and $C_{g5,M} = 0.7(1)\text{aF}$, which indicates a significantly increased separation of the charge distribution of both dots. The now only faintly visible long edges of the honeycombs also suggest a decreased tunnel coupling to the reservoirs so that cotunnelling is suppressed (De Franceschi *et al.* 2001). A finite, but very small cross capacitance of the plunger gates is also observed here, again on the order of 0.1aF .

A further increase of the interdot barrier gate to $V_{g4} = 2900\text{mV}$ completely quenches the cotunnelling current at the long honeycomb edges, so that now transport is only possible at the triple point pairs. This indicates well defined charge states confined in the quantum dots weakly coupled to the reservoirs. Again we observe a slight decrease of the gate capacitances to $C_{g3} = 5.8(2)\text{aF}$ and $C_{g5} = 5.5(2)\text{aF}$, and also the mutual capacitances decrease further to $C_{g3,M} = 0.4(1)\text{aF}$ and $C_{g5} = 0.4(1)\text{aF}$. All extracted capacitances are summarized in Table 5.2.

In Figure 5.3b we show a high-resolution stability diagram of a double quantum dot weakly coupled to the reservoirs at

Table 5.2: Capacitances for increasing voltage on the middle barrier gate g_4 of an electrostatically defined single ($V_{g4} = 0\text{ mV}$) or double quantum dot ($V_{g4} \geq 2000\text{ mV}$) as extracted from Figure 5.3a.

V_{g4} (mV)	C_{g3} (aF)	C_{g5} (aF)	$C_{g3,M}$ (aF)	$C_{g5,M}$ (aF)
0	5.8(3)	5.8(3)		
2000	6.3(2)	6.1(2)	2.3(3)	2.5(3)
2700	5.9(2)	5.7(2)	0.6(2)	0.7(2)
2900	5.8(2)	5.5(2)	0.4(1)	0.4(1)

$V_{SD} = -1.5\text{ mV}$ with barrier gate voltages of $V_{g2} = 2500\text{ mV}$, $V_{g4} = 2100\text{ mV}$, and $V_{g6} = 3180\text{ mV}$. Clearly visible is a very regular pattern of 25 bias triangle pairs, from which we extract the gate-to-dot capacitances in the same way as before. We obtain $C_{g3} = 5.9(2)\text{ aF}$ and $C_{g5} = 5.9(2)\text{ aF}$, and mutual capacitances of $C_{M,g3} = 0.9(1)\text{ aF}$ and $C_{M,g5} = 0.9(1)\text{ aF}$. The increased values for $C_{M,i}$ indicate an indeed increased capacitive coupling between the dots.

We extract the charging energies from Figure 5.3b by relating the bias triangle size to an energy of 1.5 meV . We obtain a charging energy of the left dot $U_1 = 10.6(5)\text{ meV}$ and of the right dot $U_2 = 9.3(5)\text{ meV}$. For the mutual charging energy U_M we extract $U_M = 1.5(2)\text{ meV}$. The size and shape of the bias triangles are exceptionally stable over the whole range of the measurement. This underlines the high degree of control over the electrochemical potentials of the quantum dots as well as the tunnel and capacitive couplings. Our devices are therefore exceptionally suitable for direct-transport experiments in comparison to other systems, where tunnel couplings change strongly when changing the charge occupation (Fasth *et al.* 2005; Li *et al.* 2015; Liu *et al.* 2008; Sand-Jespersen *et al.* 2008). Such experiments are relevant, because the applied bias between the two reservoirs serves as an energy scale, which, e.g., allows for the determination of the singlet-triplet splitting (Brauns *et al.* 2016a) and the Zeeman splitting (Brauns *et al.* 2016b).

In summary we demonstrate a high degree of control over the charge distribution in a double quantum dot. We have changed the mutual capacitances, a measure for the degree of separation of the dots, by a factor of six while keeping the capacitances between the left (right) dot and g_3 (g_5) almost constant. The corresponding charging energies are in agreement with the experiments on single quantum dots of the same length in D_2 .

5.5 CONCLUSION

In conclusion, we have electrostatically formed highly-tunable single and double quantum dots inside Ge-Si core-shell nanowires. We can vary the length of the single quantum dots from 60 nm to 460 nm corresponding to charging energies of the quantum dots varying from ~ 18 meV down to ~ 4 meV.

Furthermore, we have split a single quantum dot into a double quantum dot in a controlled way. Our low-cross-capacitance gate design enables us to keep the voltage on the outer barriers constant while varying the interdot barrier, i.e. it is not necessary to retune all gates. All capacitances and charging energies extracted from single and double quantum dot measurements are highly consistent. 25 bias triangle pairs form a very regular pattern in the stability diagram with constant triangle sizes, indicating an exceptional degree of control over the tunnel couplings over a large range of gate voltages.

This combination of tuneability and stability makes Ge-Si core-shell nanowires an ideal platform for further experiments towards quantum computation applications.

ELECTRIC-FIELD DEPENDENT g -FACTOR ANISOTROPY IN GE-SI CORE-SHELL NANOWIRE QUANTUM DOTS

We present angle-dependent measurements of the effective g -factor g^* in a Ge-Si core-shell nanowire quantum dot. g^* is found to be maximum when the magnetic field is pointing perpendicular to both the nanowire and the electric field induced by local gates. Alignment of the magnetic field with the electric field reduces g^* significantly. g^* is almost completely quenched when the magnetic field is aligned with the nanowire axis. These findings confirm recent calculations, where the obtained anisotropy is attributed to a Rashba-type spin-orbit interaction induced by heavy-hole light-hole mixing. In principle, this facilitates manipulation of spin-orbit qubits by means of a continuous high-frequency electric field.[†]

[†] This work was published as ‘Anisotropic Pauli spin blockade in hole quantum dots’, M. Brauns, J. Ridderbos, A. Li, E. P. A. M. Bakkers, W. G. van der Wiel and F. A. Zwanenburg, Physical Review B 93.12 (2016)

6.1 INTRODUCTION

Quantum computation (Aaronson 2013; DiVincenzo 1995; Ladd *et al.* 2010) has made an enormous leap from a far-fetched promise (Feynman *et al.* 1986) to a realistic near-future technology (Jones *et al.* 2012; Vandersypen *et al.* 2001; Veldhorst *et al.* 2015a) during the past three decades. Among others, spin systems in the solid state (Kane 1998; Loss *et al.* 1998) have been developed into a mature but still very fast-evolving research field. In recent years increased research efforts have focused on C, Si, and Ge (Amato *et al.* 2014; Laird *et al.* 2015; Zwanenburg *et al.* 2013), which can be purified to only consist of isotopes with zero nuclear spin (Itoh *et al.* 2003; Itoh *et al.* 1993) and thus exhibit exceptionally long spin lifetimes (Muhonen *et al.* 2014; Veldhorst *et al.* 2014).

The one-dimensional character of electrostatically defined quantum dots in Ge-Si core-shell nanowires leads to unique electronic properties in the valence band, where heavy and light hole states are mixed (Csontos *et al.* 2009; Csontos *et al.* 2007; Kloeffer *et al.* 2011). The band mixing gives rise to an enhanced Rashba-type spin-orbit interaction (Kloeffer *et al.* 2011), leading to strongly anisotropic and electric-field dependent g -factors (Maier *et al.* 2013). This makes quantum dots in Ge-Si core-shell nanowires promising candidates for robust spin-orbit qubits that can be electrically controlled via circuit quantum electrodynamics (Kloeffer *et al.* 2013a).

Despite these profound theoretical contributions, only few experiments in Ge-Si core-shell nanowires have been reported including Josephson junctions (Xiang *et al.* 2006b), spin-filling (Roddaro *et al.* 2008), spin relaxation (Hu *et al.* 2012), spin coherence (Higginbotham *et al.* 2014c), charge sensing (Hu *et al.* 2007) in the many-hole regime, and signatures of weak antilocalization (Higginbotham *et al.* 2014a).

In this work we experimentally explore the anisotropy of the g -factor in Ge-Si core-shell nanowires. We electrostatically define a highly-tunable, elongated hole quantum dot in the nanowire by means of local gates. We measure the Zeeman splitting of a single-particle state in the quantum dot while rotating the magnetic field around the high-symmetry axes of the system and find a strong anisotropy with respect to the nanowire as well as to the electric field, in line with theoretical predictions (Maier *et al.* 2013).

6.2 GATE-DEFINED QUANTUM DOTS

Our device in Figure 6.1a consists of a p^{++} -doped Si substrate covered with 200 nm SiO_2 , on which six bottom gates with 100 nm pitch are patterned with EBL. The gates are buried by 10 nm Al_2O_3 grown with ALD at 100°C. A single nanowire with a Si shell thickness of ~ 2.5 nm and a defect-free Ge core with a radius of ~ 8 nm (Conesa-Boj *et al.* 2017) is deterministically placed on top of the gate structure with a micromanipulator and then contacted with ohmic contacts made of 0.5/50 nm Ti/Pd. A source-drain bias voltage V_{SD} is applied to the source, the current I is measured between the drain and ground. All measurements are performed using dc electronic equipment in a dilution refrigerator with a base temperature of 8 mK and an effective hole temperature of $T_{\text{hole}} \approx 30$ mK determined by measuring the temperature dependence of the Coulomb peak width (Goldhaber-Gordon *et al.* 1998; Mueller *et al.* 2013).

We use this gate design (Fasth *et al.* 2005) to electrostatically define a single quantum dot (Fasth *et al.* 2005; Kouwenhoven *et al.* 1997). The two barrier gates g_3 and g_5 control the tunnel barriers, and a third plunger gate g_4 the electrochemical potential of the quantum dot. In Figure 6.1c we plot I versus the voltage on g_4 V_{g_4} . When applying a high $V_{\text{SD}} = 50$ mV we observe a strong suppression of I for $V_{g_4} > 2.5$ V, indicating depletion of the nanowire at $V_{g_4} \approx 2.5$ V. At low $V_{\text{SD}} = 1$ mV we observe Coulomb peaks (Kouwenhoven *et al.* 1997) with a regular spacing of $\Delta V_{g_4} \approx 30$ mV over a range of 2 V, i.e. we are able to change the hole occupation of the quantum dot by more than 60 holes. Above $V_{g_4} \approx 2$ V no regular Coulomb peaks are observed, but the high-bias gate sweep suggests that the quantum dot is not completely emptied, i.e. in this device we are unable to identify the last hole on the quantum dot. If we assume the plunger gate coupling to stay constant and the quantum dot to be empty at $V_{g_4} = 2.5$ V, we can estimate the number of remaining holes to be $N \approx 17$ at $V_{g_4} = 2$ V. Reaching the single-hole regime was not possible in our device.

A non-linear transport measurement is displayed in Figure 6.1d. In this bias spectroscopy we plot the numerical differential conductance $dI/dV \equiv dI/dV_{\text{SD}}$ vs. V_{SD} and V_{g_4} , as will be in all the following bias spectroscopy plots. Formation of a single quantum dot is indicated by regularly shaped, closing Coulomb diamonds (Kouwenhoven *et al.* 1997). The height of the Coulomb diamonds indicates an addition energy of $E_{\text{add}} \approx 8 - 10$ meV. The variations in E_{add} cannot be explained by an interacting second quantum dot, which would lead to

Figure 6.1: (a) False-color AFM image of the device. (b) Schematic cross-section displaying the p^{++} Si substrate (grey) with 200 nm of SiO_2 (dark red), six bottom gates g1-g6 (light red), each ~ 35 nm wide and with 100 nm pitch buried under 10 nm Al_2O_3 (yellow), on top of which the nanowire is placed (green) with ohmic contacts (0.5/50 nm Ti/Pd, blue). (c) Current I vs. V_{g4} with g3 and g5 forming tunnel barriers ($V_{g3} = 2060$ mV, $V_{g5} = 2260$ mV). Black curve is taken at $V_{SD} = 1$ mV, red curve at $V_{SD} = 50$ mV. (d) Numerical differential conductance dI/dV_{SD} plotted vs. V_{SD} and V_{g4} at the same barrier voltages as in (c).

non-closing diamonds. Orbital shell filling can cause the variations in E_{add} (Kouwenhoven *et al.* 2001a; Tarucha *et al.* 1996). The low number of residing holes (~ 25 -30) supports this reasoning.

The results in Figure 6.1 show a highly tunable nanowire device in which we intentionally define a very stable quantum dot. We can control the number of holes in the quantum dot over a wide range from approximately 85 down to approximately 17.

6.3 ZEEMAN SPLITTING OF THE ORBITAL GROUND STATE

We now investigate the Zeeman splitting ΔE_Z of the spin-degenerate quantum dot states (Hanson *et al.* 2007). To determine the g -factor with high accuracy, we choose a charge transition where the onset of conductance is sufficiently separated from other lines of increased conductance corresponding to, e.g., (orbital) excited states of the quantum dot, or resonances due to the low dimensionality of the leads (Escott *et al.* 2010). In Fig. 2(a) we show a bias spectroscopy of such a charge transition together with the two Coulomb diamonds adjacent to it. The number of residing holes here is approximately 35, again estimated by comparing the current plunger gate voltage ($V_{\text{g4}} \approx 1450$ mV) to the depletion voltage at high bias ($V_{\text{g4}} \approx 2.5 - 2.7$ V).

We keep the plunger-gate voltage at $V_{\text{g4}} = 1454.0$ mV and sweep V_{SD} along the green line in Figure 6.2a at different magnetic fields B while measuring the current [Figure 6.2b]. The magnetic field here is applied in the plane of the chip perpendicular to the nanowire axis. At $B = 0$, one very pronounced peak marks the onset of conductance, which splits up into a spin-ground and spin-excited state at finite magnetic fields. Note that the shifts of the two states are symmetric and linear up to at least 2 T, indicating that for magnetic fields $B < 2$ T the linear Zeeman splitting is the only relevant term, and other effects, such as a diamagnetic shift (Rinaldi *et al.* 1996; Zielke *et al.* 2014), are negligible. The spin splitting of the orbital ground state is further confirmed by the two bias spectroscopies in Figure 6.2d at $B = 0$ and $B = 1$ T. The spin-degenerate orbital ground state of the charge transition at $B = 0$ [indicated by a single green arrow Figure 6.2d] is clearly split into two lines at $B = 1$ T (indicated by two green arrows).

We extract the Zeeman splitting ΔE_Z by converting the V_{SD} scale into energy. The lever arm $\alpha \equiv C_{\text{tot-S}}/C_{\text{tot}}$ with $C_{\text{tot-S}} = C_{\text{tot}} - C_{\text{S}}$, where C_{tot} is the total capacitance of the dot, and C_{S} is the source capacit-

Figure 6.2: (a) Bias spectroscopy of the charge transition used for magnetospectroscopy measurements. The dI/dV color scale applies to all bias spectroscopies in this figure. (b) dI/dV vs. V_{SD} and B along the green line in (a). (c) ΔE_Z extracted from b) vs. B together with a linear fit (red line) that yields $g^* = 2.7 \pm 0.1$. (d) Bias spectroscopies measured at $B = 0$ (upper panel), and $B = 1$ T (lower panel). The green arrows indicate the spin-degenerate and spin-split orbital ground state. Measurements in Figure 6.3 were taken along the blue line.

ance) for this conversion is graphically extracted: the slopes of the Coulomb diamond edges from Figure 6.2a are $a \equiv |-C_G/(C - C_S)| = 2.94$ and $b \equiv C_G/C_S = 0.44$, where C_G is the gate capacitance (Hanson *et al.* 2007). By using $\alpha = 1/(1 + b/a)$ we find a lever arm of $\alpha = 0.87$. The linear increase of ΔE_Z with increasing B is shown in Figure 6.2c. We fit the slope of ΔE_Z according to $\Delta E_Z = g^* \mu_B B$, where g^* is the effective g -factor, and μ_B is the Bohr magneton [see Figure 6.2c]. This yields an effective g -factor for this transition of $g^* = 2.7 \pm 0.1$. We point out that the spin states are mixtures of heavy and light hole states and therefore $m_s \neq 1/2$, which is accounted for by the introduction of g^* as an effective g -factor. Note that g^* may differ significantly from transition to transition due to the varying heavy-hole light-hole mixing of subbands and quantum dot states (Kloeffel *et al.* 2011) at the valence band edge of the nanowire (Kloeffel *et al.* 2013a; Roddaro *et al.* 2008).

In summary, in Figure 6.2 we determine the effective g -factor g^* to be $g^* = 2.7 \pm 0.1$ for an in-plane magnetic field perpendicular to the nanowire. The corresponding Zeeman splitting is symmetric and linear up to at least 2 T.

6.4 G-FACTOR ANISOTROPY

To investigate the anisotropy of the g -factor, we measure the Zeeman splitting of the ground state at a fixed magnetic field magnitude of $B \equiv |\vec{B}| = 1$ T while changing the direction of $\vec{B}$. We choose the coordinate system in accordance with Maier *et al.* (2013), i.e. the z -axis points along the nanowire axis, the x -axis points out of the chip plane parallel to the electric field produced by the bottom gates, and the y -axis is in plane with the chip and perpendicular to the nanowire [see Fig 3(a)]. We will show measurements in three orthogonal rotation planes. Within each plane, a full 2π -rotation of $\vec{B}$ is performed in steps of $\pi/36$. For each step, I is measured vs. V_{SD} along the blue line in Figure 6.2d. The values for ΔE_Z along the different directions are obtained by fitting the line cuts with two peaks for the spin-ground and spin-excited states and calculating the distance between the two peak centers.

First we rotate the magnetic field from the y -axis to the z -axis [Figure 6.3a]. At $\phi_1 = 0$, the Zeeman splitting of $\Delta E_{Z,y} = 155 \pm 5 \mu\text{eV}$ corresponds to a g -factor of $g_y^* = 2.7 \pm 0.1$ [in agreement with Figure 6.2c]. The Zeeman splitting decreases when the magnetic field is rotated towards the nanowire axis, until it is almost completely quenched at $\phi_1 = \pi/2$ with $\Delta E_{Z,z} = (13 \pm 10) \mu\text{eV}$, corresponding to $g_z^* = 0.2 \pm 0.2$.

Figure 6.3: (a) dI/dV plotted for different $\vec{B}$ -field directions at constant $B = 1$ T measured along the blue line in (d). Rotation of the magnetic field along (a) ϕ_1 in plane of the chip (b) ϕ_2 in the plane perpendicular to the nanowire axis. (c) ϕ_3 from the electric-field axis to the nanowire axis. (d) line cuts taken from (a) at $\phi = -\pi$ (black circles) and $\phi = -\pi/2$ (green circles) plotted together with the fit (green line as the sum of the dark green lines) for the $\phi = -\pi/2$ line cut. (e) line cuts taken from (b) at $\phi = \pi$ (black circles) and $\phi = -\pi/2$ (blue circles) along with the respective fits (black and blue lines) (f) summary of the measured g -factors along the high-symmetry axes.

For the magnetic field along the z -axis $\phi_1 = -\pi/2$ the peak is approximately twice as high and also significantly broadened compared to the $\phi_1 = -\pi$ peak. This indicates that here the Zeeman splitting is too small for the two peaks of the spin-excited and spin-ground state to be resolved. However, the broadened peak can be fitted very well with two peaks that have approximately the height and width of the peak for the spin-ground state measured along the y -axis. This provides further confirmation that the broadened peak is indeed a superposition of two separate peaks.

For the second measurement $\vec{B}$ always points in a direction perpendicular to the nanowire and is rotated from the y axis at $\phi_2 = 0$ (the same field direction as for $\phi_1 = 0$ in Fig 3(a) to the x axis at $\phi_2 = \pi/2$. Along the y axis, the Zeeman splitting is again $\Delta E_{Z,y} = 154 \pm 5 \mu\text{eV}$. The Zeeman splitting decreases until it reaches $\Delta E_{Z,x} = 120 \pm 10 \mu\text{eV}$ along the x -axis, which corresponds to a g -factor of $g_x^* = 2.1 \pm 0.2$. In Figure 6.3e two line cuts from Figure 6.3b along the y -axis (red curve) and the x -axis (blue curve) are presented along with the fitted curves that were used to calculate ΔE_Z .

The third rotation plane is the x - z plane, with $\phi_3 = 0$ pointing along the x -axis, and $\phi_3 = \pi/2$ along the z -axis [see Figure 6.3c]. At $\phi_3 = 0$ we measure a spin splitting of $\Delta E_{Z,x} = 117 \pm 10 \mu\text{eV}$, corresponding to a g -factor of $g_x^* = 2.0 \pm 0.2$. Rotation of $\vec{B}$ towards the z axis again results in a Zeeman splitting of $\Delta E_{Z,z} = 17 \pm 10 \mu\text{eV}$, corresponding to $g_z^* = 0.3 \pm 0.2$.

Combining the three rotation experiments, the Zeeman splitting along each of the x , y , and z axes is measured twice with consistent values across experiments for the effective g -factor. Thus we can summarize our findings in Figure 6.3f.

