HOLE SPINS

IN GE-SI NANOWIRES

MATTHIAS BRAUNS

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DISSERTATION

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Il faut avoir l'esprit dur et le cœur tendre.

— Sophie Scholl

Dedicated to the good-hearted.

ABSTRACT

In a universal quantum computer, coherent control over the state of a quantum mechanical two-level system is needed. This requires interactions of the quantum state with its environment. Inherently, such interactions also lead to decoherence and thus limit the performance of the quantum computer. A profound knowledge of the relevant interaction mechanisms is therefore key to the realization of a quantum computer.

In this thesis we use Ge-Si core-shell nanowires to investigate holes confined to one dimension. Mixing of heavy and light hole states leads to a strong, anisotropic spin-orbit interaction in this system. We define highly stable quantum dots of different lengths in the nanowire and controllably split up longer quantum dots into double quantum dots. The effective g -factor in these one-dimensional hole quantum dots is found to be highly anisotropic with respect to the nanowire axis as well as the electric-field axis. In double quantum dots, we observe shell filling of new orbitals and Pauli spin blockade of the second hole entering the orbital. The leakage current in the spin-blocked state is highly anisotropic with spin-flip cotunnelling as the dominant leakage mechanism. At finite magnetic fields, we also find signatures of leakage current induced by spin-orbit coupling and anisotropic Coulomb effects.

*Let us think the unthinkable,
let us do the undoable,
let us prepare to grapple with the ineffable itself,
and see if we may not eff it after all.*

— Douglas Adams

PREFACE

At the time I decided to study physics, in 2006, the hype of the solar-cell industry in Germany was at its peak. Q-Cells was the largest solar-cell producer in the world, a multi-billion dollar company just around the corner of my home town. This was not the only reason for me to study physics, but I was fascinated by how such interesting stuff can also be of social and environmental value. In 2012, Q-Cells was bankrupt. What is left is the fascination of how sunlight becomes electricity. Directly, elegantly, no 19th-century-style steam turbine. In other words, my focus shifted from ‘What can we make of it?’ to ‘How does it work?’ Or in the timeless words of von Goethe (1808):

*Daß ich erkenne, was die Welt
Im Innersten zusammenhält,*

*That I may detect the inmost force
Which binds the world, and guides its course;*

Although I still acknowledge the impact of science on the world we live in, I do not consider it wise to let the hope for this impact *drive* scientific research. Trying to plan scientific research is a *contradictio in terminis* that prevents truly new knowledge coming from the elements of chaos and surprise that so often in history have lead to scientific breakthroughs.

After finishing my studies and becoming a ‘Dipl.-Phys.’, going for a PhD was the logical next step. It was the perfect opportunity to perform hands-on research on one topic over an extensive period of time, dig into the subject until the very forefront. Coming from a diploma-thesis research on a quite applied topic on metal physics in a Fraunhofer Institute, I wanted to go for the real thing. Fundamental research in quantum electronics seemed the perfect opportunity. And it was. Of course the research I did during these four years ended up being not

quite what we initially planned to do. I did not even know about nanowires when I started my PhD in 2011. But this is the beauty of it: a PhD does not have to work out as planned. It probably even should not. Is doing a PhD worth the time? Hell, yeah!

This conclusion is of course only possible because of numerous people who *make* it worth the time. So let me try to sprinkle some kind words for all these people. First of all, I want to thank *Wilfred van der Wiel* for not only giving me the opportunity to work on my PhD in his research group, but also for hinting me at the just-opened position in the first place. Furthermore, he taught me right at the beginning that the Dutch equivalent to “Sehr geehrter Herr Prof. van der Wiel” is “Hoi Wilfred”.

I want to thank *Floris Zwanenburg*. He is an extremely kind and dedicated supervisor who always encouraged me to develop my own ideas of how to tackle a problem and in which way to interpret data, while also always challenging me to stay critical of these interpretations, to rule out possible flaws, and also to write manuscripts avoiding endless run-on sentences like this one. Would I recommend him as a boss? Definitely. At least as importantly: He is really good at organising the defence in football.

Switching to nanowires soon also gave me a partner in crime: *Joost Ridderbos*. We spent countless hours together fabricating, failing, fabricating again, making a stupid mistake, drinking beer, fabricating again, measuring, blowing up devices, laughing cynically, fabricating, and finally succeeding. This last one was only possible, because I could rely on Joost, the EBL-guru, with his superior degree of organisation and immense patience. I was at the top of the list in his phone, ahead of his girlfriend. Says enough. Thanks, Joost!

Another reason why working was always fun is the rest of the *Silicon Team*. *Filipp*, thanks a lot for helping me with the first steps around the university in the beginning, the VriMi-borrel times, and of course the conference extensions in the snow as well as in the sun. *Chris*, our master programmer, thanks for bearing with my ‘it-works-for-me’ programming attitude, keeping up the paper discussions, and the countless beer times at the university, in Enschede, and beyond. *Sergey*, thank you especially for the evening discussions about new ideas for devices and all kinds of other stuff, and your healthy Russian hands-on mentality regarding equipment.

Research would be even more difficult without people who fix stuff that is broken, show people how to use equipment, and in general make sure that a lab is running smoothly. So, *Johnny, Martin, and Thijs*, thank you! Also big thanks to *Karen*, who made sure all the administration is taken care of even with people like me whose motivation for such stuff is rather limited.

Apart from these people semi-permanently present throughout my time in NanoElectronics, I had the pleasure to (co-)supervise a number of bachelor and master students. So thanks to *Anne, Joost van der Hoff, Gertjan, Yizhen, Joren, and Agung*.

The *NanoElectronics* group was my professional family throughout the last years. So thanks to all of of you for the fun times during NEvents, lunch breaks, journal clubs, the occasional chit-chat in the coffee corner and all the other stuff that makes sure research is not just somebody staring at a machine or a monitor. Although not an official NE member according to the website, I am also deeply indebted to the *NE espresso machine*, which saved my day on countless occasions.

The cleanroom was my part-time home for most of the time I was working on my thesis. Luckily, the cleanroom has a *staff* that tries to make sure impatient people like me can do their stuff. So, thanks a lot to all of you, especially *Hans* who sometimes even beat his optimistic promises for ALD runs, while keeping up his mood with only brief moments of irritation when I forgot my BAK reservation again.

I had the honour of sharing an office with *Elmer* and *Chris*. We were the first ones to have a couch in our office, the only ones ever to throw an office party. We even gave shelter to one of the plants in the coffee corner and still had enough space for a crate of Club Mate. Best office ever. Fact. *Chris*, you did your best as our music executive, and you did well. Also thanks for always making sure that we never ran out of half-broken electronics you found somewhere. *Elmer*, a salute to you for always being in for a coffee, a *Feierabendbier*, or a round of 9gag, for being really bad at saying 'no', even if asked for a translation of my summary into Dutch, and generally for being the aged sunshine you are. I also want to thank all the refugees hiding from the world (or work) on our couch. *Joost and Ksenia*, our regulars, thanks for always bringing exciting news and the latest gossip from the scary world outside our cozy office!

Ksenia, apart from sitting on our couch, thank you for the good times in bars or in your tequila-equipped living room, for sushi dinners in Carré, the Saturday lunches at the Extrablatt, and generally for being around and the good girl you are.

It took me about one and a half years, but then I started playing football again. I had the honour to be the captain of the freshly-founded *Nanoelectrics futsal team*, thanks to all of you guys! Since I needed the distraction, I was always eager to jump in if another team needed a player, so thanks to all the people I had the pleasure to receive some beautiful assists from. I even had the privilege to become a regular member of *Pi Hard* and play some glorious matches with them.

The fact that I also played for v. v. Drienerlo on Saturdays is to large extent the fault of *Bijoy*. He persuaded me to join *Veld 4* and fight some epic battles as a central midfielder (and even centre-back in times of despair). *Bijoy* was the team's captain, its mother, father, all at once. So, in a way, it was also his fault that I got to know disproportionately many mathematicians at the UT. I played football with *Bijoy*, *Edo*, *Milos*, and *Felix* for Drienerlo and I was lured into their circle somehow. Through them I also got to know the rest of the mathematics-related crowd. We had great times in the pubs, Sunday brunches at Extrablatt, playing cards at Edo's place, spending New Year's Eve in the legendary house of Felix in Scharnstein, or just hanging out in the Volkspark. So many thanks to *Bettina*, *Bijoy*, *Daniela*, *Edo*, *Felix*, *Gijs*, *Ivana*, *Koen*, *Laura*, *Mihaela*, *Milos*, *Paolo*, and *Wilbert*.

There are many people I met under various circumstances that made my life here in Enschede what it was. So thanks also to *Xenia*, *Corina*, *AnKa*, *Lina*, *Céline*, *Andras*, *Fritzi*, *Julie*, *Jasmin*, and *Dominik*.

Despite making a lot of new friends, I am also lucky enough to still have some old ones aus dem guten alten Bernburg. *Marcus*, was würde ich nur ohne deine manchmal endlosen Fragen über die Grundfesten der Physik machen? Zwei Wochen lang in einem Auto durch die neuseeländische Pampa fahren, das würde ich nicht mit vielen aushalten. Mit dir war es mir eine helle Freude. *Lydia*, deine moralische Unterstützung, wenn gerade mal wieder alles genervt hat, vergess ich dir nicht, genauso wenig, dass du nicht schreiend weggerannt bist als ich auf dem letzten Drücker noch mit meinem Cover angekrochen kam. *Caro Falke*, vielen Dank für Berlin für Insider und das gemeinsame Rumreisen, war cool! *Franz und Karo*, der Bodensee hat nur wegen euch so

viel Spaß gemacht! *Nadja*, danke für die Gastfreundschaft in Straßburg, *Guru und Collie*, *Ali*, *Calli und André*, *Paul*, und *Armin*, selbst nur ein oder zwei Mal im Jahr sehen fühlt sich an wie zu Hause.

Zum Schluss möchte ich noch meiner Familie danken. *Jana und Basti*, danke, dass ihr öfter den Weg nach Holland gefunden habt und auch sonst immer da wart. *Emil*, dem Jüngsten, darf ich zumindest indirekt für die vielen wunderbaren Whatsapp-Fotos danken, die mich täglich erreichen. Wenn du 18 bist, mach ich dir ein Album draus. *Mama und Papa*, ich bin euch ewig dankbar für die Unterstützung über all die Jahre, egal ob ich mal wieder körperlich außer Gefecht gesetzt war, oder zwar zu Besuch kam, aber dank eines furchtbaren Arbeitsrhythmus nicht wirklich ansprechbar war.

Matthias Brauns
March 2016

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ACRONYMS

AFM	atomic-force microscopy
ALD	atomic layer deposition
CPF	copper powder filter
DC	direct current
DOS	density of states
EBL	electron-beam lithography
FET	field-effect transistor
PSB	Pauli spin blockade
SEM	scanning electron microscopy
TEM	transmission electron microscopy
VLS	vapour-liquid-solid

INTRODUCTION

1.1 A MOTIVATION

We live in a classical world. Our brain is a wet chunk of organic material at a temperature above 300 K, and so is most of the material that surrounds us. We are too big, too warm, too slow, and too messy to experience quantum mechanics at work. And yet quantum mechanics is currently a gigantic threat to the single device that has turned our world upside-down within the past fifty years: the transistor.

Miniaturisation of transistors as the driving force behind digital information technology has torn apart old and built new industries, and vastly changed our lives on a scale that is inexplicable in mere words. Current semiconductor technology approaches a fundamental barrier for further miniaturization: the size of an atom. Transistors are already small enough to allow quantum mechanical effects such as quantum tunnelling of electrons to disturb their performance, which forces the semiconductor industry to play expensive tricks like FinFET technology or high- k dielectric materials to allow for further gain of performance (Waldrop, 2016). It seems that the semiconductor industry still has some more tricks up its sleeve to ensure further gain of performance for another decade or two. While this certainly is a reassuring thought, it does not forbid us to ask a fundamental question: Instead of cursing it, why not embrace quantum mechanics?

Quantum Computing

In fact, this thesis is far from being the first place where this question has been posed. Already in the 1980s, Feynman (1982, 1986) proposed a quantum computer. It makes use of a quantum mechanical two-level system (like the spin of an electron), which forms a *quantum bit* (qubit) as the equivalent of the classical bit in classical computer technology. The qubit has two eigenstates, the actual state of the qubit can be one of the two eigenstates or any quantum superposition of the two. These superposition states of the qubit *do not* have an equivalent in a classical bit, and are therefore the ‘magic ingredient’ that makes a quantum computer so much more powerful at certain tasks than a classical computer.

Soon after the proposition of the quantum computer, Deutsch (1985) proposed a first academic example of a quantum algorithm, which can be seen as the start for the ever-since growing field of theoretical quantum computation. To date, the probably most famous examples of quantum algorithms that reflect a significant speed-up compared to classical algorithms are the Shor algorithm for number factorization (Shor, 1997), and the Grover search algorithm (Grover, 1997). For readers interested in a thorough yet entertaining introduction to the theory of quantum computing I can warmly recommend ‘Quantum computing since Democritus’ by Aaronson (2013).

Especially Shor’s algorithm has generated a significant amount of interest in quantum computing, since it can efficiently find the prime factors of a given large number, which is exactly what current public-key data encryption schemes rely on. A much more substantial role future quantum computers could play is often neglected in public discussions: the simulation of quantum systems. Because quantum computers *are* quantum mechanical systems, they are way more efficient than classical computers at simulating other quantum mechanical systems such as organic molecules. This could be a real game changer e.g. in the process of designing new pharmaceutical drugs.

Physical implementation

Over the past decades, numerous implementations of qubits in very different systems have been shown to work. Early experiments include cold-ion traps (Cirac and Zoller, 1995), circuit quantum electrodynamics systems (Turchette et al., 1995), liquid-phase nuclear spins (Chuang et al., 1998), and atoms in optical lattices (Brennen et al., 1999). Although these experiments pioneered the field, their limited scalability is a major disadvantage for building a useful quantum computer.

Therefore, an increased amount of research has been focussed on solid-state systems in the last twenty years. Notable examples here are superconducting circuits (Moore et al., 1999) and spins in semiconductors, either as electron spins in electrostatically defined quantum dots as proposed by Loss and DiVincenzo (1998) or nuclear spins of dopant atoms (Kane, 1998). Especially the last two possibilities are technologically interesting because they are possibly compatible with current CMOS technology. This has two advantages: the immense experience in semiconductor industry can be used for scaling-up, and, maybe even more importantly, such quantum computers would be easy to integrate

with classical computing circuits, which is essential, because most quantum algorithms still rely on a significant overhead of classical computation.

Spin qubits

Within the realm of spin qubits in semiconductors, pioneering work has focussed on III-V heterostructures like GaAs, where Tarucha et al. (1996) used a mesa-like structure to define a few-electron quantum dot and showed atom-like shell filling. Experiments on qubit initialization and spin readout (Elzerman et al., 2004), and coherent control of spin states (Petta et al., 2005; Koppens et al., 2006) followed. Despite this progress, there is one unavoidable limiting factor for all III-V systems: nuclear spins. The net nuclear spin of the atoms in the semiconductor lattice has a random orientation and slowly fluctuates, which limits the coherence time of the qubit spin state via the hyperfine interaction between electrons and nuclei (Fermi, 1930).

Group-IV semiconductors like Si and Ge have predominantly stable isotopes with zero net nuclear spin, which limits the hyperfine interaction even in natural abundances of these material, and poses the possibility of isotope-enrichment to zero nuclear spins in the material (Itoh et al., 1993, 2003). The research towards group-IV-based spin qubits is comparably young, with the first single-electron and single-hole occupation of quantum dots reported less than ten years ago (Simmons et al., 2007; Zwanenburg et al., 2009b). After these first results, the field quickly took off and single-shot read-out of a donor-bound electron spin (Morello et al., 2010), Pauli spin blockade (Borselli et al., 2011), and Rabi oscillations of singlet-triplet states (Maune et al., 2012) have been reported in quick succession. What is possible with isotopical engineering of the Si host material has recently been shown by reports on a quantum dot-bound electron qubit with a coherence time of 28 ms (Veldhorst et al., 2014), a nuclear spin qubit with a spin coherence time of more than 30 s (Muhonen et al., 2014), and a two-qubit logic gate (Veldhorst et al., 2015).

Nanowires

An interesting alternative to these gate defined quantum dots within a planar Si wafer is starting with a system that already provides confinement in two dimensions in the first place, which is naturally the case in semiconductor nanowires. Here, gates are only needed to provide

two tunable tunnel barriers and change the electrochemical potential of the formed quantum dot. First experiments were reported again in III-V nanowires (with the above mentioned drawbacks) including gate-defined double quantum dots (Fasth et al., 2005; Fuhrer et al., 2007), determination of the spin-orbit interaction strength (Fasth et al., 2007), spin-orbit qubits (Nadj-Perge et al., 2010a), determination of the hyperfine interaction strength (Nadj-Perge et al., 2010b), and electric-dipole spin-resonance in a few-hole double quantum dot (Pribyl et al., 2013). Also group-IV nanowires attracted an increased research interest in recent years with the first few-hole quantum dots in Si nanowires (Zhong et al., 2005; Zwanenburg et al., 2009b), as well as experiments on spin filling in single quantum dots (Roddaro et al., 2008), charge sensing (Hu et al., 2007), spin relaxation times (Hu et al., 2012), and spin coherence times (Higginbotham et al., 2014b) in Ge-Si core-shell nanowires. Carbon nanotubes are very similar to nanowires regarding their geometry but will not be discussed here. A good overview is provided by Laird et al. (2015).

Ge-Si core-shell nanowires

In this thesis we will use Ge-Si core-shell nanowires as a playground to study hole spins in gate-defined quantum dots. Apart from the spatial confinement itself, nanowires have other interesting properties, since their one-dimensional character leads to unique new electronic properties as well, especially in the valence band (Csontos et al., 2009). In contrast to the two-dimensional case, there are theoretical predictions of mixing of heavy and light hole states at the valence band edge (Csontos and Zülicke, 2007; Kloeffel et al., 2011).

The band mixing gives rise to an enhanced Rashba-type spin-orbit interaction that is predicted to be particularly pronounced in Ge-Si core-shell nanowires (Kloeffel et al., 2011), leading to highly-varying effective g -factors that are strongly anisotropic and electric-field dependent (Maier et al., 2013), making them promising candidates for robust electrically controlled spin-orbit qubits via circuit quantum electrodynamics (Kloeffel et al., 2013). These theoretical studies demonstrate the outstanding potential of hole quantum dots in Ge-Si core-shell nanowires for quantum computing applications as well as for providing a platform for experimental studies of the rich and complicated properties of holes in one dimension.

1.2 THESIS OUTLINE

In this thesis, quantum transport experiments are performed on Ge-Si core-shell nanowires in order to deepen the understanding of this unique one-dimensional system, in particular we study the g -factor anisotropy, Pauli spin blockade and the anisotropic leakage through the spin-blocked system.

We will start with the relevant theory for single and double quantum dots in Chapter 2 and also introduce the symbols and nomenclature used throughout the thesis.

In Chapter 3 we will first give a description of the nanowire growth and will show the exceptional quality of the nanowires through electrical transport measurements. We will also explain how the devices were fabricated and present the measurement setup.

The flexibility of our device design and the high degree of control over the electrostatic environment of the charge carriers will be covered in Chapter 4. In particular, we will demonstrate the formation of gate-defined quantum dots of different lengths up to half a micrometer. We tune the device in a controlled way from a single to a double quantum dot and extract all relevant capacitances and energies.

The underlying idea for Chapter 5 stems from theoretical predictions about an anisotropic g -factor in Ge-Si core-shell nanowires that can be tuned by electric fields. We perform magnetospectroscopy measurements with full 360° rotation of the magnetic field around the three main axes of a single quantum dot, confirming the predictions of a significant g -factor anisotropy with respect to the electric-field axis.

In Chapter 6 we will tune our device to form a gate-defined double quantum dot in the weak-coupling regime. We will present evidence for orbital shell filling in both dots and reveal Pauli spin blockade at the charge degeneracies where we expect it based on these findings.

The scope of Chapter 7 is to elucidate the mechanisms dominating the finite leakage current in the Pauli spin blocked double quantum dot from the previous chapter. We will perform magnetospectroscopy measurements with the magnetic field pointing along three different directions and the spin-blocked double quantum dot tuned to three different regimes. Spin-flip cotunnelling dominates the leakage process

at low magnetic fields. Signatures of spin-orbit interaction induced leakage at higher magnetic fields and a modulation of the Coulomb interaction between the holes forming the spin-blocked triplet state are both visible.