Let us now compare our findings with experimental and theoretical results from the literature. An anisotropy of the effective g -factor has been measured in other systems like Si nanowire MOSFETs ($g_{\text{max}}^*/g_{\text{min}}^* \approx 1.7$) (Voisin *et al.* 2016), InAs nanowires ($g_{\text{max}}^*/g_{\text{min}}^* \approx 1.3$) (Schroer *et al.* 2011), and InSb nanowires ($g_{\text{max}}^*/g_{\text{min}}^* \approx 1.5$) (Nadj-Perge *et al.* 2012), all an order of magnitude smaller than our findings of $g_{\text{max}}^*/g_{\text{min}}^* \approx 13$ for rotations with respect to the nanowire axis. Also self-assembled SiGe islands on Si have been used for studies on the anisotropy of the effective g -factor ($g_{\text{max}}^*/g_{\text{min}}^* \approx 5$) (Katsaros *et al.* 2010), a system that is similar to ours, but lacking the one-dimensional confinement as well

as the pronounced strain of our nanowires. None of the reported anisotropies has been attributed to tunable electric fields, where we find $g_{\max}^*/g_{\min}^* \approx 1.4$ for rotation with respect to the E -field axis. For Ge/Si core-shell nanowires, Hu *et al.* (2012) reported an effective g -factor of $g^* \approx 1.02$ measured with the B -field aligned along the nanowire axis $0 \pm 30^\circ$. Roddaro *et al.* (2008) have measured g^* for different transitions ranging from 1.6 to 2.2, $\vec{B}$ was here aligned perpendicular to the nanowire. Both values are consistent with our measurements.

A g -factor anisotropy can in principle be related to the crystal direction (Nowak *et al.* 2011). In our probably $[110]$ -oriented device, we have observed a different anisotropy for holes states most probably originating from higher subbands, while we have observed qualitatively the same anisotropy in a second device tuned to the few-hole regime. Therefore we have strong evidence that the g -factor anisotropy observed here is rather related to an electric-field induced mixing between the lowest-lying subbands as discussed in the following paragraph.

Maier *et al.* (2013) theoretically investigated the g -factor in Ge-Si core-shell nanowires. They assumed elongated quantum dots, i.e. $r_{\text{core}} \ll l_{\text{dot}}$, which is very well fulfilled in our device, where $r_{\text{core}} \approx 8$ nm and $l_{\text{dot}} \approx 150$ nm. They predicted the g -factor to be highly anisotropic, with g indeed being quenched along the nanowire axis, and a maximum g -factor perpendicular to the nanowire. This is in excellent agreement with our measurements. Moreover, they predicted a lower g -factor at finite electric fields. In particular, their calculations showed a more effective diminishment for $\vec{B} \parallel \vec{E}$ than for $\vec{B} \perp \vec{E}$. Also this agrees well with our findings. Maier *et al.* (2013) show that this tunability of the g -factor with electric fields is caused by the effective coupling of different subbands through these electric fields and the mixed heavy-hole light-hole nature of the individual subbands. The combination leads to a very pronounced spin-orbit interaction (SOI) introduced as the DRSOI (Kloeffel *et al.* 2011) because of its resemblance of the standard Rashba SOI and the fact that it is a leading-order process not suppressed by the band gap and thus expected to be 10-100 times stronger than the standard Rashba SOI for geometries similar to our device.

Also quantitatively our measurements agree very well with the predictions regarding the g -factor quenching along the nanowire axis. The g -factor suppression by the electric field is less pronounced than the calculations. This can be explained by differences in the exact geometry

of the wires, the quantum dot not being in the single-hole regime, and the fact that our device is operated at significantly higher electric fields than assumed by Maier *et al.* (2013).

Let us now briefly discuss the implications of these results for quantum computation applications. A main obstacle for spin-based qubits is the fast coherent manipulation of the spin state. In principle this can be done with a pulsed high-frequency (HF) magnetic field, but this is technologically very challenging. Our results imply that it is not only possible to use pulsed HF electric fields as also used in other systems with significant SOI (Nadj-Perge *et al.* 2010a; Nowack *et al.* 2007), but with continuous HF electric fields while tuning the qubit in and out of resonance by changing the static electric field, e. g., through a combination of top and bottom gates.

6.5 CONCLUSION

In conclusion, we have demonstrated control over the hole occupancy in a Ge-Si core-shell nanowire quantum dot over 60 charge transitions. The effective g -factor has been found to be highly anisotropic with respect not only to the nanowire axis but also the electric-field direction. In particular we have found excellent qualitative agreement between our measurements and theoretical calculations (Maier *et al.* 2013). This opens the way to controlled manipulation of the spin-state with a continuous high-frequency electric field, a major technological advancement.

ANISOTROPIC PAULI SPIN BLOCKADE IN HOLE QUANTUM DOTS

We present measurements on gate-defined double quantum dots in Ge-Si core-shell nanowires, which we tune to a regime with visible shell filling in both dots. We observe a Pauli spin blockade and can assign the measured leakage current at low magnetic fields to spin-flip cotunneling, for which we measure a strong anisotropy related to an anisotropic g factor. At higher magnetic fields we see signatures for leakage current caused by spin-orbit coupling between $(1,1)$ -singlet and $(2,0)$ -triplet states. Taking into account these anisotropic spin-flip mechanisms, we can choose the magnetic field direction with the longest spin lifetime for improved spin-orbit qubits.[†]

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For quantum computation (Aaronson 2013; DiVincenzo 1995; Ladd *et al.* 2010), increasing research efforts have focused in recent years on C, Si, and Ge (Amato *et al.* 2014; Laird *et al.* 2015; Zwanenburg *et al.* 2013) because these materials can be purified to only consist of isotopes with zero nuclear spin (Itoh *et al.* 2003; Itoh *et al.* 1993) and thus exhibit exceptionally long spin lifetimes (Muhonen *et al.* 2014; Veldhorst *et al.* 2014). The one-dimensional character of Ge-Si core-shell nanowires leads to unique electronic properties in the valence band, where heavy and light hole states are mixed (Csontos *et al.* 2009; Csontos *et al.* 2007; Kloeffer *et al.* 2011). The band mixing gives rise to an enhanced Rashba-type SOI (Kloeffer *et al.* 2011), leading to strongly anisotropic and electric-field dependent g factors (Brauns *et al.* 2016b; Maier *et al.* 2013). This makes quantum dots in Ge-Si core-shell nanowires promising candidates for robust spin-orbit qubits.

Crucial steps towards high-fidelity qubits are spin readout via PSB (Ono *et al.* 2002) and understanding the spin relaxation mechanisms. For Ge-Si core-shell nanowires, there have been reports on charge sensing (Hu *et al.* 2007) spin coherence (Higginbotham *et al.* 2014c), and spin relaxation (Hu *et al.* 2012). The authors of the latter performed their experiments along a single magnetic field axis and concluded additional work was needed to pinpoint the spin relaxation mechanism.

In this chapter, we define a hole double quantum dot in a Ge-Si core-shell nanowire by means of gates. We observe shell filling and a Pauli spin blockade. The measured leakage currents strongly depend on both the magnitude and direction of the magnetic field and are assigned to spin-flip cotunneling processes for low magnetic fields. We find signatures of SOI-induced leakage current at higher magnetic fields up to 1 T.

7.1 SHELL FILLING AND PAULI SPIN BLOCKADE

Our device in Figure 7.1a consists of a p^{++} -doped Si substrate covered with 200 nm SiO_2 , on which six bottom gates with 100 nm pitch are patterned with EBL. The gates are buried by 10 nm Al_2O_3 grown with ALD at 100°C. A single nanowire with a Si shell thickness of ~ 2.5 nm and a defect-free Ge core with a radius of ~ 8 nm (Conesa-Boj *et al.* 2017), is deterministically placed on top of the gate structure with a micromanipulator and then contacted with ohmic contacts made of 0.5/50 nm Ti/Pd. Based on TEM studies of similar nanowires the wire axis is most likely pointed along the $[110]$ crystal axis. A source-drain voltage V_{SD}

Figure 7.1: (a) False-color AFM image of the device (left) and schematic depiction of the gate configuration (right). (b) Current I vs. V_{g3} and V_{g5} with $V_{g2} = 2585$ mV, $V_{g4} = 2210$ mV, $V_{g6} = 3195$ mV, and $V_{SD} = 2$ mV. White dashed lines are guides to the eyes for the honeycomb edges. Blue arrows represent E_C , and the gaps between adjacent arrows indicate an additional E_{orb} . Circles mark triangle pairs exhibiting PSB. (m, n) denotes the effective hole occupation m and n on the left and right dot, respectively.

is applied between the source and ground, and the current I is measured at the drain contact. All measurements are performed using DC electronic equipment in a dilution refrigerator with a base temperature of 8 mK.

In Figure 7.1b we plot I versus the voltage V_{g3} on gate $g3$ and the voltage V_{g5} on $g5$. We see a highly regular pattern of bias triangles (van der Wiel *et al.* 2002), indicating the formation of a double quantum dot above gates $g3$ (“left dot”) and $g5$ (“right dot”). The 16 bias triangle pairs all have very sharp edges, and the absence of current along the honeycomb edges indicates a double quantum dot weakly coupled to the reservoirs (van der Wiel *et al.* 2002). We introduce (m, n) as the effective charge occupation numbers m and n of the left and right dot, respectively.

Nine honeycomb cells are visible in Figure 7.1b. In each column from right to left a hole is added to the left dot, while in each row from top to bottom a hole is added to the right dot. The addition energy E_{add} for each added hole can be extracted from Figure 7.1b by measuring the distance between the triple points that are connected by the dashed lines and converting this distance graphically from the gate voltage into energy using the bias triangle size as a scale. The addition energy of the left dot is $E_{\text{add}} = 9.8 \pm 0.1$ meV for the middle and left column, and $E_{\text{add}} = 10.3 \pm 0.1$ meV for the right column. For the right dot $E_{\text{add}} = 9.5 \pm 0.1$ meV in the top and bottom row, but $E_{\text{add}} = 10.2 \pm 0.1$ meV in the middle row. The increased E_{add} in the right column and middle row can be readily explained by the filling of a new orbital in the corresponding dot, so that in addition to the (classical) charging energy E_C the (quantum-mechanical) orbital energy E_{orb} has to be taken into account, with $E_{\text{orb}} = 0.5 \pm 0.1$ mV in the left and $E_{\text{orb}} = 0.7 \pm 0.1$ mV in the right dot. This filling of a new orbital for both dots allows us to identify the charge occupation (m, n) with the occupation numbers of the newly filled orbitals in the left and right dot for $m, n \geq 0$.

For spin- $\frac{1}{2}$ particles filling the orbitals, the bias triangle pairs marked by red circles in Figure 7.1b should exhibit PSB in opposite V_{SD} directions. Figure 7.2a and Figure 7.2b display zooms of these triangle pairs for positive and negative V_{SD} . The lower panels of Figure 7.2a and Figure 7.2b display line cuts along the dotted lines for the V_{SD} direction with (blue) and without (green) PSB. A suppression of the current at a detuning ε lower than the singlet-triplet splitting $\Delta_{\text{S-T}}$ can be observed at negative V_{SD} in Figure 7.2a, where

the zero-detuning current at positive V_{SD} of $I_{\text{pos}}(\varepsilon = 0) = 3.6$ pA is reduced to $I_{\text{neg}}(\varepsilon = 0) = -1.6$ pA at negative V_{SD} . Current suppression at positive V_{SD} is visible in Figure 7.2b, where $I_{\text{pos}}(\varepsilon = 0) = 1.1$ pA and $I_{\text{neg}}(\varepsilon = 0) = -4.2$ pA, exactly as expected. The current is thus only partially suppressed; comparable values for the resonant ($\varepsilon = 0$) leakage current in double quantum dots have been reported in the literature (Churchill *et al.* 2009; Nadj-Perge *et al.* 2010b; Pfund *et al.* 2007b). From the line traces taken in the spin-blocked bias direction, we extract a singlet-triplet splitting $\Delta_{S,T} = 0.5 \pm 0.1$ meV. Note that this value is very close to E_{orb} (see above). This is reasonable since the (2,0) and (0,2) singlet-triplet splittings involve states originating from successive quantum dot orbitals.

We retune the device by lowering V_{g2} and V_{g6} in small, controlled steps and following the bias triangle pair at the (1,1)–(2,0) degeneracy in V_{g3} – V_{g5} gate space [Figure 7.2c]. From the line cuts (lower panel) we extract $I_{\text{pos}}(\varepsilon = 0) = 10.4$ pA and $I_{\text{neg}}(\varepsilon = 0) = 1.2$ pA, i. e., the non-blocked current is almost threefold higher after retuning, whereas the leakage current even reduced so that current rectification is now more pronounced.

In conclusion, Figure 7.1 and Figure 7.2 display the formation of a gate-defined double quantum dot, in which we show orbital shell filling for both dots and extract orbital energies of 500–700 μeV . We find PSB for two bias triangle pairs in opposite bias directions.

7.2 ANISOTROPIC LEAKAGE CURRENT

Our quantum dots are elongated along the nanowire axis $\vec{a}_{\text{NW}}$ and are exposed to a static electric field $\vec{E}$ from the bottom gates pointing out of the chip plane. We explore the origin of the leakage current I_{leak} in PSB by performing magnetospectroscopy measurements along the white dashed line in Figure 7.2c and plot I_{leak} versus the detuning and the magnetic field in Figure 7.3a. In order to investigate possible anisotropic effects, we conduct measurements along three orthogonal directions of $\vec{B}$ [see also Figure 7.3d]: (B_z) $\vec{B}$ parallel to the nanowire, (B_y) $\vec{B}$ perpendicular to both the nanowire and the electric field, and (B_x) $\vec{B}$ perpendicular to the nanowire and parallel to the electric field.

In Figure 7.3a we plot I_{leak} vs ε_0 and $B \equiv |\vec{B}|$. Here, $\varepsilon_0 \equiv \varepsilon(B = 0)$ is introduced as an absolute energy scale, since ε is only defined relative to the alignment of the (2,0) and (1,1) ground states. For all three

Figure 7.2: Zoom of the bias triangle pairs exhibiting PSB: (a) and (b) with the same barrier gate voltages as in Figure 7.1a, and (c) at $V_{g2} = 2570$ mV, $V_{g4} = 2210$ mV, and $V_{g6} = 3160$ mV. Line cuts (low-est panels) taken along the dotted lines for the V_{SD} direction with (blue) and without PSB (green).

$I_{\text{leak}}(\varepsilon_0, B)$ plots in Figure 7.3a, we scan the $I_{\text{leak}}(\varepsilon_0)$ line traces and set the center of the lowest-energy peak as $\varepsilon = 0$. In Figure 7.3b and Figure 7.3c we plot line traces for the leakage current at constant detuning as indicated in the panels. The green dashed lines in Figure 7.3a are guides to the eye for $\varepsilon(B) = 0$ ('S-onset'). The lifting of the blockade at $\varepsilon = \Delta_{\text{S-T}}$ is indicated by the blue dashed lines ('T₀-onset'). The shift of the S-onset and the T₀-onset to positive ε_0 -values for increasing B in the plots has also been observed in other experiments (Fujisawa *et al.* 2003; Johnson *et al.* 2005a; Kyriakidis *et al.* 2002) and is explained by orbital effects (Kyriakidis *et al.* 2002; Wagner *et al.* 1992; van der Wiel *et al.* 1998). A more detailed discussion can be found Section 7 B.

7.2.1 Magnetospectroscopy along B_z

We start our discussion with $\vec{B}$ parallel to the nanowire axis. The zero-detuning line cut [left panel in Figure 7.3b] has a maximum at $B = 0$ and decreases for increasing magnetic field. Similar $I_{\text{leak}}(B)$ curves with peak widths of several hundred millitesla have been reported in other systems (Lai *et al.* 2011; Li 2015; Yamahata *et al.* 2012) and were explained by spin-flip cotunneling (Coish *et al.* 2011). We can fit the peaks to

$$I_{\text{co}}(B) = I_{\text{res}} + \frac{4ecg^*\mu_B B}{3 \sinh \frac{g^*\mu_B B}{k_B T}}, \quad (7.1)$$

with $c = \frac{h}{\pi} [\{\Gamma_{\text{r}}/(\Delta - \varepsilon)\}^2 + \{\Gamma_{\text{l}}/(\Delta + \varepsilon - 2U_{\text{M}} - 2eV_{\text{SD}})\}^2]$, where I_{res} is the residual leakage current, e the electron charge, g^* the effective g factor, h Planck's constant, $\Gamma_{\text{l(r)}}$ the tunnel coupling with the left (right) reservoir, Δ the depth of the two-hole level, and U_{M} the mutual charging energy (Coish *et al.* 2011). The fit in the left panel of Figure 7.3b gives $g_z^* = 0.4 \pm 0.1$ with a hole temperature $T = 40$ mK. The finite residual current of $I_{\text{res}} = 0.8$ pA at high magnetic fields indicates a second leakage process efficient at finite magnetic field, e. g. spin-orbit interaction (Danon *et al.* 2009). The obtained g factors for a magnetic field applied parallel to the nanowire axis are consistent with our findings for a single quantum dot (Brauns *et al.* 2016b), where $g_z^* = 0.2 \pm 0.2$.

To summarize, for $\vec{B}$ pointing along the nanowire axis we can explain the peak of I_{leak} at $B = 0$ with spin-flip cotunneling and obtain $g_z^* = 0.4(1)$, and we find hints for additional leakage processes above ± 0.8 T.

Figure 7.3: (a) Magnetospectroscopy measurements for different magnetic field directions. V_{g3} is swept from 486.0 to 489.0 mV while keeping $V_{g5} = 360.0$ mV fixed and sweeping B from 1 to -1 T. (b) and (c) line cuts from (a) at fixed ϵ (open circles) alongside fitted curves (solid lines). The inset shows a high-resolution scan of the central $I(B_x)$ -peak, B_x is here plotted in mT. (d) Schematic depiction of the magnetospectroscopy directions.

7.2.2 *Magnetospectroscopy along B_y*

We now let the magnetic field point perpendicular to both the nanowire and the electric field. The zero-detuning line cut suggests the superposition of at least two peaks: a narrow peak dominates up to $B \approx 0.2$ T, and is superimposed on a broader peak that is visible over the whole range from -1 to 1 T and cannot be explained without further investigation. At finite detuning [left panel in Figure 7.3c] this broader peak is not observed, which permits us to perform a more precise fit of the central peak. By fitting to Equation (7.1)) we obtain $g_y^* = 1.2 \pm 0.2$ at $\varepsilon = 150 \mu\text{eV}$. The g factor along this magnetic field direction is significantly lower than in single quantum dot measurements (Brauns *et al.* 2016b), where $g_y^* = 2.7 \pm 0.1$ was maximal.

Because $g_y > g_z$, the spin-flip cotunneling induced leakage current is exponentially suppressed for magnetic fields above $B \approx 0.25$ T and we obtain I_{res} at $\varepsilon = 150 \mu\text{eV}$ of ~ -0.1 pA. At zero detuning, the minimum observed current is ~ -0.2 pA, which might be overestimated because of the additional features at high magnetic fields.

In summary, for $B \perp E$, $\perp \text{NW}$ we observe a peak in the leakage current at $B = 0$, which we explain with spin-flip cotunneling and find $g_z^* \approx 1.2$. The remaining leakage current is significantly lower than for $\vec{B} \parallel \vec{a}_{\text{NW}}$.

7.2.3 *Magnetospectroscopy along B_x*

The third high-symmetry direction is B_x , parallel to E and perpendicular to the nanowire. Similar to the other two magnetic field directions we find a peak around $B = 0$ in the $I_{\text{leak}}(B)$ -curve at $\varepsilon = 0$ [right panel of Figure 7.3b]. We fit Equation (7.1)) to a high-resolution magnetic field sweep at a ten times lower sweeping rate (5 mT/min instead of 50 mT/min) [inset of right panel in Figure 7.3b], which results in $g_x^* = 3.9 \pm 0.1$. g_x^* here is substantially higher than the g_y^* values obtained for $B \perp E$, as opposed to our findings in single quantum dots (Brauns *et al.* 2016b), where $g_y^* > g_x^*$. One possible explanation for this discrepancy is that it is very likely that we do not operate in the lowest-energy subband of the nanowire. Here, we estimate approximately 70 holes to reside in each quantum dot. The theoretical calculations for the g factor anisotropy (Maier *et al.* 2013) that we have confirmed experimentally take into account only the quasi-degenerate two lowest-lying subbands, and other theoretical work suggests that the heavy-hole light-hole mix-

ing is very different for different subbands (Csonotos *et al.* 2009; Csonotos *et al.* 2008). The measured angle dependence of g^* confirms that the measured g^* -factor anisotropy is not related to the crystal orientation but to the one-dimensional confinement in the nanowire and a finite electric field perpendicular to the nanowire axis.