THEORETICAL BACKGROUND

2.1 SINGLE QUANTUM DOTS

A quantum dot is, as far as we are concerned in this thesis, a solid-state structure that contains a well-defined number of charge carriers which are in well-defined quantum states. The following section explains basic principles related to quantum dots that are necessary for the understanding of the phenomena discussed in this thesis, but it is not meant to be exhaustive. More detailed descriptions and omitted aspects can be found e.g. in Kouwenhoven et al. (1997b); Hanson et al. (2007); Kouwenhoven et al. (2001). In order to form a quantum dot, two requirements have to be met:

1. The charging energy E_C , which is necessary to overcome the Coulomb repulsion between charge carriers on the dot, must be much larger than the thermal energy $k_B T$ and the tunnel coupling $\hbar\Gamma$, where k_B is Boltzmann's constant, T is the temperature, $\hbar$ is the Planck constant, and Γ the tunnelling rate: $E_C \gg k_B T, \hbar\Gamma$; so that at most a single charge carrier can tunnel on and off the dot.
2. The energy difference between the single-particle states E_{orb} must also be much larger than $k_B T$ and $\hbar\Gamma$: $E_{\text{orb}} \gg k_B T, \hbar\Gamma$; i.e. the available energy states for a given number of charge carriers on the dot must be quantised and well-defined.

The first requirement originates from the quantisation of charge, a concept still in the realm of classical physics, and leads to the so-called Coulomb blockade. Since E_C has to be paid for every charge carrier added to the dot, the electrochemical potentials μ_i for consecutive numbers of charge carriers i are separated by E_C . When applying a small bias voltage $V_{\text{SD}} \equiv \mu_S - \mu_D$ between the electrochemical potential of the source μ_S and drain μ_D with $|eV_{\text{SD}}| \ll E_C$, where e is the electron charge, and using a gate to change μ_i , a current can only flow when $\mu_S > \mu_i > \mu_D$, otherwise the system is in *Coulomb blockade*. Fulfilling this first requirement is equivalent to having a single-electron or single-hole transistor (Kastner, 1992).

The second requirement is based on a purely quantum mechanical phenomenon. The spatial confinement of the charge carriers leads to

Figure 2.1: (a) Measurement of current through a quantum dot I vs. gate voltage V_g in the linear-transport regime. Coulomb peaks spaced by ΔV_g appear. (b) Cartoon of a bias spectroscopy with V_{SD} and V_g swept. Grey areas are Coulomb diamonds where $I = 0$. N denotes the charge occupation number on the dot. The red line represents an orbital excited state.

a quantisation of the energy eigenstates of their orbital wave function very much like for the electrons in an atom, the reason for quantum dots also being called ‘artificial atoms’. Since the charge carriers considered in this thesis, electrons as well as holes, are fermions, the Pauli exclusion principle applies to them. For a spin-1/2 particle, at most two particles can occupy the same orbital state if no other quantum numbers apply. This leads to a shell-filling effect in the quantum dot: the energy E_{add} necessary to add a charge carrier does not always equal E_C but is modulated by E_{orb} , i. e. $E_{add} = E_C + E_{orb}$ if the added particle occupies an empty orbital or $E_{add} = E_C$ for the second particle in a half-filled orbital (Kouwenhoven et al., 2001).

In essence this means that we need *small* dimensions and *low* temperatures. We realize such small structure by using nanowires, which provide us with an intrinsic confinement in two dimensions. For the third dimension we use gates to locally deplete the nanowire from charge carriers by locally changing the electrostatic potential in the nanowire. Thusly, we can achieve confinement on the nano-scale in all three dimensions in the nanowire section between two such depleted regions and therefore create a quantum dot, which is tunnel-coupled to both ends of the nanowires that serve as charge carrier reservoirs.

Throughout this thesis we will concentrate on the case $E_C > E_{\text{orb}} \gg k_B T, \hbar \Gamma$ and holes as charge carriers. We perform electrical measurements in two regimes: the linear and the non-linear regime. The basics of the linear regime have already been outlined when introducing the single-electron transistor: when only applying a very small V_{SD} , i.e. $\mu_S \approx \mu_D$, continuously sweeping μ_i by means of an applied gate voltage V_g leads to a sequence of so-called Coulomb peaks (for μ_i resonant with μ_S and μ_D) separated by segments where the dot is in Coulomb blockade (see Figure 2.1a).

Applying a finite V_{SD} opens a window between μ_S and μ_D so that a current can flow for a μ_i being at any position within $\mu_S > \mu_i > \mu_D$, i.e. the sharp Coulomb peaks evolve into regions of finite current. If we now measure I within a two-dimensional space of $\{V_{\text{SD}}, V_g\}$ and plot the numerical differential conductance $dI/dV_{\text{SD}}(V_{\text{SD}}, V_g)$, we obtain a so-called *bias spectroscopy* (see Figure 2.1b for a schematic depiction). The diamond-shaped grey areas are *Coulomb diamonds* within which $I = 0$, i.e. the charge occupation number N is constant. Going to higher V_g , one empties the dot by one hole per Coulomb diamond. For $|V_{\text{SD}}|e > E_{\text{add}}$ there is no region in gate space where the current is blocked, i.e. we can obtain E_{add} from the diamond height. The Coulomb peak spacing ΔV_g (see also Figure 2.1a) is also related to E_{add} by $\alpha_g \Delta V_g = E_{\text{add}}$, where α_g is the gate lever arm, a measure for the capacitive coupling between the gate and the dot. Since we assume $E_C > E_{\text{orb}}$, at large enough $V_{\text{SD}} < E_{\text{add}}/e$ orbital excited states may contribute to the current and we observe a line of increased conductance (i.e. current step) parallel to the Coulomb diamond edge as displayed in Figure 2.1b by the red line.

2.2 DOUBLE QUANTUM DOTS

Let us now consider the situation of two tunnel-coupled quantum dots in series. The following description of charge transport through such a double quantum dot as well as the figures largely follow van der Wiel et al. (2003) as far as relevant for this thesis.

In a classical picture one can draw an equivalent electric circuit diagram as shown in Figure 2.2a as two charge islands with hole occupation numbers M and N that are tunnel-coupled (represented by a resistor) and capacitively coupled (represented by a capacitor) to each other and to the reservoirs S and D , and capacitively coupled to two gates 1 and 2. With the two gates we can independently change the

Figure 2.2: (a) Classical electric circuit diagram of two tunnel-coupled quantum dots in series capacitively coupled to two gates. (b) Electrostatic potential diagram of a double quantum dot with a finite bias voltage V_{SD} between the electrochemical potential of the source μ_S and drain μ_D reservoir.

electrochemical potentials on the dots.¹ The couplings R_M and C_M between the two dots are especially interesting here, since they are essentially what distinguishes double quantum dots qualitatively from single quantum dots. Let us first neglect C_M and only assume a finite tunnel resistance R_M : now charge transport from source to drain is possible by sequential tunnelling, but only if $\mu_S \geq \mu_1 \geq \mu_2 \geq \mu_D$ as depicted in Figure 2.2b. Thus in the linear transport regime, i. e. $\mu_S \approx \mu_D$, transport is only possible at specific point in the $\{V_1, V_2\}$ space, where V_i is the voltage on the gates used to change the electrochemical potential μ_i of the left ($i = 1$) and the right ($i = 2$) dot.

A plot of the current I through the double quantum dot versus V_1 and V_2 in the linear transport regime is shown schematically in Figure 2.3. In the left panel we have the situation $C_M \approx 0$ as discussed above: a finite current is only there where four charge states (m, n) are degenerate. At the dashed lines only one of the dots is resonant with the adjacent reservoir, so that holes can tunnel on and off this dot but cannot tunnel all the way from source to drain. If we now switch on a finite capacitive coupling C_M , the edges of the rectangular regions of (m, n) charge stability become slanted. The points of finite conductance split up into pairs of triple points where three charge states are degenerate and still allow for charge transport through the double quantum

¹ In principle there is also a cross capacitance between gate 1 and dot 2, and gate 2 and dot 1, but it is very small compared to the other capacitances in our case.

Figure 2.3: Stability diagram of a double quantum dot with three different inter-dot couplings C_M . Left panel: Uncoupled dots, current flow only at black points. (M, N) denotes charge occupation in left (M) and right dot (N). Middle panel: Intermediate coupling regime, honeycomb pattern forms, finite current only at pairs of black points. Right panel: Strongly coupled dot forming effectively one large dot, finite current along black lines.

dot. The lines, along which two charge states are degenerate, now form a honeycomb pattern instead of a square pattern (dashed lines in left and middle panel of Figure 2.3). The separation between the two triple points in terms of gate voltages is $\Delta V_{1,m}$ and $\Delta V_{2,m}$ for the left and the right dot, respectively. With increasing C_M this splitting also increases according to $\Delta V_{1,m} = \Delta V_1(C_M/C_2)$ and $\Delta V_{2,m} = \Delta V_2(C_M/C_1)$, where ΔV_i is the voltage difference between adjacent honeycomb edges that denote successive charge degeneracies of the left dot ($i = 1$) or the right dot ($i = 2$) (see Figure 2.3). Therefore it is possible to calculate C_M from other quantities in a charge stability diagram, which we will do in Chapter 4.

For very large C_M the double quantum dot effectively becomes one large single quantum dot and a finite current can be measured along diagonal lines in the charge stability diagram as shown in the right panel of Figure 2.3.

In the non-linear transport regime, i. e. at finite V_{SD} , the triple points evolve into triangles, see Figure 2.4. Along the edges of a bias triangle at least two of the electrochemical potentials μ_S , μ_1 , μ_2 , and μ_D are resonant with each other as shown by the small green sketches in Figure 2.4 that give a minimalistic impression of Figure 2.2b. Along the base line, i. e. the line connecting the two triple points, the two

Figure 2.4: Stability diagram of a double quantum dot in the non-linear transport regime. A finite current can only be measured within the grey triangles, the red line denotes an excited state entering the bias window. Green schematics sketch the alignment of electrochemical potentials at different point of the bias triangle.

dot potentials are resonant. Therefore we define the detuning $\epsilon = \mu_1(N+1, M) - \mu_2(N, M+1)$ and refer to the base line of the triangle also as the ‘zero-detuning line’. Within the bias triangles transport occurs via inelastic tunnelling (van der Wiel et al., 2003), but for $|eV_{SD}| > E_{orb}$ orbital excited states can serve as additional transport channel which leads to a current step within the bias triangle where the excited state enters the bias window (see the red line in Figure 2.4).

2.3 PAULI SPIN BLOCKADE

Current rectification caused by Pauli spin blockade (PSB) has been observed for the first time in the early 2000s (Ono et al., 2002) and has since been established as a means of detection of spin states through spin-to-charge conversion (Petta et al., 2005; Koppens et al., 2006; Maune et al., 2012; Hu et al., 2012; Prance et al., 2012).

The basic principle of PSB is sketched in Figure 2.5. The explanation here is a short summary of the relevant effects, a more elaborate description is given e.g. by Hanson et al. (2007). Without loss of generality, we assume an unpaired spin-down hole in the left quantum dot, its electrochemical potential $\mu_1(1, 0)$ well below the Fermi energy of the drain contact. The electrochemical potential of filling a second hole $\mu_1(2, 0)$ now depends on its spin state. A spin-up hole can form a singlet state S with the first hole, its electrochemical potential being $\mu_{1,S}(2, 0)$. In contrast, the electrochemical potential for the addition of a second spin-down hole $\mu_{1,T}(2, 0)$ is significantly higher. Here, a triplet

Figure 2.5: Schematic electrochemical potential diagram for current rectification by Pauli spin blockade (PSB) in the unblocked (left) and blocked direction (right).

state T has to be formed that involves the next orbital of the quantum dot because of the Pauli exclusion principle, leading to a splitting of $\Delta_{S-T} \equiv \mu_{1,T}(2,0) - \mu_{1,S}(2,0)$, which is on the order of the orbital level splitting for a single quantum dot. Such a large splitting is not observed if the second hole is added to the right dot. Only a small exchange energy mediated by the tunnel coupling of $J = 4t^2/E_C$, where t is the interdot tunnel coupling, leads to a splitting between the singlet state $S(1,1)$ and the triplet state $T(1,1)$. Since $E_C \gg t$, this splitting is much smaller than Δ_{S-T} and negligible in our experiments.

A current through the double quantum dot involves sequential tunnelling of a hole from left to right or *vice versa*. Assuming $V_{SD} > 0$ with current flowing from left to right, this changes the occupation number of the double quantum dot as follows: $(2,1) - (2,0) - (1,1) - (2,1)$ or $(1,0) - (2,0) - (1,1) - (1,0)$. Both of the two cases involve interdot-tunnelling of a hole from the $(2,0)$ and the $(1,1)$ state, which is always possible regardless of the spin state (see left panel of Figure 2.5).

If we now reverse the bias, the occupation number cycle is also reversed, which includes the transition $(1,1) - (2,0)$. As depicted in Figure 2.5, we tune our plunger gates in such a way, that the electrochemical potentials of the singlet states $S(2,0)$ and $S(1,1)$ are aligned. We define the detuning $\epsilon = 0$ to be the difference between these two electrochemical potentials: $\epsilon \equiv \mu_{2,S}(1,1) - \mu_{1,S}(2,0)$ for negative bias. Now tunnelling $S(1,1) - S(2,0)$ is still possible, but as soon as a spin-down hole tunnels onto the left dot and forms a $(1,1)$ -triplet state, the system is trapped in this configuration because tunnelling to the $(2,0)$ -triplet state is energetically forbidden. Only for $\epsilon \geq \Delta_{S-T}$, spin-blockade is lifted and a current can be measured. In a charge stability diagram like Figure 6.2, this results in a reduced bias triangle size, because a trape-

zoid is missing between the base line where $\epsilon = 0$ and the parallel line where $\epsilon = \Delta_{S,T}$.

In real experiments, a finite current in this spin-blocked region of the bias triangles may still occur due to processes that induce spin-flips and thus change the spin-state from triplet to singlet. The strength of the leakage current I_{leak} and its evolution when changing tunnel couplings or the magnetic field can give valuable insights into the processes limiting the spin lifetime in such structures and are thus important for both fundamental understanding and applicability for quantum computing.

2.4 LEAKAGE PROCESSES IN PAULI SPIN BLOCKADE

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 $T(1,1)^2$. For a current to be measured, there must exist an efficient process that leads to the system reaching the $(2,0)$ -singlet state³ $S(2,0)$, 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 and Nazarov, 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

2.4.1 Hyperfine interaction

Such spin flips can be induced by hyperfine coupling to nuclei with non-zero net spin (Jouravlev and Nazarov, 2006), which generate random magnetic fields that superimpose in semi-classical approximation

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- 2 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 $T_- = |\downarrow\downarrow\rangle$.
  - 3 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 $E_{\text{th}} \approx 3$ μ eV $\ll \Delta_{S,T}$, we consider this a fair assumption.

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 systems based on III-V semiconductors like GaAs (Koppens et al., 2005; Johnson et al., 2005b), InAs (Pfund et al., 2007a; Nadj-Perge et al., 2010b), 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 wave function, which suppresses the (in s-type bands) dominating contribution of the contact hyperfine interaction (Fermi, 1930).

2.4.2 Spin-flip cotunnelling

A second major cause for transitions between $T(1,1)$ and $S(1,1)$ is the so-called spin-flip cotunnelling (Qassemi et al., 2009; Coish and Qassemi, 2011). 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 ϵ (the overlap of $S(1,1)$ with $S(2,0)$ limits I_{leak} for large ϵ) 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 tunnelling 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 $k_B T > t$ is (Coish and Qassemi, 2011)

$$I_{co} = \frac{4}{3} ec \frac{g^* \mu_B B}{\sinh \frac{g^* \mu_B B}{k_B T}}, \quad (1)$$

where

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

Δ is the depth of the two-hole levels,⁴ and U_M is the interdot charging energy.⁵

2.4.3 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 and Nazarov (2009) for a short overview). Here we shall summarize the key features of the process, the in-depth derivation can be found in Danon and Nazarov (2009). The exact overlap of the $T(2,0)$ wave functions with the $S(1,1)$ wave functions 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 $I_{\text{leak}}^{\text{max}} = 4\Gamma_{\text{rel}}$. At zero detuning, the dip around $B = 0$ follows for $\vec{B} = (0, 0, B_Z)$ (in the following $B \equiv B_Z$)

$$I_{\text{leak}}(B) = I_{\text{leak}}^{\text{max}} \left(1 - \frac{8}{9} \frac{B_C^2}{B^2 + B_C^2} \right), \quad (3)$$

where

$$B_C = 2\sqrt{2} \frac{(1 + |\vec{\eta}|^2)}{\sqrt{\eta_x^2 + \eta_y^2}} t \sqrt{\Gamma_{\text{rel}}/\Gamma} \quad (4)$$

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 $(1,1)$ wave functions and thus very hard to predict in real systems.

⁴ See Figure 1b) in Qassemi et al. (2009) for an insightful sketch.

⁵ This is the term used by Coish and Qassemi (2011). Other sources refer to it as the ‘mutual charging energy’ (see e. g. Laird et al., 2015)).

3.1 NANOWIRE GROWTH

The Ge-Si core-shell nanowires used in this thesis are grown on a single-crystalline Ge $\langle 111 \rangle$ substrate via the vapour-liquid-solid (VLS) method (Wagner and Ellis, 1964) by our collaborators¹ at the Technical University Eindhoven². In this process, Au nanodroplets are first formed at elevated temperatures on the substrate. These catalysts then get supersaturated with Ge by the introduction of germane gas that dissociates at elevated temperatures on the Au surface into H_2 and Ge that then dissolves in the Au. At the interface between the droplet and the substrate, Ge precipitates epitaxially and a monocrystalline Ge nanowire grows whose diameter is determined by the size of the Au droplet. During this process, the pressure and temperature are tuned in such a way that growth via VLS dominates and direct precipitation of Ge at the sidewalls of the already formed nanowire part is negligible, which results in a nearly tapering-free defect-free Ge nanowire of 1-10 μm in length. In a second step, silane gas is introduced instead of germane and a separation segment of Si is grown between the Ge and the Au droplet before conditions are changed to favour sidewall-growth of the Si over VLS growth resulting in a Si shell of tunable thickness. The separation segment prevents diffusion of the Au during shell growth.

Despite the monocrystalline Ge $\langle 111 \rangle$ substrate, there are also nanowires with a $\langle 110 \rangle$ crystal axis. These nanowires are found to be thinner (diameter ~ 20 nm) on average than the $\langle 111 \rangle$ wires (diameter ≥ 30 nm) and have a nearly defect-free Si shell, whereas the $\langle 111 \rangle$ nanowires exhibit a finite defect density (Li et al., 2015a).

3.2 ELECTRICAL CHARACTERISATION

For the quantum transport measurements performed throughout this thesis, a high charge carrier mobility μ is crucial since only then we can

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¹ Ang Li, Erik Bakkers

² The exact growth conditions and exhaustive characterization of crystal structure, composition, strain, and electric transport studies are part of a shared publication between our collaborators in Eindhoven and us, which is currently in preparation, see Li et al. (2015a)

Figure 3.1: TEM micrograph of a nanowire with $\langle 110 \rangle$ crystal direction along the nanowire axis (left panel), corresponding fast Fourier transform (right panel). Courtesy A.Li and S. Conesa-Boj.

effectively change the charge carrier density by means of electric fields applied by gates. Furthermore, high mobilities are associated with low defect densities. A high μ is therefore an indicator for a system that is suitable for quantum transport since defects are effective scattering centres where charge carriers may scatter inelastically. This leads to decoherence and therefore destruction of quantum states. We build field-effect transistors from the nanowires and calculate μ by measuring the transconductance $G \equiv dI/dV_g$ and using (Wunnicke, 2006)

$$\mu = \frac{GL^2}{CV_{SD}}, \quad (5)$$

where I is the current from the drain contact to ground while applying a voltage V_{SD} to the source contact of the field-effect transistor (FET), V_g is the voltage applied to the gate, L the length of the FET channel and C the capacitance between channel and gate.

The FET devices are fabricated on a p^{++} -doped Si wafer that is conductive at low temperatures and serves as the back gate. 60 nm thermally grown SiO_2 cover the wafer as a dielectric, on top of which large metallic contact pads are patterned by structuring a resist layer with photolithography and subsequent deposition of (1/30) nm Ti/Pd by electron-beam evaporation followed by lift-off. In a second step a pattern of bitmarkers and alignment crosses³ is fabricated by means of resist-structuring with electron-beam lithography (EBL), metallisation using electron-beam evaporation of (1/30) nm Ti/Pd and lift-off. Single nanowires are picked up from the growth chip with a micromanipulator and deposited on the device chip. This deposition is performed

³ The bitmarker structure is a regular pattern of unique structures that allows the localisation of micro-sized objects on a macroscopic chip.

Figure 3.2: AFM micrograph of a nanowire device used for measuring the hole mobility (left panel) with a height profile across the nanowire as indicated by the blue dashed line (inset). Schematic cross section of the device (right panel).

under a light microscope with which a picture of the nanowire alongside surrounding bitmarkers and alignment crosses is taken. Ohmic contacts to the nanowires are patterned with an EBL after aligning the patterned design with the picture of the nanowire in a subsequent step. The contacts are metallised by depositing 0.5/50 nm of Ti/Pd directly following a 3 s dip in buffered HF that etches away the native oxide of the silicon shell, followed by lift-off. The contacts are designed to form a 800 nm channel between them. An atomic-force microscopy (AFM) micrograph of a typical device and a schematic cross section are depicted in Figure 3.2.