The leakage current at magnetic fields $B > 0.1$ T, where spin-flip cotunneling is efficiently suppressed, exhibits a qualitatively different behavior than for B_y : Up to $B \approx 0.3$ T the leakage current is minimal and increases again for $B > 0.3$ T. Up to $B = 1$ T the leakage current does not fully saturate, although the slope reduces at both $\varepsilon = 0$ and $\varepsilon = 150 \mu\text{eV}$ when B approaches 1 T, which hints at a saturation for $B > 1$ T. Such an increasing $I_{\text{leak}}(B)$ with saturation at higher B is again an indication for spin-orbit induced leakage. Taking the line trace at $\varepsilon = 0$ in the right panel of Figure 7.3b, we find a minimal leakage current of $I_{\text{min}} = 0.1 \pm 0.1$ pA. The leakage current due to spin-orbit coupling between (2,0) triplet and (1,1) singlet states can be expressed as

$$I_{\text{SOI}} = I_{\text{max}} \left(1 - \frac{8B_C^2}{9(B^2 + B_C^2)} \right), \quad (7.2)$$

where I_{max} is the maximum leakage current at high magnetic fields, and B_C the width of the characteristic dip around $B = 0$ (Danon *et al.* 2009). If we now assume the minimum leakage current I_{min} to be exclusively due to a spin-orbit interaction we expect $I_{\text{max}} = 9I_{\text{min}} = 1 \pm 1$ pA at high fields. This estimate is in reasonably good agreement with the value for I_{max} we obtain by fitting the data excluding the central peak to Equation (7.2) [see red solid line in the right panel of Figure 7.3b], where we find $I_{\text{max}} = -1.6 \pm 0.3$ pA and $B_C = 1.0 \pm 0.2$ T. Since at zero detuning $I_{\text{max}} = 4e\Gamma_{\text{rel}}$, where Γ_{rel} is the spin relaxation rate (Danon *et al.* 2009), we can calculate $\Gamma_{\text{rel}} = 2.5 \pm 0.5$ MHz. This is comparable with reports on measurements of heavy holes in intrinsic Si (Li 2015), where $\Gamma_{\text{rel}} = 3$ MHz, and electrons in InAs (Nadj-Perge *et al.* 2010b), where Γ_{rel} ranges from 0 to 5.7 MHz. We note that B_C is of the order of ~ 1 T. (Danon *et al.* 2009) neglect the Coulomb interaction effects of B , which are of relevance at such field strengths (see also the Supplemental Material S1). Therefore also other spin-orbit or Coulomb related effects can provide significant leakage paths (Bădescu *et al.* 2005; Flindt *et al.* 2006; Golovach *et al.* 2004; Khaetskii *et al.* 2001; Trif *et al.* 2007).

To sum up, the spin-flip cotunneling peak in $I_{\text{leak}}(B, \varepsilon = 0)$ can be efficiently quenched by a magnetic field due to a very high effective

g factor of $g_x^* \approx 3.9$. g_x^* is significantly higher than g_y^* , in contrast to our findings in single quantum dots, where we see the opposite behavior. At higher B we notice an increasing I_{neg} , which we assign to spin-orbit coupling induced mixing of the spin states.

Let us now briefly compare our findings with existing literature and discuss the implications for spin-orbit qubits. (Pribrig *et al.* 2013) measured the leakage current at three different angles in plane of the sample and found an almost isotropic dependence of the SOI-induced leakage current. To our knowledge there are no reports of an angle dependence in the plane perpendicular to the nanowire. Spin-flip cotunneling limited leakage current as found in our device is exponentially suppressed by a Zeeman splitting of the spin states at finite $\vec{B}$, i.e. the remaining leakage current at a given $\vec{B}$ depends on the effective g factor. Since we find g^* to be highly anisotropic with respect to both $\vec{E}$ and $\vec{a}_{\text{NW}}$, the leakage current can be minimized by pointing $\vec{B}$ along $\vec{E}$. Also the leakage current caused by SOI is anisotropic and depends on the wave function overlap between the two dots (Danon *et al.* 2009). Therefore it is possible to tune the double quantum dot so that the SOI leakage current dip around $B = 0$ is wider than the leakage current peak around $B = 0$ caused by spin-flip cotunneling, which is the case here for $\vec{B} \parallel \vec{E}$. Previously measured spin relaxation times of several hundred μs obtained with $\vec{B}$ along the nanowire (Hu *et al.* 2012) might be extended by an order of magnitude when measured parallel to $\vec{E}$ according to the data presented here.

7.3 CONCLUSION

In conclusion, we have successfully formed a gate-defined double quantum dot in a Ge-Si core-shell nanowire. We have observed shell filling and a Pauli spin blockade, and have been able to explain the observed leakage current by a combination of spin-flip cotunneling at low magnetic fields and SOI-induced coupling between singlet and triplet states at higher fields.

With these results we show that by wisely choosing the magnitude and direction of a magnetic field applied to a Pauli spin-blocked double quantum dot, one can achieve longer spin lifetimes.

APPENDIX

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7 A LEAKAGE PROCESSES IN PAULI SPIN BLOCKADE

The following section was published in Brauns (2016)

The leakage current I_{leak} can be altered by a magnetic field B because when changing B we directly influence the very property that causes PSB in the first place: the spin state. If our system is in spin blockade, it is trapped in one of the (1,1)-triplet states $T(1,1)$ ¹. For a current to be measured, there must exist an efficient process that leads to the system reaching the (2,0)-singlet state² $S(2,0)$, from which the hole can then readily tunnel out to the left lead. In first order, transitions from the trapped $T(1,1)$ to $S(2,0)$ are forbidden, only $S(1,1)$ has a finite overlap with $S(2,0)$. Transitions where the trapped $T(1,1)$ -state first relaxes to the $S(1,1)$ -state are thus one important cause for I_{leak} , the other one being processes that directly mix $T(1,1)$ and $S(2,0)$ (Danon *et al.* 2009; Nadj-Perge *et al.* 2010b). In the following we will discuss three major processes:

1. Hyperfine interaction
2. Spin-flip cotunnelling
3. Spin-orbit interaction

7 A.1 Hyperfine interaction

Such spin flips can be induced by hyperfine coupling to nuclei with non-zero net spin (Jouravlev *et al.* 2006) which generate random magnetic fields that superimpose in semiclassical approximation to an effective magnetic field B_N at the site of the quantum dot. Hyperfine-induced leakage current can be suppressed by applying an external magnetic field that exceeds B_N and has been observed in numerous

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- ¹ The triplet state T is threefold degenerate, the three states being T_+ , T_0 , and T_- . In terms of the single-particle spins, they can be defined as $T_+ = |\uparrow\uparrow\rangle$, $T_0 = 1/\sqrt{2}|\uparrow\downarrow + \downarrow\uparrow\rangle$, and $T_- = |\downarrow\downarrow\rangle$.
- ² This assumes a hole temperature low enough to exclude thermal excitation to the (2,0)-triplet state. With $\Delta_{S-T} \approx 0.5$ meV and a hole temperature of $T \approx 30$ mK, i. e., a thermal energy $E_{\text{th}} \approx 3$ $\mu\text{eV} \ll \Delta_{S-T}$, we consider this a fair assumption.

systems based on III-V semiconductors like GaAs (Johnson *et al.* 2005b; Koppens *et al.* 2005), InAs (Nadj-Perge *et al.* 2010b; Pfund *et al.* 2007a), and InSb (Nadj-Perge *et al.* 2012). There are two reasons why we consider the effect from nuclei negligible in our case: First and foremost, both Si and Ge are made up mostly of isotopes with no nuclear spin. The only Si isotope with a net spin is ^{29}Si (4.7 at % in natural Si) (Itoh *et al.* 2003). For Ge, the only isotope carrying nuclear spin is ^{73}Ge with an abundance of 7.8 at % in natural Ge (Itoh *et al.* 1993). Furthermore our quantum dot states originate from valence band states with a p-type character of the wavefunction, which suppresses the (in s-type bands) dominating contribution of the contact hyperfine interaction (Fermi 1930).

7 A.2 Spin-flip cotunnelling

A second major cause for transitions between $T(1,1)$ and $S(1,1)$ is the so-called spin-flip cotunnelling (Coish *et al.* 2011; Qassemi *et al.* 2009). Here the spin of the trapped hole in the right dot is flipped by a spin-exchange process with the closest lead. The leakage current is dependent on e (the overlap of $S(1,1)$ with $S(2,0)$ limits I_{leak} for large e) as well as on B , because B splits $T_+(1,1)$ and $T_-(1,1)$ from $S(1,1)$ and $T_0(1,1)$ by the Zeeman energy E_Z . The increasing Zeeman splitting makes the process inefficient, because both the virtual intermediate state and the state of the hole tunneling into the dot (which are split by E_Z) have to originate from the temperature-broadened Fermi level in the lead. So the rate of this spin-flip process is exponentially suppressed when $E_Z > k_B T$, where k_B is the Boltzmann constant and T is the hole temperature. The leakage current due to spin-flip cotunnelling for $k_B T > t$ (Coish *et al.* 2011)

$$I_{co} = \frac{4}{3} ec \frac{g^* \mu_B B}{\sinh \frac{g^* \mu_B B}{k_B T}} \quad (7.3)$$

where

$$c = \frac{h}{\pi} \left[\left(\frac{\Gamma_R}{\Delta - \varepsilon} \right)^2 + \left(\frac{\Gamma_L}{\Delta + \varepsilon - 2U_M - 2eV_{SD}} \right) \right], \quad (7.4)$$

Δ is the depth of the two-hole levels,³ and U_M is the interdot charging energy.⁴

³ See Figure 1b) in Qassemi *et al.* (2009) for an insightful sketch.

⁴ This is the term used by Coish *et al.* (2011). Other sources refer to it as the 'mutual charging energy' (see e. g. (Laird *et al.* 2015))

7 A.3 Spin-orbit interaction

A third mechanism for I_{leak} is the spin-orbit interaction, which in combination with the finite interdot tunnel coupling mixes the $T(2,0)$ -states with the $S(1,1)$ -state (among other effects, see e.g. the introductory part of Danon *et al.* (2009) for a short overview). Here we shall summarize the key features of the process, the in-depth derivation can be found in Danon *et al.* (2009). The exact overlap of the $T(2,0)$ wavefunctions with the $S(1,1)$ wavefunctions is crucial for the efficiency of this mechanism, which is described by a non-spin conserving tunnel coupling *vector* $\vec{t}_{\text{SO}}$, in contrast to the usual spin-conserving tunnel coupling *scalar* t . The modulation of the tunnel coupling by the spin-orbit interaction leads to a mixing of all four $(1,1)$ spin states S , T_+ , T_0 , and T_- . Three out of four new eigenstates are blocked at $B = 0$ and I_{leak} is limited by the spin relaxation rate Γ_{rel} , which here is the transition rate between these four states with $I_{\text{leak}}^{\text{min}} = 4/9\Gamma_{\text{rel}}$. At high magnetic fields, only one out of four eigenstates is blocked, which leads to a nine times higher $I_{\text{leak}}^{\text{min}} = 4\Gamma_{\text{rel}}$. At zero detuning, the dip around $B = 0$ follows for $B = (0, 0, B_Z)$ (in the following $B \equiv B_Z$)

$$I_{\text{leak}}(B) = I_{\text{leak}}^{\text{min}} \left(1 - \frac{8}{9} \frac{B_C^2}{B^2 + B_C^2} \right), \quad (7.5)$$

where

$$B_C = 2\sqrt{2} \frac{(1 + |\vec{\eta}|^2)}{\sqrt{\eta_x^2 + \eta_y^2}} t \sqrt{\Gamma_{\text{rel}}/\Gamma} \quad (7.6)$$

with $\vec{\eta} \equiv \vec{t}_{\text{SO}}/t$. Here, η_x and η_y are the components of the normalized tunnel coupling vector perpendicular to the magnetic field. The relative orientation of $\vec{t}_{\text{SO}}$ and B is thus crucial for the width of the dip in I_{leak} . The direction of $\vec{t}_{\text{SO}}$ is dependent on the exact overlap of the $(2,0)$ and $(1,1)$ wavefunctions and thus very hard to predict in real systems.

7 B ANISOTROPIC COULOMB INTERACTION

We start our discussion with the magnetospectroscopy measurements for B_z and B_y (left and middle column in Figure 7.3a of the main text). In Figure 7 A.1 we plot $I(\varepsilon_0)$ line cuts at $B = 0$ T and $B = 1$ T. The T_0 -onset only changes minimally between 0 and 1 T, whereas the S -onset shifts to positive ε_0 -values by $\Delta S = 100 \pm 10 \mu\text{eV}$ for B_z and by $\Delta S = 200 \pm 10 \mu\text{eV}$ for B_y . As stated in the main text this behaviour has also been observed in other experiments (Fujisawa *et al.* 2003; Johnson

Figure 7 A.1: $I(\varepsilon_0)$ line cuts from Figure 7.3b in the main text at $B = 0$ T (black curve) and $B = 1$ T (red) for all three magnetic field directions. ΔS denotes the shift of the S-onset, ΔT_0 the shift of the T_0 -onset.

et al. 2005a; Kyriakidis *et al.* 2002). It can be explained by orbital effects of the magnetic field (Kyriakidis *et al.* 2002; Wagner *et al.* 1992; van der Wiel *et al.* 1998), where B leads to a shrinking of the wave function which increases the Coulomb energy cost of singlet states as compared to triplet states and therefore leads to a shift of the S-onset to higher energies.

A peculiarity of the measurements for B_x is the T_0 -onset, which, in contrast to the measurements along B_y and B_z , shifts to higher ε_0 for increasing B . Between $B = 0$ and $B = 1$ T this shift amounts to $\Delta T_0 = 280 \pm 10 \mu\text{eV}$, whereas the shift of the S-onset sums up to $\Delta S = 330 \pm 10 \mu\text{eV}$. For B_z and B_y , we only find $\Delta S = 100 \pm 10 \mu\text{eV}$ and $\Delta S = 200 \pm 10 \mu\text{eV}$, respectively. This enhanced ΔS along with the pronounced ΔT_0 for relatively small magnetic fields $B \leq 1$ T indicate a change in the Coulomb interaction between the two holes as compared to the measurements for B_y and B_z (Kouwenhoven *et al.* 2001a; Kyriakidis *et al.* 2002). Theoretical calculations suggest an anisotropic character of hole-hole interactions with respect to the electric-field axis in Ge-Si core-shell nanowires (Maier *et al.* 2014b), but further studies are necessary for a definite conclusion on the cause for the observed anisotropic B -dependence of the two-hole spin states.

7 C TUNNEL COUPLINGS

From the $I(\varepsilon)$ -curves in the non-blocked bias direction (lower panels of Figure 7.2a in the main text), we can extract the tunnel coupling Γ

Figure 7 A.2: Zooms of the $I(\varepsilon)$ line cuts for the unblocked bias direction in the lower panels of Figure 7.2a in the main text.

to the reservoirs and the interdot tunnel coupling t by fitting to the expression

$$I(\varepsilon) = (\Gamma|t|^2) / ((\varepsilon/h)^2 + \Gamma^2/4 + 3|t|^2), \quad (7.7)$$

where h is the Planck constant (van der Wiel *et al.* 2002). We fit the $I(\varepsilon)$ -curves to Equation (7.7) only for $\varepsilon \leq 0$, because for $\varepsilon > 0$ inelastic processes that are not taken into account lead to an increased current, as can be inferred from the asymmetry of $I(\varepsilon)$ around $\varepsilon = 0$ in Figure 7 A.2 where we plot the fits alongside the data (the same approach has been used e. g. by (Li 2015)). The extracted values for Γ and t are:

a) $\Gamma = 100 \pm 10 \mu\text{eV}, t = 1.6 \pm 0.2 \mu\text{eV}$

b) $\Gamma = 90 \pm 10 \mu\text{eV}, t = 1.6 \pm 0.2 \mu\text{eV}$

c) $\Gamma = 100 \pm 10 \mu\text{eV}, t = 2.7 \pm 0.2 \mu\text{eV}$

These values suggest that by changing the barrier voltages between the left and the right panel the interdot tunnel coupling t increases significantly from $1.6 \mu\text{eV}$ to $2.7 \mu\text{eV}$. This increase reflects an increased wavefunction overlap between states in the left and the right dot, which can be explained by the finite cross capacitance between g_2/g_6 and the quantum dots which may affect the shape of the wave functions. In all three cases t is more than an order of magnitude smaller than Γ and thus the dominant factor for the measured current.

Let us now take a more careful look at how the tunnel couplings relate to the leakage current in Pauli spin blockade. Although t , which is the tunnel coupling limiting the current through the double quantum

dot, is significantly higher in the right panel than in the left panel, I_{leak} even *decreases* (see Figure 7.2a in the main text). This means that an increased overlap between $(1,1)$ and $(2,0)$ states does not play a major role in the process that determines the leakage current.

MULTIPLE ANDREEV REFLECTIONS AND SHAPIRO STEPS IN A GE-SI NANOWIRE JOSEPHSON JUNCTION

8.1 INTRODUCTION

Josephson junctions are created by coupling two superconductors via a weak link. Josephson junctions exhibit the remarkable property that they allow a supercurrent to be transported through intrinsically non-superconducting materials by means of proximity induced superconductivity, and their discovery (Josephson 1962) therefore resulted in the Nobel prize in physics in 1973.

While early Josephson junctions realised a weak link by using thin layers of oxide, micro-constrictions, point contacts or grain boundaries (Fagaly 2006; Grimes *et al.* 1968; Grossman 1994; Likharev 1979; Shapiro 1963), access to complex mesoscopic semiconducting materials have led to Josephson junction employing 1- and 2-dimensional nanostructures where control over the charge carrier density plays a crucial role (Jarillo-Herrero *et al.* 2006; Katsaros *et al.* 2010; Krive *et al.* 2004; Mizuno *et al.* 2013; Taupin *et al.* 2016; Wiedenmann *et al.* 2016). These devices have been used in a wide range of applications (De Franceschi *et al.* 2010) such as superconducting quantum interference devices (SQUIDS) (Cleuziou *et al.* 2006; van Dam *et al.* 2006; Kim *et al.* 2016), superconducting qubits (Larsen *et al.* 2015; Mooij 2004; van der Wal *et al.* 2000), Cooper pair splitters (Hofstetter *et al.* 2009; Tan *et al.* 2015) and π -junctions (van Dam *et al.* 2006; Jørgensen *et al.* 2007).

In this work we present a Josephson junction based on semiconducting Ge-Si core-shell nanowires. This system has proven itself in the realm of normal-state quantum dots (Brauns *et al.* 2016a; Brauns *et al.* 2016b; Brauns *et al.* 2016c; Higginbotham *et al.* 2014a; Higginbotham *et al.* 2014c; Hu *et al.* 2007; Hu *et al.* 2012; Zarassi *et al.* 2017), but apart

Figure 8.1: **Nanowire Josephson FET.** (a) False color SEM image of the device under investigation. A nanowire with a 20 nm diameter (yellow) lies on the SiO_2 covered substrate (blue) and is contacted by an Al source and drain (green). (b) V_{SD} vs I_S for two $V_{BG} = -15$ V and $V_{BG} = -7.6$ V. I_S is swept from left to right (dark) and successively from right to left (light) denoted by the coloured arrows. I_C and I_R are shown for $V_{BG} = -7.6$ V. Horizontal black arrows point at ‘wiggles’ in the curve corresponding to MAR of the n^{th} order.

from a limited number of reports (Su *et al.* 2016; Xiang *et al.* 2006a), research involving superconductivity is relatively unexplored.

We will present the experimental observation of the DC Josephson effect: a finite switching current I_C^1 through the nanowire Josephson junction. We will also look at MAR (Flensberg *et al.* 1989; Kuhlmann *et al.* 1994) and analyse the position of the resulting conductance peaks inside the superconducting gap of Al Δ_{Al} . Additionally, we look at the temperature dependence of MAR and I_C . Finally, we irradiate our junction with microwaves resulting in Shapiro steps, a manifestation of the alternating current (AC) Josephson effect which has not been shown in this system before. The observation of both the DC and the AC Josephson effect confirms we have a true Josephson junction using Ge-Si nanowires, which marks the beginning for exploring more complicated systems such as SQUIDs (Cleuziou *et al.* 2006) or coupling a quantum dot to superconducting contacts (van Dam *et al.* 2006; De Franceschi *et al.* 2010).

¹ When measuring a Josephson junction in this configuration, one obtains the switching current and not its critical current which requires measuring a distribution of switching currents at several temperatures (Wallraff *et al.* 2003).