The channel length of 800 nm ensures applicability of Equation 5 since this equation is valid for $L \gg d_{\text{ox}}$, where d_{ox} is the thickness of the gate dielectric. The transconductance measurements are performed inside a liquid helium dewar at a temperature of 4 K. A typical I versus V_g plot is displayed in Figure 3.3a. The transconductance used for calculating μ is extracted by fitting the linear part, which leaves only the capacitance C as an unknown in Equation 5. In principle one can estimate C by using the analytical formula given by the cylinder-on-plane model, but this has been shown to significantly overestimate C (Wunnicke, 2006) because it assumes an infinitely long metallic cylinder embedded in the dielectric rather than placed on top of it. Therefore we use a finite-element solver software (COMSOL Multiphysics™) to numerically calculate the capacitances with which we then estimate μ . The mobilities of eleven devices characterized this way are plotted in Figure 3.3b versus the diameter measured by AFM. A clear trend of higher mobility for wires with diameters smaller than 20 nm is visible. The four coloured data points refer to nanowire devices of which the crystal orientation is determined after the electrical measurements. The two coloured data points with a diameter smaller than 20 nm exhibit a

Figure 3.3: (a) I vs. gate voltage V_g of a typical device used for calculation of μ . (b) μ versus the wire diameter d_w of 11 devices. The coloured data points are from nanowires whose crystal structure is determined after the electrical measurements.

$\langle 110 \rangle$ crystal direction along the nanowire axis, whereas the ones with bigger diameter have a $\langle 111 \rangle$ crystal orientation. The maximum μ of more than $1800 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ is much higher than previously reported values of $730 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ (Xiang et al., 2006a), $600 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ (Hao et al., 2010), and $500 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ (Nguyen et al., 2014).

3.3 QUANTUM DOT DEVICE FABRICATION

The devices used in Chapters 4 to 7 consist of a p^{++} -doped Si substrate covered with 200 nm thermally grown SiO_2 , on which six bottom gates with a 100 nm pitch are patterned with EBL. The gates are buried by 10 nm Al_2O_3 grown with atomic layer deposition at 100°C . A single nanowire with a Si shell thickness of approximately 2.5 nm and a Ge core radius of approximately 8 nm 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. All contacts are defined by EBL. The nanowire part above the bottom gates is at no point exposed to the electron beam, which prevents carbon deposition and introduction of defects into the otherwise defect-free Ge core (Li et al., 2015a).

3.4 QUANTUM DOT MEASUREMENT SETUP

All measurements in Chapters 4 to 7 are carried out in a dilution refrigerator (Triton200 from Oxford Instruments) with a base temperature of

Figure 3.4: I vs. relative plunger gate voltage V_{rel} measurement at $V_{\text{SD}} = 4 \mu\text{V}$ of a Coulomb peak at different temperatures without (upper panel) and with metal powder filters (lower panel) (b) peak widths FWHM extracted from (a) and corrected by the gate lever arm α versus the set temperature at the mixing chamber plate.

$\sim 10 \text{ mK}$. For the electronic measurements we use a battery-powered IVVI rack custom-built at the TU Delft that comprises low-noise voltage sources and I/V -converters for current measurements. This system is galvanically decoupled through an optical fibre from the computer that is used for controlling the measurements. The electric wiring between the IVVI rack and the sample hooked up to the cold finger comprises three stages of electric high-frequency noise filtering: room-temperature pi filters, as well as low-temperature RC-filters and copper powder filters. The dilution refrigerator also includes a superconducting vector magnet with a maximal magnetic field of (6–1–1) T along three orthogonal axes.

3.5 DETERMINATION OF THE ELECTRON AND HOLE TEMPERATURE

We use a single quantum dot in a silicon MOSFET structure (we employ the device design introduced by Angus et al. (2007)) to measure the effective electron temperature T_e of a high-impedance device in our

dilution refrigerator setup.⁴ The temperature of the electrons in an electrical device can differ significantly from the refrigerator temperature T_p , mainly due to noise and pick-up of interference. Our setup comprises several stages of electrical filtering (see Section 3.4) to minimise electrical noise, one essential part of which are copper powder filter (CPF) designed and built in our research group. In order to evaluate the effectiveness of these in-house built filters we measure the electron temperature with and without the CPF.

The shape and width of the Coulomb peak of a single-level tunnelling resonance in a quantum dot is dominated by two mechanisms: Tunnel broadening of the resonant electrochemical potential level on the dot, and thermal broadening. Both processes lead to distinct peak shapes, which, if both processes are relevant, form a convolution of a Lorentzian function (tunnel broadening) with the derivative of the Fermi-Dirac distribution function (in the following called *F-D function*, thermal broadening), for details see e.g. Beenakker (1991). The peak width due to thermal broadening is directly proportional to temperature and considering single-level tunnelling gives: $FWHM = 3.52k_B T_e$, where $FWHM$ is the full width at half maximum of the Coulomb peak, k_B is Boltzmann's constant, and T_e the electron temperature. The proportionality factor of 3.52 is only valid for single-level tunnelling, if one has to take into account tunnelling through multiple quantum levels, this factor changes to 4.35. We employ this by measuring the same Coulomb peak at different bath temperatures, which we can control with a heater. This whole procedure is performed during two different cool-downs of the setup, once with and once without the metal powder filters connected. In Figure 3.4 we plot I versus the gate voltage relative to the peak centre at $V_{SD} = 4 \mu V$ for different temperatures we set at the heater system of the mixing chamber plate (which is thermally anchored to the sample holder).

We extract the full width at half maximum $FWHM$ for each scan and plot $FWHM$ versus T_p in Figure 3.4b. For both with and without CPF we observe the same general trend: At very low temperatures the $FWHM$ stays constant for increasing temperature up to a critical temperature above which it increases linearly. We explain this by the discrepancy between T_p and T_e : As long as $T_e > T_p$, increasing T_p has no effect on the Coulomb peak width, because the effective temperature of the tunnelling electrons remains constant. Only when $T_e = T_p$ a further increase of T_p will also increase T_e . We apply a linear fit to the high-

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⁴ The results presented here are part of Mueller et al. (2013)

temperature regime and a constant fit to the low temperature regime. The crossing point of the two fits represents the minimal temperature at which $T_e = T_p$, i. e. this temperature is also the electron temperature at the base temperature of the refrigerator. By doing this we obtain $T_e = (35 \pm 7)$ mK without and $T_e = (22 \pm 2)$ mK with CPF. There are two deviations from the theoretical situation described above: For the measurements without CPF we found a slope of $5.0k_B$ instead of $3.52k_B$ which cannot be explained by multi-level tunnelling and the cause remains unknown. The second deviation is also visible in Figure 3.4b: For the measurements with CPF, the low-temperature extrapolation of the thermal peak width does not result in $FWHM(T_e = 0) = 0$, but a significant residual peak width remains corresponding to ~ 10 mK. Nevertheless we find a significantly lower electron temperature in realistic measurement situations when the CPF is installed.

HIGHLY TUNEABLE SINGLE AND DOUBLE QUANTUM DOTS IN GE-SI CORE-SHELL NANOWIRES

4.1 INTRODUCTION

In this chapter we will introduce a highly flexible quantum dot device based on Ge-Si core-shell nanowires. We will show and characterize single quantum dots of varying lengths up to 450 nm and tune them to stepwise form double quantum dots with different interdot tunnel couplings.

4.2 DEVICE DESIGN

We will discuss measurements in two different devices $D1$ and $D2$ on two different chips, which have been fabricated in the same way. The devices (an AFM micrograph of $D1$ is shown in Figure 4.1a) consist of a p^{++} -doped Si substrate covered with SiO_2 , on which six bottom gates $g1$ - $g6$ with 100 nm pitch are patterned with EBL. The gates are buried by 10 nm Al_2O_3 grown with atomic layer deposition (ALD) at 100 °C. A single nanowire with a Si shell thickness of approximately 2.5 nm and a Ge core radius of approximately 8 nm ($D1$) and 10 nm ($D2$) is deterministically placed on top of the gate structure with a micromanipulator. Subsequently we define ohmic contacts to the nanowire and gate contacts made of Ti/Pd (0.5/50 nm) with EBL. The nanowire part above the bottom gates is at no point exposed to the electron beam, which prevents carbon deposition and introduction of defects into the otherwise defect-free Ge core. All measurements are performed in a dilution refrigerator with a base temperature of 8 mK using direct current (DC) electronic equipment. A source-drain 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 the device by measuring the temperature dependence of the Coulomb peak width (Mueller et al., 2013; Goldhaber-Gordon et al., 1998).

Figure 4.1: (a) False-colour atomic-force microscopy (AFM) image of the device. (b) Schematic cross-section displaying the p++-doped Si substrate (grey) with 200 nm of SiO₂ (dark red), six bottom gates g1-g6 (light red), each approximately 35 nm wide and with a pitch of 100 nm. The bottom gates are buried under 10 nm of Al₂O₃ (yellow), on top of which the nanowire is deposited (green). Ohmic contacts (0.5/50 nm Ti/Pd, blue) are defined by means of EBL.

4.3 SINGLE QUANTUM DOTS OF VARYING LENGTH

By using different gates as barriers we can form quantum dots in our nanowire with lengths varying from very long quantum dots (using g1 and g6 to form tunnel barriers) to very short dots (using adjacent gates as barrier gates). The formation of tunnel barriers for quantum dots in nanowires is also possible by using lateral heterostructures (Fuhrer et al., 2007; Roddaro et al., 2011) or taking advantage of Schottky barrier formation at the nanowire-metal interface with the contacts (Zwanenburg et al., 2009a; Nilsson et al., 2011). Defining tunnel barriers electrostatically using local gates we have two great advantages:

1. We can vary the length of the quantum dot *in situ*.
2. We can change the tunnel coupling from the quantum dot to the reservoirs *in situ*.

The ability to change these properties during measurements is particularly important, because quantitative comparison between different device is difficult and not always conclusive due to the inevitable variations in the device properties after fabrication (e.g. nanowire diameter, thickness of the dielectric). Imperfections in the crystal structure of the nanowire, so-called defects, can lead to fluctuations in the potential profile or serve as trapping centres for charge carriers. While potential fluctuations can e.g. lead to the formation of unintentional quantum dots

l (nm)	E_C (meV)	C (aF)	ΔV_g (mV)	C_g (aF)
60	18.3 ± 0.2	8.8 ± 0.2	104 ± 1	1.54 ± 0.02
160	10.2 ± 0.2	15.7 ± 0.3	31.5 ± 0.2	5.09 ± 0.03
260	6.8 ± 0.2	23.5 ± 0.5	29.6 ± 0.4	5.41 ± 0.07
360	5.2 ± 0.1	30.8 ± 0.6	28.6 ± 0.4	5.63 ± 0.08
460	4.2 ± 0.1	38.6 ± 0.9	29.7 ± 0.4	5.39 ± 0.07

Table 4.1: Parameters for electrostatically defined quantum dots of varying length as extracted from Figure 4.2.

(Spruijtenburg et al., 2013; Llobet et al., 2015), charge traps may perturb measurements as a source of telegraph noise (Culcer et al., 2009). The formation of very long quantum dots is challenging because given a certain defect density in the nanowire, longer quantum dots will have a higher chance to suffer from such defects.

In Figure 4.2 we plot $dI/dV \equiv dI/dV_{SD}$ while sweeping V_{SD} and the voltage on the plunger gate V_p that we use to control the electrochemical potential of the quantum dot. We show five of these bias spectroscopies for five different gate configurations in two different devices. Figure 4.2a shows a quantum dot formed in $D1$, the quantum dots displayed in Figures 4.2b to 4.2e are formed in $D2$. Determination of the actual length of the respective quantum dot is challenging because it would involve knowledge of the exact confinement potential (for a harmonic potential one could then use the orbital level spacing, see e.g. Biercuk et al. (2006)) or numerical calculation of the gate capacitance (Zwanenburg et al., 2009b), which is beyond the scope of this chapter. Also geometrically it is possible to at least get an estimate of the quantum dot length by using the distance between the barrier gates. In this chapter we define the quantum dot length as the distance of the inner edges of the barrier gates. Assuming a gate width of ~ 40 nm this results in quantum dot lengths of ~ 60 nm for adjacent barrier gates (Figure 4.2a), ~ 160 nm for barrier gates with one plunger gate in between (Figure 4.2b), ~ 260 nm for two plunger gates (Figure 4.2c), ~ 360 nm for three plunger gates (Figure 4.2d), and ~ 460 nm for four plunger gates (Figure 4.2e), i.e. we are able to tune the dot length over almost and order of magnitude.

Figure 4.2: Bias spectroscopy of gate-defined single quantum dots formed with (a) gates, (b) one gate, (c) two gates, (d) three gates, (e) four gates between the barrier gates.

Charging energies

The formation of quantum dots of five different lengths in Figures 4.2a to 4.2e is reflected in the clear Coulomb diamonds. We extract the respective charging energies E_C from the height of the Coulomb diamonds and find a decreasing E_C from 18.3 meV to 4.2 meV for increasing dot length (see Table 4.1). We observe two distinct evolutions of E_C for the different gate configurations: E_C increases significantly over the three Coulomb diamonds from 16.7 meV to 20.0 meV in Figure 4.2a, where we use the left barrier gate to change the electrochemical potential μ of the quantum dot. For the quantum dot configurations where we use a dedicated plunger gate to shift μ (Figures 4.2b to 4.2e) E_C stays constant over several Coulomb diamonds. Since the charging energy of the quantum dot is directly linked to its total capacitance via $E_C = e^2/C$, C is also not constant in Figure 4.2a and therefore the constant interaction model is not valid here. This can be readily explained by the fact that we use the barrier adjacent to the drain contact to control the electrochemical potential of the quantum dot, which also changes the tunnel barrier and therefore the capacitance to drain and the gate. For this configuration, we extract the values in Table 4.1 from the middle Coulomb diamond because it represents the highest occupation number where we do not observe significant cotunnelling and therefore have well-defined tunnel barriers as also observed for the other configurations.

Capacitances

The constant charging energies over several Coulomb diamonds in Figures 4.2b to 4.2e are accompanied by constant voltage differences between adjacent Coulomb peaks ΔV_g at $V_{SD} = 0$, indicating a constant gate capacitance $C_g = e/\Delta V_g$ over several charge transitions. Together with the total capacitance $C = e^2/E_C$ of the quantum dot staying constant as well we infer that the constant interaction model is valid for these configurations. If we now compare the plunger gate capacitances between Figures 4.2b to 4.2e, we find them to be all very similar, (~ 5.5 aF), while the total capacitance of the quantum dots increases linearly by ~ 7.5 aF per additional plunger gate. The discrepancy of ~ 2 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 capacitance indicates equal coupling of all the gates, which is consistent with the gate geometry (equal width and distance to the nanowire), and therefore demon-

strates a high degree of control over the electrostatic environment of the gate-defined quantum dot.

Extremely long bias spectroscopy

We will now focus on the stability of the quantum dots. As stated earlier, very long quantum dots have a high chance of being in the direct vicinity of any kind of defect that gives rise to unintended changes in the quantum dot, either by an unstable electrostatic environment (e.g. telegraph noise due to bistable charge traps in the Si shell or the gate oxide) or the formation of unintentional quantum dots. In order to investigate the stability and cleanliness of our nanowire devices we tune the gates of $D1$ to form a quantum dot of maximum length between $g1$ and $g6$ similar to the configuration in Figure 4.2e for device $D2$. The corresponding bias spectroscopy, where we sweep $g3$ as a plunger gate over a range of 4000 mV, is shown in Figure 4.3a (split up into four panels). Like for $D2$, we again obtain regular Coulomb diamonds, here with a charging energy of $E_C = 4.4$ meV. This is slightly higher than for $D2$, which we link to the smaller radius of the nanowire in $D1$. In a simple picture one might expect E_C to change indirectly proportional to the quantum dot volume, i.e. to r_{wire}^2 for a cylinder-shaped quantum dot of constant length, which would give $E_{C,D2}/E_{C,D1} = r_{D1}^2/r_{D2}^2 = 0.64 \neq 4.2 \text{ meV}/4.4 \text{ meV}$. In reality, the situation is more complicated, because this picture only holds for the self-capacitance of the dot, but not for the capacitances between the dot and gates or reservoirs, for which again numerical modelling might provide further insight that is beyond the scope of this chapter.

In the V_{g3} range from -2000 mV to 1000 mV the Coulomb diamonds are extremely regular in size and shape and also the current outside the Coulomb diamonds does not change significantly while depleting the quantum dot by 112 holes. Above $V_{g3} \approx 1000$ mV the size of Coulomb diamonds varies and above $V_{g3} \approx 1850$ mV the Coulomb diamonds do not close any more. The distance between adjacent Coulomb peak ΔV_g is plotted in Figure 4.3b versus the number of holes by which the quantum dot is depleted n compared to the number of holes N on the quantum dot at $V_{g3} = -2000$ mV. ΔV_g remains very stable at $\Delta V_g = (25 \pm 2)$ mV up to a hole occupation of $N - 112$. Above $V_{g3} \approx 1000$ mV the variation of ΔV_g increases which we assign to shell filling effects when reaching the few-hole regime (Tarucha et al., 1996; Hanson et al., 2007). Below $V_{g3} \approx 1000$ mV we observe a significantly smaller ΔV_g for three Coulomb diamonds at $n = \{72, 93, 105\}$. A

closer look at Figure 4.3a reveals switches in these Coulomb diamonds, i.e. sudden shifts in the electrochemical potential of the dot, which can be caused by e.g. charge traps (Pioro-Ladrière et al., 2005). The non-closing diamonds above $V_{g3} \approx 1850$ mV are also observed in the same gate range for shorter dots and across both devices. They are attributed to splitting of the single quantum dot into a double dot with the plunger gate as the interdot tunnel barrier. In Section 4.4 we will make use of this effect to intentionally form a double quantum dot.

The whole measurement in Figure 4.3a takes ~ 32 h and depletes the quantum dot by 152 holes, which equals approximately one switching event per ten hours over this extremely large range of the occupation number. This is a strong indication for a quantum dot with a very clean and stable electrostatic environment, i.e. low defect density in the nanowire and a high-quality gate dielectric.

4.4 TUNABLE DOUBLE QUANTUM DOTS

In order to form a highly tunable 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. Starting from the situation in Figure 4.2d, where we have tuned our device to form a single quantum dot above three gates, we can increase the voltage on the middle gate V_{g4} . When approaching the pinch-off voltage, it will form a tunnel barrier and the single quantum dot splits up into two tunnel-coupled quantum dots. By performing gate-versus-gate measurements with the two plunger gates $g3$ and $g5$ while step-wise increasing V_{g4} we obtain charge stability diagrams from which we extract relevant capacitances.

The stability diagrams at four different V_{g4} are plotted in Figure 4.4a. There we keep the outer barrier gates at constant voltages ($V_{g2} = 2490$ mV, $V_{g6} = 2940$ mV) and plot $\log |I|$ at a fixed $V_{SD} = 1$ mV while sweeping the voltage on the left plunger gate V_{g3} versus the voltage on the right plunger gate V_{g5} . Between each measurement in the panels from left to right we increase the voltage on the middle barrier gate V_{g4} from $V_{g4} = 0$ mV up to $V_{g4} = 2900$ mV. For $V_{g4} = 0$ mV we observe the typical stability diagram of a single quantum dot:¹ The diagonal, parallel lines of finite current are Coulomb peaks. In the regions between them the quantum dot is in Coulomb blockade and current is blocked.

¹ The different regimes described in the following reproduce experimentally the schematics in Figure 2 of van der Wiel et al. (2003).

Figure 4.3: (a) Bias spectroscopy of gate-defined single quantum dot formed with barrier gates $g1$ and $g6$ (similar to Figure 4.2e) in $D1$. (b) Coulomb peak spacing ΔV_g plotted versus number of depleted holes n .

The spacing between the Coulomb peaks along the respective plunger gate axis is directly related to the capacitance between the quantum dot and the gate: $C_g = e/\Delta V_g$. Here the spacing is $\Delta V_g \approx 27$ mV for both g_3 and g_5 , which means both gates have the same capacitance $C_g \approx (5.8 \pm 0.3)$ aF to the quantum dot. This indicates that the quantum dot indeed stretches over the whole distance between the tunnel barriers formed directly above gates g_2 and g_6 and is also very similar to the gate capacitances measured in Section 4.3.