8.2 NANOWIRE JOSEPHSON JUNCTION

Figure 8.1a shows a SEM image of the device with a channel length of ~ 150 nm, designed for 4-terminal measurements. We plot the sourced current I_S versus the measured voltage between source and drain V_{SD} in Figure 8.1b. Sweeping I_S forward, i.e., from 0 to finite bias, we find that the junction switches from the superconducting state to a dissipative state at a switching current of $I_C = 44$ at $V_{BG} = -15$ and $I_C = 32$ nA at -7.6 V. When sweeping backwards, i.e., from finite I_S to 0, the junction returns to its superconducting state at the current I_R , resulting in hysteretic behaviour. For a backgate voltage $V_{BG} = -15$ V, we find $I_R = 34$ nA and a ratio $I_C/I_R = 1.3$, while for $V_{BG} = -7.6$ V, $I_R = 10$ nA and a ratio $I_C/I_R = 3.4$. This indicates that our junction is underdamped (Stewart 1968) and that I_C , as well as the damping, changes for different V_{BG} , mainly due to the changing number of subbands participating in transport and their position relative to the Fermi energy of the Al contacts (for a more detailed analysis of I_C and I_S as a function of V_{BG} see Chapter 9).

8.3 MULTIPLE ANDREEV REFLECTIONS

Small ‘wiggles’ are visible in the V_{SD} versus I_S curve for $V_{BG} = -7.6$ V in Figure 8.1b, which are a signature of MAR. This becomes more clear in the numerical differential conductance $\partial I_D / \partial V_{SD}$ for a range of V_{BG} in Figure 8.2a: the ‘wiggles’ in V_{SD} translate to conductance peaks showing at specific values of V_{SD} corresponding to $V_{SD}(n) = 2\Delta_{Al}/en$ (Tinkham 1996) with n an integer denoting the MAR order. $n = 1-5$ are indicated by the green arrows in Figure 8.2a for positive bias (orders $n = 2-5$ are indicated in Figure 8.1b). The strong conductance peak at $V_{SD} = 0$ corresponds to a supercurrent and is a direct consequence of the inversion of the I_S and V_{SD} axes, which maps I_S onto the corresponding value of V_{SD} . Since a supercurrent implies $V_{SD} = 0$ for a range of I_S , this results in a strong peak in $\partial I_D / \partial V_{SD}$. The height of the oscillating black regions for $V_{SD} < 0.05$ mV as a function of V_{BG} is a measure for the magnitude of I_R (see Section 8 c) and the oscillatory pattern indicates varying occupation of the subbands in the wire (see Chapter 9).

The MAR conductance peaks can be more clearly distinguished as individual linetraces in Figure 8.2b and we focus on the blue trace at $V_{BG} = -7.6$ V. The finite width of the MAR peaks reflects the distribution of the DOS peak at $V_{SD} = 2\Delta_{Al}$ and is additionally broadened by

Figure 8.2: Multiple Andreev reflections up to the 6th order. (a) Numerical differential conductance $\partial I_S / \partial V_{SD}$ versus V_{SD} and V_{BG} . Horizontal equipotential lines of increased conductance indicated by the green arrows correspond to MAR. Current biased measurement where the I_S and V_{SD} axes were inverted (see Section 8 B) before numerical derivation. Only return current was measured (see black arrows) in a highly hysteretic regime (see blue curve in Figure 8.1b) to reach the low-voltage regime. (b) Single traces of $\partial I_S / \partial V_{SD}$ versus V_{SD} for three values of V_{BG} . Green, orange and blue (offset by 200 μS) taken at $V_{BG} = -8.3$, -8 and -7.6 V (see (a), dotted lines). Vertical grey dotted lines denote expected MAR peak positions calculated by $n = 2\Delta_{Al}/eV_{SD}$ for $n = 1 - 10$. Inset: MAR peak positions (P.P.) vs inverse MAR order $1/n$ at $V_{BG} = -7.6$ (blue trace) for positive bias. The black line is a linear fit through zero. (c) Same data as (b) now plotted versus n , only positive V_{SD} is shown. The vertical dotted grey lines show integers of n which can be matched with the MAR peaks up to $n = 6$.

phase decoherence and inelastic processes when quasiparticles traverse the channel (Nilsson *et al.* 2012; Tinkham 1996). We extract the peak positions (P.P.) in V_{SD} of the first 6 orders and plot them versus the inverse MAR order in the inset in Figure 8.2b. We expect the second order MAR peak to be at the position of our superconducting gap, i. e., for $n = 2$, $V_{SD} = \Delta_{Al}$. For a more accurate estimate of Δ_{Al} we perform a linear fit through zero for the six MAR peak positions and find $\Delta_{Al} = 0.212$ mV which translates to a critical temperature of our Al $T_{C,Al} = \Delta_{Al}/1.764 * k_B T = 1.39$ K (Tinkham 1996). This is confirmed by an independent measurement of the T_C of a Al stripline (not shown) and to the critical temperature observed in Figure 8.4.

In Figure 8.1b higher-order MAR peaks become increasingly hard to resolve because of the hyperbolic relation of V_{SD} with n . In Figure 8.2c, we therefore convert the x-axis from V_{SD} to units of $n = 2\Delta_{Al}/eV_{SD}$, resulting in evenly spaced orders of n , and plot the conductance for positive bias for the same three V_{BG} as in Figure 8.2b. We see that for $n > 6$, the conductance peaks can no longer be unambiguously assigned and they span multiple n . Possibly, the peak patterns for high-order MAR are a superposition of many interfering MAR processes.

Comparing the blue curve in Figure 8.2c with the orange and green curve taken at different V_{BG} we observe that peak positions for lower order MAR do not perfectly reproduce, for instance, the orange curve does not show a clear peak at $n = 4$. We partly explain this by considering our interface transparencies. Although interface transparency is estimated to be $> 80\%$ (see Chapter 9), the interfaces act as a weak confinement potential resulting in highly broadened energy levels in the wire. The relative position of these levels with respect to the Fermi level changes the resonant condition for MAR, resulting in a shift of the MAR peak positions (Buitelaar *et al.* 2003). Inspecting Figure 8.2a, the high-order MAR peaks ($n > 6$) are indeed modulated, both in intensity as in position in V_{SD} , by the black regions at low bias ($V_{SD} < 0.05$ mV).

We conclude from Figure 8.2 that the resolvability of MAR up to $n = 6$, indicates that quasiparticles can elastically scatter at least 6 times on the interfaces, each time traversing the nanowire channel. This indicates that our Al-nanowire interface is homogeneous (no inelastic processes) and has a high transparency (Flensberg *et al.* 1988).

Figure 8.3: **Temperature dependence of MAR and I_C .** (a) Numerical differential resistance $\partial V_{SD}/\partial I_S$ vs I_S and T for $V_{BG} = -13.35$ V. I_S is swept from negative to positive bias. I_C and I_R are denoted by the white arrows. (b) Numerical differential conductance $\partial I_S/\partial V_{SD}$ vs V_{SD} and T . Same data as (a) with I_S and V_{SD} axes reversed before numerical derivation (see Section 8 b). Black dotted lines are fits to $2\Delta(T)/n$.

8.4 TEMPERATURE DEPENDENCE OF I_C AND MAR

We will now investigate the temperature dependence of the switching current and the multiple Andreev reflections. In Figure 8.3a we plot the differential resistance $\partial V_{SD}/\partial I_S$ versus I_S as a function of temperature T . The black region $\partial V_{SD}/\partial I_S = 0$ indicates superconductivity and we can see the decrease of I_C for increasing T until I_C disappears at $T \approx 1.4$ K, in agreement with $T_{C,AI} = 1.39$ K calculated from Δ_{AI} . Attempts to fit I_C as a function of T using models describing a Josephson junction with two interface barriers (Usadel 1970) do not allow for a correct description of our junction. One reason might be that our junction exhibits an additional superconducting phase inside, or in close proximity to, the nanowire (see Chapter 10) which influences I_C in a non-trivial way.

The MAR signatures visible outside the superconducting region, scale with Δ_{AI} and therefore gradually decrease and converge to $V_{SD} = 0$ for $T \rightarrow T_{C,AI}$. Figure 8.3b shows the same dataset as Figure 8.3a converted to a voltage-biased plot (see Section 8 b) and since for MAR order $n = 2$, $V_{SD} = \Delta_{AI}$, we have a direct measure of $\Delta_{AI}(T)$.

To compare with theory, we use the Bardeen-Cooper-Schrieffer (BCS) interpolation formula (Kajimura *et al.* 2012; Senapati *et al.* 2011):

$$\Delta(T) = \frac{2\Delta_{\text{Al},0}}{n} \tanh\left(1.74\sqrt{\frac{\Delta_{\text{Al},0}}{1.76k_B T} - 1}\right) \quad (8.1)$$

where we replaced the pre-factor $\Delta_{\text{Al},0}$ with $2\Delta_{\text{Al},0}/n$ where $\Delta_{\text{Al},0}$ is the superconducting gap of Al at $T \approx 0$ as determined in Figure 8.2b. We plot this theoretical curve in Figure 8.3b and find excellent agreement for the $n = 2$ MAR peak. Additionally, $n = 3$ results in a good fit. Together with the fact that the value of $\Delta_{\text{Al},0}$ corresponds to the observed $T_{\text{C,Al}}$, the MAR are indeed an excellent measure for the superconducting gap of the contacts.

8.5 SHAPIRO STEPS

We now look at the AC Josephson effect by irradiating our junction with an antenna located above the chip with frequencies ranging from 0.8 to 4.4 GHz. Figure 8.4a shows V_{SD} versus I_{S} for three different frequencies at finite microwave amplitudes V_{rms} , revealing a step-like current-voltage relation. These so-called Shapiro steps (Shapiro *et al.* 1964) are a direct manifestation of the AC Josephson effect where phase locking occurs between the quasiparticles in the junction and the applied microwaves. The height of the DC steps ΔV is determined by the applied frequency f as $\Delta V = hf/2e$ (Tinkham 1996). The extracted step height for various frequencies shows good agreement in Figure 8.4b. We attribute the qualitative variation in the rounding of the steps as a function of frequency to spectral broadening of the microwaves, in turn caused by the microwave antenna properties and the coupling to the Faraday cage in which the sample resides.

We now fix the applied frequency at 2.65 GHz and plot $\partial V_{\text{SD}}/\partial I_{\text{S}}$ versus I_{S} and V_{rms} at $V_{\text{BG}} = -15$ V in Figure 8.4c. Upon increasing V_{rms} from left to right, clear lines of differential resistance enter the superconducting bias window defined by steps of ΔV . V_{SD} remains constant in the regions enclosed by the steps resulting in plateaus of constant I_{S} ($\partial V_{\text{SD}}/\partial I_{\text{S}} = 0$), which increase with ΔV for every step as seen Figure 8.4a. The reason more steps enter the bias window for increasing V_{rms} , is that higher AC amplitudes result in a stepwise increase of the phase evolution of quasiparticles in the junction, which increases V_{SD} according to the AC Josephson relation (Gross *et al.* 2005; Tinkham 1996).

Figure 8.4: AC Josephson effect, up to 23 shapiro steps. (a) V_{SD} versus I_S for frequencies $f = 1.51, 2.80$ and 4.40 GHz at respective amplitudes $V_{rms} = 0.11, 0.13$ and 0.13 V. V_{rms} values are AC amplitudes applied before filtering. $f = 2.8$ GHz results in a step height of $\Delta V = 5.8 \mu V$ (black arrow). (b) Step height ΔV versus microwave frequency f extracted from data (blue boxes). The black line is a plot of $\Delta V = hf/2e$. (c) Numerical differential conductance $\partial V_{SD}/\partial I_S$ versus I_S and microwave rms voltage V_{rms} applied at a frequency of 2.65 GHz at $V_{BG} = -15$ V (red curve in Figure 8.1b). (d) Left: $\partial I_S/\partial V_{SD}$ versus V_{SD} and V_{rms} , same measurement data as (c) with I_S and V_{SD} axes reversed before numerical derivation (see Section 8 B). V_{SD} shown in units of $hf/2e$. Right: linecut at $V_{rms} = 0.66$ V showing 23 peaks.

In these experiments, V_{BG} was fixed at -15 V, corresponding to a region with a high I_C and low hysteresis. Figure 8 A.1 shows a complete backgate dependence at fixed f and V_{rms} where we see that hysteresis results in missing Shapiro steps.

As in most experimental setups, our microwave source and antenna have a much higher impedance than our superconducting Josephson junction (Gross *et al.* 2005; Tinkham 1996) and it therefore acts mainly as an AC current source instead of a voltage source. In this case, steps of ΔV occur when the phase change of the particle synchronises with the AC source. This does not change the observed values ΔV , but mainly changes the width of the current plateaus (Gross *et al.* 2005). Although the widths of the current plateaus still show a ‘Bessel-like’ character as a function of V_{rms} (see Figure 8.4c), the evolution of their width cannot be solved analytically (Russer 1972; Tinkham 1996).

To gain insight in the number of Shapiro steps and their corresponding plateau heights in Figure 8.4c, we show a V_{SD} biased plot in units of $\Delta V = hf/2e$ of the same measurement data in Figure 8.4d. The plateaus of constant I_S in Figure 8.4c are now represented as the horizontal peaks in $\partial I_D / \partial V_{SD}$ at their respective value of V_{SD} . Looking at the linecut on the right we see that up to 23 steps are visible, all aligned with integer values of $\Delta V = hf/2e$. This clear demonstration of the AC Josephson effect in Figure 8.3, together with DC effects such as MAR and finite I_C is proof that our junction is, indeed, a Josephson junction.

8.6 JUNCTION CHARACTERISTICS

We will now try to establish whether our junction is in the ballistic or the diffusive limit. In the ballistic case, particles traversing the junction do not scatter, except on the interfaces. In the diffusive case, particles encounter scattering sites inside the junction which leads, for example, to suppression of I_C (Du *et al.* 2008). For a ballistic junction and completely transparent interfaces, one expects the normal state conductance to appear in multiples of the conductance quantum $G_0 = 2e^2/h$, and the critical current in multiples of the maximum critical current for a single subband $I_{C,MAX} = e\Delta_{Al}/\hbar = 51$ nA (Tinkham 1996). In our case, the finite interface transparency leads to lower observed values of both G_0 and $I_{C,MAX}$, where $I_{C,MAX}$ is suppressed by additional mechanisms such as electromagnetic coupling with the environment (Jarillo-Herrero *et al.* 2006), premature switching and heating effects (Tinkham 1996; Xiang *et al.* 2006b). From experiments it is therefore not trivial to

conclude whether our junction is diffusive or ballistic and we therefore make a quantitative estimation based on calculations.

We start with estimating the elastic scattering length using $l_e = \mu m^* v_F / e$ (Davies 1998) with μ the hole mobility, m^* the effective hole mass and v_F the Fermi velocity. We use $\mu \approx 3500 \text{ cm}^2/\text{Vs}$ (see Chapter 4) and $m^* \approx 0.5m_e$ for the mixed heavy and light holes (Kloeffel *et al.* 2011; Maier *et al.* 2014b) with m_e the electron mass. To obtain the Fermi velocity we use the solutions of the Schrödinger equation for a cylindrical potential well and find the expression for the Fermi energy of the n^{th} subband with quantum number l as $E_{n,l} = \hbar^2 \alpha_{n-1,l}^2 / 2m^* R^2$ (Griffiths 1996) with $\alpha_{n,l}$ the l^{th} root of the n^{th} order Bessel function, R the wire radius and m^* the hole effective mass. For the first subband this gives $E_{1,1} \approx 15 \text{ meV}$ corresponding to a Fermi velocity $v_F \approx 1 \cdot 10^5 \text{ m/s}$. This results in an estimated elastic scattering length of $l_e \approx 100 \text{ nm}$, which increases for higher energy subbands.

Since our channel length $L = 150 \text{ nm}$, we are neither purely in the ballistic limit ($l_e \gg L$), nor in the diffusive limit ($l_e \ll L$). For the coherence length in the diffusive limit we obtain $\xi_{diff} = \sqrt{\hbar D / \pi \Delta_{Al}} \approx 90 \text{ nm}$ with $D = v_F l_e$. The ballistic limit does not depend on l_e and we find $\xi_{ball} = \hbar v_F / \pi \Delta_{Al} \approx 170 \text{ nm}$ (Tinkham 1996). From these estimations it appears our junction is in an intermediate quasi-ballistic regime where only a few scattering sites are present. As we will see in Chapter 9, we form a quantum dot of $\sim 150 \text{ nm}$ inside the nanowire channel, which excludes that the junction is in the diffusive limit.

8.7 CONCLUSION

We show a Josephson junction where the high contact transparency combined with an abrupt interface between the superconducting leads and the nanowire results in multiple Andreev reflections being observed up to the 6th order. We additionally show up to 23 Shapiro steps, clearly demonstrating the AC Josephson effect which has not been shown before in previous work on this system (Su *et al.* 2016; Xiang *et al.* 2006b) and is an important proof of the realisation of a Josephson junction. The temperature dependence of I_C is non-trivial, which we cannot fully explain, and from calculations we estimate the junction to be in the quasi-ballistic regime.

APPENDIX

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8 A SHAPIRO STEPS VERSUS V_{BG}

In Figure 8 A.1a we show a current-sourced backgate dependence of Shapiro steps at fixed microwave frequency and power. The junction is hysteretic for regions with lower I_C observed in this figure as a white speckle pattern (see for instance between $V_{BG} = -10$ and -8 V), which is a direct result of reversing the bias direction of I_S for every step in V_{BG} .

Figure 8 A.1b shows the voltage-biased backgate dependence of the same measurement data as Figure 8 A.1a where plateaus in current are translated to peaks in $\partial I_S / \partial V_{SD}$ (for a more detailed explanation see Figure 8.4c and Figure 8.4d). We identify 5 Shapiro steps which partially disappears when the junction is hysteretic. For this specific V_{rms} , step 0, 1 and 4 disappear in the hysteretic regions but this depends on their respective current plateau widths. Since the plateau width is a function of V_{rms} (see Figure 8.4c), the missing steps also change as a function of V_{rms} . At $V_{BG} = -15$ V, all steps are visible which is the reason this specific voltage was used in Figure 8.4.

8 B POST-PROCESSING OF MEASUREMENT DATA

All measurements in this chapter are performed using a 3-probe measurements. A series resistance of 3.46 k Ω was subtracted from all measurement data. In Figure 8.2a, Figure 8.4d and Figure 8.3b, the datasets are obtained using a current source I_S with V_{SD} readout after which a software routine is used to invert the source and measurement axis. For a 2D color plot it is required that the points in the new V_{SD} source axis are equidistantly spaced. This is realised by recalculating the points by interpolation in I_S on a grid with predetermined V_{SD} stepsize. The resolution of V_{SD} is chosen high enough so that no features in the original measurement of V_{SD} are lost. In Figure 8.4c and Figure 8.4d a similar grid interpolation procedure was used to convert the x-axis from units of dBm to V.

Figure 8 A.1: **Shapiro steps versus backgate voltage.** (a) Numerical differential resistance $\partial V_{SD}/\partial I_S$ versus I_S and V_{BG} at fixed microwave frequency $f = 2.65$ GHz and $V_{rms} = 0.1$ V. For each step in V_{BG} , $\text{step} = 25$ mV, the sweep direction in which I_S is acquired is reversed. The white dashed line denotes $V_{BG} = -15$ V used in Figure 8.4 (b) Numerical differential conductance $\partial I_S/\partial V_{SD}$ vs V_{SD} (units of $hf/2e$) vs V_{BG} . Same data set as (a) where I_S and V_{SD} axes were inverted before numerical derivation (see Section 8 B). We identify up to 5 Shapiro steps, i.e., current plateaus $s = 0 - 5$. The ‘wavy’ pattern, i.e., the gradual shift of the lines in V_{SD} as a function of V_{BG} , is caused by small variations in the leakage current at different V_{BG} .

8 C MAR DATASET ACQUISITION

In the open regime MAR peaks can only be seen when the junction is in the dissipative current state and since higher-order ($n > 6$) MAR

reside close to 0 bias, they can be obscured by the superconducting 'blind spot' of the junction. We use the bi-stable current-voltage relation (hysteresis) of our underdamped Josephson junction in Figure 8.2a, where we choose a region of V_{BG} with a low I_R . In order to measure the current-voltage relation of the junction way below I_C , the low I_R is exploited by sweeping from finite $|I_S|$ to 0 in both bias directions. We note that I_R is still finite which is reflected in the small black oscillating 'blind spot' region around $|V_{SD}| = 0$ and a sufficient range V_{SD} can now be probed. The visibility of MAR also depends on G_N : a higher G_N results in a lower voltage drop over the same I_S , thus effectively enhancing measurement resolution of the equipotential MAR peaks. This is especially important for higher order MAR (> 4) since its hyperbolic relation with V_{SD} means that the corresponding peaks become very closely spaced.

JOSEPHSON EFFECT IN A FEW-HOLE QUANTUM DOT

We use a Ge-Si core-shell nanowire to realise a Josephson field-effect transistor with highly transparent contacts to superconducting leads. By changing the electric field we gain access to two distinct regimes not combined before in a single device: In the accumulation mode the device is highly transparent and the supercurrent is carried by multiple subbands, while near depletion supercurrent is carried by single-particle levels of a strongly coupled quantum dot operating in the few-hole regime.

9.1 INTRODUCTION

Combining low-dimensional mesoscopic semiconductors with superconducting leads results in a special class of Josephson junctions where the charge carrier density and the critical current of the junction material can be changed by an applied electric field (Doh 2005; Morpurgo 1999). In the quantum regime, the energy levels of the semiconductor become discrete and superconducting transport is therefore carried by a well-defined number of modes (Beenakker 1992). An even more interesting situation arises when charge is localised on the semiconducting junction, resulting in a quantum dot coupled to superconducting leads. In these hybrid devices, the discrete levels in the dot are well resolved and their interaction with the macroscopic wavefunction of the superconductor depends on the strength of the coupling between the dot and the leads (De Franceschi *et al.* 2010). A renewed interest in these hybrid devices has been sparked by proposal from Sau *et al.* (2012) where it is predicted that a chain of several strongly coupled superconducting nanowire quantum dots results in a system which contains Majorana zero modes that are highly robust against disorder.

Interaction between quantum dots and superconducting leads has been explored in a range of experiments performed on e. g. carbon nanotubes (CNTs) (Cleuziou *et al.* 2006; Eichler *et al.* 2007; Jarillo-Herrero *et al.* 2006; Jørgensen *et al.* 2009) and graphene (Du *et al.* 2008; Heersche *et al.* 2007; Kumaravadivel *et al.* 2016; Lee *et al.* 2015; Mizuno *et al.* 2013) showing Kondo physics (Buitelaar *et al.* 2002; Eichler *et al.* 2009; Kim *et al.* 2013) as well as Andreev bound state (ABS) (Dirks *et al.* 2011; Kim *et al.* 2013; Pillet *et al.* 2010). InSb (Buizert *et al.* 2007; van Dam *et al.* 2006; Doh 2005; Kanai *et al.* 2010; Sand-Jespersen *et al.* 2007), InAs / InP nanowire devices (Doh *et al.* 2008; Günel *et al.* 2014; Lee *et al.* 2014) and to a lesser extent PbS nanowire devices (Kim *et al.* 2017; Kim *et al.* 2016) embody other successful platforms for studying quantum dots with superconducting leads. Germanium and/or silicon systems, however, are still relatively unexplored (Katsaros *et al.* 2010; Su *et al.* 2016; Xiang *et al.* 2006a).

In this work, we use Ge-Si core-shell nanowires with diameters under 25 nm, exhibiting a very low defect density which allows us to form intentional quantum dots over several hundreds of nanometers (Brauns *et al.* 2016c). These nanowires have proven to be a versatile platform for spin-based experiments in normal-state quantum dots (Brauns *et al.* 2016a; Brauns *et al.* 2016b; Brauns *et al.* 2016c; Higginbotham *et al.* 2014a; Higginbotham *et al.* 2014c; Hu *et al.* 2007; Hu

Figure 9.1: **Josephson field-effect transistor with a gate-tunable switching current.** (a) False-colour SEM image of the device. A nanowire with a 20 nm diameter (yellow) lays on top of SiO_2 (blue). Al source and drain contacts (green) define a 150 nm long nanowire channel. (b) V_{SD} versus I_{S} for five different V_{BG} . The blue arrows indicate the switching currents I_{C} while the black arrows indicate the retrapping currents I_{R} . The grey arrows indicate the sweep direction of I_{S} .

et al. 2012; Zarassi *et al.* 2017). Furthermore, they have an electric-field tunable g -factor (Brauns *et al.* 2016b; Maier *et al.* 2013) and are predicted to exhibit an exceptionally strong spin-orbit interaction (Kloeffel *et al.* 2011). When combined with superconductivity, this makes these wires highly suitable for observing Majorana zero modes (Maier *et al.* 2014a; Thakurathi *et al.* 2017). However, due to the difficulty to form transparent contacts to most superconducting materials, experiments in this field have been limited (Su *et al.* 2016; Xiang *et al.* 2006a).

We use a non-trivial but straightforward fabrication technique to obtain highly transparent superconducting contacts from Al to Ge-Si nanowires and show a Josephson FET. Next, we observe two regimes in the junction's current-voltage relation as a function of applied electric field: (1) a highly transparent Josephson junction (JJ) regime with multiple subbands contributing to transport, and (2) a superconducting quantum dot (QD) regime near depletion, with lower interface transparencies where superconducting transport takes place through a strongly coupled quantum dot (De Franceschi *et al.* 2010) operating in the few-hole regime. For the first time we show access to both regimes in a single device. Additionally, this is the first observation of proximity-induced superconductivity in a few-hole quantum dot.

9.2 DEVICE AND MEASUREMENT SETUP

A SEM image of the device is shown in Figure 9.1a where a Ge-Si core-shell nanowire with a diameter of 20 nm lies on top of 100 nm SiO_2 covering the p^{++} Si substrate which serves as a backgate. We use electron-beam lithography and metal evaporation to define contacts with a 50 nm thick Al layer compatible with a 4-terminal measurement configuration. Before metal evaporation, a 3 second etch with BHF is performed to remove native SiO_2 from the nanowire.

Transparent superconducting contacts are obtained in a final crucial fabrication step: we perform a 10 min. thermal anneal at 180°C on a hotplate in ambient conditions resulting in a drastic decrease of contact resistance from several $\text{M}\Omega$ to several $\text{k}\Omega$ for about 50% of the devices, out of which about 75% show superconducting transport. As described in detail in Chapter 10, our junction exhibits a superconducting silicide or germanide (Chevrier *et al.* 1987; Deutscher *et al.* 1971; Kuan *et al.* 1982; Lesueur *et al.* 1988; Tsuei *et al.* 1974; Xi *et al.* 1987a) caused by Al diffusion from the leads into the nanowire channel. We believe that the resulting modified Al-nanowire interface is essential to obtain transparent contacts.

All measurements are performed in a dilution refrigerator at base temperature (~ 15 mK), which is equipped with copper powder filters to reach electron temperatures as low as 25 mK (Mueller *et al.* 2013). For the presented device, a 3-probe configuration is used and a series resistance of 3.46 $\text{k}\Omega$ due to the measurement lines is subtracted, unless stated otherwise. We have measured 3 other Josephson field-effect transistors of which 2 exhibit a quantum dot regime but have lower switching currents in accumulation mode. The other device is highly transparent but could not be fully depleted.

9.3 JOSEPHSON FIELD-EFFECT TRANSISTOR: GATE TUNABLE SUPERCURRENT

Figure 9.1b shows the measured voltage V_{SD} as a function of the sourced current I_{S} at five different backgate voltages V_{BG} . A zero-voltage current, i.e., a supercurrent, can be seen for all V_{BG} while I_{S} is swept from negative to positive bias. In all curves, we see the first occurrence of supercurrent at the retrapping current I_{R} . Upon increasing I_{S} , the device switches again to the normal state at the

switching current I_C .¹ Since Si-Ge nanowires are hole conductors, a more negative V_{BG} increases the number of subbands, i.e., parallel conduction channels, resulting in higher switching currents I_C and an increased conductance in the normal state. In Figure 9.1b, the ability to tune I_C with an applied electric field is clearly demonstrated.

For a more in-depth investigation of the current-voltage relation as a function of electric field, we turn to the numerical differential resistance $\partial V_{SD}/\partial I_S$ versus I_S and V_{BG} in Figure 9.2a. The black region corresponds to a transport regime of dissipationless current measured from negative to positive I_S . The transition to the normal state is observed as a very sharp peak in $\partial V_{SD}/\partial I_S$ at both $I_S = I_C$ and $I_S = I_R$.

Two distinct regimes can be identified as a function of V_{BG} : the Josephson junction regime with finite switching currents throughout $-20 < V_{BG} < 1.5$ V and the quantum dot regime where I_C repeatedly vanishes and reappears for $1.7 < V_{BG} < 4.2$ V. While in the JJ the device is highly transparent, transport in the QD regime is characterised by an interplay between proximity-induced superconductivity and Coulomb interaction (De Franceschi *et al.* 2010; Katsaros *et al.* 2010) as we will discuss in detail in Figure 9.3 and Figure 9.4.

We extract both $|I_C|$ and $|I_R|$ from Figure 9.2a and plot them together with the normal state conductance G_N as a function of V_{BG} in Figure 9.2b. G_N is obtained as an inverse of $\partial V_{SD}/\partial I_S$ at $I_S = 0$ and $|B| = 2$ T to suppress superconductivity. In the JJ regime, we see an overall trend of decreasing I_C and G_N for increasing V_{BG} which decreases the charge carrier density in the wire. Consequently, the correlated oscillatory pattern between I_C and G_N is explained by the depopulation of 1D-subbands for increasing V_{BG} (Kotekar-Patil *et al.* 2017; Xiang *et al.* 2006a).

To quantify the correlation between I_C and G_N we plot the product $I_C R_N$ (with $R_N = 1/G_N$). For $V_{BG} < -5$ V we see that $I_C R_N$ becomes almost constant and has an average value $\langle I_C R_N \rangle = 217 \pm 23$ μeV . This means that I_C changes proportionally to G_N , indicating that we alter the number of parallel conduction channels. Integer plateaus of $2e^2/h$ in G_N cannot be clearly distinguished which we attribute to the presence of the confinement potential of the quantum dot in the nanowire channel (see Figure 9.3). We do stress however, that $\langle I_C R_N \rangle = 217 \pm 23$ μeV is approximately equal to the superconducting gap of our alu-

¹ When measuring a Josephson junction in this configuration, one obtains the switching current and not its critical current which requires measuring a distribution of switching currents at several temperatures (Wallraff *et al.* 2003).

Figure 9.2: **Josephson FET in the Josephson junction regime and the quantum dot regime.** (a) Numerical differential resistance $\partial I_S / \partial V_{SD}$ versus I_S and V_{BG} . Like in Figure 9.1b I_S is swept from negative to positive bias. The black region corresponds to zero resistance and indicates superconductivity. Vertical dashed lines indicate the traces in Figure 9.1a of the same colour. (b) Top panel: I_C (blue) and I_R (black) versus V_{BG} extracted from (a). The normal-state conductance G_N (orange) measured at $B = 2$ T when superconductivity is suppressed and is shown in units of $2e^2/h$. Bottom panel: The $I_C R_N$ product (green) is calculated from I_C and $R_N = 1/G_N$. The red trace represents the I_{exc} normalised by multiplying with the ratio $e R_N^* / \Delta_{Al}$.

minimum $\Delta_{\text{Al}} = 212 \mu\text{eV}$ and is an indication of a high interface transparency between the superconductor and the nanowire (Tinkham 1996).

In the bottom panel of Figure 9.2b we look at the normalised excess current $eI_{\text{exc}}R_{\text{N}}^*/\Delta_{\text{Al}}$ which I_{exc} can be considered as the extra current carried by the superconducting state of the junction. When normalised by multiplying with $eR_{\text{N}}^*/\Delta_{\text{Al}}$, it gives a direct measure for the scattering parameter Z of an S-N-S junction in the BTK model (Blonder *et al.* 1982; Flensberg *et al.* 1988). I_{exc} is calculated from the zero crossing of a linear fit for $I_{\text{S}} - V_{\text{SD}}$ at high bias (300-500 nA) well away from I_{C} and R_{N}^* is the averaged normal-state resistance obtained at high bias. We find the average normalised excess current in the JJ regime to be 1.48 ± 0.39 , which translates to a scattering parameter $Z = 0.4 \pm 0.14$ and a total junction transparency $T = 86 \pm 8 \%$. Using the on-resonance transmission probability $T = 4\Gamma_{\text{S}}\Gamma_{\text{D}}/(\Gamma_{\text{S}} + \Gamma_{\text{D}})^2$ with $\Gamma_{\text{S(D)}}$ the tunnel coupling to the source (drain), we estimate a tunnelling asymmetry of a factor ~ 2 (Stone *et al.* 1985). Throughout the JJ regime, $0 < eI_{\text{exc}}R_{\text{N}}^*/\Delta_{\text{Al}} < 8/3$ as expected for a superconducting junction in the BTK theory (Blonder *et al.* 1982; Flensberg *et al.* 1988).

Multiple Andreev reflections are prominently featured in Figure 9.2a as a wavy pattern of lines of increased conductance in the dissipative state. These lines denote steps of $V_{\text{SD}} = 2\Delta/n$ with n an integer denoting the MAR order (see Figure 9 A.1), and are therefore correlated to the device conductance G (and G_{N}), explaining their oscillations as a function of V_{BG} . A requirement for observing MAR is a sudden and homogeneous interface between superconductor and nanowire, albeit with a transmission probability lower than unity (Flensberg *et al.* 1988), corresponding well to the obtained value for the junction transparency.

Inspecting Figure 9.2b, we notice I_{R} to oscillate much stronger than I_{C} or G_{N} as a function of V_{BG} . Small ratios $I_{\text{R}}/I_{\text{C}}$ correspond to an underdamped Josephson junction. In this case, the Stewart-McCumber parameter β_{C} is proportional to $(I_{\text{C}}/I_{\text{R}})^2$ (Zappe 1973) and the retrapping current is not inversely proportional to changes in R_{N} but rather to the subgap resistance, R_{sg} . The subgap resistance is very sensitively depending on the presence of states in the gap, for example from MAR as illustrated by their clear correlation to I_{R} , and might strongly depend on the applied back gate voltage. To summarize, in the JJ regime, the highly transparent superconductor-nanowire interfaces allows for continuous Josephson current as function of V_{BG} while $I_{\text{C}}R_{\text{N}} \approx \Delta_{\text{Al}}$.

Figure 9.3: **Depletion in the quantum dot regime.** Numerical differential conductance $\partial I_D / \partial V_{SD}$ versus V_{SD} and V_{BG} in the QD regime and $|B| = 2$ T. A bias up to $|V_{SD}| = 80$ mV is applied only where G_N is low; the grey area therefore contains no measurement data. A series resistance of 9.1 k Ω in the measurement lines and the IV-converter is subtracted from the data.

9.4 THE SUPERCONDUCTING QUANTUM DOT REGIME

We now tune our device into the QD regime in Figure 9.3 and Figure 9.4. Here, the nanowire has a low charge carrier density and transport is governed by an interplay between superconductivity and Coulomb interactions (Doh *et al.* 2008; Eiles *et al.* 1994). We first look at a voltage-biased measurement in the normal state in Figure 9.3 where we apply a $|V_{SD}|$ of up to 80 mV to the source and measure the current I_D from drain to ground and calculate the numerical differential conductance $\partial I_D / \partial V_{SD}$. We observe a smooth turn-off between $V_{BG} = 4.4 - 4.9$ V as $|V_{SD}|$ increases. The charge switches (black-white pixelated region) right before pinch-off are caused by a nearby charge defect. To verify depletion of the nanowire at higher V_{BG} , we continued this measurement up to $V_{BG} = 8$ V: no current above the noise level of

Figure 9.4: **Few-hole quantum dot strongly coupled to superconducting leads.** (a) numerical differential conductance $\partial I_D / \partial V_{SD}$ versus V_{SD} and V_{BG} in the QD regime and $|B| = 2$ T. Zoom of Figure 9.3 with an applied bias up to $|V_{SD}| = 12$ mV. A series resistance of 9.1 k Ω in the measurement lines of the IV-converter is subtracted from the data in both. We observe Coulomb diamonds of alternating heights indicating an even (E) and odd (O) filling of a strongly coupled quantum dot. White dashed lines are guides to the eye. (for the same figure at $|B| = 0$ T see Figure 9 A.2b, or without guides to the eye see Figure 9 A.2a). (b) $\partial V_{SD} / \partial I_S$ versus I_S and V_{BG} . Regions of supercurrent align with the Coulomb peaks in (a) and (c). (c) Top panel: I_C (blue) extracted from a) and G_N (orange) versus V_{BG} . Bottom panel: $I_C R_N$ product (green) and $e I_{exc} R_N^* / \Delta_{AI}$ (red). Black dashed lines in the top (bottom) panel is a fit of I_C to G_N ($I_C R_N$ to G_N).

our measurement setup (≈ 20 fA) was detected and we therefore label $V_{BG} > 5$ V as depleted ($N = 0$).

The zoom in Figure 9.4a reveals a strongly coupled quantum dot and we identify six Coulomb diamonds, numbered $D1 - D6$ of alternating size, characteristic for even-odd filling of orbital levels (Eichler *et al.* 2007). The crossing of two adjacent diamonds marks the charge degeneracy point where transport takes place via resonant single-particle tunnelling (Kouwenhoven *et al.* 2001a). Figure 9 A.2c shows Fig. 10.3a at $B = 0$ T for a much smaller V_{SD} and here the Coulomb peaks can be identified by their modulation of the MAR pattern (Buitelaar *et al.* 2003; Buitelaar *et al.* 2002).

The best resolved diamond, $D4$, has an addition energy $E_{add} \approx 10$ meV and serves to find the gate lever arm $\alpha \approx 0.02$ eV/V, which we use to estimate the size of the other diamonds in correspondence with their Coulomb peak spacing. We estimate the charging energy E_C of the quantum dot from the average height of the smaller uneven diamonds $D1$, $D3$ and $D5$ for which we find $E_C \approx 4$ meV. For the larger even diamonds, $E_{add} = E_C + \Delta E$ where ΔE the orbital energy and E_{add} the total energy to add an unpaired hole on the dot. $D2$, $D4$ and $D6$ give an average $E_{add} \approx 10$ meV, resulting in $\Delta E = E_{add} - E_C \approx 6.0$ meV, comparable to the first energy level in a 1-dimensional particle-in-a-box equal to our channel length of 150 nm. We are confident that the device operates in the few-hole regime, because we observe charge transitions up to depletion and a corresponding smooth pinch-off above $V_{BG} = 4.3$ V. Due to the large tunnel broadening and the distortion caused by the bi-stable charge defect, we cannot be completely sure that only a single hole occupies the dot in $D1$.

We also note the presence of a zero-bias ridge inside the diamonds with uneven charge occupation which we ascribe to the Kondo effect, as observed in numerous other studies of strongly coupled quantum dots (Buitelaar *et al.* 2003; Buitelaar *et al.* 2002; Buizert *et al.* 2007; Eichler *et al.* 2007; Grove-Rasmussen *et al.* 2012; Grove-Rasmussen *et al.* 2007; Jespersen *et al.* 2006; Lee *et al.* 2014; Nilsson *et al.* 2009; Sand-Jespersen *et al.* 2008; Sand-Jespersen *et al.* 2007; Wu *et al.* 2009). In the current-sourced plot at $B = 0$ T, multiple regions of superconductivity coincide with the resonant single-particle levels of the strongly coupled quantum dot in Figure 9.4a. An exception is $D3$, where in addition to single-particle levels, the strong Kondo ridge ($T_k \approx 20$ K as approximated from its full width half maximum (FWHM) as $T_k = eFWHM/k_B$ (van der Wiel 2000)) facilitates a supercurrent in-

side the diamond (Eichler *et al.* 2009). This is reflected in the corresponding peak in I_C and G_N in Figure 9.4c, which is a superposition of two diamond crossings and the Kondo ridge. Inside $D3$, the maximum normal state conductance of the Kondo ridge $\sim 0.8 \cdot 2e^2/h$ and since $T \ll T_k$, this indicates a tunnel barrier asymmetry smaller than a factor of 3 (Kouwenhoven *et al.* 2001b), consistent with our findings in the JJ regime (see Figure 9 A.2b for a clearer Kondo ridge).

The tunnel coupling $\Gamma = (\Gamma_S + \Gamma_D/\Gamma_S\Gamma_D)$ of the dot to the leads is estimated by taking α FWHM of the peaks in G_N . For the Coulomb peaks on the $D1/D2$ crossing and the $D4/D5$ crossing we find $\Gamma = 1.5 \pm 0.3$ meV and 5.5 ± 0.8 meV respectively. Comparison of the relevant energy scales $\Gamma \approx E_C, \Delta E > \Delta_{Al}$ shows the quantum dot is strongly coupled to the superconducting leads (De Franceschi *et al.* 2010), confirmed by the finite I_C through the single-particle levels.

The maximum switching current carried by the single-particle levels is in the order of $I_C \approx 10$ nA in Figure 9.4c, considerably lower than the theoretical maximum $I_{C,MAX} = e\Delta_{Al}/\hbar = 51$ nA (Beenakker *et al.* 1991), but comparable to results found in literature (Jarillo-Herrero *et al.* 2006; Xiang *et al.* 2006a). We attribute this decreased I_C to finite barrier transparencies and coupling to the electromagnetic environment (Jarillo-Herrero *et al.* 2006; Joyez *et al.* 1994; Vion *et al.* 1996), known to suppress the measured I_C up to an order of magnitude in underdamped junctions. For wide resonances of a single-particle level $\hbar\Gamma \gg \Delta$ including coupling to the electromagnetic environment we expect $I_C \propto (1 - (1 - G_N)^{1/2})^{3/2}$ (Galaktionov *et al.* 2002; Jarillo-Herrero *et al.* 2006). Using this relation we fit I_C and $I_C R_N$ to G_N for the broadened peak at $V_{BG} = 3$ V and even though the shape of G_N is determined by the Kondo ridge (non-Lorentzian), we obtain reasonable qualitative agreement.