If we now set $V_{g4} = 2000$ mV and perform the same measurement, we see that the straight lines have evolved into a regular honeycomb pattern with two distinct slopes that form the long edges of each honeycomb. The honeycomb formation with a large current along the edges indicates the formation of a strongly coupled double quantum dot, where the more vertical (horizontal) set of parallel lines refers to sweeping through the electrochemical potential of the left (right) dot with fixed alignment of the electrochemical potential of the right (left) dot with the right (left) reservoir. From the distance between adjacent lines we extract the voltages needed to add a hole to the left (right) dot ΔV_{g3} , (ΔV_{g5}) and calculate the respective capacitances. For the left dot we find $\Delta V_{g3} = (25.1 \pm 0.5)$ mV and for the right dot $\Delta V_{g5} = (26.1 \pm 0.5)$ mV, from which we calculate capacitances $C_{g3} = (6.3 \pm 0.2)$ aF and $C_{g5} = (6.1 \pm 0.2)$ aF. The sets of honeycomb edges representing the addition of a hole to either the left or the right dot are both significantly slanted as compared to the charge stability diagram of an uncoupled double quantum dot (see Figure 2a in van der Wiel et al. (2003)), which can be explained predominantly by the significant mutual capacitive coupling between the two dot C_M that leads to a separation between the two triple points (the short sides of each honeycomb, see van der Wiel et al. (2003)). We express this shift in terms of gate voltages and find $\Delta V_{g3,M} = (9.2 \pm 0.5)$ mV and $\Delta V_{g5,M} = (10.9 \pm 0.5)$ mV. Using the expression $C_{gi,M} = C_{gi}\Delta V_{gi,M}/\Delta V_{gi}$ (van der Wiel et al., 2003), we obtain $C_{g3,M} = (2.3 \pm 0.3)$ aF and $C_{g5,M} = (2.5 \pm 0.3)$ aF. A second mechanism that can affect the slopes of the honeycomb edges is the finite cross capacitance between g_3 and the right dot $C_{g3,C}$, and g_5 and the left dot $C_{g5,C}$. This cross capacitance leads to a shift of the triple points along the g_3 gate axis while changing the charge occupation of the right dot, and along the g_5 gate axis while changing the number of holes on the left dot. This effect is very weak, and we extract $C_{g3,C} \approx C_{g5,C} \approx 0.1$ aF.

Increasing the voltage on the interdot barrier gate to $V_{g4} = 2700$ mV only slightly changes the gate capacitances to $C_{g3} = (5.9 \pm 0.2)$ aF

Figure 4.4: (a) Charge stability diagrams with $\log I$ 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 are performed in device $D1$.

V_{g4} (mV)	C_{g3} (aF)	C_{g5} (aF)	$C_{g3,M}$ (aF)	$C_{g5,M}$ (aF)
0	5.8 ± 0.3	5.8 ± 0.3		
2000	6.3 ± 0.2	6.1 ± 0.2	2.3 ± 0.3	2.5 ± 0.3
2700	5.9 ± 0.2	5.7 ± 0.2	0.6 ± 0.2	0.7 ± 0.2
2900	5.8 ± 0.2	5.5 ± 0.2	0.4 ± 0.1	0.4 ± 0.1

Table 4.2: Capacitances for increasing voltage on the middle barrier gate g_4 of an electrostatically defined single ($V_{g4} = 0$ mV) or double quantum dot ($V_{g4} \geq 2000$ mV) as extracted from Figure 4.4a.

and $C_{g5} = (5.7 \pm 0.2)$ aF. For the mutual gate capacitances we find a much stronger relative change to $C_{g3,M} = (0.6 \pm 0.1)$ aF and $C_{g5,M} = (0.7 \pm 0.1)$ aF, which indicates a significantly increased separation of the charge distribution of both dots. The strongly suppressed current along the long edges of the honeycombs, which are now only faintly visible, also suggests 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 can also be observed for this more weakly tunnel-coupled double quantum dot, again on the order of 0.1 aF.

A further increase of the interdot barrier gate to $V_{g4} = 2900$ mV completely quenches to cotunnelling lines along 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 leads, so that cotunnelling processes can not lead to a finite current any more. Again we observe a slight decrease of the gate capacitances to $C_{g3} = (5.8 \pm 0.2)$ aF and $C_{g5} = (5.5 \pm 0.2)$ aF, and also the mutual capacitances decrease further to $C_{g3,M} = (0.4 \pm 0.1)$ aF and $C_{g5} = (0.4 \pm 0.1)$ aF.

In summary we have demonstrated a high degree of control over the charge distribution in a quantum dot system, where we have split up a long single quantum dot into a tunnel-coupled pair of quantum dots in a controlled way. In the double quantum dot regime, 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. All extracted capacitances are summarized in Table 4.2.

In order to extract the charging energies U_i as well as the mutual charging energy U_M , we need an energy scale which can be provided by the size of the bias triangles at finite bias (van der Wiel et al., 2003). Therefore in Figure 4.4b we show a high-resolution stability diagram of a double quantum dot at a bias of $V_{SD} = -1.5$ mV with barrier gate voltages of $V_{g2} = 2500$ mV, $V_{g4} = 2100$ mV, and $V_{g6} = 3180$ mV, i. e. the outer barriers are set to a higher voltage than before in order to prevent cotunnelling while keeping the interdot tunnel coupling relatively high. 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 $C_{g3} = (5.9 \pm 0.2)$ aF and $C_{g5} = (5.9 \pm 0.2)$ aF, and mutual capacitances of $C_{M,g3} = (0.9 \pm 0.1)$ aF and $C_{M,g5} = (0.9 \pm 0.1)$ aF. The increased values for $C_{M,i}$ is an indication for an indeed increased (capacitive) coupling between the dots.

Since the bias triangle size corresponds to an energy of 1.5 meV, we can extract the charging energies from Figure 4.4b by relating this triangle size to the distance between two adjacent triple point corresponding to successive charge states of the left and right dot. We obtain a charging energy of the left dot $U_1 = (10.6 \pm 0.5)$ meV and of the right dot $U_2 = (9.3 \pm 0.5)$ meV. For the mutual charging energy U_M we extract $U_M = (1.5 \pm 0.2)$ meV. The charging energies of the single dots are very similar to our findings in Section 4.3. Over the whole range of the measurement, the size and shape of the bias triangles is exceptionally stable², which reveals the high degree of control we have over the electrochemical potentials of the quantum dots while keeping the tunnel and capacitive couplings constant owing to the almost negligible cross capacitances.

4.5 CONCLUSION

In conclusion, we have demonstrated the electrostatic formation of highly-tunable single and double quantum dots inside a Ge-Si core-shell nanowire by means of gates. With our gate design we can vary the length of the single quantum dots between 60 nm and 460 nm corresponding to charging energies of the quantum dots varying from ~ 18 meV down to ~ 4 meV. Even for a quantum dot of 460 nm length we have shown the extreme stability of our device in a 32-hour measurement where we have changed the gate voltage over a range of 4 V

² Many reports of gate-defined double quantum dots in literature display strongly changing tunnel couplings when changing the charge occupation, see e.g. Fasth et al. (2005); Liu et al. (2008); Sand-Jespersen et al. (2008); Li et al. (2015b).

while changing the number of holes on the quantum dot by more than 150.

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 highly regular pattern in the stability diagram with constant triangle sizes, indicating again a high degree of control over the tunnel couplings over a large range of gate voltages. This combination of tunability and stability makes this system an ideal platform for further quantum transport studies, see Chapters 5 to 7.

ELECTRIC-FIELD DEPENDENT G-FACTOR ANISOTROPY IN GE-SI CORE-SHELL NANOWIRES

5.1 INTRODUCTION

Quantum computation (Aaronson, 2013; DiVincenzo, 1995; Ladd et al., 2010) has made an enormous leap from a far-fetched promise (Feynman, 1986) to a realistic near-future technology (Vandersypen et al., 2001; Jones et al., 2012; Veldhorst et al., 2015) during the past three decades. Among others, spin systems in the solid state (Loss and DiVincenzo, 1998; Kane, 1998) have been developed into a mature but still very fast-evolving research field. First pioneering results have been achieved in III-V material systems like GaAs (Meirav et al., 1991; Tarucha et al., 1996; Koppens et al., 2006), which suffer from decoherence of the spin states by hyperfine coupling to the nuclear spin bath (Merkulov et al., 2002). Thus in recent years increased research efforts have focussed on group-IV materials like C, Si, and Ge (Laird et al., 2015; Zwanenburg et al., 2013; Amato et al., 2014) that can be purified to only consist of isotopes with zero nuclear spin (Itoh et al., 2003, 1993) and thus exhibit exceptionally long spin lifetimes up to 30 s for nuclear spins (Muhonen et al., 2014) and 28 ms for electron spins (Veldhorst et al., 2014).

Within the realm of group-IV materials, Si-Ge heterostructures with their atomically sharp and defect-free interfaces exhibit exceptionally high charge carrier mobilities (Ismail et al., 1995; Schaffler et al., 1999; Lauhon et al., 2002; Xiang et al., 2006b) and are therefore highly suitable for quantum computation applications (Maune et al., 2012; Kim et al., 2014). Such heterostructures have the intrinsic advantage of spatially separating the charge carriers from disordered surfaces, which puts them ahead of e.g. quantum dots electrostatically defined at the Si/SiO₂ interface in planar silicon systems which intrinsically suffer from the inevitable disorder at the interface between the crystalline Si and the amorphous SiO₂ (de Sousa, 2007).

While sharing the material-inherent advantages with their two-dimensional counterparts, electrostatically defined quantum dots in Ge-Si core-shell nanowires combine intrinsic confinement in two dimen-

sions with *in situ* tunable tunnel barriers in the third dimension (Hu et al., 2007; Roddaro et al., 2008), as also demonstrated in other nanowire systems (Fasth et al., 2005; Nadj-Perge et al., 2010a). Their one-dimensional character leads to unique new electronic properties, especially in the valence band (Csontos et al., 2009). In contrast to the two-dimensional case, there are theoretical predictions of mixing of heavy and light hole states at the valence band edge (Csontos and Zülicke, 2007; Kloeffer et al., 2011). The band mixing gives rise to an enhanced Rashba-type spin-orbit interaction that is predicted to be particularly pronounced in Ge-Si core-shell nanowires (Kloeffer et al., 2011), leading to highly-varying effective g -factors that are strongly anisotropic and electric-field dependent (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., 2013).

Despite these profound theoretical contributions, only few experiments in Ge-Si core-shell nanowires have been reported in recent years. Josephson junctions have been built by combining the nanowires with superconducting contacts (Xiang et al., 2006c). Spin-filling in a many-hole single quantum dot has been shown (Roddaro et al., 2008) and charge sensing has been successfully demonstrated by capacitively coupling a double quantum dot to a single quantum dot (Hu et al., 2007). Spin relaxation (Hu et al., 2012) and spin coherence (Higginbotham et al., 2014b) times have been measured in many-hole double quantum dots and finally indication for spin-orbit coupling has been reported by observing antilocalisation of Coulomb peaks (Higginbotham et al., 2014a). To our knowledge no experiments have been reported that investigate anisotropic effects arising from the one-dimensional character of the nanowires.

In this work we experimentally explore the theoretically predicted 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 spin 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).

Figure 5.1: (a) False-colour atomic-force microscopy (AFM) image of the device (left), and schematic cross-section (right) displaying the p^{++} -doped Si substrate (grey) with 200 nm of SiO₂ (dark red), six bottom gates g_1 - g_6 (light red), each approximately 35 nm wide and with a pitch of 100 nm. The bottom gates are buried under 10 nm of Al₂O₃ (yellow), on top of which the nanowire is placed (green). Ohmic contacts (Ti/Pd 0.5/50 nm, blue) are defined by means of EBL. (b) Current I vs V_{g4} with g_3 and g_5 forming tunnel barriers ($V_{g3} = 2060$ mV, $V_{g5} = 2260$ mV). The curve taken at $V_{SD} = 1$ mV (black) exhibits clear and regular Coulomb oscillations up to $V_{g4} = 2000$ mV. At $V_{SD} = 50$ mV (red curve) a sudden suppression of current above $V_{g4} = 2000$ mV is visible. (c) Numerical differential conductance dI/dV_{SD} plotted vs. V_{SD} and V_{g4} at the same barrier voltages as in (b), displaying several very well-defined, closing Coulomb diamonds.

5.2 GATE-DEFINED QUANTUM DOTS

Our device in Figure 5.1a consists of a p^{++} -doped Si substrate covered with 200 nm thermally grown SiO_2 , on which six bottom gates with a 100 nm pitch are patterned with EBL. The gates are buried by 10 nm Al_2O_3 grown with atomic layer deposition at 100 °C. A single nanowire with a Si shell thickness of approximately 2.5 nm and a Ge core radius of approximately 8 nm 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. All contacts are defined by EBL. The nanowire part above the bottom gates is at no point exposed to the electron beam, which prevents carbon deposition and introduction of defects into the otherwise defect-free Ge core (Ang Li *et al.*, in preparation). All measurements are performed in a dilution refrigerator with a base temperature of 8 mK using DC electronic equipment. A source-drain bias voltage V_{SD} is applied to source, the current I is measured between drain and ground. An effective hole temperature of $T_{\text{hole}} \approx 30$ mK (see also Chapter 3) has been determined in the device by measuring the temperature dependence of the Coulomb peak width (Mueller *et al.*, 2013; Goldhaber-Gordon *et al.*, 1998).

We use this highly flexible and versatile gate design Fasth *et al.* (2005) to electrostatically define a single quantum dot (Kouwenhoven *et al.*, 1997b; Fasth *et al.*, 2005). The two barrier gates g_3 and g_5 give individual control over the tunnel barriers, and a third plunger gate g_4 over the electrochemical potential of the quantum dot. In Figure 5.1b we plot I versus the voltage on g_4 V_{g4} at constant voltages V_{g3} and V_{g5} on the left barrier and right barrier gate, respectively. When applying a high $V_{\text{SD}} = 50$ mV we observe a strong suppression of I for $V_{g4} > 2.5$ V, indicating depletion of the nanowire at $V_{g4} \approx 2.5$ V. At low $V_{\text{SD}} = 1$ mV we observe Coulomb peaks (Kouwenhoven *et al.*, 1997b) with a regular spacing of $\Delta V_{g4} \approx 30$ 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 $V_{g4} \approx 2$ 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 $V_{g4} = 2.5$ V, we can estimate the number of remaining holes to be $N \approx 17$ at $V_{g4} = 2$ V.

A non-linear transport measurement is displayed in Figure 5.1c. In this bias spectroscopy we plot the numerical differential conductance

$dI/dV \equiv dI/dV_{SD}$ versus V_{SD} and V_{g4} , as will be in all following bias spectroscopy plots. The barrier gate voltages V_{g3} and V_{g5} are kept at the same constant voltages as in Figure 5.1b. Formation of a single quantum dot is indicated by regularly-shaped, closing Coulomb diamonds (Kouwenhoven et al., 1997a). This indicates a very clean nanowire device with a very stable electrostatic environment (Dahl Nissen et al., 2012). A number of lines of increased conductance inside the regions where the current is not blocked suggest the existence of several quantized single-particle conduction paths (Kouwenhoven et al., 2001). Their exact nature, however, cannot be determined without further investigation (Escott et al., 2010). The height of the Coulomb diamonds indicates an addition energy of $E_{add} \approx 8 - 10$ meV. The variations in E_{add} cannot be explained by an interacting second quantum dot, which would lead to non-closing diamonds. Orbital shell filling, however, can very well cause such variations (Tarucha et al., 1996; Kouwenhoven et al., 2001). The low number of residing holes ($\approx 25 - 30$) supports this reasoning. Nevertheless, the complicated subband structure of the valence band edge in low-dimensional structures (Winkler, 2003; Kloeffel et al., 2011) in combination with the quasi-one-dimensional orbitals in the quantum dot do not allow for a definitive conclusion without reaching the single-hole regime, which was not possible in our device.

The results in Figure 5.1 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.

5.3 ZEEMAN SPLITTING OF THE ORBITAL GROUND STATE

We now investigate the Zeeman splitting ΔE_Z 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 Figure 5.2a 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 ($V_{g4} \approx 1450$ mV) to the depletion voltage at high bias ($V_{g4} \approx 2.5 - 2.7$ V).

Figure 5.2: (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) (left panel) and ΔE_Z extracted from (b) vs. B together with a linear fit (red line) that yields $g^* = 2.7 \pm 0.1$ (right panel). (c) 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 5.3 were taken along the blue line.

We keep the plunger-gate voltage at a fixed value $V_{g4} = 1454.0$ mV and sweep V_{SD} along the green line in Figure 5.2a at different magnetic fields B while measuring the current (Figure 5.2b). The magnetic field here is applied in plane of the chip perpendicular to the nanowire axis. At $B = 0$, one very pronounced conduction 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 5.2c at $B = 0$ (left plot) and $B = 1$ T (right plot). The spin-degenerate orbital ground state at $B = 0$ (indicated by a single green arrow in Figure 5.2c) 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 in the left panel of Figure 5.2b into energy. The lever arm $\alpha \equiv C_{\text{tot-S}}/C_{\text{tot}}$ with $C_{\text{tot-S}} = C_{\text{tot}} - C_S$, where C_{tot} is the total capacitance of the dot, and C_S is the source capacitance) for this conversion is graphically extracted: the slopes of the Coulomb diamond edges from Figure 5.2a 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 the right panel of Figure 5.2b. 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. 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. This yields an effective g -factor for this transition of $g^* = 2.7 \pm 0.1$. 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 Kloeffer et al. (2011) at the valence band edge of the nanowire Roddaro et al. (2008); Kloeffer et al. (2013).

To summarise, in Figure 5.2 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.

Figure 5.3: dI/dV plotted for different $\vec{B}$ -field directions at constant $B = 1$ T measured along the blue line in Figure 5.2c. 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 (e) 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.

5.4 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.* (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 3a). 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 versus V_{SD} along the blue line in Figure 5.2c. 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 state and calculating the distance between the two peak centres.

First we rotate the magnetic field from the y -axis (perpendicular to the nanowire and in the chip plane, the same direction as in Figure 5.2b) to the z -axis (Figure 5.3a). At $\phi_1 = 0$, the Zeeman splitting of $\Delta E_{Z,y} = (155 \pm 5)$ μeV corresponds to a g -factor of $g_y^* = 2.7 \pm 0.1$ (in agreement with Figure 5.2b). 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 $\Delta E_{Z,z} = (13 \pm 10)$ μeV , corresponding to $g_z^* = 0.2 \pm 0.2$. Figure 5.3d shows a line cut with $\vec{B}$ along the y -axis at $\phi_1 = -\pi$ (black) and $\vec{B}$ along the z -axis at $\phi_1 = -\pi/2$ (green). For the magnetic field along the z -axis the conduction peak is approximately twice as high and also significantly broadened with respect 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 (green curve) with two peaks (dark green) 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 conduction peaks.

For the measurement in Figure 5.3b $\vec{B}$ always points in a direction perpendicular to the nanowire and is rotated from being in the sample plane at $\phi_2 = 0$ (the same field direction as for $\phi_1 = 0$ in Figure 5.3a) to perpendicular to the sample plane at $\phi_2 = \pi/2$. At $\phi_1 = 0$, i.e. along the y -axis, the Zeeman splitting is again $\Delta E_{Z,y} = (154 \pm 5)$ μeV . The Zeeman splitting decreases until it reaches $\Delta E_{Z,x} = (120 \pm 10)$ μeV

along the x -axis, which corresponds to a g -factor of $g_x^* = 2.1 \pm 0.2$. In Figure 5.3e two linecuts from Figure 5.3b 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, i. e. along the direction of the electric field lines, and $\phi_3 = \pi/2$ along the z -axis (see Figure 5.3c). 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 (along the nanowire) again results in a Zeeman splitting of $\Delta E_{Z,z} = (17 \pm 10) \mu\text{eV}$, corresponding to $g_z^* = 0.3 \pm 0.2$.

If we now combine all three rotation experiments, the Zeeman splitting along each of the x -, y -, and z -axis is measured twice with consistent values across experiments for the Zeeman splitting and the effective g -factor. Thus we can summarize our findings from Figure 5.3 as follows: The g -factor for magnetic fields along the nanowire axis is almost zero, $g_z^* = 0.2 \pm 0.2$. The g -factor is maximal for $\vec{B}$ being perpendicular to the nanowire axis and also perpendicular to the electric field from the gates, $g_y^* = 2.7 \pm 0.1$. The g -factor for $\vec{B}$ perpendicular to the nanowire and parallel to $\vec{E}$ is still non-zero, $g_x^* = 2.1 \pm 0.2$, but significantly lower than g_y^* .

Let us now compare our findings with experimental and theoretical results from 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., 2015), 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 of them are 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 $g_{\text{max}}^*/g_{\text{min}}^* \approx 1.4$ for rotation with respect to the E -field axis. For Ge/Si core-shell nanowires, Hu et al. Hu et al. (2012) report 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. 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.

Maier et al. (2013) have investigated the g -factor in Ge-Si core-shell nanowires theoretically. 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. With this geometry, they predict 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 predict a lower g -factor at finite electric fields. In particular, their calculations show a more effective diminishment for $\vec{B} \parallel \vec{E}$ than for $\vec{B} \perp \vec{E}$. Also this is in superb agreement with our findings. Maier *et al.* 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 ‘Direct Rashba Spin Orbit Interaction’ (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.