In the QD regime, the normalized excess current $eI_{exc}R_N^*/\Delta_{Al}$ gives qualitative information about the state of the junction: it becomes negative inside the Coulomb diamonds and indicates, as expected, transport over a potential barrier (Blonder *et al.* 1982). On the other hand, in the superconducting regions around the Coulomb peaks, $eI_{exc}R_N^*/\Delta_{Al}$ is positive and appears to exceed the theoretical limit of $8/3$. This is caused by resistance variations persisting at high bias due to the presence of the quantum dot. At the diamond edges of $D3$, $eI_{exc}R_N^*/\Delta_{Al}$ shows a double peak representing the two charge degeneracy points which are obscured at low bias by the Kondo ridge.

9.5 CONCLUSION

We have demonstrated a Ge-Si nanowire based Josephson junction, where the switching current is tunable with an electric field. A new straightforward fabrication technique provides high interface transparencies in the Josephson junction regime, confirmed by an $I_C R_N$ product comparable to the superconducting gap of our aluminium. Near depletion, we observe the superconducting quantum dot regime, where supercurrents are carried by single-particle levels in a strongly coupled quantum dot, operating in the few-hole regime. We demonstrate, for the first time, access to both regimes in a single device. These results pave the way for a wide range of follow-up experiments and, combined with the predicted strong spin-orbit interaction (Kloeffel *et al.* 2011), provide a promising platform for Majorana fermions (Maier *et al.* 2014a).

APPENDIX

Figure 9 A.1: **Josephson FET**: Numerical differential conductance $\partial I_S / \partial V_{SD}$ vs V_{SD} and V_{BG} . Same dataset as in Figure 9.2a with I_S and V_{SD} reversed. Horizontal lines correspond to multiple Andreev reflection (MAR) at values $|V_{SD}| = 2\Delta_{A1}/n$ with n an integer denoting the order of MAR. The first three MAR orders are shown in the figure with higher orders visible for decreasing V_{SD} .

Figure 9 A.2: **QD regime:** a) Numerical differential conductance $\partial I_D / \partial V_{SD}$ vs V_{SD} and V_{BG} for $B = 0$ T. The superconducting Al opens a gap cutting through the diamonds for $|V_{SD}| < 2\Delta_{Al}$. b) $\partial I_D / \partial V_{SD}$ vs V_{SD} and V_{BG} for $|B| = 2$ T. Same plot as Figure 9.4a without the guides to the eye for the diamonds. c) $\partial I_D / \partial V_{SD}$ vs V_{SD} and V_{BG} for $|V_{SD}| < 1$ mV at $B = 0$. The positions of the Coulomb peaks are shown by the green dashed lines. Odd (O) and even (E) dot occupancies are denoted based on diamond size and the presence of a Kondo peak. Horizontal lines at values $|V_{SD}| = 2\Delta_{Al}/n$ correspond to multiple Andreev reflection modulated by interaction with Coulomb peaks.

HARD SUPERCONDUCTING GAP AND ADDITIONAL SUPERCONDUCTING PHASES IN GE-SI NANOWIRES

We show a hard induced gap up to magnetic fields of 250 mT in a Ge-Si nanowire Josephson transistor. The highly homogeneous and abrupt interface between the superconducting Al contacts and the semiconducting nanowire are obtained by annealing devices at 180°C. During annealing, aluminium diffuses and alloys with Ge and/or Si forming a second superconducting binary and/or ternary alloy inside or in close proximity with the nanowire which induces a proximity supercurrent in the nanowire even when Al is normal. Compared to pure Al, this alloy has a lower critical temperature ($T_C = 0.9$ K) and a higher critical field ($B_C \approx 1$ T). In another device where the diffusion of Al rendered the nanowire completely metallic, a superconducting alloy with a much higher critical temperature ($T_C = 2.9$ K) and critical field ($B_C = 3.4$ T) is found. No previous reports exist on the diffusion induced formation of superconducting alloys inside nanowires and they may be of particular interest for applications requiring proximity-induced superconductivity in high magnetic fields. A hard gap at finite magnetic fields is an important step towards creating and detecting Majorana fermions.

10.1 INTRODUCTION

The discovery that Majorana fermions offer a route towards an inherently topologically protected fault-tolerant quantum computer (Das Sarma *et al.* 2005; Nayak *et al.* 2008; Read *et al.* 2000) marked the beginning of a quickly growing field of research to achieve their experimental realisation. Majoranas require a topological superconducting material, which in practice can be realised by coupling a conventional *s*-wave superconductor to a 2-dimensional topological insulator (Bocquillon *et al.* 2016; Deacon *et al.* 2017; Drachmann *et al.* 2017; He *et al.* 2017; Wiedenmann *et al.* 2016) or to a 1-dimensional nanowire with high spin-orbit coupling and *g*-factor (Kitaev 2000; Lutchyn *et al.* 2010; Oreg *et al.* 2010). In the 1-dimensional case, signatures of Majorana fermions are expected to arise as a conductance peak at zero bias and finite magnetic fields. The first reports showing these zero-bias conductance peaks (Churchill *et al.* 2013; Das *et al.* 2012; Finck *et al.* 2013; Lee *et al.* 2012; Mourik *et al.* 2012) in InAs and InSb nanowires suffered from sizeable sub-gap conductivity attributed to inhomogeneities in the nanowire-superconductor interface (Cole *et al.* 2015; Takei *et al.* 2013). This quasiparticle poisoning in the induced gap decoheres Majorana states since they will participate in braiding operations (Albrecht *et al.* 2017; Higginbotham *et al.* 2015), and additionally obscure the Majorana signatures at zero bias. Great efforts have been made to improve these interfaces, i.e., induce a hard gap, using epitaxially grown Al (Chang *et al.* 2015; Kjaergaard *et al.* 2016) or specialised surface treatments methods (Gül *et al.* 2017; Zhang *et al.* 2017), resulting in much better resolved Majorana signatures (Albrecht *et al.* 2016; Deng *et al.* 2016; Higginbotham *et al.* 2015).

In contrast to the group III-V materials used in previous work, we use Ge-Si core-shell nanowires, a candidate for observing Majoranas (Maier *et al.* 2014a; Thakurathi *et al.* 2017) for which interaction with a superconductor is still relatively unexplored (Katsaros *et al.* 2010; Ridderbos 2018; Su *et al.* 2016; Xiang *et al.* 2006b). These wires are predicted to have a strong first-order Rashba type spin-orbit coupling (Kloeffel *et al.* 2011) and have a gate-tunable *g*-factor (Brauns *et al.* 2016b; Maier *et al.* 2013). Our devices consist of a nanowire with superconducting Al source and drain on an oxidised Si substrate and exhibit two highly interesting properties: we find a very hard induced gap and we observe the presence of a second superconducting phase in the nanowire channel.

In the first part of this chapter, we investigate this newly found superconducting phase which arises during thermal annealing. We focus on two devices containing a diffusion-induced superconducting phase with very different T_C : device A is a nanowire Josephson junction with a superconducting phase we call X1 with a critical temperature $T_{C,X1} = 0.9$ K, lower than of our Al ($T_{C,Al} = 1.4$ K), but a much higher critical field in the order of 1 T. In device B, Al has diffused through the entire nanowire channel, rendering it metallic. This second superconducting phase (X2) has both a higher critical temperature $T_{C,X2} = 2.9$ K and a much higher $B_{C,X1} \approx 3.4$ T (see Table 10.2).

In the second part we investigate the hardness of the superconducting gap (Chang *et al.* 2015; Gül *et al.* 2017) induced in the semiconducting nanowire of device A where we find an in-gap current suppression of a factor ~ 1000 . We believe that the diffusion of Al is essential in obtaining the required homogeneous Al-nanowire interface (Takei *et al.* 2013), although the exact role of the second superconductor X1 is not fully understood. Furthermore, Al diffusion is allegedly responsible for obtaining highly transparent contacts (Ridderbos 2018; Xiang *et al.* 2006a).

10.2 SUPERCONDUCTING AL/GE AL/SI ALLOYS

The proposed mechanism for the formation of the observed superconducting alloys is the diffusion of Al into the Ge-Si nanowire where its stoichiometric composition depends on the microscopic details of the diffusion process. Preliminary results from a TEM study in combination with energy-dispersive X-ray spectroscopy (EDX) indeed suggest there is significant Al diffusion into the Ge core of the nanowire but we have not yet been able to resolve the consistency of the resulting alloy and are unable to provide more concise information at this moment.

Superconducting alloys of Al/Si, Al/Ge and Al/Ge/Si have been studied in thin films and in bulk for various methods of preparing the alloy: high pressures (Degtyareva *et al.* 1985), co-evaporation (Deutscher *et al.* 1971; Deutscher *et al.* 1979), ion beam mixing (Xi *et al.* 1987a; Xi *et al.* 1987b; Xi *et al.* 1987c) and rapid quenching in after melting (Chevrier *et al.* 1987; Kuan *et al.* 1982; Lesueur *et al.* 1988; Tsuei *et al.* 1974). For all preparation techniques, alloys show a wide range of T_C depending on their stoichiometry, and T_C is generally higher than of pure Al. Reported record values are $T_C = 11.0$ K for Al/Si formed under high pressure and $T_C = 7.35$ K for

Al/Ge using low-temperature ion implantation (Lamoise *et al.* 1976), both for semiconductor to metal ratios of $\sim 30\%$. A lower T_C than Al has been realised for quenched Al/Ge alloys with metal to semiconductor ratios close to the metal-insulator-transition of $\sim 50\%$ (Deutscher *et al.* 1979; Lesueur *et al.* 1988).

Since our alloys are formed when Al diffuses into our nanowires during annealing, we cannot make a direct comparison with the alloys reported in literature. Nevertheless, both the T_C for X1 and X2 fall within reported values of T_C .

10.3 TWO SUPERCONDUCTING PHASES IN A NANOWIRE JOSEPHSON JUNCTION

In Figure 10.1a we show a magneto-spectroscopy of device A, the Josephson junction: we plot the differential resistance $\partial V_{SD}/\partial I_S$ versus the sourced current I_S and the out-of-plane magnetic field $B_\perp$ (see inset Figure 10.2b). The backgate is fixed at $V_{BG} = -4.7$ V where multiple subbands contribute to transport and the junction is highly transparent (see Chapter 9). The superconducting region (black) is bounded by $I_R < I_S < I_C$ with I_R the retrapping current at negative bias and I_C the switching current at positive bias. Upon increasing $B_\perp$ from 0, I_C decreases gradually until aluminium becomes normal at the critical out-of-plane field $B_{C\perp,Al} \approx 40$ mT after which a finite I_C remains.

When increasing $B_\perp$ further, I_C slowly decreases and finally disappears. Observing a proximity-induced supercurrent above $|B_{C\perp,Al}|$ implies the presence of a second superconducting material in or near the nanowire channel, X1, with a critical field $B_{C\perp,X1} \approx 950$ mT.

We will now provide additional arguments to support this hypothesis. To confirm that our Al contacts are normal for $B_\perp > |B_{C\perp,Al}|$, we look at the background resistance R_B in the superconducting region as a function of $B_\perp$ in the bottom panel of Figure 10.1b. $R_B = 0$ for $B_\perp < |B_{C\perp,Al}|$, while for $B_\perp > |B_{C\perp,Al}|$ the background resistance gradually increases to $R_B \approx 0.25$ k Ω attributed to a series resistance of the Al contacts, thus confirming they are normal. Additionally, the out-of-plane critical field found for a strip of pure Al of the same geometry (not shown here) matches $B_{C\perp,Al}$ found for the Josephson junction.

In contrast to the critical temperature T_C , the critical field of a superconductor is not a material property and depends strongly on its

Figure 10.1: **Josephson junction with two superconducting phases:** (a) Numerical differential resistance $\partial V_{SD}/\partial I_S$ versus I_S and $B_{\perp}$ taken at $T = 100$ mK. Black region corresponds to superconductivity. The white dashed line indicates $B_{C,\text{Al}}$. Arrows indicate I_C and I_R . (b) Top panel: Same as (a) for a larger range of $B_{\perp}$. The vertical black dashed line indicates $B_{C,\text{X1}}$. Bottom panel: Horizontal cross-section showing $\partial V_{SD}/\partial I_S$ vs $B_{\perp}$ taken at $I_S = 0$. (c) Same as (a) taken at $T = 900$ mK. (d) Linecuts showing V_{SD} vs I_S taken at the corresponding symbols in (a) and (c). In all figures, configuration I-IV are combinations of Al and X1 in the superconducting/normal state indicated by the green (superconducting) and red (normal) boxes. In all figures $V_{BG} = -4.7$ V and I_S is swept from negative to positive bias.

Table 10.1: Configurations of device A with two superconducting phases and the corresponding conditions for the magnetic field $\mathbf{B}$ and temperature T , SC refers to superconductivity. The last column refers to the plots in Figure 10.1d.

Conf.	SC	Norm.	Conditions	I_C / color Fig. 10.1d
I	Al,X1		$\mathbf{B} < B_{C,Al}, B_{C,X1}$ & $T < T_{C,X1}, T_{C,Al}$	$I_C \approx 36$ nA blue
II	X1	Al	$B_{C,Al} < \mathbf{B} < B_{C,X1}$ & $T < T_{C,X1}, T_{C,Al}$	$I_C \approx 17$ nA purple
III	Al	X1	$\mathbf{B} < B_{C,Al}, B_{C,X1}$ & $T_{C,X1} < T < T_{C,Al}$	$I_C \approx 24$ nA brown
IV		Al,X1	$\mathbf{B} > B_{C,X1}$ or $T > T_{C,Al}$ or $B_{C,Al} < \mathbf{B} < B_{C,X1}$ & $T_{C,X1} < T < T_{C,Al}$	$I_C = 0$ grey

geometry (Tinkham 1996), i. e., the two critical fields can be explained by two different shapes of the same superconductor. In Figure 10.1a, we show the same magneto-spectroscopy at 900 mK and observe that X1 is quenched for all $B_\perp$, while Al still induces a supercurrent for $B_\perp < |25|$ mT. Since the X1 clearly has a lower T_C and a higher B_C than Al, we conclude that X1 must have a different composition as well as shape.

Because X1 has a higher B_C and a lower T_C than Al, we can selectively switch either superconductor to the normal state resulting in four possible device configurations as illustrated in Figure 10.1. These four configurations are decribed in Table 10.1.¹

10.4 JUNCTION I_C VERSUS B AND T

Since for each superconductor the critical field and critical temperature are inter-dependent variables and may have a non-trivial relation, the boundaries of the configurations I-IV in terms of B_C and T_C cannot directly be deduced from the data in Figure 10.1. We therefore collect I_C versus B from magneto-spectroscopies such as in Figure 10.1 for a large number of temperatures and the three main magnetic field axes

¹ For generality the field direction is removed from the subscripts

Figure 10.2: I_C , T_C and B_C of a Josephson FET and a metallic device: (a) I_C versus T and B_* for device A. The green (red) boxes indicate whether the material is superconducting (normal) and show the configurations as defined in the main text. (b) T_C vs B_C for Al and X1 for three main field axis $B_\perp$, $B_\parallel$ and B_* as illustrated by the inset. Curves are extracted from plots such as (a) (see main text). Shaded areas are standard deviations in B_C . (c) I_C versus T and B_Z for completely metallised nanowire consisting of alloy X2. (d) T_C vs B_C for X2 extracted from (c). Inset shows the in-plane B_Z field direction which is rotated $\sim 10^\circ$ with respect to the nanowire. B_Z corresponds to the z-axis of the vector magnet, the only axis capable of fields > 1 T. For $B_Z < 300$ mT and $T < 1$ K a slight enhancement of I_C can be observed caused by the superconducting Al contacts.

Table 10.2: Maximum values for T_C , B_C of Al, X1 and X2 as determined in Figure 10.2. We take $T_{C,Al}(B_C = 0 \text{ mT})$, $T_{C,X1}(B_\perp = 50 \text{ (mT)})$ and $B_C(T_C \approx 0)$ to obtain their respective maximum values. The BCS superconducting gap is calculated as $\Delta = 1.764k_B T_C$ (Tinkham 1996).

	T_C (K)	Δ (μV)	$B_{C\star}$ (mT)	$B_{C\perp}$ (mT)	$B_{C\parallel}$ (mT)
Al	1.4 ± 0.05	212 ± 6	293 ± 10	41 ± 2	282 ± 10
X1	0.9 ± 0.05	133 ± 8	$1\,230 \pm 10$	909 ± 11	$1\,010 \pm 20$

	T_C (K)	Δ (μV)	$B_{C,Z}$ (T)
X2	2.9 ± 0.1	441 ± 14	3.4 ± 0.1

$B_\star$, $B_\perp$ and $B_\parallel$ which are illustrated by the inset in Figure 10.2b. For the in-plane field perpendicular to the nanowire, I_C has two clearly distinct overlapping shapes as a function of T and $B_\star$ in Figure 10.2a: The ‘peak’ extending to $T \approx 1\,400 \text{ mK}$ at $B = 0$ with a width of $|B_\star| \approx 300 \text{ mT}$ at $T = 0$ is attributed to the superconducting state of Al, while the second shape (the ‘tail’), extending up to $\approx 1\,000 \text{ mT}$ at $T = 0$, belongs to the superconducting phase of X1. We can thus place the four configurations in Table 10.1 that describe the state Al and X1 in the colour plot on the T vs $B_\star$ axes.

We now extract both the T_C - $B_{C\star,Al}$ and T_C - $B_{C\star,X1}$ curves from Figure 10.2a, i.e., the critical temperature - critical field relation for Al and X1. We define $B_{C\star,Al}$ ($B_{C\star,X1}$) as the field where Al (X1) no longer induces an I_C . We cannot directly observe $B_{C\star,Al}$ for $0 > T > 800 \text{ mK}$ where the ‘peak’ of Al and the ‘tail’ of X1 overlap (for $230 < B_\star < 300 \text{ mT}$) and we therefore use the following method: (1) We take each individual I_C - $B_\star$ curve, i.e., horizontal linecuts in Figure 10.2a for all temperatures, (2) we select only I_C where $B_\star < B_{C\star,Al}$ (3) we fit I_C to an empirical polynomial of the form $I_C(B_\star) = aB_\star^4 + bB_\star^2 + c$ with a and b only allowed negative while c is always positive, (4) we find $B_{C\star,Al}$ for each temperature as the roots of $I_C(B_\star)$ (i.e., zero crossings). For X1 we use the same method except in (2) we select only I_C for $B_\star > B_{C\star,Al}$. We perform the same procedure for field directions $B_\perp$ and $B_\parallel$ (see Supplementary Information, Figure 10 A.1 for I_C versus T and $B_\parallel$ and $B_\perp$) and plot all resulting T_C - B_C curves in Figure 10.2b.

In Table 10.2 we summarize the maximum T_C , the resulting superconducting gap Δ and B_C in the three field directions for Al and X1.

Comparing $B_{C\perp,Al} = 41$ mT with $B_{C\star,Al} = 293$ mT and $B_{C\parallel,Al} = 282$ mT we notice a factor ~ 7 difference. This strong anisotropy for the out-of-plane field direction is clearly present in the $T_{C,Al}$ - $B_{C,Al}$ curves in Figure 10.2b and is expected for the large aspect ratio of the Al contacts.

The $T_{C,X1}$ - $B_{C,X1}$ curves show a less prominent magnetic field anisotropy from which we can roughly deduce the shape of X1 by assuming the normal surface of the material is inversely proportional to the critical field, i. e., a larger superconducting normal surface requires to expel more flux (Tinkham 1996). Using the respective ratios of $B_{C\star,X1}$, $B_{C\perp,X1}$ and $B_{C\parallel,X1}$ we find that X1 is to a small degree compressed in the out-of-plane direction and is slightly elongated along the nanowire.

We now switch to the completely metallised device B in Figure 10.2c which shows I_C vs T and B_Z . The corresponding $T_{C,X2}$ - $B_{C,X1}$ relation in Figure 10.2d is obtained by the previously described polynomial fitting method. We see a critical temperature $T_{C,X2} = 2.9$ K at $B = 0$ and critical field $B_{C,X1} = 3.4$ T at $T \approx 0$, both much higher than for X1. $I_C = 1.5$ μ A, two orders of magnitude higher compared to device A. Comparing T_C and Δ of X1 and X2, we conclude that the diffusion of Al into Ge-Si nanowires gives rise to different superconductors with lower and higher T_C than pure Al, most likely depending on the relative ratios of Al, Si and Ge.