Even 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.*, which can lead to coupling of more than two subbands by the electric field.

5.5 CONCLUSION

In conclusion, we have fabricated a highly tunable Ge-Si core-shell nanowire device in which we were able to control the hole occupancy of an electrostatically defined quantum dot over a wide range. We have tuned the quantum dot to a charge transition where the spin-degenerate ground state is well-separated from other states. The effective g -factor has been found to be highly anisotropic not only with respect to the nanowire axis but also the electric-field direction. In particular we have found excellent qualitative agreement between our mea-

surements and theoretical calculations Maier et al. (2013). This opens the way to controlled manipulation of the spin-state by simply changing the electric field across the quantum dot by means of gate voltages and thus tuning the two-level system in and out of resonance with a continuous high-frequency electric field.

PAULI SPIN BLOCKADE AND SHELL FILLING IN DOUBLE QUANTUM DOTS

6.1 INTRODUCTION

In this chapter we will tune our device to form a gate-defined double quantum dot, in which we will demonstrate orbital shell filling. We will show current rectification at low source-drain voltages due to Pauli-spin blockade.

6.2 DEVICE DESIGN

Our device is the same as described in Section 5.2. Here, we use five of the six bottom gates to electrostatically define a highly tunable double quantum dot similar to the one in Chapter 4. The gates labelled g_2 , g_4 , and g_6 are initially tuned in such a way that tunnel barriers are formed in the nanowire ($V_{g_2} = 2585 \text{ mV}$, $V_{g_4} = 2210 \text{ mV}$, $V_{g_6} = 3195 \text{ mV}$), while gates g_3 and g_5 serve as plunger gates for individual control of the electrostatic potential in the left and the right quantum dot, respectively (see Figure 6.1). We apply a DC bias across the double quantum

Figure 6.1: The device used in this chapter is the same one as in Chapter 4 and Chapter 5. Gates g_2 , g_4 , and g_6 are tuned to induced tunnel barriers in the nanowire. Gates g_3 and g_5 serve as plunger gates.

dot by applying a voltage V_{SD} ¹ to the source contact and measure the current I between drain and ground.

6.3 FORMATION OF A DOUBLE QUANTUM DOT

In Figure 6.2 we sweep the voltage V_{g3} on gate $g3$ vs. the voltage V_{g5} on $g5$ while applying a bias voltage of $V_{SD} = 2$ mV to the source contact and measuring the current I from drain to ground. This stability diagram clearly shows a highly regular pattern of bias triangles (van der Wiel et al., 2003), indicating the formation of a double quantum dot above gates $g3$ ² and $g5$.³ Note that the review by van der Wiel et al. (2003) is for electron transport, which is why there the honeycomb patterns have a different slope. The 16 bias triangle pairs visible in Figure 6.2 all have very clear and sharp edges. In particular there is no finite current visible along the white $\mu_i(M, N)$ -lines. These lines are guides to the eye added to point out the edges of the typical honeycomb structure of weakly coupled quantum dots (van der Wiel et al., 2003) and denote the lines in gate space where the electrochemical potential of the i^{th} dot ($i = 1$ for left, $i = 2$ right dot) for having M holes in the left and N holes in the right dot is resonant with the Fermi energy μ_j in the adjacent reservoir ($j \in \{S, D\}$ for source and drain reservoir). Crossing the $\mu_1(M, N)$ -lines from right to left, the left dot is filled with one hole at a time, while the $\mu_2(M, N)$ -lines demarcate from top to bottom the addition of one hole at a time to the right dot. Hole transport through the whole double dot structure is only possible when also the electrochemical potential for addition of a hole to the other dot is within the bias window (which is only the case within the bias triangles). Therefore the edges of the honeycombs are not visible in transport experiments⁴. Only in the case of strongly coupled dots cotunnelling processes could still lead to a finite current via a virtual state (van der Wiel et al., 2003), which indicates that our double quantum dot is weakly coupled to the reservoirs. All bias triangles are identical in size and shape, but for all eight bias triangle pairs above $V_{g3} = 500$ mV the interdot charging energy U_M is reduced compared to that of the bias triangle pairs below $V_{g3} = 500$ mV from $U_M(V_{g3} < 500 \text{ mV}) = 1.31 \text{ meV}$ to $U_M(V_{g3} > 500 \text{ mV}) = 1.06 \text{ meV}$. This change in U_M indicates a change in the strength of the interdot tunnel

¹ Throughout this chapter, the term ‘bias’ will be used to refer to the voltage applied to the source contact.

² In the following we will refer to this quantum dot as the ‘left dot’.

³ This quantum dot will be referred to as the ‘right dot’ in the remainder of this chapter.

⁴ Using charge sensing techniques, however, it is possible to map out the honeycomb edges (see e.g. Simmons et al. (2009)).

Figure 6.2: Stability diagram with I plotted in colour scale while sweeping g_5 vs. g_3 at $V_{SD} = 2$ mV. Gates g_2 , g_4 , and g_6 are set to $V_{g2} = 2585$ mV, $V_{g4} = 2210$ mV, and $V_{g6} = 3195$ mV, respectively. 16 regularly shaped bias triangle pairs are visible with effective occupation number (M, N) between them labelled in light blue. White dashed lines are guides to the eye for the honeycomb edges. Blue arrows show distance between adjacent bias triangle pairs. Pink circles mark bias triangles where Pauli spin blockade is observed (see Section 6.4).

coupling between the $\mu_1(1, n)$ - and $\mu_1(2, n)$ -line. In line with this observation, also the $\mu_i(M, N)$ honeycomb edges are more slanted for $M \geq 2$, a signature of an increased cross-capacitance of the left plunger gate (g_3) to the right dot and g_5 to the left dot. We do not observe such a change when altering the occupation number of the right dot.

Let us now focus on the energy that is needed to add the m^{th} hole to the left ($i = 1$) or right ($i = 2$) dot, the so-called addition energy $E_{i,m}^{\text{add}}$. The addition energy for the m^{th} hole to the left dot can be extracted from Figure 6.2 by measuring the distance between the triple points that are connected by the $\mu_2(m - 1, n)$ -line. To convert this distance in gate space into $E_{1,m}^{\text{add}}$ we need an absolute energy scale. For this purpose we use the length of the edge of the bias triangle, which corresponds to the 2 meV bias window. By relating the distance between the triple points to the length of the bias triangle edge we can extract an addition energy of $E_{1,m}^{\text{add}} = 9.8 \text{ meV}$ for $m \in \{2, 3\}$ and $E_{1,m}^{\text{add}} = 10.3 \text{ meV}$ for $m = 1$. The discrepancy between $E_{1,m}^{\text{add}}$ for $m = 1$ and $m \in \{2, 3\}$ is independent of the number of holes in the right dot. This strongly hints on $E_{i,m}^{\text{add}}$ being composed of two major contributions: The (classical) charging energy E_C , and the (quantum-mechanical) orbital energy $E_{\text{orb},i}$. While E_C originates from the Coulomb repulsion between the (charge-carrying) holes and thus always has to be provided, $E_{\text{orb},i}$ is due to a combination of the confinement-induced quantization of the available energy states and the fact that holes are fermions and thus follow Fermi-Dirac statistics. $E_{\text{orb},i}$ therefore is only non-zero if the added hole occupies a different orbital from the holes already residing in the quantum dot. This reasoning gives us strong evidence that the hole numbering for the left dot in Figure 6.2 represents the effective hole number in the highest-energy orbital that gets occupied in this measurement. The energy difference between the $(k - 1)^{\text{th}}$ and the k^{th} orbital in the left dot can thus be calculated by $E_{\text{orb},1} = E_{1,1}^{\text{add}} - E_{1,2}^{\text{add}} = 0.5 \text{ meV}$. A similar value for the orbital energy ($E_{\text{orb}}(380 \pm 60) \mu\text{eV}$) has been extracted from single quantum dot measurements in a Ge-Si core-shell nanowire with comparable quantum dot size (Roddaro et al., 2008).

We find a nearly identical situation for adding holes to the right dot. Following the same analysis as for the left dot, we obtain an addition energy for adding the n^{th} hole of $E_{2,n}^{\text{add}} = 9.5 \text{ meV}$ for $n \in \{0, 2\}$, and $E_{2,n}^{\text{add}} = 10.2 \text{ meV}$ for $n = 1$, resulting in $E_{\text{orb},2} = 0.7 \text{ meV}$.

In summary, the observation of an increased addition energy due to filling of a new orbital for both dots lets us choose the effective number

of holes (m, n) on each dot as displayed in Figure 6.2 in such a way that for $m, n > 0$ they are identical with the occupation numbers for the highest-lying orbitals that get filled in Figure 6.2. We will use this knowledge in the following section to identify bias triangle pairs for which we expect to observe current rectification.

6.4 PAULI SPIN BLOCKADE

Following the reasoning in Section 2.3, in our device with the barriers tuned as in Figure 6.2, the encircled bias triangle pair at the degeneracy $(2,0) - (1,1)$ should exhibit PSB if we apply a negative bias. In Figure 6.3a a zoom of this bias triangle pair for both positive (left panel) and negative (right panel) bias displays a clear asymmetry in the current around zero detuning. A line cut taken along the white dashed lines is plotted in Figure 6.3c (lines with black symbols)⁵. The maximum current for zero detuning at positive bias is $I_{\text{pos}}(\epsilon = 0) = 3.6 \text{ pA}$, at negative bias the current is reduced to $I_{\text{leak}} \equiv I_{\text{neg}}(\epsilon = 0) = 1.6 \text{ pA}$. The current is thus only partially suppressed, comparable values for the resonant ($\epsilon = 0$) leakage current in double quantum dots have been reported in literature (see e.g. Pfund et al. (2007b); Churchill et al. (2008); Nadj-Perge et al. (2010b)). From the line traces taken in the spin-blocked bias direction, we can also extract $\Delta_{\text{S-T}} = 0.5 \text{ meV}$. Note that this value is very close to the E_{orb} we have determined in Section 6.3. This is reasonable since the $(2,0)$ -singlet-triplet splitting involves states originating from successive quantum dot orbitals.

Spin-filling into the orbitals of our double quantum dot predicts PSB for a second bias triangle pair visible in Figure 6.2. Indeed the bias triangle pair marked by the lower right pink circle exhibits current rectification, as shown in Figure 6.3d. As expected, current around zero detuning is here blocked for positive V_{SD} .

Because of the high tunability of our device, we can get a first hint on the origin of the leakage current by changing the tunnel couplings. We do this by following the bias triangle pair at the $(1,1)$ – $(2,0)$ degeneracy while changing V_{g2} and V_{g6} in small, controlled steps. This approach ensures that we are still measuring the same bias triangle pair that is shifted in V_{g3} – V_{g5} gate space due to the finite cross-capacitance between $g2$ ($g6$) and the left (right) dot. In Figure 6.3b we plot the bias triangle

⁵ The energy scale for the detuning axis is, similarly to Section 6.3, obtained by projecting the distance in gate space from the zero-detuning line to the triangle tip onto the 2 meV window opened by applying $V_{\text{SD}} = 2 \text{ mV}$ across the device.

Figure 6.3: (a) Zoom-in of the stability diagram at the $(2,0)$ - $(1,1)$ degeneracy in Figure 6.2 with the same barrier voltages ($V_{g^2} = 2585$ mV, $V_{g^4} = 2210$ mV, $V_{g^6} = 3195$ mV). (b) the same bias triangle pair for lower voltages on the outer barriers ($V_{g^2} = 2570$ mV, $V_{g^6} = 3160$ mV). (c) line cuts along the dashed white lines in (a) and (b). (d) Zoom-in of the stability diagram at the $(1,1)$ - $(0,2)$ degeneracy with the same barrier voltages as in (a). $V_{SD} = 2$ mV for all measurements.

LINE	$I_{\text{neg}}(\epsilon = 0)$ (pA)	$I_{\text{pos}}(\epsilon = 0)$ (pA)	Γ (μeV)	t (μeV)
(1)	3.6	1.6	80	1.4
(2)	10.4	1.2	100	2.6
(3)	5.8	3.3	88	1.9

Table 6.1: Zero-detuning currents for non-blocked (I_{pos}) and blocked (I_{neg}) bias direction, and tunnel couplings extracted from fitting Equation 7 to the line cuts in the left panel of Figure 6.3c.

pair at the $(2,0) - (1,1)$ degeneracy after decreasing V_{g2} by 15 mV and V_{g6} by 35 mV. As expected the bias triangles have shifted to higher V_{g3} and V_{g5} . Furthermore, the strongly enhanced overall current within the non-blocked bias triangle regions suggests significantly increased tunnel couplings. At zero detuning, the situation is more complicated than in Figure 6.3a, since the current varies significantly along the $\epsilon = 0$ line for both bias directions. More specifically, the current at $\epsilon = 0$ in the bias triangle representing the $(2,1) - (2,0) - (1,1) - (2,1)$ charge cycle (line cuts (3) in Figure 6.3c) is $I_{\text{pos}}(\epsilon = 0) = 5.8$ pA in the non-blocked direction and $I_{\text{leak}} = 3.3$ pA in the blocked direction, whereas in the $(1,0) - (1,1) - (2,0) - (1,0)$ bias triangle (line cuts (2) in Figure 6.3c) $I_{\text{pos}}(\epsilon = 0) = 10.4$ pA and $I_{\text{leak}} = 1.2$ pA.

From the $I(\epsilon)$ -curves in the non-blocked bias direction, we can extract the tunnel couplings to the left and right lead $\Gamma_{L,R}$ and the inter-dot tunnel coupling t by fitting with the expression (van der Wiel et al., 2003)

$$I(\epsilon) = \frac{\Gamma_{\text{r}}|t|^2}{(\epsilon/h)^2 + \Gamma_{\text{r}}^2/4 + |t|^2(2 + \frac{\Gamma_{\text{r}}}{\Gamma_{\text{l}}})}, \quad (6)$$

where h is the Planck constant. Although $I(\epsilon = 0)$ is different for the two bias triangles in Figure 6.3b, within a bias triangle the current does not vary a lot, so that we can assume $\Gamma_{\text{l}} = \Gamma_{\text{r}} \equiv \Gamma$, which simplifies Equation 6 to

$$I(\epsilon) = \frac{\Gamma|t|^2}{(\epsilon/h)^2 + \Gamma^2/4 + 3|t|^2}. \quad (7)$$

In order to extract Γ and t , we fit Equation 7 to the $I(\epsilon)$ -curves in Figure 6.3c only for $\epsilon \leq 0$, because for $\epsilon > 0$ inelastic processes that are

not considered in Equation 6 lead to an increased current, as can be inferred from the asymmetry of $I(\epsilon)$ around $\epsilon = 0$ in the left panel of Figure 6.3c (the same approach has been used e. g. by Li et al. (2015b)). The values for Γ and t extracted from these fit are summarized in Table 6.1. Although the tunnel coupling to the leads Γ exhibits the biggest change in absolute terms between line cuts (1) and (2) from 80 μeV to 100 μeV , the relative change for the interdot tunnel coupling t is much higher (from 1.4 μeV to 2.6 μeV). The change in Γ can be readily explained by the tunnel barrier change that we have induced by changing V_{g2} and V_{g6} . The increased value for t reflects an increased wave function overlap between states in the left and the right dot. This can be explained (as mentioned before) by the finite cross capacitance between $g2/g6$ and the quantum dots which may affect the shape of the wave functions. For all configurations in this chapter, 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 the leakage current in Table 6.1 and how it relates to the tunnel couplings. Although both Γ and t increase significantly by tuning the device from configuration (1) to (2), I_{leak} even *decreases*. 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. On the other hand, comparing configuration (1) to (3) we observe an increase of I_{leak} by a factor of two, along with an increased t , even though the latter grows only by a factor of 1.4.

In summary, we have observed PSB in the bias triangle pairs for the effective (2,0) – (1,1) and (1,1) – (2,0) charge degeneracy points, as expected from the shell filling picture obtained in Section 6.3. From line traces along the detuning axis, we have determined tunnel rates as well as values for the zero-detuning current in both blocked and unblocked bias direction.

6.5 CONCLUSION

In this chapter, we have successfully formed a gate-defined double quantum dot, in which we have separate control over the tunnel couplings and the electrochemical potential of the coupled quantum dots and. We are able to change the charge occupation of each dot by several holes while maintaining exceptionally stable tunnel couplings. Capitalizing on this we have been able to tune the dots to a regime where we observe shell filling in both quantum dots and therefore identify

effective charge occupation numbers for the highest-lying orbitals in each quantum dot. Based on this we have identified and successfully measured current rectification by Pauli spin blockade for both the effective $(2,0)-(1,1)$ and $(1,1)-(0,2)$ charge occupations. For the $(2,0)-(1,1)$ occupation degeneracy we have observed a change in the leakage current through the spin-blocked double quantum dot when changing the tunnel couplings.

The results in this chapter pave the way for using Pauli spin blockade as a means of spin detection by spin-to charge conversion, for which first experiments have been performed (Hu et al., 2012; Higginbotham et al., 2014b), although there is still room for improvement. In order to optimize spin relaxation and decoherence times, a more thorough analysis of the leakage current processes is needed, which we will carry out in Chapter 7.

ANISOTROPIC LEAKAGE CURRENT IN THE PAULI SPIN BLOCKADE REGIME

7.1 INTRODUCTION

In Section 6.4 we have conclusively established PSB in our double quantum dot. Yet the leakage current I_{leak} is not perfectly zero in the blocked state, and is related to Γ and t in a non-trivial way. In this section, we will measure the magnetic-field dependence of the leakage current for different field directions. An anisotropy in the dependence of I_{leak} on B can be expected, because, as shown in Chapter 5, two other axes are of importance for the interaction of magnetic fields with spin states in our system: the nanowire axis $\vec{a}_{\text{NW}}$ itself, but also the electric fields $\vec{E}$ induced by our gates that point perpendicular to the nanowire axis and out-of-plane of the chip. A short overview of possible causes for I_{leak} is given in Section 2.4.

In this chapter we will perform magnetospectroscopy measurements on the bias triangles shown in Figures 6.3a to 6.3b with the magnetic field pointing along different directions. We will demonstrate a highly anisotropic evolution of the leakage current in the magnetic field and identify spin-flip cotunnelling as the dominating leakage mechanism at low magnetic fields. We will make use of this to extract a g -factor anisotropy that differs qualitatively from our findings in Chapter 5. Furthermore we will argue that spin-orbit coupling leads to a finite leakage current at high magnetic fields. Finally we will present signatures of an anisotropic Coulomb interaction between the holes forming the spin-blocked state.

7.2 DEVICE DESIGN

Our device is the same as described in Section 5.2. Here, we use five of the six bottom gates to electrostatically define a highly tunable double quantum dot as demonstrated in Chapter 6. The gates labelled g_2 , g_4 , and g_6 are initially tuned in such a way that tunnel barriers are formed in the nanowire ($V_{g_2} = 2585 \text{ mV}$, $V_{g_4} = 2210 \text{ mV}$, $V_{g_6} = 3195 \text{ mV}$), while gates g_3 and g_5 serve as plunger gates for individual control of the electrostatic potential in the left and the right quantum dot, respec-

Figure 7.1: The device used in this chapter is the same one as in Chapter 4 and Chapter 5. Gates g_2 , g_4 , and g_6 are tuned to induce tunnel barriers in the nanowire. Gates g_3 and g_5 serve as plunger gates.

tively (see Figure 7.1). We apply a DC bias across the double quantum dot by applying a voltage V_{SD} ¹ between the source contact and ground, and measure the current I between drain and ground.

7.3 MAGNETOSPECTROSCOPY OF THE LEAKAGE CURRENT

For the magnetospectroscopy measurements that we will discuss now, we adopt the numbering introduced in Figure 6.2 and Table 6.1 for measurements of the same bias triangle pair at ‘high’ barrier voltages (configuration (1’)), and ‘low’ barrier voltages with ‘low’ (2’) and ‘high’ leakage current (3’). We do not exactly follow the line cuts in Figure 6.3, but instead only sweep one gate, which gives us a slightly higher resolution and shorter sweep times. The new line cut positions here are

(1’) $V_{g3} = 479.8 \text{ mV}$ fixed, $V_{g5} [343.5, 346.5] \text{ mV}$, Figure 7.2a

(2’) $V_{g5} = 360.0 \text{ mV}$ fixed, $V_{g3} [486.0, 489.0] \text{ mV}$, Figure 7.2b

(3’) $V_{g3} = 483.8 \text{ mV}$ fixed, $V_{g5} [354.7, 357.7] \text{ mV}$, Figure 7.2c.

In Figures 7.2a to 7.2c we plot I versus ϵ_0 and $B \equiv |\vec{B}|$ for (1’)-(3’). We conduct magnetospectroscopy measurements along three different directions of $\vec{B}$ for each configuration:

- $\vec{B} \parallel \vec{a}_{NW}$ and $\vec{B} \perp \vec{E}$ (left column of Figure 7.2),
- $\vec{B} \perp \vec{a}_{NW}$ and $\vec{B} \perp \vec{E}$ (middle column),

¹ Throughout this chapter, the term ‘bias’ will be used to refer to the voltage applied to the source contact with respect to ground.

- $\vec{B} \perp \vec{a}_{\text{NW}}$ and $\vec{B} \parallel \vec{E}$ (right column)

Here $\epsilon_0 \equiv \epsilon(B = 0)$ 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. Therefore $\epsilon = 0$ shifts in terms of energy when the electrochemical potentials of these two states change, which will be visible as a shifted zero-detuning point with respect to the fixed energy scale ϵ_0 . The green dashed lines in Figure 7.2 are guides to the eye for $\epsilon(B) = 0$, which we will refer to in the following as the ‘S-onset’. The lifting of blockade at $\epsilon = \Delta_{\text{S,T}}$ is indicated by the blue dashed lines, we will refer to this as the ‘T₀-onset’ in the remainder of the chapter. We sweep the magnetic field both in the positive and negative direction to distinguish dot-intrinsic effects from others, e. g. thermal drift because of heating due to sweeping the magnet.

For all $I_{\text{leak}}(\epsilon_0, B)$ plots in Figures 7.2a to 7.2c, we scan the $I(\epsilon)$ line traces and detect the maximum of the lowest-energy peak as $\epsilon = 0$. We plot line traces for the leakage current at constant detuning in Figure 7.3: Figure 7.3a shows the leakage current for all magnetic field directions and all configurations at zero detuning $I_{\text{leak}}(B) \equiv I_{\text{leak}}(B, \epsilon = 0)$, in Figure 7.3b we plot line traces at $\epsilon = 150 \mu\text{eV}$ for $\vec{B} \perp \vec{a}_{\text{NW}}, \perp \vec{E}$, and in Figure 7.3c for $\vec{B} \perp \vec{a}_{\text{NW}}, \parallel \vec{E}$ line cuts are shown at $\epsilon = 150 \mu\text{eV}$ (left panel), and a high-resolution scan at $\epsilon = 0 \mu\text{eV}$ for (2') (right panel). The average background current of the Coulomb blocked region ($\sim 0.4 \text{ pA}$) in each plot of Figure 7.2 has been subtracted from all line traces in Figure 7.3.

In the following sections we will discuss the magnetospectroscopy measurements displayed in Figures 7.2 to 7.3 for each magnetic field direction.

7.4 MAGNETOSPECTROSCOPY ALONG $\vec{B} \parallel \vec{a}_{\text{NW}}$ AND $\vec{B} \perp \vec{E}$

We start our discussion with the measurements for $\vec{B}$ parallel to the nanowire axis (left column in Figure 7.2). For all three configurations (1')–(3'), the T₀-onset does not change significantly with increasing $B \equiv |\vec{B}|$, whereas the S-onset shifts to positive ϵ_0 -values for increasing B . This behaviour has also been observed in other experiments (Kyriakidis et al., 2002; Fujisawa et al., 2003; Johnson et al., 2005a) and can be explained by orbital effects of the magnetic field (Wagner et al., 1992; van der Wiel et al., 1998; Kyriakidis et al., 2002), where an interplay of the Coulomb interaction between the two holes in the (2,0)

Figure 7.2: $I(\epsilon, B)$ plots in the spin-blocked bias direction for the three different configurations discussed in Section 6.4 and different magnetic field directions. In the left column, B is parallel to the nanowire axis, in the centre column B is perpendicular to the nanowire and parallel to the electric field, and perpendicular to the nanowire and parallel to the electric field in the right column. Configuration (1') is plotted in (a), (2') in (b), and (3') in (c).

charge state and the different symmetries of the wave function for S- and T-states induces much stronger shifts of the electrochemical potentials of the S(2,0) states than the T(2,0) states.² The zero-detuning line cut in Figure 7.3a shows a qualitatively identical dependence of I_{leak} on B for all three configurations: The leakage current has a maximum at $B = 0$ and decreases for increasing magnetic field. Within the measurement window, no clear saturation behaviour is observed, so that conclusive statements about a finite minimum leakage current $I_{\text{leak}}^{\text{min}}$ at high magnetic fields³ can only be extracted from a peak fit. Similar $I_{\text{leak}}(B)$ -curves with peak widths of several hundred millitesla have been reported in other systems (Lai et al., 2011; Yamahata et al., 2012; Li et al., 2015b) and were explained by spin-flip cotunnelling. By fitting Equation 1 to the zero-detuning line traces we find for the three configurations

$$(1') \quad g_z^* = 0.3 \pm 0.1, \quad I_{\text{leak}}^{\text{min}} = (-0.7 \pm 0.1) \text{ pA}$$

$$(2') \quad g_z^* = 0.4 \pm 0.1, \quad I_{\text{leak}}^{\text{min}} = (-0.8 \pm 0.1) \text{ pA}$$

$$(3') \quad g_z^* = 0.3 \pm 0.1, \quad I_{\text{leak}}^{\text{min}} = (-1.2 \pm 0.1) \text{ pA}$$

if we set the hole temperature as $T_{\text{hole}} = 40 \text{ mK}$ in the model. This slightly higher temperature than measured in Section 3.5 is generally observed during magnetic-field sweeps and probably originates from additional heat load from the magnet coils. The fits are plotted alongside the data in the left panel of Figure 7.3a). Also at finite detuning the trend is the same, but here we operate at the noise limit which obscures the very broad peak (not shown). We note that at both zero and finite detuning a residual current $I_{\text{leak}}^{\text{min}}$ remains at high magnetic fields. This indicates a second leakage process that is efficient at finite magnetic field, e.g. spin-orbit interaction, but further evidence is needed for profound conclusions. The obtained g-factors for a magnetic field applied parallel to the nanowire axis are consistent with our findings for a single quantum dot in Chapter 5, where $g_z^* = 0.2 \pm 0.2$. By extracting U_{M} and Δ from Figures 6.3a to 6.3b, and using Equation 2 we calculate from the c -values the following tunnel coupling to the leads Γ :

$$(1') \quad U_{\text{M}} = 1.2 \text{ meV}, \quad \Delta = 0.9 \text{ meV}, \quad \Gamma = 113 \text{ } \mu\text{eV}$$

$$(2') \quad U_{\text{M}} = 1.1 \text{ meV}, \quad \Delta = 1.7 \text{ meV}, \quad \Gamma = 204 \text{ } \mu\text{eV}$$

² This process affects (2,0) states much more than (1,1) states because of the significantly higher Coulomb interaction.

³ In this context, ‘high’ refers to magnetic fields high enough to quench the spin-flip cotunnelling induced leakage current at low B , see the remainder of the paragraph.

$$(3') \ U_M = 1.1 \text{ meV}, \Delta = 0.5 \text{ meV}, \Gamma = 96 \mu\text{eV}$$

If we compare these values to the values extracted from the non-blocked current ((1) $\Gamma = 80 \mu\text{eV}$, (2) $\Gamma = 100 \mu\text{eV}$, (3) $\Gamma = 88 \mu\text{eV}$, see also Table 6.1), we observe a good agreement for (1') and (3'), but a discrepancy by a factor of two for (2'). To weigh the two different methods for obtaining Γ , we point out that the I_{leak} should depend on Δ for a given Γ , i.e. vary along the baseline of the triangle. We do not observe this for our bias triangle pair, and also other groups have reported spin-flip cotunnelling dominated leakage currents with no change in I_{leak} along the zero-detuning line (Yamahata et al., 2012). Therefore we deem the Γ 's that we calculate here with the spin-flip cotunnelling formalism merely a rough estimate.

Two findings are at the core of this first part of our spin-blockade magnetospectroscopy: we can explain the I_{leak} -peak found at $B = 0$ with spin-flip cotunnelling and obtain $g_z^* = 0.3 \pm 0.1$ (consistent with Chapter 5), and we find a residual leakage current that hints at additional leakage processes.

7.5 MAGNETOSPECTROSCOPY ALONG $\vec{B} \perp \vec{a}_{\text{NW}}$ AND $\vec{B} \perp \vec{E}$

We now let the magnetic field point perpendicular to both the nanowire and the electric field. The T_0 -onset stays at constant ϵ_0 when changing the magnetic field whereas the S-onset shifts to higher ϵ_0 for increasing B , qualitatively the same behaviour as for B parallel to the nanowire. Quantitatively, the S-onset shift is bigger here than for $B \parallel \vec{a}_{\text{NW}}$, which indicates a stronger Coulomb interaction for this magnetic-field direction. The zero-detuning line cuts (see middle panel in Figure 7.3) exhibit a more complex structure than for B parallel to the nanowire: while we observe one broad peak for (3'), the curves for configurations (1') and (2') suggest the superposition of at least two peaks: a narrow peak dominates up to $B \approx 0.2 \text{ T}$, and is superimposed on a broader peak that is visible up to $B \approx 0.5 \text{ T}$ in (1') and over the whole range in (2'). At finite detuning (Figure 7.3b) these broader peaks are not observed, which permits us to perform more precise fits of the central peaks. Assuming again spin-flip cotunnelling as the cause for the leakage current we obtain with Equation 1

$$(1') \ g_y^* = 1.5 \pm 0.2 \text{ at } \epsilon = 150 \mu\text{eV},$$

$$(2') \ g_y^* = 1.2 \pm 0.2 \text{ at } \epsilon = 150 \mu\text{eV},$$

$$(3') \ g_y^* = 0.3 \pm 0.1 \text{ at } \epsilon = 0 \mu\text{eV}.$$

Figure 7.3: Line cuts at fixed ϵ from Figure 7.2 (open circles) alongside fitted curves (solid lines, see text). (a) $\epsilon = 0$ line cuts for configurations (1')-(3') and $\vec{B}$ pointing along different axes. (b) line cuts taken at $\epsilon = 150 \mu\text{V}$ for $\vec{B} \perp \vec{a}_{NW} \perp \vec{E}$, and (c) at $\epsilon = 150 \mu\text{V}$ for $\vec{B} \parallel \vec{a}_{NW} \parallel \vec{E}$ (left panel) and high-resolution line trace at $\epsilon = 0 \mu\text{V}$ for (2') with $\vec{B} \parallel \vec{a}_{NW} \parallel \vec{E}$ (right panel).

The g -factors of (1') and (2') are consistent with each other but significantly lower than what we have found for this magnetic field direction in Chapter 5, where $g_y^* = 2.7 \pm 0.1$ was maximal. Taking also into account the Γ -values we have obtained in Section 6.4, which are of the same order or even higher than in other reports of spin-flip cotunnelling-induced leakage current (see Lai et al. (2011); Yamahata et al. (2012); Li et al. (2015b)), we conclude that spin-flip cotunnelling is also consistent with these measurements. The much lower g^* indicated by the broader peak in curve (3') of Figure 7.3 is observed for all ϵ and its cause remains unknown. In principle, a possible explanation would be an increased temperature, because the peak width for $I_{\text{leak}}(B)$ due to spin-flip cotunnelling is actually dependent on both g^* and T . For all fits we have set $T \equiv 40$ mK, as measured by the thermometer thermally anchored to the cold finger. But if we now assume a fixed $g^* \equiv 1.2$ (as for (2')), we obtain a temperature $T = 165$ mK. As there is no reason for such a significantly increased T_{hole} in this particular measurement, we consider this possibility highly unlikely.

Due to the higher effective g -factor in (1') and (2') than was the case for $\vec{B} \parallel \vec{a}_{\text{NW}}$, the spin-flip cotunnelling induced leakage current is exponentially suppressed for magnetic fields above $B \approx 0.25$ T and we obtain a constant residual leakage current at $\epsilon = 150$ μeV of ~ -0.3 pA for (1') and ~ -0.1 pA for (2') using the fits with Equation 1. At zero detuning, the minimum observed current is ~ -0.7 pA for (1') and ~ -0.2 pA for (2'), but both values might be overestimated because of the additional features at high magnetic fields. For (3') the residual current at zero detuning as extracted from the fit is ~ -0.8 pA. These features might also have the same origin as the exceptionally broad peak for (3'), which we cannot explain solely with spin-flip cotunnelling.

In summary, for $B \perp E$, $\perp$ NW we observe a peak in the leakage current for $B = 0$, which we explain with spin-flip cotunnelling and find $g_z^* \approx 1.4$ for (1') and (2'), and $g_z^* \approx 0.3$ for (3'). We cannot reproduce all features visible in the zero-detuning line traces assuming only spin-flip cotunnelling as a leakage process. The remaining leakage current is lower than for $\vec{B} \parallel \vec{a}_{\text{NW}}$, although obscured by additional broader peak-like features.

7.6 MAGNETOSPECTROSCOPY ALONG $\vec{B} \perp \vec{a}_{\text{NW}}$ AND $\vec{B} \parallel \vec{E}$

The third high-symmetry direction for B in our system is parallel to E and perpendicular to the nanowire. As for the other two magnetic field

directions we find a peak around $B = 0$ in the $I_{\text{leak}}(B)$ -curves at $\epsilon = 0$ for all three configurations (1')-(3'). Just like in Section 7.5 we observe a non-constant background that is hard to distinguish from the actual peak in the case of (1') and (3') (see right panel of Figure 7.3a). At $\epsilon = 150 \mu\text{eV}$ the central peaks are well-distinguished from the background for both configuration. For all three configurations (1')-(3') the very narrow central peaks exhibit a noticeable asymmetry, which we assign to the sweeping rate of the magnet. This assumption is supported by a high-resolution magnetic field sweep at a ten times lower sweeping rate (5 mT min^{-1} instead of 50 mT min^{-1}) for (2'), which we plot in the right panel of Figure 7.3c. At this low sweeping rate the peak is indeed more symmetric and we can utilize this high-resolution measurement to also perform the fitting of the central peak.

Following this reasoning, we fit the spin-flip cotunnelling formula Equation 1 to the experimental data as follows: we use the data obtained at $\epsilon = 150 \mu\text{eV}$ (left panel of Figure 7.3c) for (1') and (3'), and the high-resolution sweep at $\epsilon = 0$ for (2'), which results in

$$(1') \quad g_x^* = 3.7 \pm 0.5,$$

$$(2') \quad g_x^* = 3.9 \pm 0.1,$$

$$(3') \quad g_x^* = 4.0 \pm 0.5.$$

These values are substantially higher than the g_y^* values obtained for $B \perp E$ (see Section 7.5), as opposed to our findings in Chapter 5, where $g_y^* > g_x^*$. One possible explanation for this discrepancy is that, in contrast to the measurements performed in Chapter 5, it is very likely that we do not operate in the lowest-energy subband of the nanowire. By using the same estimate as for the single quantum dot in Chapter 5, 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 in Chapter 5 take into account only the quasi-degenerate two lowest-lying subbands, and other theoretical work suggests that properties such as the heavy-hole light-hole mixing is very different for different subbands (Csontos and Zülicke, 2008; Csontos et al., 2009). Note also the significantly lower error for g_x^* obtained from (2'), which reflects the higher measurement resolution, but still results in the same g_x^* well within the measurement errors, which justifies our utilization of the data obtained at higher sweep rates despite the asymmetry of the peaks versus B .

The leakage current at magnetic fields exceeding $B \approx 0.1 \text{ T}$, where spin-flip cotunnelling is efficiently suppressed because of the high effec-

tive g -factor, exhibits a qualitatively different behaviour than for $B \perp E$, $\perp \vec{a}_{\text{NW}}$ (Section 7.5): up to $B \approx 0.3$ T the leakage current is minimal and increases again for $B > 0.3$ T. This can be observed at finite ϵ in the left panel of Figure 7.3c for all three configurations, whereas at $\epsilon = 0$ we only see this behaviour for (2'), since there we do not observe the broader peaks up to ± 0.5 T for (1') and (3') that obscure the current minimum for these configurations (and which we can not explain without further investigation). Up to $B = 1$ T the leakage current does not fully saturate, although the slope reduces at least for (2') at both $\epsilon = 0$ and $\epsilon = 150 \mu\text{eV}$ 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 as described in Section 2.4.3. If we take the line trace for (2') at $\epsilon = 0$ in the right panel of Figure 7.3a, we find a minimal leakage current of $I_{\text{leak}}^{\text{min}} = (0.1 \pm 0.1)$ pA. From Section 2.4.3 we know that $I_{\text{leak}}^{\text{max}} = 9I_{\text{leak}}^{\text{min}}$ for spin-orbit induced leakage. If we now assume the minimum leakage current for (2') to be exclusively due to spin-orbit interaction we expect $I_{\text{leak}}^{\text{max}} = (1 \pm 1)$ pA at high fields. This estimate is in reasonable agreement with the value for $I_{\text{leak}}^{\text{max}}$ we obtain by fitting the data excluding the central peak (see red line in the right panel of Figure 7.3a) with Equation 3, where we find $I_{\text{leak}}^{\text{max}} = (-1.6 \pm 0.3)$ pA and $B_C = (1.0 \pm 0.2)$ T. Since at $\epsilon = 0$ $I_{\text{leak}}^{\text{max}} = 4e\Gamma_{\text{rel}}$ applies, where Γ_{rel} is the spin relaxation rate (see Section 2.4.3), 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 et al., 2015b), 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 emphasise again that this is a mere estimate, because we do not directly measure current saturation at high B , and also note that the critical magnetic field, which we obtain applying the theory from Section 2.4.3, is at the order of ~ 1 T. Danon and Nazarov (2009) neglect Coulomb interaction effects of B , which are of relevance in our case at such field strengths (see also next paragraph). Therefore also other spin-orbit related effects can provide a significant contribution to the increasing leakage current we observe, e. g. Coulomb-interaction mediated spin-spin coupling which scales with B^2 (Flindt et al., 2006; Trif et al., 2007), or coupling between spins and phonons (Khaetskii and Nazarov, 2001; Golovach et al., 2004).

Another peculiarity of the measurements for $B \parallel E$ is the T_0 -onset at $\epsilon = \Delta_{\text{S-T}}$, as indicated by the blue dashed lines in Figure 7.2a. In contrast to the measurements along the other magnetic field directions, for the T_0 -onset we observe two different scenarios when $B \parallel E$: In configuration (1') we see a two-step onset of current above the S-onset,

Figure 7.4: Line traces from Figure 7.2 at constant magnetic field $\vec{B} \parallel \vec{E}$ for configuration (1') at $B = 0$ (black) and $B = 0.7$ T (grey) in the left panel, and for configuration (2') at $B = 0$ (red) and $B = 0.7$ T (magenta) in the right panel. S marks the S-onset, T_0 the T-onset, and $T_{\pm}$ the alignment between T_+ (1,1) and T_0 (2,0) or T_0 (1,1) and T_- (2,0) that is split from the T-onset by E_Z .

with a first step that shifts to lower ϵ_0 (as indicated by the black dashed line in Figure 7.2a), and a second current step (blue dashed line) that remains constant at $\epsilon_0 \approx 0.5$ meV. In (2') and (3') we only observe one current step (like in all measurements for the other B directions), which we again identify as the T_0 -onset, but which now shifts to higher ϵ_0 for increasing B . For more clarity we plot $I(\epsilon)$ line traces for configurations (1') and (2') at both $B = 0$ and $B = 0.7$ T in Figure 7.4. In the first scenario (1') (left panel in Figure 7.4), we observe an additional current step for finite magnetic fields (gray line) between the S-onset and the T_0 -onset. The current at this step (~ 1 pA) is much lower than at the T_0 -onset (~ 6 pA), which indicates that the first current step is due to leakage from alignment between either $T_+(1,1)$ and $T_0(2,0)$ or $T_0(1,1)$ and $T_-(2,0)$, while the T_0 -onset (i.e. alignment of the $T_0(1,1)$ and $T_0(2,0)$ states) stays at constant energy. For both processes we expect a lowering of the necessary detuning with respect to $\epsilon = \Delta_{S-T}$ that equals the Zeeman energy. We observe a splitting between the first and second step of $\Delta_{T_0-T_{\pm}} \approx (150 \pm 20) \mu\text{eV}$ at $B = 0.70$ T (as indicated in Figure 7.4, left panel). Assuming this corresponds to the Zeeman splitting between T_0 and $T_{\pm}$ we calculate $g^* = 3.7 \pm 0.4$, which is in very good agreement with g^* as extracted from the spin-flip cotunnelling fit of $I_{\text{leak}}(B)$. No such additional leakage path is observed for the other two magnetic-field directions, which hints at a mixing of the triplet states that is related to the electric field direction. Such mixing can be caused by spin-

orbit coupling (Danon and Nazarov, 2009), which couples subbands of different spin character at finite electric fields, and the coupling is indeed expected to be anisotropic with respect to $\vec{E}$ (Kloeffel et al., 2011) and also causes the g-factor anisotropy we observe in Chapter 5.