In device B, we believe Al has diffused completely through the channel, effectively making the nanowire a metallic superconductor and the device is thus no longer a Josephson junction. The high T_C and B_C of X2 makes it a promising material for super-semi devices with high B_C where the challenge is to control Al diffusion in the longitudinal direction of the nanowire.

To sum up, Figure 10.2b shows two superconductors: X1 with $T_{C,X1} = 0.9$ K in a Josephson junction and X2 with $T_{C,X2} = 2.9$ K in a metallic device. Both superconductors are formed during annealing and are able to induce a supercurrent in the nanowire channel.

10.5 HARD SUPERCONDUCTING GAP

We now focus on device A and tune V_{BG} to a regime where the nanowire is near depletion. Figure 10.3a shows the differential conductance $\partial I_D / \partial V_{SD}$ versus the applied voltage bias V_{SD} and the backgate voltage V_{BG} . We notice a prominent MAR pattern for

Figure 10.3: **Hard superconducting gap in device A:** (a) Differential conductance $\partial I_D / \partial V_{SD}$ vs V_{SD} and V_{BG} . Odd (O) and even (E) hole occupation are indicated. (b) Vertical linecuts from (a) showing $\partial I_D / \partial V_{SD}$ vs V_{SD} at the denoted V_{BG} . Curves are offset by 15 μS . (c) Averaged in-gap conductance $\langle G_G \rangle$ and outside-gap conductance $\langle G_O \rangle$ and the ratio $\langle G_G \rangle / \langle G_O \rangle$ versus V_{BG} . $\langle G_G \rangle$ and $\langle G_O \rangle$ are averaged over a range V_{SD} for every V_{BG} as indicated by grey areas in (b)). (d) $\partial I_D / \partial V_{SD}$ vs V_{SD} for B_* from 0 to 1000 mT, curves are offset by 1 μS . Black (grey) dotted lines show the expected position of the quasiparticle peak for Al (X1). (e) Ratio $\langle G_G \rangle / \langle G_O \rangle$ for the three main field axes $B_\perp$, $B_\parallel$ and B_* at $V_{BG} = 4.45$ V (blue dotted line in (a)). Ranges in V_{SD} where $\langle G_G \rangle$ and $\langle G_O \rangle$ are determined are shown in (c).

$V_{\text{BG}} = 3\text{--}4$ V and a zero-bias conductance peak caused by superconducting transport. A reduced barrier transparency near depletion confines charges in the nanowire channel, and allows us to see odd and even charge occupation in a quantum dot in the wire (see Chapter 9), which is supported by a Kondo peak on the odd transition (Kim *et al.* 2013; Ridderbos 2018) (see Supplementary Information, Figure 10 A.2). Above $V_{\text{BG}} = 4.4$ V, the MAR and zero-bias peak disappear while the onset of quasiparticle transport is visible at the induced gap at $V_{\text{SD}} = |2\Delta_{\text{Al}}|$ in Figure 10.3a. This trend is also present in the $\partial I_{\text{D}}/\partial V_{\text{SD}}$ linecuts for V_{BG} between 4.3 and 4.5 V in Figure 10.3b.

A measure for the number of quasiparticle states inside the gap is the in-gap suppression of conductance, also termed as the hardness of the induced gap. We therefore investigate the ratio $\langle G_{\text{G}} \rangle / \langle G_{\text{O}} \rangle$ where $\langle G_{\text{G}} \rangle$ ($\langle G_{\text{O}} \rangle$) is an averaged conductance value inside (outside) the gap. Figure 10.3c shows $\langle G_{\text{G}} \rangle$, $\langle G_{\text{O}} \rangle$ and the ratio $\langle G_{\text{G}} \rangle / \langle G_{\text{O}} \rangle$ versus V_{BG} and we find the conductance is suppressed by a factor > 1000 for $V_{\text{BG}} \approx 4.4$ V. For higher V_{BG} , $\langle G_{\text{G}} \rangle$ is limited by the noise floor of our measurement setup and remains constant. The increase of the ratio $\langle G_{\text{G}} \rangle / \langle G_{\text{O}} \rangle$ is caused by the gradually decreasing $\langle G_{\text{O}} \rangle$, and not due to an increase in in-gap quasiparticle states.

Since we cannot accurately determine $\langle G_{\text{G}} \rangle$ above $V_{\text{BG}} = 4.4$ V, the ratio $\langle G_{\text{G}} \rangle / \langle G_{\text{O}} \rangle$ is an upper limit and the actual conductance suppression may be lower (Zhang *et al.* 2017). Moreover, MAR and the zero-bias peak, both characteristic for Josephson junctions, cause conductance peaks inside the gap which may lead to an increased ratio $\langle G_{\text{G}} \rangle / \langle G_{\text{O}} \rangle$. Devices with one superconducting contact and one normal contact will not exhibit these peaks and may therefore result in a lower ratio $\langle G_{\text{G}} \rangle / \langle G_{\text{O}} \rangle$ and give a better approximation of the quasiparticle density in the gap. Although we cannot directly compare suppression factors because of the difference in device configuration (single vs two superconducting contacts) the ratio $\langle G_{\text{G}} \rangle / \langle G_{\text{O}} \rangle$ suggests that our semiconductor-nanowire interface homogeneity is equal or better than devices using epitaxial growth techniques (Chang *et al.* 2015) or specialised surface treatments (Gül *et al.* 2017).

We will now look at the magnetic field dependence of the hardness of the gap. We fix V_{BG} at 4.45 V and plot $\partial I_{\text{D}}/\partial V_{\text{SD}}$ versus V_{SD} for several $B_{\star}$ in Figure 10.3d. Upon increasing $B_{\star}$, the sharp quasiparticle peaks at $2\Delta_{\text{Al}}$ reduce in height and broaden towards $B_{\text{C}\star, \text{Al}} \approx 300$ mT. Instead of observing a second quasiparticle peak for $2\Delta_{\text{X1}}$ above $B_{\text{C}\star, \text{Al}}$,

the peaks keep broadening until a background conductance is present in the entire gap for $B_\star > B_{C\star,Al}$.

In Figure 10.3e we plot the ratio $\langle G_G \rangle / \langle G_O \rangle$ for the three main field directions. The initial ratio is $\sim 1 \cdot 10^{-3}$ in configuration **I** as defined in Table 10.1 and the gap remains hard until we approach the critical field of Al for the respective field direction (see Table 10.2). The highest field where the gap remains hard, $B_\parallel \approx 250$ mT, is slightly lower than $B_{C\parallel,Al}$ because of the strongly reduced Δ_{AL} at this field. The much softer gap in configuration **II** induced by X1 leads to a $\langle G_G \rangle / \langle G_O \rangle \approx 1 \cdot 10^{-1}$ which gradually increases to 1 approaching $B_{C,X1}$.

The results in Figure 10.3e show that Al needs to be superconducting in order to observe a hard gap. On the other hand, when going from configuration **I** to **III**, i. e., $T > T_{C,X1}$ at $B = 0$, we see no change in G_G that can be attributed to X1 becoming normal (see Supplementary Information, Figure 10 A.3 for the temperature dependence of the differential conductance at $V_{BG} = 4.45$ V and $B = 0$). This suggests that X1 being a superconductor is not required to observe a hard gap. However, X1 itself is essential for the hard gap since it is created by Al diffusion into the nanowire channel and therefore ensures a highly transparent contact. This results in a sudden and homogeneous interface at Al-X1 (there is a very sudden transition from pure Al to X1) and a more gradual transition between X1 and the Ge-Si channel with a transparency close to unity. Another indication that the interface properties change when Al becomes normal is seen in Figure 10.1a and Figure 10.1c. Here, the fringes outside the superconducting region attributed to MAR are only visible for $B_\perp < B_{C\perp,Al}$. For $B_\perp > B_{C\perp,Al}$, the conductance outside the gap shows no features.

In Figure 10.3a, between $V_{BG} = 4.2$ V and $V_{BG} = 4.4$ V, we observe a conductance peak in both bias directions smoothly moving from $V_{SD} = \Delta_{AL}$ to $2\Delta_{AL}$ which we attribute to an ABS signature. Observing ABS in a junction with two tunnel barriers requires a high asymmetry in transparency where the most opaque barrier is used to probe the ABS residing at the Fermi energy (Kim *et al.* 2013). In contrast to the bias-symmetric MAR features, the asymmetric barriers are reflected in the intensity of the ABS signatures which is highlighted by the purple linecut in Figure 10.3b taken at $V_{BG} = 4.3$ V. Depending on the bias direction, there are two different rate-determining tunnel sequences: (1) tunnelling through an opaque barrier onto a single available ABS or (2) tunnelling from an ABS through an opaque barrier into the Fermi

sea. Sequence (2) has a much higher tunnel probability than (1), which results in the observed asymmetry in conductance.

Additional evidence for ABS presents itself in the form of a region of negative differential conductance (NDC) between $V_{SD} = \Delta_{AL}$ and $V_{SD} = 2\Delta_{AL}$, around $V_{BG} = 4.15$ V (odd occupancy). Such a region of NDC is expected when probing an ABS with a highly asymmetric barrier (Kim *et al.* 2013). A high barrier asymmetry in our devices can be expected, since the final interface properties are determined by microscopic details on the Al-nanowire interface during annealing. Since for lower V_{BG} our barriers quickly become highly transparent (see Chapter 9), we are only able to observe an ABS signature near depletion.

10.6 CONCLUSION

Ge-Si nanowire devices with Al contacts show a second superconducting phase after annealing most likely caused by diffusion of Al into the nanowire channel. The resulting superconducting phase consists of an alloy of Al/Ge, Al/Si or Al/Si/Ge. We identify different compositions of this alloy: X1 is present in a Josephson FET and X2 resides in a metallic nanowire channel. X1 and X2 have a very different critical temperature and thus most likely a different composition. Since both X1 and X2 remain superconducting for magnetic fields much higher than Al, they are suitable materials for applications where proximity-induced superconductivity is required in high magnetic fields.

In the Josephson junction we can selectively switch Al or X1 to either the normal or superconducting state to independently assess their effect on the junction.

Close to depletion, the Josephson FET exhibits a hard superconducting gap where the current is suppressed by a factor ~ 1000 , indicating a very homogeneous interface which resides between Al and X1. We use the method of selectively switching Al and X1 to the normal state to find that Al needs to be superconducting in order to observe a hard gap, while X1 appears essential in obtaining transparent contacts. Additionally we find an ABS signature on the last odd to even transition before depletion.

APPENDIX

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10 A FABRICATION

Ge-Si core-shell nanowires are deposited on a p^{++} doped Si substrate covered with 100 nm SiO_2 and contacted after AFM imaging. Source and drain contacts are defined using EBL and after developing, a 3 second BHF dip is performed to remove native SiO_2 from the Si shell of the nanowire. The contacts are metallised using electron-beam evaporation of Al, resulting in a 150 nm nanowire channel. As a last step, devices are annealed for 10 minutes on a hotplate in ambient at 180°C , during which Al diffuses into the wire, reducing contact resistance from several $\text{M}\Omega$ to several $\text{k}\Omega$.

In total, 7 out of 15 devices tested at low temperature showed a supercurrent with 4 devices showing a superconducting phase with a T_C between 600 and 900 mK, comparable to X1. Three devices showed a metallised nanowire with comparable I_C to device B, we have no information on their respective T_C or B_C .

10 B ADDITIONAL FIGURES

Figure 10 A.1: I_C vs T and B of device A: (a) I_C vs $B_{\parallel}$ vs T , (b) I_C vs $B_{\perp}$ vs T and (c) I_C vs $B_{\star}$ vs T (same as Figure 10.2a in the main text). Configuration I-IV are denoted in all figures in which green indicates superconductivity while red indicates the normal state.

Figure 10 A.2: **Kondo effect at finite magnetic field:** $\partial I_D/\partial V_{SD}$ vs V_{SD} and V_{BG} for $|B| = 2$ T. Odd (O) and even (E) hole occupation of the nanowire quantum dot is indicated. The conductance peak around $V_{SD} = 0$ visible for odd occupation is attributed to the Kondo effect.

Figure 10 A.3: **Hard superconducting gap versus T:** $\partial I_D/\partial V_{SD}$ vs T taken at $V_{BG} = 4.45$ (blue dotted line in Figure 10.3a.). The white dotted line indicates the critical temperature of X1 $T_{C,X1}$

CONCLUSION & OUTLOOK

The experimental results in this thesis illustrate the versatility and high quality of the Ge-Si core-shell nanowire system and demonstrate their suitability for both spin-qubits as well as Majorana fermions.

11.1 NORMAL-STATE DEVICES

A clean electrostatic environment in the form of a smooth potential along the nanowires is the result of an extremely low defect density. In turn, this leads to very high hole mobilities which are enhanced by the intrinsic strain generated by the Ge-Si heterostructure. As we have shown, these properties allow us to create extremely stable and highly tunable quantum dots.

The above is underlined by our Pauli-spin blockade results where the electrostatic environment of a double quantum remains unchanged for a duration of at least several hours and where the hole occupancy in both dots can be independently changed with highly regular charging and orbital energies. The identification of the different spin-flip mechanisms and their relative contribution allows us to carefully select the magnitude and direction of the magnetic and electric fields, which will likely lead to increased spin relaxation times compared to current results (Hu *et al.* 2012). Together with the predicted DRSOI, the experimentally verified tunable g -factor, and the fact that group-IV materials have very low (potentially zero) hyperfine interaction, we possess a promising platform for spin-orbit qubits. A next step would be to apply microwaves to one of the capacitively coupled gates and demonstrate spin rotations using EDSR by lifting spin blockade (Elzerman *et al.* 2004; Pribiag *et al.* 2013). By showing Rabi oscillations and performing a Ramsay experiment, universal spin control can be demonstrated which would result in the first Ge-Si nanowire qubit (Nadj-Perge *et al.* 2010a). Long-range coupling of multiple nanowire devices could be realized using circuit quantum electrodynamics to realize a multiple

qubit setup (Kloeffel *et al.* 2013b), where the anisotropic and electric field-dependent g -factor can be exploited to change EDSR frequencies and address individual qubits.

From a physics point of view, the valence band in the 1-dimensional Ge-Si nanowires is a rich system where the mixing of heavy and light holes depends on the subband population (Csontos *et al.* 2009; Kloeffel *et al.* 2011; Maier *et al.* 2014b). Additionally, in double quantum dots, an anisotropic Coulomb interaction which also sensitively depends on the interdot coupling is observed (Maier *et al.* 2013; Maier *et al.* 2014b) but the mechanism cannot yet be fully explained. To this purpose, it is important that we are able to reach the single-hole regime in these devices. Full control over the dot and subband occupation allows us to observe changes to the described anisotropic effects as holes are added to the system, hopefully revealing more about the underlying mechanisms that cause it.

For the first generation of devices as described in Chapter 3, reaching the single-hole regime has not been possible since the gates couple very locally to the nanowire. When going towards the few-hole regime, the plunger gate therefore induces a tunnel barrier inside the dot, effectively splitting the single dot into an unintentional (not highly tunable) double dot. Preliminary experiments on the second or third generation devices which try to solve this problem by decreasing the gate pitch, suggest this problem still persists. A possible solution in Figure 11.1a presents a modified quantum dot bottom gate design: The wider designated plunger gates now couple to the complete dot defined by the higher lying barriers. This design requires two highly aligned subsequent EBL fabrication steps with in-between ALD of an oxide: one for the plunger gates and another for the barrier gates. On the other hand, the gates do not need to be embedded anymore since the stack of metal and oxide is of approximately equal height below the complete nanowire channel. It would be wise to perform electrostatic simulations on this design to ensure the higher lying gates do not screen the plunger gates and to optimise the relative widths of the plunger and barrier gates.

Ge-Si nanowires could also be of high interest for metrology in the quantized charge pumping community where the main goal is to define a new current standard based on the elementary charge (Blumenthal *et al.* 2007). For this purpose, single-charge tunnelling devices are operated at GHz frequencies, shuttling a single charge through the device for every cycle. This needs to result in a sizeable current in order to

Figure 11.1: (a) Cross-section of a double layer gate design for reaching the single-hole regime. Plunger gates $p1$ and $p1$ are defined first, barrier gates $b1$ - $b5$ are defined in a second fabrication step. (b) Cross-section of a Kitaev chain device design. Electrically floating Al patches on top of the barrier gates extend proximity induced superconductivity in the complete wire.

obtain a high enough accuracy and therefore high switching speeds in combination with low error rates are required. Multiple GHz operation frequencies have been realised, for instance, in Si (Jehl *et al.* 2013; Yamahata *et al.* 2014) and InAs nanowires (D’Hollosy *et al.* 2015). Our extremely stable quantum dots in high-mobility nanowires should result in robust high-frequency operation and low error rates (Kaestner *et al.* 2015). The combination with the large gate structure, as shown in Figure 3.4, allows multiple wires to be controlled in parallel by the same set of gates which, in turn, would increase the measured current by a factor equal to the number of parallel wires.

11.2 SUPERCONDUCTING DEVICES

As we have shown, the ability to reproducibly induce superconductivity in Ge-Si nanowires opens up new possibilities for a wide range of experiments and makes Ge-Si a new interesting system where all ingredients for Majorana fermions are present. However, we find one issue when we look at the magnitude and the anisotropy of our g -factor. Compared to group III/IV materials it is an order of magnitude lower, meaning we need an order of magnitude higher magnetic fields to obtain equal Zeeman splitting. Let us consider the condition for obtaining a topological phase $E_Z > (\Delta_I^2 + \mu^2)^{1/2}$ (Mourik *et al.* 2012) with Δ_I the induced superconducting gap, μ the chemical potential and E_Z the Zeeman splitting. Assuming $\mu = 0$, we see that the Zeeman splitting must at least equal the induced superconducting gap. Letting $\Delta_I = \Delta_{AL} = 212 \mu\text{eV}$ and taking $g = 4$, we require $B = 1.8 \text{ T}$, a field

at which Al is completely quenched. To make matters worse, Majorana fermions require a B field component perpendicular to the spin-orbit field, i.e., along the nanowire where our g -factor is minimal, which means we need even higher effective fields. Furthermore, Δ_I decreases with increasing B , meaning we require a material with a B_C in the direction of the applied field that is much higher than the applied B . Moving to a higher T_C superconductor such as Nb will not solve this particular issue since this increases Δ and the required B proportionally.

Reaching the Majorana regime might still be accomplished by carefully engineering an anisotropic geometry of the superconductor to sustain higher magnetic fields or by using the superconducting Al/Si_x/Ge_y described in Chapter 10, and by making sure the last subband is reached, where the highest g -factors are predicted (Maier *et al.* 2013).

An alternative approach to create Majorana fermions was proposed by Sau *et al.* (2012). Here, a chain of strongly coupled superconducting quantum dots effectively forms a Kitaev chain (Kitaev 2000) and can be tuned to host Majorana fermions that are highly robust against disorder. A strong spin-orbit interaction and a sufficiently high g -factor in the quantum dot host material are still required, but the only demands on the dots are that only one charge resides at the chemical potential and that the dots are sufficiently strongly coupled so that the proximity-induced superconductivity extends through the entire chain. Figure 11.1b shows a proposed slightly modified device design based on the highly-tunable normal-state quantum dots as described in Section 3.2.3. Al patches are placed on regular distances on the nanowire above the barrier gates to extend the superconducting proximity effect throughout the wire. The patches are electronically floating so that they do not screen the effect of the barrier gates. Adding more quantum dots in the chain results in an exponential increase in the robustness of the Majorana fermions against disorder. The design shown allows four quantum dots in series which is reported to be sufficient to compensate for most disorder-induced decoherence.

11.3 SCALABILITY

A subject that is mostly of less concern to the fundamental physicist is scalability, i.e., taking the scientific concepts out of the laboratory and integrating them into a more complex device that can be routinely fabricated in an automated way for use in an application or

product. Scalability is a major challenge for devices based on bottom-up grown nanowires, especially considering quantum computation applications, where many wires must be integrated on a single chip in very close proximity. Nanowires need to be grown while free standing because of the required interaction with gases (Wagner *et al.* 1964), which means that there are always additional fabrication steps needed to transfer these wires to a measurement substrate. A first step towards a more scalable process for nanowires has been realised by growing InAs (Mandl *et al.* 2011), InP and GaP nanowires (Pierret *et al.* 2010) from a regularly spaced pre-patterned grid of nucleation sites on the growth substrate. This idea has been developed further to grow wires on the slopes of small trenches resulting in nanocrosses and hastags with in-situ grown metal layers (Gazibegovic *et al.* 2017), highly relevant for Majorana braiding operations. However, to apply this technique to the much thinner Ge-Si wires poses challenges. First, three different crystal directions have been identified to grow randomly while only the $[110]$ direction results in the desired defect-free wires. Second, multiple wires are often seen to grow from a single nucleation site and seldom grow straight up which makes a systematic approach to harvest these wires more difficult. Even if these issues could be solved, automated harvesting and depositing 25 nm diameter wires with nanometer accuracy using purely non-invasive techniques would pose the next difficulty.