Let us now turn to the right panel of Figure 7.4, where the $I(\epsilon)$ line traces at $B = 0$ and $B = 0.7$ T are plotted for configuration (2'). We observe a significant shift to higher energies for the T_0 -onset, as opposed to (1') in the left panel where the T_0 -onset does not shift. Between $B = 0$ and $B = 1$ T this shift amounts to $\Delta T = (280 \pm 10)$ μ eV in (2'), whereas the shift of the S-onset sums up to $\Delta S = (330 \pm 10)$ μ eV. For the same direction of B , in (1') we only find $\Delta S = (230 \pm 10)$ μ eV. This enhanced ΔS along with the pronounced ΔT 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 other directions of B (Kyriakidis et al., 2002; Kouwenhoven et al., 2001). 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. The fact that we only observe this enhanced Coulomb interaction only in (2') and (3') but not in (1') can be explained by the different interdot tunnel couplings (see Table 6.1), which change the effective charge distribution of the (1,1) and (2,0) states (van der Wiel et al., 2003).

To sum up, we observe a peak in I_{leak} that we interpret as spin-flip cotunnelling induced leakage current. We obtain a very high effective g-factor of $g_x^* \approx 3.9$ that is significantly higher than g_y^* , in contrast to our findings in Chapter 5, which we explain by the occupation of different subbands with different spin character. At higher B we notice an increasing I_{leak} , which we assign to spin-orbit coupling induced mixing of the spin states. We also observe an enhanced Coulomb interaction for the measurements configurations with increased tunnel coupling when $\vec{B} \parallel \vec{E}$.

Let us now briefly compare our findings with existing literature and discuss the implications for spin-orbit qubits. Pribiag 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 co-

tunneling 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 and Nazarov, 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 B along the nanowire (Hu et al., 2012) might be extended by an order of magnitude when measured parallel to E according to the data presented here.

7.7 CONCLUSION

In this chapter, we have analysed several aspects of the leakage current through a Pauli spin-blocked double quantum dot, by applying magnetic fields up to 1 T along three different axes for three different configurations of the tunnel couplings. We have found the leakage at small magnetic fields to be dominated by spin-flip cotunnelling for all directions and configurations. This has enabled us to extract effective g-factors, which we have found to be highly anisotropic. We have found the g-factor to be maximized for $B \parallel E$ and $B \perp \vec{a}_{\text{NW}}$, where $g_x^* = 3.9 \pm 0.1$ is exceptionally high, also compared to literature on Ge-Si core-shell nanowires (Roddaro et al., 2008; Hu et al., 2012) ($g^* = 1.0 - 2.2$). This is different from our findings in Chapter 5, where g^* is maximal for $B \perp E, \perp \vec{a}_{\text{NW}}$. We explain this difference by the significantly higher occupation number of each quantum dot in this chapter (~ 70) than in Chapter 5 (~ 35) resulting in orbital states of different subbands with a different spin character being involved.

At magnetic fields high enough to quench the spin-flip cotunnelling induced leakage current, we have found a residual leakage current. While for $B \perp E, \perp \vec{a}_{\text{NW}}$ the leakage current appears to be constant and very low ($\sim 0.1 \text{ pA}$), we observe an increasing leakage current in B for $B \parallel E, \perp \vec{a}_{\text{NW}}$, which we can reproduce reasonably assuming the tunnel coupling being modulated by spin-orbit coupling (as described in Section 2.4.3). This signature of a significant spin-orbit interaction strength in Ge-Si core-shell nanowires is in line with calculations (Kloeffel et al., 2011). Also the very low I_{leak} for magnetic fields $\vec{B} \parallel \vec{E}, \perp \vec{a}_{\text{NW}}$ up to

1 T is in agreement with this mechanism, since the critical field for spin-orbit coupling to be effective is expected to be highly anisotropic (Danon and Nazarov, 2009).

A third aspect we have investigated in this chapter is the energy shift of the S-onset (i.e. $\epsilon = 0$) and the T_0 -onset (i.e. $\epsilon = \Delta_{S-T}$) in a finite magnetic field. Theory predicts a much stronger shift for the S-onset than for the T_0 -onset due to the higher Coulomb interaction (Merkel et al., 1991; Kyriakidis et al., 2002; Kouwenhoven et al., 2001), which is in very good agreement with what we observe in our experiments. For $B \perp E, \perp \vec{a}_{NW}$ and $B \perp E, \parallel \vec{a}_{NW}$ we find the T_0 -onset to not shift at all within the measurement resolution, while the S-onset shifts significantly to higher energies. With $B \parallel E, \perp \vec{a}_{NW}$ we observe two different behaviours: For the double quantum dot configuration with higher barriers to the leads we observe qualitatively the same as for the other B -directions with an additional T_0 - $T_{\pm}$ leakage path. For the configuration with lower lead barriers we observe an increased shift for the S-onset and also a significant shift of the T_0 -onset, which suggests an increased Coulomb interaction. This has the trivial but important consequence that in the latter case one needs higher magnetic fields for Δ_{S-T} to become zero than for the other situations, which means that Pauli spin blockade can be observed for a much bigger B -range.

The complex and anisotropic interplay of spin-flip cotunnelling, spin-orbit interaction, and hole-hole Coulomb interactions massively influences spin relaxation and spin decoherence and is therefore of high relevance for possible quantum computing schemes relying on read-out of the spin-state via spin-to-charge conversion by PSB (Petta et al., 2005; Koppens et al., 2006; Pribyl et al., 2013; Higginbotham et al., 2014b). Our work suggests that the unique properties of the one-dimensional hole states in Ge-Si core-shell nanowires provide the possibility to optimize the different parameters in order to reach long spin lifetimes while retaining efficient spin manipulation with electric fields.

CONCLUSION AND OUTLOOK

Ge-Si core-shell nanowires are extremely suitable for the formation of gate defined quantum dots, as we have conclusively shown in Chapter 4. This has allowed us to study single hole states under the influence of electric and magnetic fields (Chapters 5 to 7). We have found properties like the effective g-factor to be strongly anisotropic due to the one-dimensional confinement of the holes inside the nanowire and, even more interestingly, with respect to a static electric field.

8.1 IMPLICATIONS FOR FUNDAMENTAL PHYSICS

From a physicist's point of view, these findings are of intrinsic interest because they shed light on the peculiarities of the valence band edge in one dimension. Even in bulk, the valence band exhibits interesting behaviour due to its p-like character that leads to the heavy-hole light-hole degeneracy at the valence band edge for $\vec{k} = 0$. Confinement into two dimensions already causes new effects due to mixing of the two subbands at finite $\vec{k}$. But only if we further reduce the spatial degree of freedom to one, we end up with subbands occupied by mixtures of heavy holes and light holes for all $\vec{k}$. Moreover, the mixing varies strongly between the different subbands, giving rise to, e. g., a different effective g-factor for every wire subband (Csontos et al., 2009).

Varying subband properties

From the viewpoint of fundamental physics, Chapter 7 is the richest chapter within this dissertation. There, we have been able to show that the leakage current in a Pauli spin-blocked double quantum dot is dominated by the spin-flip cotunnelling mechanism at low magnetic fields and also have observed signatures of leakage current caused by spin-orbit coupling. Not surprisingly, given the g-factor anisotropy we have studied extensively in Chapter 5, we have found a strong anisotropy in how effectively spin-flip cotunnelling is suppressed by a magnetic field. More surprising is the fact that the g-factor anisotropy observed in Chapter 7 is, with respect to the electric field, opposite of what we have seen in Chapter 5. This can be seen as an indirect confirmation of

Figure 8.1: Gate structure before deposition of the dielectric with a 100 nm pitch (left) and 60 nm pitch (right).

the calculations of Csontos et al. (2009), since the g -factor anisotropy as predicted by Maier et al. (2013) is caused by an anisotropic mixing of different subbands by the electric field and therefore heavily depends on the character of the heavy-hole light-hole mixing in the respective subbands. This complete change of the hole properties in different nanowire subbands makes the physics of 1D-hole conductors extremely rich and complicated.

The complicated subband structure can be partly avoided by reaching the single-hole regime in quantum dots, since this is the most elegant way to ensure that the quantum dot states one operates in originate from the lowest-energy nanowire subband. In the devices presented in this thesis, this is unfortunately not possible. The main reason for this has been identified to be the gate pitch. When approaching the few-hole regime, the gate operated as a plunger forms a third tunnel barrier in the nanowire, effectively splitting up the single quantum dot into a double quantum dot. Similar results by Fasth et al. (2005) suggest that a reduction of the gate pitch from 100 nm to 60 nm is a promising approach to overcome this limitation. As can be seen in Figure 8.1, we already have made substantial progress in this direction.

Other 1D-hole systems

As shown by Csontos et al. (2009), the properties of one-dimensional hole subbands are to some extent robust against the exact cross-sectional shape and strength of the confinement. Therefore our findings may also be of relevance for a very similar system: self-assembled Ge nanowires on a silicon substrate as realized by Zhang et al. (2012).

Also other one-dimensional hole conductors have been of experimental interest in recent years, including InSb nanowires (Pribyl et al., 2013) and Si nanowires (Zwanenburg et al., 2009b). The theoretical predictions for the enhanced spin-orbit interaction (Kloeffel et al., 2011) and anisotropic, electric-field dependent g-factor anisotropy (Maier et al., 2013) consider also the strain between the Ge core and Si shell, which will be very different in the case of the other nanowire systems. Therefore, quantitatively one may expect different results for them but qualitatively the predictions should still hold. Although Ge-Si core-shell nanowires exhibit the largest coupling constants between the two lowest-lying subbands, qualitatively the found anisotropies should also hold in other material systems. Please note that this still assumes *elongated* quantum dots, which is usually not the regime where the relatively thick III-V nanowires in the literature are operated.

Coulomb interactions

The Zeeman-splitting is probably the most prominent magnetic-field effect in our research field. Nevertheless, also other magnetic-field effects can play an important role in experiments, certainly at high magnetic fields on the order of or exceeding 1 T. At the end of Chapter 7 we have discussed orbital effects of the magnetic field, where we have seen signatures of an increased Coulomb interaction even for triplet states, but only for the magnetic field aligned with the electric field. So also here we have found a significant anisotropy worth investigating since it hints on an interplay between spin-spin interactions and Coulomb effects mediated by the electric field, possibly again via coupling of different nanowire subbands. If this turns out to be true, also this anisotropic Coulomb interaction might differ between nanowire subbands. Such anisotropic Coulomb interactions have been predicted in this system by Maier et al. (2014b), who have calculated a stabilization of Majorana-bound states in superconducting hybrid devices.

Superconducting hybrid devices

Devices with superconducting contacts are therefore another promising line of research for Ge-Si core-shell nanowires. Unfortunately, our hero metals for ohmic contacts, Palladium with a thin, not covering layer of Titanium underneath, is not superconducting. Even worse, palladium as a quite heavy metal with pronounced spin-orbit coupling will quench proximitised superconductivity from neighbouring superconducting metal layers and is therefore not suitable as an intermediate

layer for improved ohmic contact. Aluminium has been reported to give ohmic contacts (Xiang et al., 2006c), but has the disadvantage of a comparably small superconducting gap, which limits the strength of the magnetic field. First tests with sputtered niobium have revealed a large Schottky barrier between the contacts and the nanowire. Therefore this technological challenge remains to be solved if Ge-Si core-shell nanowires with their possibly very large spin-orbit coupling (Kloeffel et al., 2011) shall be used to e.g. form topological qubits (Maier et al., 2014a).

8.2 ASPECTS REGARDING QUANTUM COMPUTING

The solid-state quantum computing research field has the beauty of being at the tipping point between fundamental science and the engineering of useful devices. In principle, the single aspects of a qubit suitable for quantum computation as demanded by the famous ‘DiVincenzo criteria’ (DiVincenzo, 2000) have been shown to work in experiments. So, what is there left to do?

Integration into qubit circuits

A first obvious answer to this question: integration of (a lot of) single qubits into quantum circuits of *entangled* qubits to form a quantum computer. At first encounter, this sounds more like a task for nano-electronics engineers rather than for scientists. And to some extent this is correct. There are many technological challenges to be overcome, also¹ if one considers the system that stands at the centre of this thesis: Ge-Si core-shell nanowires. One of the biggest is the growth and the deposition of the nanowire on gate structures. Instead of growing the nanowires vertically out of a flat substrate and transferring them manually to a second chip with the gate structures, one could think of ways to grow the nanowires horizontally on a pre-patterned substrate with a mesa-like structure to which the gold seed is attached (see Figure 8.2). The qubits in a two-dimensional grid of nanowires can then be coupled via floating gates (Trifunovic et al., 2012) or circuit-QED (Kloeffel et al., 2013) to allow for universal quantum gate operations. One could even think about integrating this idea with the method of Hauge et al. (2015) for the growth of hexagonal silicon, i.e. using the side walls of auxiliary nanowires as growth sites for the Ge-Si core-shell nanowires.

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¹ and compared to the case of planar CMOS-based qubits in Si (Veldhorst et al., 2015) maybe even more so

Figure 8.2: Schematic depiction of a possibility to integrate nanowire growth and gate structures. Side view of a single nanowire device (top), top view of a 2D-integrated array of nanowire devices (bottom).

The growth of group-IV nanowires with a hexagonal structure is very interesting by itself, since hexagonal Ge is predicted to exhibit a direct bandgap (De and Pryor, 2014). In principle, this opens the possibility for the integration of spin-based qubits, which are very suitable for computation, with so-called ‘flying’ qubits, which are suitable for quantum communication, e.g. implemented as the polarization of a photon. In a quantum dot structure within a direct-bandgap material, this could be realized by the combination of electron and hole quantum dots. Such structures may also be interesting for the formation of long-lived excitons because of the possibility to tune the interaction strength between the electron and hole quantum dot by means of gates.

Qubit manipulation with continuous high-frequency electric fields

A very interesting possibility for quantum computing applications follows from the electric-field dependent g-factor anisotropy. As shown by Maier et al. (2013), the effective g-factor g^* will be maximal, if the mag-

Figure 8.3: Schematic depiction of the integration of side gates into the nanowire device to provide a static electric field E_{SG} : Changing E_{SG} changes the Zeeman splitting between the two spin states at a constant magnetic field B_0 . If the Zeeman splitting is resonant with the continuous high-frequency electric field E_{HF} , the hole spin undergoes Rabi oscillations. Spin rotations can therefore be switched on and off by changing E_{SG} .

netic field points perpendicular to the nanowire. When a static electric field is switched on along the same axis as the magnetic field, g^* is reduced. This behaviour lets us build a recipe for a qubit: we need an elongated quantum dot in a Ge-Si core-shell nanowire (see Figure 8.3), in which we split a spin-orbit doublet state by a static magnetic field B_0 in the substrate plane perpendicular to the nanowire. A continuous high-frequency electric field E_{HF} of frequency f_0 drives coherent oscillations between the two spin-orbit states (so-called Rabi oscillations). In order to switch the oscillations on or off, we use two additional side gates on opposite sides of the nanowire (similar to (Roddaro et al., 2011)), with which we can produce a static electric field E_{SG} across the quantum dot in the magnetic field direction by applying opposite voltages $\pm V_{SG}$ on the two gates. By changing V_{SG} we can now change E_{SG} without changing the dot potential. Changing E_{SG} also changes the Zeeman-splitting of the spin-orbit states without the need to B_0 . Therefore the Zeeman-split states are in resonance with E_{HF} only for a certain value of E_{SG} , so that we can switch on and off the Rabi oscillations by simply changing V_{SG} .

This approach becomes particularly interesting, if we do *not* apply E_{HF} by means of a local gate but by a superconducting cavity resonator,

as also theoretically investigated by Kloeffel et al. (2013). With several qubits within one resonator, it is perfectly possible to drive qubit manipulation by switching the side gate voltage of the desired qubit to the ‘on’ mode during gate operations, while all other qubits remain in their ‘idle’ mode and stay unaffected. This is an enormous technological advantage over using local, pulsed high-frequency magnetic fields for each qubit (Koppens et al., 2006).

Combination with nuclear spins

Due to their humongously long coherence time of up to more than 30 s (Muhonen et al., 2014), single nuclear spins are extremely interesting for the storage of quantum information such as the state of a qubit. Unfortunately, these long decoherence times are linked to their weak coupling to the environment which makes it inherently difficult to manipulate the nuclear spin state. As discussed in the previous paragraphs, spin-orbit qubits in Ge-Si core shell nanowires on the other hand offer fast and efficient means of quantum state manipulation.

Therefore, optimal performance can only be reached by using the best of both worlds: qubit manipulation in the nanowire quantum dots, qubit storage in the nucleus. This can in principle be realized by implanting an acceptor atom into the nanowire (or the side branch of a nanocross, see Plissard et al. (2013)) and forming the gate-defined quantum dot in the direct vicinity of it. In this case one could establish interaction between the nuclear spin and the quantum dot via the acceptor hole state. The disadvantage of this approach is the necessary high-temperature annealing after implantation, which will most probably at least deteriorate the atomically sharp interface between Ge and Si. A more elegant way could be the *in-situ* implementation of the acceptor atom during the nanowire core growth. In principle, one could even think about not implanting the acceptor atom into the nanowire at all, but in a planar structure (Muhonen et al., 2014) and establish coupling via circuit quantum electrodynamics (QED), similar to what we have discussed in the previous section.

8.3 CLOSING REMARKS

In the grand picture of quantum computing nobody can say today, if Ge-Si core-shell nanowires as we have used them throughout this thesis will be the building blocks for a future quantum computer. But in Chapters 4 to 7 we have discussed experimental results that are very rel-

evant to further improve current qubit performance as well as deepen the knowledge of the fundamental processes governing it. In this last Chapter we have given a brief outlook on the vast realm of possibilities for short- and long-term research directions that can further push the border of our knowledge of quantum mechanics as well as get us closer to the ultimate goal: building a universal quantum computer.

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SUMMARY

In a classical computer, information is processed in the form of bits: either a current is flowing in a transistor (logical 1) or no current is flowing (logical 0). An electrical current is a directed, macroscopic flow of charge carriers, usually electrons. Electrons have other properties than just the charge they carry, e.g., they carry a spin. The spin is a quantum mechanical property and can be seen as a magnetic moment that is either parallel or anti-parallel aligned with an external magnetic field. Just like the charge, also the spin of electrons is used in information technology. In a commercial hard disk, reading the stored information is based on spin-dependent scattering of electrons at the interface of a ferromagnet.

The spin of a single electron is a quantum mechanical two-level system, which can be used to perform computation in a so-called quantum computer. Instead of bits, which are either zero or one, such a system represents quantum bits, or qubits, which can be either zero, or one, or any superposition of the two. These additional quantum superposition states make a quantum computer very powerful at certain tasks. But quantum mechanical systems are also very delicate, and a quantum state is very susceptible to any kind of disturbance from its environment and thus easily destroyed. This makes building an actual quantum computer an immensely difficult task and numerous technological and scientific challenges have to be tackled in order to achieve this goal.

In this thesis, we perform experiments that contribute to a deeper understanding of spins and charges under the influence of electric and magnetic fields for future quantum computing applications. We utilize holes (a representation of a 'missing electron' in the valence band) instead of electrons and we confine them in so-called quantum dots, nano-scale regions small enough to observe quantum effects. We form such quantum dots inside Ge-Si core-shell nanowires, which provide tight confinement in two dimensions, so that the holes can only move freely along the nanowire axis. We place the nanowires on top of a gate structure, with which we can influence the local hole density in the nanowire by applying a voltage to them. We cool the devices to extremely low temperatures of approximately 20 mK to minimize ther-

mal noise and smearing, and use two gates to form tunnel barriers in the nanowire with a quantum dot in between them, and influence the number of holes on the quantum dot with a third gate.

We define quantum dots of different lengths and change the number of holes on them one by one over a large range while keeping the tunnel barriers well-defined. As expected we find that the addition energy for a hole scales inversely with the length of the quantum dot and the capacitances of the different gates to the dot is constant. Even for dots of almost 500 nm in length the dots are stable over a very long time even when changing the number of holes on the dot over a large range. For longer dots we controllably use a gate in the middle of the quantum dot to split up the quantum dot into two. The coupling between these two dots is fully tunable from strong coupling to very weak coupling. These results show that we have a very high control over the tunnel couplings and electrochemical potentials of quantum dots inside a nanowire by means of gates, which makes them an ideal platform for the study of quantum effects in one-dimensional hole systems.

By applying a magnetic field, we separate the two spin states of each hole in energy, and we show that this so-called Zeeman splitting depends not only on the strength of the magnetic field but also on its direction. A peculiarity of our system is that there is a second axis that breaks the symmetry for the Zeeman splitting: the direction of the static electric field induced by the gates. This has been predicted theoretically and explained by heavy-hole light-hole mixing in the nanowire subbands. The electric field couples different subbands which gives rise to a very pronounced coupling of the spin and orbital degree of freedom of the holes. This in turn alters the g-factor, the proportionality constant between magnetic field and Zeeman splitting. The prediction therefore implies that the g-factor can be altered by changing the electric field.

Our experiments, in which we map the g-factor in all high-symmetry directions, are in very good agreement with the theoretical predictions and thus confirm the electric-field dependence of the g-factor. This implies that it is possible to manipulate a hole spin qubit with a continuous high-frequency electric field by tuning the Zeeman splitting in and out of resonance through simply changing the electric field. If the high-frequency electric field is not applied by a local gate but e.g. by a superconducting cavity, it is even possible to individually address many qubits by switching them between a ‘manipulation’ and an ‘idle’

mode. This is a great technological advantage for a scaled-up quantum computer with thousands of qubits.

A second important step towards a quantum computer is the read-out of the hole spin at the end of the computation. Since the magnetic moment associated with a single hole spin is extremely small, direct detection of the spin state is extremely difficult. A more feasible approach is spin-to-charge conversion by Pauli spin blockade. Here, the possibility of a hole to tunnel through a double quantum dot and generate a measurable current is dependent on its spin state. In this thesis we tune a double quantum dot in such a way that we observe shell filling of new orbitals in both quantum dots with orbital energies of approximately $500\text{ }\mu\text{eV}$. We identify the two charge degeneracy points where a hole is trapped in one of the dots and observe Pauli spin blockade of a second hole entering the double dot in both situations in agreement with the shell filling picture.

Even though there is in principle no current flow in Pauli spin blockade, we still measure a finite leakage current. Therefore, a mechanism must exist that lifts the spin blockade by flipping the spin of the blocked hole. We conduct magnetic-field dependent measurements and find a suppression of the leakage current at finite fields leading to a characteristic leakage current peak around zero magnetic field. By examining the peak shape we identify spin-flip cotunnelling as the dominant leakage mechanism, where the spin is flipped via a virtual state in the reservoir adjacent to the quantum dot. Again we observe a strong anisotropy in the width of the leakage current peak associated with a strongly anisotropic effective g -factor. The direction of the maximized g -factor is parallel to the electric field lines induced from the gate, in contrast to our earlier findings. This can be explained by the hole states here originating from a different nanowire subband. At higher magnetic fields up to 1 T we find signatures of an anisotropic leakage current induced by spin-orbit coupling of singlet and triplet states as well as hints at anisotropic Coulomb effects that are not well understood yet.

These results demonstrate the suitability of Ge-Si core-shell nanowires as a playground for the exploration of one-dimensional hole states, where spatial confinement and electric fields lead to strongly anisotropic effects that can be exploited for further fundamental studies as well as quantum computing and related spintronics applications.

SAMENVATTING

In een klassieke computer wordt informatie verwerkt in de vorm van bits: er loopt wel (logische één) of geen (logische nul) elektrische stroom door een transistor. Een elektrische stroom is een gerichte, macroscopische stroom van ladingsdragers, meestal elektronen. Naast de lading die ze dragen hebben elektronen nog andere interessante eigenschappen, zo dragen ze bijvoorbeeld ook spin. Spin is een kwantummechanische eigenschap die zich manifesteert als een magnetisch dipoolmoment dat zich richt in dezelfde dan wel tegengestelde richting als een extern magneetveld. De spin van elektronen wordt, net als hun lading, gebruikt voor toepassingen in de informatietechnologie: In commerciële harde schijven wordt de opgeslagen informatie uitgelezen met behulp van de spinafhankelijke verstrooiing van elektronen aan het grensvlak van een ferromagneet.

De spin van één enkel elektron is een kwantummechanisch systeem dat twee waarden kan aannemen en gebruikt kan worden om berekeningen uit te voeren in een zogeheten kwantumcomputer. In plaats van een bit, die de waarde één of de waarde nul kan hebben, representeert een dergelijk systeem een kwantumbit, oftewel qubit, die naast de waarde één of de waarde nul ook iedere superpositie van beide kan hebben. De extra mogelijkheid van deze superpositietoestanden zorgt ervoor dat een kwantumcomputer zeer krachtig is voor bepaalde toepassingen. Kwantummechanische systemen zijn echter zeer delicaat, en een kwantumtoestand is erg gevoelig voor alle mogelijke verstoringen van buitenaf en daardoor gemakkelijk te vernietigen. Hierdoor is het immens lastig om daadwerkelijk een kwantumcomputer te maken en zijn er talloze technologische en wetenschappelijke uitdagingen die overwonnen moeten worden om dit doel te verwezenlijken.

In dit proefschrift voeren we experimenten uit die bijdragen aan een dieper begrip van spins en ladingen onder de invloed van elektrische en magnetische velden met het oog op toekomstige kwantumcomputer toepassingen. We gebruiken gaten (een representatie van een 'ontbrekend elektron' in de valentieband) in plaats van elektronen en sluiten deze op in zogeheten kwantumdots, nanoscopische gebiedjes klein genoeg om kwantumeffecten waar te kunnen nemen. We vormen zulke kwantumdots in Ge-Si kern-mantel nanodraden die voor strakke op-

sluiting in twee dimensies zorgen, zodat de gaten alleen nog vrij kunnen bewegen langs de as van de nanodraad. De nanodraden plaatsen we op een structuur van gates waarmee we lokaal de gatendichtheid kunnen beïnvloeden door een voltage aan te brengen. We koelen onze devices af naar extreem lage temperaturen van ongeveer 20 mK om thermische ruis en verbreding te minimaliseren. Twee gates worden gebruikt om tunnelbarrières aan te leggen in de nanodraad waartussen zo een kwantumdot ontstaat en met een derde gate kunnen we het aantal gaten in deze kwantumdot regelen.

We definiëren kwantumdots met verschillende lengtes en variëren het aantal gaten op deze dots over een groot bereik met stappen van één enkel gat terwijl we de tunnelbarrières goed gedefinieerd houden. Geheel volgens verwachting vinden we dat de energie die nodig is om één volgend gat toe te voegen omgekeerd evenredig is aan de lengte van de kwantumdot en dat de capaciteit van de verschillende gates met de kwantumdot gelijk blijft. Zelfs kwantumdots met een lengte van bijna 500 nm blijven lange tijd stabiel, ook wanneer het aantal gaten over een groot bereik wordt gevarieerd. Grotere kwantumdots kunnen we met behulp van een gate midden onder de dot gecontroleerd in tweeën splitsen. De interactie tussen de twee gevormde kwantumdots is volledig regelbaar van sterke tot zeer zwakke wisselwerking. Deze resultaten tonen aan dat we door middel van gates zeer goede controle hebben over de tunnelkoppelingen en elektrochemische potentialen van kwantumdots in nanodraden. Hierdoor vormen ze een ideaal platform voor het bestuderen van kwantumeffecten in eendimensionale gatensystemen.

Door een magnetisch veld aan te leggen splitsen we de energieën van de twee spintoestanden van ieder gat en laten we zien dat deze zogeheten Zeemansplitsing niet alleen afhangt van de sterkte van het aangelegde magnetisch veld maar ook van de richting hiervan. Een eigenaardigheid van ons systeem is dat er sprake is van een tweede as die de symmetrie van de Zeemansplitsing breekt: namelijk de richting van het statische elektrisch veld dat wordt veroorzaakt door de gates. Dit effect is theoretisch voorspeld en wordt verklaard door vermenging van lichte en zware gaten in de subbanden van de nanodraad. Het elektrisch veld koppelt verschillende subbanden aan elkaar waardoor er een uitgesproken koppeling ontstaat tussen de spin- en orbitaalvrijheidsgraden van de gaten. Dit beïnvloedt op haar beurt de g -factor, de constante verhouding tussen het magnetisch veld en de Zeemansplits-

ing. De voorspelling impliceert dus dat de g-factor kan worden veranderd door het elektrisch veld te variëren.

Onze experimenten, waarin we de g-factor in alle richtingen van hoge symmetrie in kaart brengen, zijn in zeer goede overeenstemming met de theoretische voorspellingen en bevestigen dus dat de g-factor afhankelijk is van het elektrisch veld. Dit impliceert dat het mogelijk is om een qubit die bestaat uit de spin van een gat te manipuleren door met een statisch elektrisch veld de Zeemansplitsing in en uit resonantie te brengen met een aanhoudend hoogfrequent elektrisch veld. Wanneer het hoogfrequente veld niet wordt gegenereerd door een lokale gate maar, bijvoorbeeld, door een supergeleidende holte is het zelfs mogelijk om vele qubits individueel te beïnvloeden door ze selectief te schakelen tussen een ‘actieve’ modus, waarin ze gemanipuleerd kunnen worden, en een ‘passieve’ modus. Dit biedt grote technologische voordelen voor het opschalen naar een kwantumcomputer met duizenden qubits.

Een tweede belangrijke stap voor de ontwikkeling van een kwantumcomputer is het kunnen uitlezen van de spin van een gat aan het einde van een berekening. Omdat het magnetisch dipoolmoment van de spin van één enkel gat extreem klein is, is het buitengewoon moeilijk om de spintoestand direct uit te lezen. Een meer realistische aanpak is het omzetten van het spinsignaal naar een ladingsignaal door middel van Pauli-spinblokkade. Bij deze methode hangt de mogelijkheid van een gat om door een dubbele kwantumdot te tunnelen en zo een meetbare elektrische stroom te veroorzaken af van zijn spintoestand. In dit proefschrift stellen we een systeem met een dubbele-kwantumdot zodanig in dat we in beide dots het vullen van de atoomschillen in nieuwe orbitalen waarnemen met orbitaalenergieën van ongeveer 500 μeV . We zoeken de twee toestanden met gelijke lading op waarbij er een gat in een van beide kwantumdots opgesloten zit en observeren Pauli-spinblokkade van een tweede gat dat de dubbele kwantumdot binnenkomt in beide toestanden en in overeenstemming met het atoomschillen-plaatje.

Alhoewel er in principe geen stroom loopt wanneer er sprake is van Pauli-spinblokkade, meten we toch een eindige lekstroom. Er moet dus een mechanisme zijn dat de spinblokkade opheft door de spin van het geblokkeerde gat om te draaien. We voeren metingen uit waarbij we het magnetisch veld variëren en nemen een onderdrukking van de lekstroom bij eindig veld waar met een karakteristieke piek in de lekstroom rond een veldsterkte van nul. Door de vorm van de

piek te analyseren kunnen we spin-flip cotunneling, waarbij de spin wordt omgedraaid via een virtuele toestand in het reservoir aangrenzend aan de kwantumdot, aanwijzen als het voornaamste lekmechanisme. Wederom zien we een sterke anisotropie in de breedte van de lekstroompiek die wordt geassocieerd met een sterk anisotrope effectieve g -factor. De richting met de grootste g -factor is, in tegenstelling tot onze eerdere bevindingen, evenwijdig aan de door de gates veroorzaakte elektrische veldlijnen. Een verklaring hiervoor is dat de gatentoestanden die hier gebruikt worden ontstaan zijn uit een andere subbanden van de nanodraad. Bij sterkere magnetische velden tot 1 T zien we zowel tekenen van een anisotrope lekstroom door spinbaankoppeling van singlet en triplet toestanden als aanwijzingen van anisotrope Coulombeffecten, die nog niet goed begrepen worden.

Deze resultaten tonen aan dat Ge-Si kern-mantel nanodraden een geschikte proeftuin vormen voor het bestuderen van eendimensionale gatentoestanden, waar ruimtelijke opsluiting en elektrische velden leiden tot sterk anisotrope effecten die zowel ingezet kunnen worden bij verder fundamenteel onderzoek als voor kwantumberekeningen en daaraan gerelateerde spintronica-toepassingen.

ZUSAMMENFASSUNG

In einem klassischen Computer werden Informationen in Form von Bits verarbeitet: Entweder ein Strom fließt in einem Transistor (logische Eins) oder es fließt kein Strom (logische Null). Ein elektrischer Strom ist ein gerichteter, makroskopischer Fluss von Ladungsträgern, normalerweise Elektronen. Elektronen haben abgesehen von ihrer Ladung noch andere Eigenschaften, z. B. tragen sie einen Spin. Dieser Spin ist eine quantenmechanische Eigenschaft und kann als magnetisches Moment betrachtet werden, das sich entweder parallel oder antiparallel zu einem äußeren Magnetfeld ausrichtet. Genauso wie die Ladung findet der Spin von Elektronen in der Informationstechnologie Verwendung. In kommerziellen Festplatten werden die gespeicherten Informationen mit Hilfe von spinabhängiger Streuung von Elektronen an der Grenzfläche eines Ferromagneten ausgelesen.

Der Spin eines einzelnen Elektrons stellt ein quantenmechanisches Zweiniveau-System dar, das benutzt werden kann um Berechnungen in einem sogenannten Quantencomputer auszuführen. Anstelle eines Bits, die entweder Null oder Eins sind, repräsentiert ein solches System ein Quantenbit (Qubit), das entweder Null, oder Eins, oder eine Überlagerung dieser beiden Zustände sein kann. Diese zusätzlichen quantenmechanischen Überlagerungszustände machen einen Quantencomputer sehr leistungsfähig bei bestimmten Aufgaben. Allerdings sind quantenmechanische Systeme auch sehr empfindlich, und ein Quantenzustand ist sehr anfällig für jede Art von Störung und wird deshalb leicht zerstört. Dadurch ist der Bau eines Quantencomputers eine immens schwierige Aufgabe und eine Vielzahl von technologischen und wissenschaftlichen Herausforderungen muss überwunden werden um dieses Ziel zu erreichen.

In dieser Arbeit führen wir Experimente durch, die einen Beitrag zum tieferen Verständnis von Spins und Ladungen unter dem Einfluss von elektrischen und magnetischen Feldern für zukünftige Quantencomputer-Anwendungen leisten. Wir benutzen Löcher (eine Darstellung für ein 'fehlendes Elektron' im Valenzband, auch 'Defektelektron' genannt) anstelle von Elektronen und sperren sie in sogenannten Quantenpunkten ein, nanoskalige Bereiche, die klein genug sind um Quanteneffekte beobachten zu können. Wir formen solche Quantenpunkte in

Ge-Si Kern-Mantel-Nanodrähten, die einen starken Einschluss in zwei Dimensionen bieten, sodass die Löcher sich nur entlang der Nanodraht-Achse bewegen können. Wir platzieren die Nanodrähte auf einer sogenannten Gatestruktur, mit der wir lokal die Löcherdichte im Nanodraht beeinflussen können, indem wir eine Spannung an sie anlegen. Wir kühlen die Proben auf extrem niedrige Temperaturen von etwa 20 mK herab um thermisches Rauschen und Verbreiterung zu minimieren, und benutzen zwei Gates um Tunnelbarrieren im Nanodraht zu formen mit einem Quantenpunkt zwischen ihnen. Wir beeinflussen die Anzahl an Löchern im Quantenpunkt mit einem dritten Gate.

Wir formen Quantenpunkte verschiedener Längen und verändern die Zahl der Löcher in ihnen über einen weiten Bereich, die Tunnelbarrieren bleiben dabei wohldefiniert. Wie erwartet verändert sich die für die Hinzugabe eines zusätzlichen Lochs nötige Energie indirekt proportional zur Länge des Quantenpunkts und die elektrische Kapazität der verschiedenen Gates bleibt konstant. Selbst Quantenpunkte mit einer Länge von fast 500 nm sind über einen sehr langen Zeitraum und über eine große Bandbreite der Löcheranzahl stabil. Wir benutzen ein Gate auf kontrollierte Weise um einen langen Quantenpunkt in zwei kürzere zu spalten. Die Kopplungsstärke zwischen diesen beiden Quantenpunkten ist vollständig veränderbar von starker bis hin zu sehr schwacher Kopplung. Diese Ergebnisse zeigen, dass wir ein sehr hohes Maß an Kontrolle über die Tunnelkopplungsstärken und elektrochemischen Potentiale von Quantenpunkten in einem Nanodraht durch Nutzung von Gates haben, was sie zu einer idealen Plattform für die Untersuchung von Quanteneffekten in eindimensionalen Löchersystemen macht.

Durch das Anlegen eines magnetischen Feldes separieren wir die zwei Spinzustände jedes Lochs energetisch und zeigen, dass diese sogenannte Zeeman-Aufspaltung nicht nur von der Stärke des Magnetfeldes abhängt, sondern auch von seiner Richtung. Eine Besonderheit unseres Systems stellt die zweite Achse dar, die, neben der Nanodraht-Achse, die Symmetry unseres Systems bricht: Die Richtung des statischen elektrischen Feldes, das von den Gates induziert wird. Dies wurde theoretisch vorhergesagt und erklärt mit einem Mischen der leichten und schweren Lochzuständen in den Subbändern des Nanodrahtes. Das elektrische Feld koppelt verschiedene Subbänder, was zu einer sehr ausgeprägten Kopplung der Spin- und Bahn-Freiheitsgrade der Löcher führt. Dies wiederum verändert den g-Faktor, die Proportionalitätskonstante zwischen dem magnetischen Feld und der Zeeman-

Aufspaltung. Die Vorhersage impliziert also die Möglichkeit, den g-Faktor durch Verändern des elektrischen Feldes zu beeinflussen.

Unsere Experimente, in denen wir den g-Faktor entlang aller hochsymmetrischen Richtungen messen, zeigen eine sehr gute Übereinstimmung mit den theoretischen Vorhersagen und bestätigen so die Abhängigkeit des g-Faktors vom elektrischen Feld. Dies bedeutet, dass es möglich ist, ein Lochspin-Qubit mit einem kontinuierlichen, hochfrequenten elektrischen Feld zu manipulieren, indem man die Zeeman-Aufspaltung so verschiebt, dass sie resonant mit dem elektrischen Feld ist oder nicht. Wird das hochfrequente, elektrische Feld nicht durch lokale Gates erzeugt, sondern z. B. mit Hilfe eines supraleitenden Resonators, dann ist es sogar möglich, viele Qubits individuell zu adressieren, indem man sie zwischen einem 'aktiven' und einem 'inaktiven' Zustand hin und her schaltet. Dies stellt einen großen technologischen Vorteil für einen großskaligen Quantencomputer mit Tausenden Qubits dar.

Ein zweiter wichtiger Schritt hin zu einem Quantencomputer ist das Auslesen des Lochspins am Ende der Berechnung. Da das magnetische Moment eines einzelnen Lochspins extrem klein ist, ist die direkte Messung des Spinzustandes immens schwierig. Eine praktikablere Möglichkeit stellt die Spin-zu-Ladungs-Umwandlung durch Pauli-Spinblockade dar. Hierbei hängt die Möglichkeit, dass ein Loch durch einen Doppelquantenpunkt tunnelt und einen messbaren Strom verursacht, von seinem Spinzustand ab. In dieser Arbeit stellen wir einen Doppelquantenpunkt so ein, dass wir das Füllen neuer Orbitale in beiden Quantenpunkten mit Orbitalenergien von etwa $500 \mu\text{eV}$ beobachten. Wir identifizieren die zwei Ladungsentartungspunkte, bei denen ein Loch in einem der Quantenpunkte gefangen ist und beobachten Pauli-Spinblockade des zweiten Lochs, das in den Doppelquantenpunkt eintritt, in beiden Fällen in Übereinstimmung mit dem Orbitalbild.

Obwohl in Pauli-Spinblockade prinzipiell kein Strom fließt, messen wir doch einen endlichen Leckstrom. Es muss also einen Mechanismus geben, der die Spinblockade aufhebt durch Umdrehen des Spins des blockierten Lochs. Wir nehmen magnetfeldabhängige Messungen vor und beobachten eine Unterdrückung des Leckstroms bei endlichem Magnetfeld, was zu einer charakteristischen Spitze des Leckstroms bei Null Magnetfeld führt. Durch Untersuchung der Form der Spitze identifizieren wir Spinflip-Kotunneln als den dominierenden Leckmechanismus, bei dem der Spin gedreht wird über einen virtuellen Zustand

im Reservoir neben dem Quantenpunkt. Auch hier beobachten wir eine starke Anisotropie der Breite der Leckstromspitze, was auf einen anisotropen g -Faktor zurückzuführen ist. Die Richtung des maximalen g -Faktors ist hier parallel zum elektrischen Feld, im Gegensatz zu unseren vorherigen Beobachtungen. Dies lässt sich dadurch erklären, dass die Lochzustände hier einem höheren Nanodraht-Subband entstammen. Bei höheren Magnetfeldern bis zu 1 T finden wir Hinweise auf einen anisotropen Leckstrom, der durch Spin-Bahnkopplung von Singlet- und Tripletzuständen verursacht wird, sowie auf anisotrope Coulomb-Wechselwirkungen, die noch nicht in Gänze verstanden sind.

Diese Ergebnisse demonstrieren die Eignung von Ge-Si Kern-Mantel-Nanodrähten als Spielwiese für die Erforschung von eindimensionalen Lochzuständen, bei denen räumlicher Einschluss und elektrische Felder zu stark anisotropen Effekten führen, die für weitere Grundlagenforschung genauso wie für Quantencomputer-gestützte und verwandte spintronische Anwendungen genutzt werden können.

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in preparation

Newton: »Verrückt, aber weise.«

Einstein: »Gefangen, aber frei.«

Möbius: »Physiker, aber unschuldig.«

— Friedrich Dürrenmatt, *Die Physiker*