For now, most importantly, these wires serve as an ideal test bed to explore the physics involved which is closest to the heart of the experimental scientist. Additionally, they may very well yield results that can be applied to more scalable systems when the time comes.

11.4 FINAL WORDS

Ge-Si nanowires are a highly versatile and rich physical system where many aspects are still unexplored. From the existing literature, combined with the results in this thesis, it might seem that both spin-qubits and Majorana fermions are just the next logical step to be realised in this system. Still, research efforts are limited to a few groups worldwide and even reproducing many of the existing results has not proven trivial. One reason is that Ge-Si nanowires are notoriously difficult to work with during fabrication due to their very small diameters, especially during wire deposition. Luck and statistics therefore still play an important role in obtaining a successful device, especially for the bottom gated devices, where every single nanowire deposition requires

a few hours of labour operating at the limits of both the optical microscope as well as the operators ocular capabilities. Together with the fact that there is no reliable method for selecting the lowest diameter wires for deposition, finding a 'hero' device can take a significant amount of time.

Limited availability of Ge-Si nanowires is also a major factor and to our knowledge, new high-quality wires have not been fabricated in the last few years, even though there is considerable interest. Because of this, not all parties have access to the highest quality wires and are forced to work with inferior ones.

Having said that, I strongly believe that follow-up research will result in many more beautiful results with these wires, and if there will ever be a realistic approach to a scalable fabrication technique, Ge-Si nanowires might even see an application in quantum computing.

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SUMMARY

All information in classical computers is processed with discrete bits which can be either '0' or '1', which are physically represented by macroscopic currents and charges. Quantum computers work in a fundamentally different way and employs quantum mechanical bits, or *qubits*, as their smallest building blocks. A qubit must consist of a quantum mechanical two-level system which can be fully and coherently controlled. For instance, for an electron spin qubit, the spin of a single electron represents the two-level system, and full control over the position and spin state of the electron is required. Qubits are in a superposition of their two eigenstates $|0\rangle$ and $|1\rangle$ which must be mapped to physical properties (for instance spin-up or spin-down). For n qubits, a quantum computer can therefore be in 2^n states simultaneously. This property can be exploited using specific quantum algorithms for which problems scale differently with increasing complexity and can thus be solved in a completely different timescale than on a classical computer.

The quest for a quantum computer is a major research effort, since it requires an unprecedented level of control on a nanoscale and it was only fifteen years ago when the first single qubits appeared. Nowadays, qubits have been realised in a large variety of systems but it is yet unclear which will be used for the first universal quantum computer.

We investigate Ge-Si core-shell nanowires as a possible candidate for spin qubits and Majorana qubits. Ge-Si nanowires are hole conductors and for the spin qubits, we localise these holes in the nanowires to create quantum dots. The 1-dimensional nanowires provide confinement in the radial direction, while gates oriented perpendicular to the nanowire provide confinement in the direction of transport. This allows us to 'trap' the holes in specific segments of the nanowire and investigate the effect of the electrostatic potential and magnetic fields. For the Majorana zero modes we use superconducting contacts to induce a Josephson current in the nanowires and investigate the effects of the charge occupancy, magnetic field, microwave field and temperature.

Ge-Si nanowires exhibit highly promising properties: (1) A free hole gas exists in the Ge core without applying external potentials or dop-

ing, (2) they consist of group IV materials with potentially zero hyperfine interaction providing long coherence times, (3) they have a predicted enhanced Rashba type direct spin-orbit coupling, a crucial ingredient for Majorana zero modes and enables spin rotations with AC electrical fields, (4) they have a tunable, electric field dependent, g -factor, (5) they can be grown with virtually no defects on lengths scales relevant for qubits, (6) the heavy-hole light-hole mixing in the valence band edge is an interesting unexplored physical system.

To characterise the quality of the nanowires we determine the hole mobility in the linear regime of operation of nanowire field-effect transistors (FETs) with a wide variety of nanowire diameters. By performing statistical averaging on the measurement data, we obtain well defined mobility values, at 300 K and 4 K. A clear trend is observed between crystal direction and wire diameter where we see higher mobilities for smaller wire diameters. We furthermore find that the small-diameter nanowires grow predominantly in the $[110]$ crystal direction, which corresponds well to a transmission electron microscopy (TEM) study showing that $[110]$ wires have extremely low defect densities. It is therefore important to select small diameter nanowires for further experiments.

To fabricate quantum dot devices, we deposit nanowires on pre-fabricated bottom-gates and demonstrate the ability to make highly tunable quantum dots of varying lengths between 60 nm and 460 nm determined by the pitch of our gate design. Next, we show a double quantum dot with tunable interdot coupling strength and show that the dot occupancies can be changed with little effect to the tunnel couplings due to negligible gate cross-coupling. As a result, we obtain a set of highly regular bias triangles which underlines the stability of the device.

Before continuing on double quantum dots, we return to a single dot where we use excited state magneto-spectroscopy to measure the Zeeman splitting and map the g -factor on three 360° planes on the three main axis with respect to the nanowire. We find that the g -factor is highly anisotropic and depends on the electric field, as predicted by our collaborators. We obtain the highest g -factor when the magnetic field is oriented perpendicular to both the nanowire and the electric field, while the g -factor is almost completely quenched when the magnetic field is oriented parallel to the nanowire. The predicted high spin-orbit coupling of holes in our nanowires enables the use of AC electric fields to perform spin rotations on the Zeeman-split levels. The ability

to change the g -factor using local electric fields from gates, can thus be utilised to tune quantum dots in and out of resonance of a constant high frequency electric field. This makes quantum dots separately addressable, which is an important step for quantum computation applications.

We then return to the double quantum dot configuration and we analyse a highly regular set of bias triangles. By comparing the energy difference between bias triangle pairs, we observe orbital shell filling and extract the charging and orbital energy. We observe Pauli spin blockade (PSB) in charge transitions in the double dot, when transport takes place from partially filled orbitals towards filled orbitals. PSB is therefore only observed in the corresponding bias direction and we observe a clear increase in tunnel current when the bias direction is reversed, corresponding to the non-blocked case. Next, we investigate the finite leakage current in PSB and we test against three well-known spin-flip mechanisms: hyperfine interaction, spin-flip cotunnelling and spin-orbit interaction. By performing a magnetospectroscopy versus detuning on the three main axes with respect to the nanowire, we find that the leakage current is highly anisotropic with respect to the magnetic field direction and that spin-flip cotunnelling, as well as spin-orbit interaction give a significant contribution. It is therefore possible to carefully choose the magnitude and direction of the applied magnetic, corresponding to the longest spin-relaxation time.

Surprisingly, we now find the highest g -factor when the magnetic field is *parallel* to the electric field, which contrasts our result for the single quantum dot where the g -factor was the largest when the magnetic field is oriented perpendicular to both the nanowire and the electric field. This difference is partly explained by considering the higher subband population in the case of our double quantum dot, which significantly changes the light-hole heavy-hole mixing.

By using aluminium as superconducting contacts, a Josephson current can be induced in Ge-Si nanowires. A straightforward annealing process is employed to obtain the required highly transparent superconducting contacts. We observe multiple Andreev reflection (MAR) up to the sixth order and show that they follow a BCS temperature dependence, as they are directly related to the superconducting gap of our Al. Upon irradiating our junction with microwaves, Shapiro steps are observed, which is a manifestation of the AC Josephson effect and the height of the steps is equal to the energy of the applied microwaves. Increasing the microwave amplitude ultimately results in 23 Shapiro

steps. This clear demonstration of both the DC and the AC Josephson effect is proof that our nanowire device with superconducting contacts is indeed a Josephson junction.

Next, the backgate dependence of the Josephson junction is investigated. For decreasing backgate voltages, we observe a general trend of increasing switching current, while our $I_C R_N$ product remains constant and assumes a near-ideal value that is equal to the superconducting gap of our Al. For positive backgate voltages, near depletion, we find a strongly coupled quantum dot in the few-hole regime, where we observe finite supercurrents on the Coulomb diamond crossings, i. e., through the single-particle levels. This result can be used to construct a Kitaev chain of strongly coupled superconducting quantum dots, a system that supports highly robust Majorana fermions.

A magnetospectroscopy of the Josephson junction reveals a second superconducting phase being present inside, or in close proximity to, the nanowire. This superconducting phase has a critical magnetic field much higher than that of Al, while its critical temperature is lower. Our hypothesis is that during annealing, Al diffuses and forms a superconducting Al/Si_x/Ge_y alloy inside the wire. This is also suggested by a TEM study where Al was found inside the nanowire channel. In another device, we find that Al has diffused into the whole nanowire channel which has become metallic and superconducting, with a critical temperature and critical field a few times higher than that of pure Al. This superconducting alloy is an interesting material for applications requiring the combination of superconductivity with high magnetic fields, such as Majorana fermions. Near depletion, a hard superconducting gap is observed, which indicates highly homogeneous interfaces between nanowire and superconductor and we believe this interface is formed between pure Al and the sudden transition to the superconducting alloy. The hardness of the gap in our device is at least comparable to epitaxially grown Al on group III-V material wires. The hardness of the gap is a measure for number of quasiparticle states in the gap, a major source of decoherence for Majorana zero modes.

The results for both the normal-state quantum dot devices and the superconducting devices, illustrate the versatility and suitability of Ge-Si nanowires for quantum computation applications.

SAMENVATTING

Alle informatie in klassieke computers wordt verwerkt met discrete bits welke de waarden '0' of '1' kunnen aannemen en fysiek gerepresenteerd door macroscopische stromen en ladingen. Kwantumcomputers functioneren op een fundamenteel andere wijze en gebruiken hiervoor als bouwstenen kwantummechanische bits, oftewel *qubits*. Een qubit moet bestaan uit een kwantummechanisch systeem met twee niveaus waarbij de staat op een coherente manier kunnen worden gecontroleerd. In het voorbeeld van een elektron spin-qubit worden de twee niveaus vertegenwoordigd door de spin toestand. Hierbij moet er sprake zijn van volledige controle over de positie van het elektron en zijn spintoestand. Qubits bevinden zich in een superpositie van hun twee eigentoestanden $|0\rangle$ en $|1\rangle$ welke een fysieke representatie moeten hebben (bijvoorbeeld spin-up en spin-down voor een spin-qubit). Voor n qubits kan een kwantumcomputer zich daarom in een superpositie van 2^n staten tegelijk bevinden. Deze eigenschap kan worden gebruikt door specifiek ontworpen kwantum algoritmen waarbij problemen anders schalen bij toenemende complexiteit en deze problemen kunnen daarom worden opgelost in een volledig andere tijdsschaal dan wanneer een klassieke computer gebruikt zou worden.

Het streven naar een kwantumcomputer vereist veel wetenschappelijke inzet omdat het een zeer grote mate van controle op de nanoschaal vereist: slechts vijftien jaar geleden verschenen de eerste enkele qubits. Tegenwoordig zijn er qubits gerealiseerd in een grote verscheidenheid aan materialen en systemen, maar het is vooralsnog onduidelijk hoe de eerste universele kwantumcomputer gaat worden gerealiseerd.

Ge-Si kern-schil nanodraden worden onderzocht als mogelijke kandidaten voor spin qubits en Majorana qubits. Deze nanodraden zijn gaten geleiders en voor de spin qubits lokaliseren we de gaten in de nanodraden om kwantum dots te maken. Deze 1-dimensionale nanodraden zorgen voor bewegingsbegrenzing van de gaten in de radiale richting terwijl opsluiting in de transportrichting wordt bewerkstelligd door middel van gates die loodrecht georiënteerd zijn ten opzichte van de nanodraad. Op deze manier kunnen we de gaten 'vangen' in specifieke segmenten van de draad en kunnen we het effect van de elektrostatische potentiaal en magnetische velden. Voor het onderzoek richting de

Majorana fermionen gebruiken we supergeleidende kontakten om een Josephson stroom te induceren om vervolgens onderzoek te doen naar het effect van de ladingsbezetting, magnetische velden, microgolfvelden en temperatuur.

Ge-Si nanodraden hebben veelbelovende eigenschappen: (1) Er bevindt zich een vrij gaten-gas in de Ge kern zonder externe elektrostatische potentiaal of doping, (2) ze bestaan uit groep-IV materialen met de mogelijkheid tot het elimineren van de hyperfijninteractie wat resulteert in langere spin-coherentietijden, (3) voorspeld is dat ze beschikken over een versterkt Rashba spin-baaneffect, een cruciaal ingrediënt voor Majorana fermionen en het uitvoeren van spin rotaties door middel van AC elektrische velden, (4) ze hebben een tunable, elektrisch veld afhankelijke g -factor, (5) ze kunnen worden gegroeid met nagenoeg nul defecten binnen de relevante lengteschalen voor qubits en (6) het mixen van de lichte en zware gaten in de valentieband is een interessant, vrijwel onverkend fysisch systeem.

Om de kwaliteit van de nanodraden te karakteriseren bepalen we de gatenmobiliteit in het lineaire regime van een nanodraad field-effect transistor (FET) voor uiteenlopende draaddiameters. Door een statistische middelingsprocedure verkrijgen we goed gedefinieerde mobiliteitswaarden bij zowel 300 K als 4 K. Er is een duidelijke relatie zichtbaar tussen de kristalrichting en de draaddiameter, waar hogere mobiliteiten worden gemeten voor kleine diameters. Daarnaast zien we dat de draden met kleine diameter voornamelijk in de $[110]$ kristalrichting groeien en dat deze tevens een extreem lage dichtheid aan defecten hebben. Het is daarom van groot belang om draden met een kleine diameter te selecteren voor verdere experimenten.

Om kwantum dot devices te fabriceren deponeren we nanodraden op voor-gefabriceerde bottom-gates en hiermee laten we zien dat we zeer-tunable dots kunnen maken van variabele lengten van 60 nm tot 460 nm. Deze lengte wordt mede bepaald door de afstand tussen de gates. Vervolgens laten we een dubbele kwantum dot zien met tunable interdot koppelingen, waarbij de we de ladingsbezetting in beide dots kunnen veranderen zonder deze interdot koppeling te beïnvloeden, doordat de gates zeer weinig onderlinge koppeling hebben. Mede hierdoor verkrijgen we een set bias-driehoeken wat de stabiliteit van het device benadrukt.

Voordat we verdergaan met dubbele kwantum dots, keren we terug naar een enkele kwantum dot en meten we via een aangeslagen toe-

stand magneto-spectroscopie de Zeeman splitsing en brengen we de g -factor in kaart in drie 360° graden vlakken op de drie hoofdassen met betrekking tot de nanodraad. We observeren dat de g -factor zeer anisotroop is en tevens afhangt van het elektrische veld, zoals voorspeld door onze collega wetenschappers. We observeren de hoogste g -factor wanneer het magnetisch veld loodrecht georiënteerd is ten opzichte van zowel de draad als het elektrische veld, terwijl de g -factor bijna volledig wordt onderdrukt wanneer het magnetisch veld parallel aan de draad is georiënteerd. De voorspelde hoge spin-baan koppeling voor gaten maakt het mogelijk om met behulp van hoogfrequente AC elektrische velden spin rotaties uit te voeren op de Zeeman-gesplitste niveaus. De mogelijkheid de g -factor te veranderen met lokale elektrische velden kan daarom gebruikt worden om kwantum dots in- en uit-resonantie te tunen bij een constant microgolf veld. Hierdoor kunnen individuele kwantum dots geadresseerd worden, wat een belangrijke stap is in de richting van een kwantumcomputer.

We keren nu terug naar de configuratie van de dubbele kwantum dot en we analyseren een set van zeer regelmatige bias driehoeken. We zien hoe orbitalen van de dot gevuld worden en we leiden de charging energie en de orbitale energie af. Tevens zien we Pauli-spin blokkade PSB bij ladingsovergangen in de dubbele dot bij transport van een gedeeltelijk gevulde orbitaal naar een gevulde orbitaal. PSB is daarom alleen te zien in de bijbehorende bias richting en bij het omkeren van de bias richting zien we een significante toename in de gemeten stroom, in overeenstemming met de niet-geblokkeerde toestand. Vervolgens onderzoeken we de eindige lekstroom in PSB en zoeken we naar drie bekende spin-flip mechanismen: hyperfijn interactie, spin-flip cotunnelling en spin-baan interactie. Door middel van een magneto-spectroscopie versus detuning op de drie hoofdassen van de nanodraad zien we dat de lekstroom zeer anisotroop is ten opzichte van het magnetisch veld en dat spin-flip cotunnelling en spin-baan interactie de voornaamste bijdrage leveren. Het is hierdoor mogelijk om zorgvuldig de grootte en richting van het magnetisch veld te kiezen welke overeenkomt met de langste spin-relaxatie tijd.

Verrassenderwijs zien we dat de g -factor nu het hoogst is wanneer het magnetisch veld *parallel* is aan het elektrische veld, wat contrasteert met het resultaat voor de enkele kwantum dot wanneer de g -factor het grootst was wanneer het magnetisch veld loodrecht is georiënteerd aan zowel de nanodraad als het elektrische veld. Dit verschil kan deels worden verklaard door de significante verandering in de menging van lichte en zware gaten bij populatie van hogere subbanden.

Door het gebruik van aluminium als supergeleidende contacten, kan een Josephson stroom worden geïnduceerd in Ge-Si nanodraden. We gebruiken een relatief eenvoudige anneal methode om zeer transparante supergeleidende contacten te verkrijgen. We zien multiple Andreev reflection (MAR) tot de zesde orde en laten zien dat deze een Bardeen-Cooper-Schrieffer (BCS) temperatuurafhankelijkheid volgen aangezien ze direct gerelateerd zijn aan de supergeleidende gap van het aluminium. Wanneer we een microgolf veld aanbieden zien we Shapiro stappen, een direct gevolg van het AC Josephson effect. De hoogte van de stappen komt overeen met de energie van de aangeboden microgolven en door het verhogen van de amplitude zien we uiteindelijk 23 Shapiro stappen. Deze duidelijke demonstratie van zowel het DC als het AC Josephson effect is bewijs dat ons nanodraad-device daadwerkelijk een Josephson junctie is.

Hierna onderzoeken we de backgate afhankelijkheid van de nanodraad Josephson junctie. Voor afnemende backgate voltages zien we een trend van toenemende kritische stroom terwijl het $I_C R_N$ product constant blijft op een bijna ideale waarde die gelijk is aan de supergeleidende gap van ons Al. Voor positieve backgate voltages in de buurt van depletie vinden we een sterk gekoppelde kwantum dot in het enkele-gaten regime. In deze dot zien we eindige superstromen door de kruising van de Coulomb diamanten, i. e., door single-particle niveaus. Dit resultaat kan gebruikt worden om een Kitaev ketting van sterk gekoppelde kwantum dots te creëren, een systeem waarin op een zeer robuuste manier Majorana fermionen kunnen worden gerealiseerd.

Een magnetospectroscopie van de Josephson junctie onthult een tweede supergeleidende fase die zich binnen, of vlakbij de nanodraad bevindt. Deze supergeleidende fase heeft een kritisch magnetisch veld dat veel hoger is dan van Al terwijl de kritische temperatuur lager is. Onze hypothese is dat Al diffundeert tijdens de annealing procedure en daarbij een supergeleidende Al/Si_x/Ge_y legering vormt. Dit wordt ondersteund door een transmission electron microscopy (TEM) studie waarbij Al is aangetroffen binnen de nanodraad. In een ander device zien we dat Al door het gehele kanaal is gediffundeerd wat resulteert in een supergeleidend metallisch kanaal met een kritisch veld en kritische temperatuur die veel hoger zijn dan voor puur Al. Deze supergeleidende legering is een interessant materiaal voor toepassingen die zowel supergeleiding als hoge magnetische velden vereisen, zoals Majorana fermionen. Vlakbij depletie nemen we een zeer harde geïnduceerde supergeleidende gap waar, wat op een zeer homogene interface tussen supergeleider en nanodraad duidt. Het is waarschijnlijk dat dit

interface zich tussen puur Al en de supergeleidende legering bevindt. De hardheid van de gap in ons device is ten minste vergelijkbaar met epitaxiaal gegroeid Al op groep III-V draden. De hardheid van de gap is een maat voor de hoeveelheid quasiparticle states in de gap, een zeer belangrijke bron die zorgt voor decoherentie van Majorana fermionen.

De resultaten in zowel de normale-staat devices evenals in de supergeleidende devices, benadrukken de veelzijdigheid van Ge-Si en laat zien de ze een geschikte kandidaat zijn voor toepassingen met betrekking tot kwantum berekeningen.

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*Time you enjoy wasting
is not wasted time.*

- Marthe Troly-Curtin